Effects of polymers on short- and long-term dewatering of oil sands tailings

by

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Abstract

Each year, while extracting bitumen from the oil sands ores, the oil sands industry generates a large volume of tailings. Although the polymers have been found to improve the release of water from the tailings and accelerate the dewatering and settling processes in the short-term, longer-term dewatering processes such as consolidation are crucial for making the tailings deposits safe and strong enough for reclamation. Unamended tailings have been found to demonstrate time-dependent creep and thixotropic behaviour. However, whether such behaviour influences the long-term dewatering and consolidation performance of polymer amended/flocculated tailings deposits is unknown. This research investigated the creep, thixotropy, and structuration in polymer amended tailings from an experimental perspective using different polymer dosages, mixing methods, and boundary conditions. Flocculated tailings developed a thixotropic strength and also a higher preconsolidation pressure and reduced compressibility, which was clearly a structuration effect that impeded the self-weight consolidation in longer-term by limiting the creep compression. Similar experiments performed on the Leda clay showed that the structuration effect observed in flocculated tailings is not atypical of the structuration effect observed in natural clays. The potential of the UV-vis spectrophotometry and torque rheometry techniques for determining the optimum polymer dosage was investigated. The results indicated that the techniques could determine the optimum polymer dosage and help to minimize the operational costs and improve the dewaterability in the short-term. Longer-term column dewatering tests showed that the choice of polymer dosage could influence the dewatering and settlement behaviour of tailings in the long-term. Different polymer dosages resulted in different degrees of structuration with the higher dosages showing a relatively prominent

structuration behaviour but also a suboptimal dewaterability. Hence, if the structuration effect is considered, the optimum polymer dosage for the long-term dewatering mechanisms may not be the same as the polymer dosage that produces optimum dewatering performance in the short-term. Based on the experimental findings, an empirical method was proposed for predicting the degree of structuration in a particular tailings-polymer mixture; such a method would allow the operators to minimize the negative effects of structuration through the selection of an appropriate polymer dosage or deposition management plan.

Dedication

To my mother Begum Rowshan Ara Salam

It is for you. You will be remembered, always.

To my father Md Abdus Salam

You have always been and will always be the greatest influence in my life.

To my wife Shamima Nasrin

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List of Abbreviations

AER Alberta Energy Regulator

AFD Atmospheric fines drying

BSED Backscattered electron detectors

CAPP Canadian Association of Petroleum Producers

CCD Charge-coupled device

CEC Cation exchange capacity

CHWE Clark hot water extraction

COF Cyclone overflow

COSIA Canada's Oil Sands Innovation Alliance

CPSD Cumulative pore size distribution

CRD Constant rate of deformation

CRS Constant rate of strain

CST Capillary suction time

CT Composite/consolidated tailings

CWR Clay-to-water ratio

d Day

DDA Dedicated disposal area

DLVO Derjaguin, Landau, Vervy, and Overbeek

dm³ Cubic decimetre

DS Dry solids

EDL Electrical double layer

EICL Extended intrinsic compression line

ESEM Environment scanning electron microscopy

EVP Elastic visco-Plastic

FFT Fluid fine tailings

fFFT Flocculated fluid fine tailings

FTFC Fine Tailings Fundamentals Consortium

g Gram

GC Gas chromatography

GSD Grain size distribution

GSED Gaseous secondary electron detector

GWC Gravimetric water content

HPLC High-performance liquid chromatography

hr Hour

ICL Intrinsic compression line

ILTT In-line thickened tailings

kg Kilogram

km² Square kilometre

kPa Kilopascal

KPI Key performance indicator

LI Liquidity index

LL Liquid limit

m Metre

m³ Cubic metre

MDL Method detection limit

meq Milliequivalent

MFT Mature fine tailings

MIP Mercury intrusion porosimetry

mL Milliliter

MPa Mega Pascal

MS Mass spectrometry

nm Nanometre

NMR Nuclear magnetic resonance

NWR Net water release

PAM Polyacrylamide

PI Plasticity index

PL Plastic limit

PP Polypropylene

ppm Parts per millions

PSD Pore size distribution

PT Paste tailings

rpm Revolution per minute

s Second

SCL Sedimentation compression line

SCO Synthetic crude oil

SEC Size exclusion chromatography

SED Secondary electron detector

SEM Scanning electron microscopy

SFR Sands-to-fine ratio

SSC Soft soil creep

TFT Thin fine tailings

TMF Tailings Management Framework for Mineable Athabasca Oil Sands

TRO Tailings reduction operations

TSF Tailings storage facilities

TT Thickened Tailings

TTQ Totalized torque

UV-vis Ultraviolet-visible

vs. versus

WT Whole tailings

XRD X-ray diffraction

YSR Yield stress ratio

3ITT Three interval thixotropy test

List of Acronyms

C_c Compression index

C_s Solids content

C_v Coefficient of consolidation

$^{\circ}C$ Degree centigrade

d Diameter of the particle/entrance or pore throat diameter

e Void ratio

g Gravitational acceleration

G_{max} Elastic shear stiffness (maximum elastic modulus)

G_s Specific gravity of soil solids

G' Elastic modulus (Storage modulus)

G'' Viscous modulus (Loss modulus)

k Hydraulic conductivity (permeability)/cone factor (fall cone)

m_v Coefficient of compressibility

n porosity

P Pressure applied to the mercury in porosimeter

P_G Pressure of gas

P_L Absolute pressure causing intrusion

R Correlation factor in regression analysis

S Degree of saturation

s_u Undrained shear strength

u Particle settling velocity

u_a Pore air pressure

u_e Excess pore water pressure

u_w Pore water pressure

V_s Volume of solids

V_v Volume of voids

w Gravimetric water content

γ_c Shear Strain

γ_w Unit weight of water

γ_s Unit weight of solids

θ Contact angle between mercury and the pore wall

λ Wavelength

μ Viscosity of the fluid

μm Micrometre

$\mu S/cm$ microsiemens per centimetre

ρ_f Density of fluids

ρ_s Density of FFT solids

σ Normal stress

σ'_p Preconsolidation pressure

τ_c Shear stress

τ_f Shear strength

τ_y Yield stress

γ Surface tension of the mercury

Ψ Total suction

ω Frequency

% Percentage

@ At the rate of

Chapter 1: Introduction

1.1 Background

Following the mining of oil sands ores and the thermal-chemical-hydraulic separation process of bitumen extraction from these ores, the by-product material called tailings, which is a mixture of water, sands, fine clays, silts, and residual hydrocarbons is hydraulically transported from the extraction plant to the dedicated disposal areas or dammed impoundment structures with substantial footprints known as tailings ponds (Oil Sands Discovery Centre, 2009; Canadian Association of Petroleum Producers, 2019).

Figure 1.1. (a) The image of the Athabasca oil sands area published by NASA in 1984; and, (b) the image of the same Athabasca oil sands area published in 2016 (from Nasa Earth Observatory, 2019).

Since 1967, because of the rapid development of the oil sands projects, there has been a continuous accumulation of tailings in the tailings ponds. According to the latest statistics published by the Alberta Energy Regulator (AER), there are more than 30 active

tailings ponds holding more than 1,200 million cubic metres of FFT (AER, 2019). Although these ponds were meant to be temporary in the beginning, they have grown so large over the years that currently they cover an area of 220 km² (Alberta Environment and Sustainable Resource Development, 2014) (Figure 1.1). Figure 1.1 shows significant expansion of the oil sands mining operations over the years as well as the formation of new tailings ponds with only one of them reclaimed so far. Almost 900 km² of land have been affected by the oil sands mining activities and tailings ponds, and it is still growing (Natural Resources Canada, 2016). Hence, the tailings ponds are causing a considerable land disturbance as well as posing serious environmental and economic concerns (McWhinney, 2014). How to reduce the volume and increase the density and strength of these tailings deposits is probably the most important environmental as well as an engineering challenge faced by the industry. However, due to the slow rates of dewatering and consolidation of the tailings material, it is also a very difficult undertaking (Aubertin and McKenna, 2016).

The recent Alberta Energy Regulator (AER) regulation (Directive 085) requires the oil sands operators to develop a plan to manage both existing fluid tailings and future tailings production (AER, 2019). Under the Directive 085, the operators must set project-specific targets to ensure that the fluid fine tailings are in a ready-to-reclaim state within ten years of the end of the mine's life (AER, 2019). The tailings ponds typically have a maximum height of 50 ~ 100 metres and a footprint of 1,500 hectares and above. For example, at the design stage, the Jackpine surface tailings pond had an initial height of 50 metres, a footprint of 1,600 hectares, and a holding capacity of 278 million cubic metres of fluid fine tailings (Devenny, 2010). Assuming that such a 50-metre tailings deposit has

reached its capacity (at the end of the active phase of its design life), if we estimate the time required for its final settlement (based on the typical hydraulic conductivity-void ratio relationship for tailings at an average void ratio of 3 and the compressibility relationship derived from the laboratory-scale tests), the results indicate that the deposit would finally settle after approximately 160 years. For the same tailings deposit, when the same permeability relationship is used with another compressibility relationship (obtained from another tailings sample with a different ageing period, and therefore, different compressibility characteristics), it would take approximately 170 years to attain the final settlement. Even when a tailings deposit of smaller height (i.e. 20-meter) is considered and the same sets of permeability and compressibility relationships are used, the deposit would require 60 years and 70 years respectively to settle completely. Hence, depending on the height of the tailings deposit as well as permeability and compressibility, the settlement at any specific time or the time required for final settlement may vary considerably and may also differ from the projected settlement or the time frame for settlement. Nevertheless, in general, the rate of consolidation in the tailings ponds is so slow that it is only appropriate to measure it on a timescale of decades, rather than the years (Botha and Soares, 2015).

A pressing need for overcoming the challenges associated with the slow dewatering and consolidation of tailings deposits and their reclamation as well as the enactment of new tailings regulation has accelerated the development of tailings treatment technologies (Canadian Association of Petroleum Producers, 2019). Some of the improved tailings treatment technologies such as in-line flocculation, tank thickening, and centrifugation technique use polymers to promote flocculation for faster dewatering. Polymers are large

organic molecules made of repeating patterns of small molecules (monomers) bonded together. Field trials have shown that the application of polymers allows such technologies to increase solids content to at least 50% in the short-term, usually within days (Matthews et al., 2011; Wells, 2011). However, to meet the regulatory standards and develop a deposit safe and strong enough for reclamation, it is thought that a solids content of at least 70% (which is equivalent to a gravimetric water content of 43%) is required, which is close to the plastic limit of the tailings material (McKenna et al., 2016). To take the tailings deposits to such a state, it is necessary to optimize the tailings deposition and management strategies, which again warrants a comprehensive understanding of the longer-term dewatering processes such as consolidation and creep. The recent changes in tailings regulation (Directive 085) also encourage consideration of dewatering over longer timescales, up to decades (AER, 2019).

The consolidation of oil sands tailings involves the densification of fine-grained tailings material by the release of excess pore water pressure and a slow settlement over time in response to the self-weight of tailings or a vertical surcharge from a capping layer (BGC Engineering Inc., 2010). During consolidation, although very slowly, the tailings increase in effective stress and decrease in volume as the excess pore water pressure dissipates. Creep refers to that time-dependent deformation that occurs at constant effective stress. When a consolidation test is conducted on a saturated clay sample and the measured compression is plotted as a function of the logarithm of time, the sample is often observed to continue settling even after the complete dissipation of the excess pore water pressure (at constant effective stress). Such secondary consolidation is an example of creep.

In clays, the permeability of the material, which controls the flow of pore water also controls the rate at which the excess pore water can dissipate. In fact, the hydrodynamic lag or the time required for the escape of the water from the pores in clays (i.e. a complete dissipation of excess pore water pressure) governs the rate of primary consolidation (Soga, 2005). Hence, in high void ratio materials such as tailings, hydraulic conductivity-void ratio relationship is the property that dictates the rate and magnitude of settlement during primary consolidation (Suthaker and Scott, 1994; Gholami, 2014). While the excess pore water pressure dissipates, the tailings increase in effective stress and subsequently go through a deformation, and therefore, the void ratio changes. During creep compression or secondary consolidation, the hydraulic conductivity of the material still governs the rate of flow of water out of a deposit, however, the rate of change of effective stress is also affected by the viscous deformation of the soil structure (Soga, 2005; Sorensen, 2006). Such viscous deformation typically manifests as a time-dependent compressibility curve, where the apparent preconsolidation pressure shifts with the strain rate and such a shift in the apparent compressibility curve due to creep are well-defined in natural clays. Several models in the literature have quantitatively reproduced this effect (Yin and Graham, 1994; Vermeer and Neher, 1999; Hinchberger and Rowe, 2005).

Dr. Scott and his research group at the University of Alberta worked extensively on the creep behaviour in unamended oil sands tailings and published their findings. The lab-scale experiments, which were conducted over a larger time scale (e.g. 10- metre standpipe tests) showed that a creep compression occurred in tailings deposits over time at constant effective stress. In the 10-metre standpipe filled with oil sands tailings, over a period of 25

years, there was very little effective stress development with no dissipation of excess pore water pressure. However, there appeared to be a decrease in void ratio with a settlement occurring at a uniform rate over time (Jeeravipoolvarn, 2005; Jeeravipoolvarn et al., 2009).

Many natural clays show a time-dependent behaviour called ‘structuration’, also called ageing, which is typically manifested in a change in clay microstructure and an associated increase in apparent preconsolidation pressure or yield strength; subsequently, the clays show a decrease in compressibility and increased resistance to the compression pressure (Locat and Lefebvre, 1985; Leroueil and Vaughan, 1990; Burland, 1990; Liu and Carter, 1999; Delage, 2010; Sorensen, 2006). Inherent ageing effects such as thixotropy, interparticle bonding, and cementation (which involve no external influence) as well as viscous effects such as creep have been thought to be the processes responsible for such microstructural changes and the subsequent increase in apparent preconsolidation pressure (Locat and Lefebvre, 1986; Burland, 1990; Sorensen, 2006).

Because of the structuration effect, most natural clays in their undisturbed or intact state exhibit higher preconsolidation pressure and lower compressibility in comparison to the reconstituted or remoulded samples of the same clays. However, since the structuration effect is reversible, the reconstituted or remoulded samples regain a component of this preconsolidation pressure over time (Skempton and Northey, 1952; Leonards and Ramiah, 1959; Leonards and Altschaeffl, 1964). The thixotropic behaviour of clays is considered to be the primary source of such a structuration effect in clays. Although the term ‘structuration’ has rarely been used in oil sands tailings research, the thixotropy, which is also a time-dependent process has been reported in the unamended FFT samples

(Jeeravipoolvam, 2005; Jeeravipoolvarn et al., 2009; Miller, 2010). Structuration effect leads to a gradual recovery of stiffness in a reconstituted or remoulded clay sample (Leonards and Ramiah, 1959; Leonards and Altschaeffl, 1964), whereas thixotropy results in recovery of strength with time (Skempton and Northey, 1952). Skempton and Northey (1952) attributed the thixotropic effect in the soil to the gradual rearrangement of the particles under the influence of bonding forces (i.e. van der Waals forces) and suggested that thixotropy increases the mechanical stability. The effects of thixotropy are clearly manifested in the time-dependent strength recovery of clays after remoulding at a constant density (Figure 1.2).

Figure 1.2. Thixotropic strength regain in remoulded clays (adapted from Skempton and Northey, 1952, and Mitchell and Soga, 2005).

Typically, two basic constitutive relationships are used in large strain consolidation modelling: (a) a void ratio-effective stress relationship; and, (b) a void ratio-hydraulic conductivity relationship. For the fluid fine tailings, these relationships are measured by conducting the large strain consolidation tests in the laboratory. Then, the long-term consolidation behaviour of tailings is predicted by using the experimental data with the numerical models (Jeeravipoolvarn, 2005). An accurate prediction of consolidation behaviour is crucial to the sustainable management of tailings during operation, closure, and reclamation; however, it also requires an in-depth understanding of the large strain consolidation behaviour of the tailings deposits (Ito and Azam, 2013).

Both creep and thixotropic behaviour have been observed to influence the final consolidation or settlement behaviour of unamended tailings deposits (Jeeravipoolvarn et al., 2009). Although creep is commonly modelled through the concept of rate-dependent compressibility, the conventional large strain consolidation models do not incorporate the effects of thixotropy or structuration. To incorporate the effects of creep, thixotropy, and structuration into the large strain consolidation theory, reformulate the governing equations, and modify the numerical models, it requires an extensive investigation and assessment of the time-dependent phenomena in fluid fine tailings.

This study aims to investigate the nature of creep, thixotropy, and structuration in oil sands tailings, specifically in polymer amended tailings, from an experimental perspective. The objective is to examine whether the polymer amended tailings show a time-dependent structuration behaviour similar to the natural clays and whether such behaviour has any implications for the long-term dewatering and consolidation

performance of the tailings deposits. The experiments included a combination of long-term column dewatering tests with pore water pressure measurements, rheological tests, conventional and modified oedometer tests, fall cone tests, low and high vacuum scanning electron microscopy, optical microscopy, and mercury intrusion porosimetry. Both consolidation and non-consolidation volume change behaviour of tailings were characterized under saturated conditions. The changes in tailings fabric over time and their effects on the compressibility and strength characteristics of tailings were investigated. The implications of such time-dependent effects for the final settlement of tailings deposits were examined. Also, the influences of different polymer dosages and polymer mixing methods on the dewatering, consolidation, and strength development behaviour of tailings under different boundary conditions were studied.

1.2 The objective and scope of the research work

The goal of this experimental study was to examine the effects of polymers on the dewatering and consolidation performance of oil sands tailings both in the short-term and long-term and also to investigate the creep, thixotropy, and structuration in polymer amended tailings and evaluate their implications for the dewatering performance of tailings deposits in longer-term. It was accomplished through a combination of laboratory-scale experiments using different polymer dosages, polymer mixing methods, boundary conditions, and scales of tests. The following scopes of work were pursued:

- Developing analytical methods for determining the optimum polymer dosage for the dewatering of oil sands tailings; the methods include absorbance-based UV-vis spectrophotometry and torque rheometry. The goal is to improve the short-term

dewatering behaviour of oil sands tailings by using methods/tools that can determine the optimum polymer dosage and can also be used in developing process control systems for correcting or adjusting the polymer dosage in real or near real-time.

- Measuring the dewatering and consolidation properties of flocculated tailings in the longer-term by using columns with pore water pressure sensors installed at different heights (for measuring the pore water pressures, both positive and negative). The changes in water-tailings interface height and the changes in gravimetric water content at different sample heights were measured using a set of replicate columns. Generated data is to help the investigation of consolidation, creep, thixotropy, and structuration in flocculated tailings, and the calibration and validation of the numerical modelling of creep, thixotropy, and structuration in the large strain consolidation model.
- Measuring the change in compression behaviour of flocculated tailings by performing one-dimensional consolidation tests (oedometer tests) on the tailings samples of different ages. Consolidation tests include both undisturbed and remoulded samples to determine how the tailings compressibility curve may change over time due to the structuration effect (based on the knowledge of the difference between the compressibility of the remoulded samples and their natural intact compressibility).
- Measuring the creep in flocculated tailings by performing modified one-dimensional consolidation tests on the tailings samples. Modifications include installing a pore water pressure sensor at the base of the consolidation cell for measuring the changes in pore water pressure in a sample that is subjected to a constant vertical loading (so that the resulting creep compression or the decrease in void ratio with time at constant

- effective stress can be differentiated from the primary consolidation; a complete dissipation of excess pore water pressure indicates the end of primary consolidation).
- Tracking the development of undrained shear strength in flocculated tailings over time (using a fall cone) to establish a relationship between the thixotropic strength gain in the samples and the change in apparent preconsolidation pressure (the recovery of stiffness due to structuration) measured from the one-dimensional consolidation tests.
 - Examining the mechanical behaviour of flocculated tailings in response to the small strain shear by conducting oscillatory rheometry tests (using an Anton Paar Physica Rheometer with vane fixture). The oscillatory rheometry tests include amplitude sweep tests and three interval thixotropy tests to determine the time-dependent thixotropic strength buildup in tailings and the recovery of thixotropic strength after remoulding.
 - Imaging the changes in size, shape, and arrangement of the grains/flocs and pores in tailings immediately after the mixing of polymer and during the flocculation, and also at specific time intervals using optical microscopy and scanning electron microscopy; the objective is to study the relationship between the microstructural changes and the changes in compressibility and strength characteristics qualitatively. Generated micrographs are also to be processed and analyzed using the digital image processing software for evaluating the changes in floc/grain size and pore size quantitatively.
 - Determining the changes in pore size distribution in flocculated tailings over time by performing the mercury intrusion porosimetry analysis on the freeze-dried samples; the

aim is to establish a relationship between the changes in pore size distribution and the changes in dewaterability, compressibility and strength characteristics of tailings.

- Determining the effects of changes in the compressibility behaviour of flocculated tailings on the final settlement behaviour or final density of a tailings deposit by using a simple spreadsheet-based numerical analysis tool and also by using an analytical function; the goal is to determine the implications of structuration effect for the dewatering and consolidation performance of the tailings deposits in longer-term.
- Evaluating the influences of different polymer dosages and mixing methods on the time-dependent creep, thixotropic, and structuration behaviour in flocculated tailings; the objective is to determine the link between polymer dosage and time-dependent behaviour and select the best performing polymer dosage for the long-term dewatering.
- Proposing a method for predicting the degree of structuration that will result from a particular tailings-polymer mixture; the goal is to formulate an empirical method for determining the thixotropic strength development in flocculated tailings over time based on the measurements of short-term key performance indicators (i.e. settlement, net water release, water released, etc.) and undrained shear strength and the relationship between them, thereby reducing the uncertainty in tailings settlement prediction.

1.3 The novelty of the research

Although most of the current research on polymer optimization is directed towards maximizing short-term dewatering and focus on developing new polymers, relatively little research has been conducted on the impacts of process variables such as polymer dosages

and mixing conditions (i.e. mixing duration and intensity) on the flocculation and dewatering performance of the polymers. Especially, there has been no research work focusing on developing optimization technologies that can quantify the dewatering efficacy immediately after the mixing of polymer with the tailings and/or provide feedback information to adjust or correct the polymer dosage or the mixing conditions in real-time on a continuous basis. This research presented UV-vis spectrophotometry, an absorbance-based analytical method for determining the residual polymer concentration in tailings bleed water based on the absorbance measurements. The lowest measured absorbance corresponded to the minimum polymer concentration, which in turn corresponded to the optimum polymer dosage. The test results suggested that the dewatering operation of oil sands tailings could be optimized by using an in-line UV-vis spectrophotometer that is capable of generating real-time information.

The research also proposed torque-rheology based methods that use a torque rheometer for determining the peak torque and totalized torque in tailings during the mixing of the polymer. When the peak torques and totalized torques were measured for different polymer dosages and compared, they were observed to increase with the increasing dosage, which was then followed by a local decrease at the optimum polymer dosage. Both UV-vis spectrophotometry and torque rheometry demonstrated the potential for emerging as an effective tool for on-site optimization and automation of dewatering operations of tailings and for reducing the polymer consumption, thereby causing major savings.

The structuration behaviour in natural clays has been reported in the literature and found to manifest in a time-dependent change in clay microstructure and an associated

increase in apparent preconsolidation pressure. The effect of structuration is typically reflected in a change in the clay compressibility curves over time. However, yet, there exists no consolidation model to describe the time-dependent structuration effect in tailings or predict the full range of consolidation behaviour of tailings deposits. Unless such time-dependent changes are taken into account, it is highly probable that the long-term consolidation behaviour predicted by a model could fall short on accuracy, especially in those cases where the model is trying to simulate the time-dependent behaviour of soft soils or deposits (such as oil sands tailings) at low effective stresses (Jeeravipoolvarn et al., 2009). Nevertheless, so far, no attempt has been taken to study the effects of time-dependent phenomena in tailings deposits, especially the structuration effect. Also, little quantitative information is available in the literature on the creep, thixotropy, and structuration behaviour of polymer amended tailings.

In this study, it was demonstrated that the time-dependent phenomena such as creep, thixotropy, and structuration, which were observed in the flocculated tailings are not atypical of clay behaviour. It was evident in the experimental data obtained from the column dewatering tests. It also became apparent that creep, thixotropy, and structuration effect play a significant role in the long-term dewatering and consolidation performance of polymer amended tailings, at least in some types of oil sands tailings (Salam et al., 2017, 2018a, 2018b). Based on the experimental findings, an empirical method was proposed for predicting the degree of structuration in a particular tailings-polymer mixture. Such knowledge may allow the operators to minimize the negative effects of structuration through the selection of an appropriate deposition management plan or an appropriate

polymer or polymer dosage. Generated data could also help in the investigation of creep in flocculated tailings and the calibration and validation of the numerical modelling.

Although the polymer-aided flocculation was thought to be a minor influence on the consolidation characteristics of tailings (Hogg et al., 1993), this study showed that the polymer dosages have significant influences on the time-dependent behaviour of flocculated tailings. The results also implied that any specific polymer dosage that produces optimum dewatering performance in the short-term may not be the optimum polymer dosage for the long-term dewatering mechanisms. In other words, the short-term dewatering optimization may not result in optimization for the long-term dewatering. Different polymer dosages resulted in different degrees of structuration effect with the higher dosages showing a relatively prominent structuration behaviour but also a suboptimal dewaterability. By contrast, lower polymer dosages were found less susceptible to the negative effects of structuration and appeared to be an appropriate choice for producing optimal long-term dewatering and consolidation performance for the tailings deposits. Such choices of polymer dosage could also translate into reduced polymer consumption, and therefore, into major savings for the industry. By reducing 100 ppm of polymer dosage, we would be able to save at least CAD 0.1 million on treating 1 million cubic meters of tailings. If we assume that all the tailings that were deposited in the tailings ponds (there are more than 1,200 million cubic metres of tailings at a solids content $\geq 35\%$) would be treated with the polymers, the total cost savings would be CAD 120 million.

When this research program was undertaken, there was no other work on the time-dependent phenomena of creep, thixotropy, and structuration in polymer amended tailings

conducted at such an extensive level. During the duration of the research program, to the best of this author's knowledge, no other work on creep, thixotropy, and structuration in polymer amended oil sands tailings has been published in the journals or reported in the conference papers.

1.4 The organization of the thesis

The outline of the thesis is presented as follow:

Chapter 2: Presents a brief background of the research topics and includes the following:

- The challenges associated with tailings management and an overview of the common tailings treatment technologies that involve a dewatering step using a polymer.
- A review of the role of the fine components of tailings in tailings management since the flocculation-dispersion behaviour of fine particles governs the settling behaviour.
- A detailed description of the time-dependent creep, thixotropy, and structuration in natural clays and the case studies of creep, thixotropy, and structuration in natural clay deposits and/or clay slurries under field conditions and also in oil sands tailings.

Chapter 3: Provides a description of the materials used and the methods and techniques employed throughout the course of this research project.

Chapter 4: Focuses on the short-term dewatering of polymer amended oil sands tailings. After explaining the effects of polymer dosage on tailings dewatering efficiency and the need for polymer optimization, a detailed description of UV-vis spectrophotometry was presented. The technique was used for determining the residual polymer concentration in tailings bleed water based on absorbance measurements. The lowest measured absorbance

corresponded to the minimum polymer concentration, which again corresponded to the optimum polymer dosage.

Chapter 5: Presents a description of the torque-rheology based methods used for determining the optimum polymer dosage. A torque rheometer was used for determining the peak torque and totalized torque in the tailings samples while the samples were being sheared and dosed with polymer simultaneously. A local decrease in the measured values was found to correspond to the optimum polymer dosage. Standard dewaterability tests such as capillary suction time tests were performed to validate the torque rheometry results.

Chapter 6: Presents a detailed description of the investigation of creep, thixotropy, and structuration in flocculated tailings. The data presented include the results from the column dewatering tests conducted on tailings samples dosed with different polymer dosages under different boundary conditions, and also the results from the conventional/modified one-dimensional consolidation tests, fall cone tests, oscillatory rheometry tests, and microstructural investigations including mercury intrusion porosimetry, high/low vacuum scanning electron microscopy, and optical microscopy. The changes in macroscopic behaviour of flocculated tailings were compared to the changes in the tailings fabric and both were explained in light of the concept of time-dependent creep, thixotropy, and structuration behaviour. Also, the long-term implications of creep, thixotropy, and structuration effect for the dewatering performance of flocculated tailings were discussed.

Chapter 7: Provides a detailed description of the investigation of creep, thixotropy, and structuration in Leda clay. Leda clay deposits are known to display a structuration effect in their natural state. The data presented include the results from the column dewatering tests,

oedometer tests, fall cone tests, oscillatory rheometry tests, and low vacuum scanning electron microscopy. The experimental data obtained were explained in light of the concept of creep, thixotropy, and structuration, and the time-dependent behaviour observed in clay was compared to the time-dependent behaviour observed in polymer amended tailings.

Chapter 8: Provides a detailed description of the investigation of creep, thixotropy, and structuration effect in tailings samples that were dosed with different polymer dosages and mixed using different mixing methods. The data presented include the results from the column dewatering tests, oedometer tests, fall cone tests, and low vacuum scanning electron microscopy. The implications of different polymer dosages and polymer mixing methods for the time-dependent behaviour in flocculated tailings, and subsequently, for the long-term dewatering and consolidation performance of tailings deposits were discussed. Based on the experimental data, an empirical method was proposed for predicting the structuration effect in a particular tailings-polymer mixture.

Chapter 9: Synthesizes the experimental results with a focus on the criteria for the selection of an optimum polymer dosage for the long-term performance of tailings deposits.

Chapter 10: Provides a summary of the significant findings and observations. In conclusion, a set of recommendations were put forward for future research.

1.5 The publications related to the dissertation

The following section presents a summary of the publications that have been submitted to and published in journals and presented at conferences related to the research carried out for this thesis.

Journal papers:

Salam, A. M., Örmeci, B., & Simms, P. H. (2016). Determination of the optimum polymer dose for dewatering of oil sands tailings using UV-vis spectrophotometry. *Journal of Petroleum Science and Engineering*, 147, 68–76. [This paper is presented as part of this Ph.D. thesis in Chapter 4]

Salam, A. M., Örmeci, B., & Simms, P. H. (2020). *Determination of optimum polymer dosage for dewatering of oil sands tailings using torque rheology*. Manuscript submitted for publication. [This paper is presented as part of this Ph.D. thesis in Chapter 5]

Salam, A. M., Simms, P. H., & Örmeci, B. (2020). *Ageing in Leda clay and in polymer amended clayey oil sands tailings*. Unpublished manuscript. [This paper is presented as part of this Ph.D. thesis in Chapters 7, 8 and 9]

Conference papers:

Salam, A. M., Örmeci, B., & Simms, P. H. (2017). A new method for real-time optimization of polymer dose and dewatering of oil sands tailings. *Proceedings of the 70th Canadian Geotechnical Conference*. Ottawa, Ontario, Canada. [This paper is presented as part of this Ph.D. thesis in Chapter 4]

Salam, A. M., Simms, P. H., & Örmeci, B. (2017). Investigation of creep in polymer amended oil sands tailings. *Proceedings of GeoOttawa 2017, 70th Canadian*

Geotechnical Conference. Ottawa, Ontario, Canada. [This paper is presented as part of this Ph.D. thesis in Chapter 6]

Salam, A. M., Simms, P. H., & Örmeci, B. (2018a). Structuration in polymer amended oil sands fine tailings. *Proceedings of GeoEdmonton 2018, 71st Canadian Geotechnical Conference*. Edmonton, Alberta, Canada. [This paper is presented as part of this Ph.D. thesis in Chapter 6]

Salam, A. M., Simms, P. H., & Örmeci, B. (2018b). Evidence of creep & structuration in polymer amended oil sands tailings. *Proceedings of the 6th International Oil Sands Tailings Conference*. Edmonton, Alberta, Canada. [This paper is presented as part of this Ph.D. thesis in Chapter 6]

Chapter 2: Literature review

2.1 Oil sands tailings

2.1.1 Alberta's oil sands

Oil sands are a natural mixture of sand, clay, water, and bitumen, which is a relatively dense hydrocarbon ($\approx 1.0 \text{ kg/dm}^3$). After bitumen is extracted from the oil sands, it is refined to produce gasoline and other petroleum products (Natural Resources Canada, 2019). In Canada, there are three regions within the provinces of Alberta and Saskatchewan, namely, Athabasca, Cold Lake, and Peace River, which have oil sands deposits (Figure 2.1) (Canadian Association of Petroleum Producers, 2019). If we consider the proven oil reserves that can be recovered economically with the current technologies, Canada has the third-largest reserve in the world with 10.3% of the total reserves, and 98% of Canada's oil reserves are hosted in these deposits (Natural Resources Canada, 2019).

Figure 2.1. Oil sands areas in northeastern Alberta (from Alberta Energy, 2016).

Current oil sands operations are substantial and contribute largely to the national economy (Canadian Association of Petroleum Producers, 2019). The oil sands development provides significant economic benefits across Canada as billions of dollars are invested in capital projects every year promoting a range of economic activities and creating thousands of direct and indirect jobs (Canadian Association of Petroleum Producers, 2019). In 2017 alone, the oil sands industry contributed \$13 billion to the economy. Over the next two decades, the industry is expected to contribute \$4 trillion more (Canadian Association of Petroleum Producers, 2019).

2.1.2 Bitumen extraction process and basic properties of tailings

A typical oil sands ore deposit comprises bitumen (~12% by mass), sand, silts, clays (~85% mineral content by mass), and water (3% ~ 6% by mass). Bitumen, which is a thick, highly viscous fluid composed of complex, heavy hydrocarbons is the main product of the oil sands extraction process (BGC Engineering Inc., 2010). Generally, kaolinite (50% ~ 60%) and illite (30% ~ 50%) are the prominent clay minerals in oil sands; often, a small % of montmorillonite is also present (FTFC, 1995; Chalaturnyk et al., 2002; Kaminsky et al., 2009; Beier et al., 2013). Once the oil sands ores are mined using the open-pit or surface mining technique, the steps that are usually followed in producing petroleum products from the oil sands are: (a) extraction of bitumen from the oil sands ores through the removal of the solids and water by employing the Clark hot water extraction (CHWE) process (Clark and Pasternack, 1932); (b) the chemical treatment for converting heavy, thick bitumen into a lighter, intermediate crude oil product or synthetic crude oil (SCO) through the partial removal of the heavy hydrocarbon fractions (upgrading of the bitumen); and, (c) refining

of the crude oil into the higher-value petroleum products such as gasoline, diesel, aviation fuel, lubricants, and diluents (Natural Resources Canada, 2019; Oil sands magazine, 2019).

Clark hot water extraction (CHWE) process, a warm, caustic water-based extraction process used by the oil sands operators for the recovery of bitumen from the oil sands through flotation is carried out at an elevated temperature (70°C). The process uses a large amount of hot water, steam, and process additives such as sodium hydroxide and calcium citrate (Masliyah et al., 2004). Although the process is very efficient in that it recovers 90% to 92% of the bitumen, the process also results in a huge volume of fluid wastes known as oil sands tailings, which is a mixture of water, sands, fine clays, silts, and residual bitumen (Beier et al., 2013). On average, approximately 0.25 cubic metre of tailings is produced for every barrel of crude oil produced during the CHWE process (FTFC, 1995; Mikula et al., 1996).

According to the Canadian Oil Sands Classification System, the size fractions of solid constituents in tailings have been classified based on the following classification criteria: Sand (coarse particles) > 44 µm; silt (fines) 2 ~ 44 µm; and, clay (fines) < 2 µm (Masliyah et al., 2011). Typically, the fines are defined as the solid particles smaller than 44 µm in diameter or the material that can pass through a 44 µm sieve (Beier et al., 2013). The fines fraction includes silts and clays (Masliyah et al., 2011; Beier et al., 2013). The tailings that originate directly from the bitumen extraction process, which are also known as ‘whole tailings’ (WT) have a solids content (by mass) of 55% and the solids typically comprise coarse sand particles (82% by mass), fines (<44 µm) (17% by mass), and residual bitumen (1% by mass) (Chalaturnyk et al., 2002).

Figure 2.2. Schematic diagram of Clark hot water extraction (CHWE) process for bitumen extraction and formation of tailings ponds (adapted from BGC Engineering Inc., 2010).

Tailings are typically disposed of in some dedicated disposal ponds with large engineered dam and dyke systems, commonly known as tailings ponds (Oil Sands Discovery Centre, 2009; Canadian Association of Petroleum Producers, 2019). Once the

tailings are deposited into the ponds, the coarse sand particles settle out to form beaches, but the clay particles along with some fine solids such as silts remain in a suspension and form a layer (Figure 2.2). After years, these deposits tend to stabilize at water contents greater than twice their liquid limit ($> 200\%$, whereas liquid limits are typically $< 80\%$). At this stage, the tailings are called fluid fine tailings or FFT. However, they do not settle or consolidate appreciably afterwards. Even after many years, they may not attain a significant strength that would allow for successful land reclamation; even significant dam height (~ 80 m) and considerable period of containment (~ 50 years) have been proved ineffective in bringing any significant improvement (Sobkowicz, 2013; Beier et al., 2013).

According to the definition suggested by the Tailings Management Framework (TMF) for the Mineable Athabasca Oil Sands, fluid fine tailings refer to any fluid waste from the bitumen extraction facilities having more than 5% of suspended solids (by mass) and less than 5 kPa of undrained shear strength (Government of Alberta, 2015). The typical ranges for the geotechnical properties of unamended oil sands tailings are summarized in Table 2.1 (based on the studies on tailings available in literature and FTFC, 1995).

Table 2.1 Tailings properties (based on the studies on tailings and FTFC, 1995)

Parameters	Average value
Average solids content (%)	29 ~ 36
Average water content (%)	178 ~ 245
Mean particle size (μm)	5 ~ 10
Hydraulic conductivity (cm/s)	$1 \times 10^{-9} \sim 1 \times 10^{-4}$
Average void ratio	5 (at an average solids content of 33%)

Specific gravity	2.12 ~ 2.55
Liquid limit (%)	40 ~ 75
Plastic limit (%)	10 ~ 30
Shear strength (kPa)	< 1 kPa (at a solids content range of 30% ~35%)

Figure 2.3. Hydraulic conductivity of FFT (from Jeeravipoolvarn, 2005).

Since fluid fine tailings demonstrate considerable changes in hydraulic conductivity with void ratio, it is more appropriate to present the hydraulic conductivity-void ratio relationship for tailings over a wide range of void ratio (Figure 2.3).

2.1.3 Tailings regulations

To regulate the tailings treatment and management in the oil sands and to accelerate the dewatering processes and reduce the volume of fine tailings deposits so that they can be reclaimed, the Alberta Energy Regulator has adopted some guidelines, which were set out in the ‘Directive 085: Fluid Tailings Management for Oil Sands Mining Projects’. The directive describes the minimum acceptable undrained shear strength, solids content, and sand to fines ratio that the treated tailings must attain before they are ready to be reclaimed (Hripko et al., 2018; AER, 2019).

The previous directive, ‘Directive 074: Tailings Performance Criteria and Requirements for Oil Sands Mining Schemes’ was introduced in 2009 with a focus on slowing the growth of tailings ponds by creating standard requirements for all projects. The Directive 074 set rules for the number of fine particles ($<44\ \mu\text{m}$) that must be captured and required fine tailings to achieve specific strengths at specific times following depositions (5 kPa after 1 year and 10 kPa within 5 years) (AER, 2009; Simieritsch et al., 2009). However, the current directive (Directive 085) does not set standard requirements, rather it requires the oil sands operators to develop a plan to manage both existing fluid tailings and future tailings production (AER, 2019). Under Directive 085, the operators must set project-specific targets to ensure that the fluid fine tailings are ready-to-reclaim within ten years of the end of the mine’s life and the operators need to work towards this target over

the mine life (AER, 2019). Directive 085 measures the progress in tailings reduction using the overall volume of fluid fine tailings. The new directive has exceeded Directive 074 in that it handles both existing fluid tailings (also known as legacy tailings) and new fluid tailings growth (AER, 2019).

2.1.4 Conventional dewatering technologies

The dewaterability of oil sands tailings can be influenced by many different aspects of the mining and milling processes including extraction process (chemistry, temperature, bitumen recovery, recycle water use, etc.), production rate, material transport methods and distances, site conditions (weather, overburden, topography, surface area, surface water and groundwater, etc.), and also by the composition and different properties of tailings materials including fines content, overall size range, shape of particles, specific gravity, mineralogy, etc. (Wislesky et al., 2013).

In recent times, considerable effort and resources have been invested in tailings research to develop technically innovative and economically feasible technologies for accelerating the dewatering processes and increasing the solids settling rate of FFT (so that the liquid-solid separation can occur within a reasonable period) and for decreasing the volume and increasing the strength of FFT (so that they can be reclaimed) (BGC Engineering Inc., 2010). Usually, a combination of physical and/or chemical processes are utilized for the dewatering of tailings (Watson et al., 2011). Chemical processes help the fine clay particles to overcome the repulsive electrostatic forces between them and to aggregate, and thus, help in the destabilization of the highly dispersed clay particles in FFT (Watson et al., 2011). By contrast, physical processes involve the mechanical separation of

the fine solids from the water. Chemical processes typically involve some form of chemical amendments such as coagulants and polymers. Adding coagulants helps to neutralize the repulsive charges, and therefore, helps the floc formation. Polymers help the fine particles or flocs to aggregate and form larger flocs (Watson et al., 2011).

Variations on ‘composite’ or ‘consolidated’ tailings (CT) technology are commonly used by the industry to reduce the tailings inventory. In composite/consolidate tailings (CT) method, densified extraction tailings (coarse sand from cyclone underflow tailings or COF) and FFT are mixed with a coagulant (typically gypsum) to generate non-segregating tailings. Then, these tailings are deposited into a tailings pond so that they can form a rapidly consolidating deposit (BGC Engineering Inc., 2010). Although CT has always been one of the primary tailings management techniques, the approach has a few drawbacks as well. Gypsum, which is typically used in a large volume as a coagulant alters the chemistry of the recycled water through the buildup of a high concentration of divalent ions including calcium and sulphate, and therefore, affects the bitumen extraction process adversely (BGC Engineering Inc., 2010).

The tailings treatment technologies that use polymers as flocculants add polymer to the FFT through mainly two different methods: (a) adding polymer using physical-mechanical methods; and, (b) in-line flocculation (Beier et al., 2013). Common physical-mechanical methods include thickening in a conventional gravity thickener and centrifugation. In the thickening operation, the fine tailings stream that originates from the CHWE bitumen extraction process enters the thickener as cyclone overflow (COF). The polymer is added to these sand-depleted tailings streams to facilitate the settling of the fine

particles and increase the solids content. Prior to the deposition, these tailings streams are dewatered to a final solids content of 40% (by mass). Then, they are referred to as thickened tailings (TT) (Hyndman and Sobkowicz, 2010). However, when improved polymers and high-density or deep-cone thickeners are used for dewatering and the final solids content reaches up to 60% (by mass), they are referred to as paste tailings (PT) (Yuan and Lahaie, 2009; Masala, Matthews, and Berger, 2010).

Figure 2.4. (a) Deep-cone thickener used for producing paste tailings (<http://www.mining-technology.com/contractors/filtering/flsmidth/flsmidth5.html>); and, (b) a centrifuge vessel (<https://www.cosia.ca/initiatives/tailings/projects/tailings-centrifugation>).

In centrifugation operations, the tailings are dredged from a tailings pond, diluted, and then, treated with gypsum and polymer. The flocculated FFT (fFFT) is then transferred to a centrifuge vessel that uses the centrifugal force for rapid solid-liquid separation. After the fFFT is processed in a centrifuge vessel, a high solids content (55%) centrifuge cake is formed as a final product, which is disposed of in a designated facility (Sobkowicz, 2012; Beier et al., 2013). The centrifugation method of treating FFT has the potential for reducing the tailings inventory as well as eliminating the need for the storage of FFT, thereby

minimizing the time required between the deposition and reclamation processes (Mikula et al., 2009).

In the case of in-line flocculation or in-line thickened tailings (ILTT), the polymer stock solutions, which are usually made of an anionic polymer are injected directly into the transfer pipeline containing dredged FFT at a distance of a few to several hundred metres from the deposition point, and the combined product is re-deposited into the DDA (Matthews et al., 2011; Wells et al., 2011). In this technique, the efficiency of dewatering typically depends on a successful combination of polymer addition and strategic deposition (Beier et al., 2013). Field trials have shown that the application of polymers allows such technologies to increase the solids content (by mass) to at least 45% ~ 55% from 30% ~ 35% within 24 hours following deposition in thin lifts of 0.60 m (Matthews et al., 2011). For dense fines slurries such as FFT, the in-line flocculation technology has been observed to produce better dewatering performance in comparison to the thickeners (Wells, 2014).

Generally, two depositional techniques are available for the in-line fFFT. After the polymer is added to the FFT within the pipeline and mixed, the combined product is deposited onto a mildly sloped beach at a solids content of 30% ~ 35% (Kolstad et al., 2012). Immediately after the deposition, the fFFT releases water as the fine particles aggregate to form flocs and settle by gravity. The fFFT is typically deposited in thin layers so that additional dewatering can be achieved through a combination of settlement, seepage, and environmental dewatering. Depending on the commercial operators, this disposal technology is also known as ‘tailings reduction operations’ (TRO) or ‘atmospheric fines drying’ (AFD) (Kolstad et al., 2012; Beier et al., 2013; Dunmola et al., 2013b).

In another disposal technique, the fFFT is discharged into large, deep depositional cells to induce self-weight consolidation and environmental dewatering through a combination of surface evaporation and freeze-thaw dewatering. Additionally, the decant systems and the mechanical channelling (perimeter ditching) systems are installed for facilitating the drainage of the water that is released to the surface and for accelerating the process of dewatering and development of strength. At the commercial scale, this depositional technique is known as ‘rim-ditching’ or ‘accelerated dewatering’ (Lahaie et al., 2010; Beier et al., 2013; Dunmola et al., 2013b).

2.1.5 Flocculation and flocculants

Over the last decade, accelerating the dewatering and strength gain of the tailings streams through the polymer-aided flocculation of fine clay particles has been researched and trialled by the oil sands industry extensively (Beier et al., 2013). Flocculation refers to the process by which high molecular weight polymers help the colloidal-sized fine particles to aggregate from dispersions into larger flocs through the formation of bridges between those particles or by charge neutralization (Vajihinejad and Soares, 2018; Vajihinejad et al., 2019). The larger aggregates or flocs settle more quickly than the individual fine particles that would not normally settle within an economically reasonable time frame (Yao, 2016). Hence, polymer-assisted flocculation is often deemed an important pre-treatment step in the dewatering of tailings streams containing significant quantities of very fine particles ($<44\ \mu\text{m}$) and the efficiency of the flocculation step ultimately governs the performance of the dewatering system (Hogg, 2000).

Depending on the properties of the polymer used as well as tailings composition and characteristics, the flocculation process could involve different destabilization and flocculation mechanisms such as bridging, charge patching and charge neutralization (Hogg, 1999, 2000). Destabilization mechanisms involve the compression of the double layer and the suppression of interparticle repulsive forces so that attractive forces can become dominant and the particles can come closer (Hogg, 2000). Bridging flocculation is the most prevalent form of flocculation in which small quantities of large chain polymers are adsorbed over many particles simultaneously, thereby developing molecular ‘bridges’ between the individual particles and helping them to aggregate (Figure 2.5) (Hogg, 1999). Since the adsorption of polymers on the surfaces of suspended solid particles initiates the formation of such bridges, the extent and the efficiency of the polymer adsorption at solid-liquid interfaces play a crucial role in the formation of the particle-particle bridges. The polymer-particle collision frequencies govern the kinetics of adsorption, which again determines the distribution of adsorbed polymers on the individual solid particle surfaces and also the adhesion efficiency of the particle-particle collisions that ultimately lead to the bridging flocculation (Hogg, 1999).

Figure 2.5. Bridging flocculation of particles (adapted from Kitchener, 1972).

Polyacrylamide (PAM) flocculants and its ionic copolymers (the synthetic polymers based on the repeating units of acrylamide and its derivatives), which typically have a molecular weight in the order of several million Daltons are extensively used with the existing tailings dewatering technologies as they are capable of forming larger flocs, inducing effective solid-liquid separation, and producing high settling rates (Vedoy and Soares, 2015). The mutual charge repulsion between the negatively charged anionic PAM and the negatively charged clay particles helps the rearrangement of adsorbed polymer chains on the solid surfaces, which increases the polymer-particle collision probability, and subsequently, promotes the adsorption of polymer molecules on different particle surfaces simultaneously, thereby initiating a network of the polymer molecules and clay particles that results in flocculation (Hogg, 1999; Agarwal, 2002; Tripathy and De, 2006).

In the industrial application of high molecular weight anionic polyacrylamides, when an optimal range of polymer dosage is used, it causes substantial flocculation of the negatively charged particles and leads to the maximum dewatering performance (Hogg, 1999, 2000). However, if an inadequate polymer dosage is used, failure to establish sufficient bridging links between the particles could result in dispersion (Tripathy and De, 2006). By contrast, using polymer in the overdose range hinders the adsorption of the polymer by reducing their access to the particle surfaces, thereby reducing the efficiency of the bridging process (Moody, 1992). In fact, during flocculation, polymer adsorption and floc growth occur simultaneously as they are not any separate or sequential processes. Rather, these two mechanisms depend on each other since floc growth requires adsorption and adsorption is limited by the floc growth (Hogg, 1999). An optimum flocculation occurs

only at a polymer dosage that corresponds to a partial particle coverage since such incomplete surface coverage ensures that there is adequate unoccupied surface available on each particle for the adsorption of other polymer molecules and polymer bridging of two or more particles will occur during the collisions of the segments of flocculant chains attached to the particles (Hogg, 1984).

2.2. Clay-water-electrolyte system

2.2.1 Clays in FFT and flocculation

The clay particles do not constitute the bulk part of the tailings streams, rather they comprise a relatively small percentage of tailings by weight. Nevertheless, the clay-water interactions influence the tailings properties and play a crucial role in tailings dewatering behaviour by inhibiting the consolidation of the tailings materials into trafficable deposits (Mikula and Omotoso, 2006; Mikula, 2011). The tailings retain a significant amount of water at alkaline pH and show a poor dewaterability, which has been attributed to the high surface area of the clay particles. Higher surface area increases the charge density on the fine clay particles, thereby increasing the repulsion and the distance between them, and subsequently, forming a stable colloidal suspension where the particles remain in a highly dispersed state (Vedoy and Soares, 2015; Gumfekar, Vajihinejad, and Soares, 2019).

The dispersed or flocculated state of the clay particles depends on how the plate-shaped clay particles orient themselves in the clay-water suspensions and whether the structure formed by the clay particles are ordered or random (Figure 2.6). The faces and the edges of the clay particles have different charge distributions. When the clay particles

organize themselves in a face-to-face pattern, the interactions between them are generally repulsive; however, when they are arranged in a face-to-edge pattern, the interactions are generally attractive. When the clay particles are in a dispersed state, the individual clay particles orient themselves in a face-to-face pattern. By contrast, a more random structure is prevalent in the flocculated clays, where face-to-edge orientations are more common (Mitchell and Soga, 2005).

Figure 2.6. Different arrangements of the clay particles in clay-water suspensions (from Firoozi et al., 2015).

When polymers are added to the FFT, they help the negatively charged, highly dispersed fine clay particles to overcome the potential energy barrier between them through the suppression of double layer and trigger the destabilization of the suspended clay particles so that they can flocculate (Tripathy and De, 2006). To ensure that the polymer

will act most efficiently on the dispersed clay particles in the suspension, it is necessary to select the best performing polymer or polymer dosage experimentally. However, prior to polymer optimization, it is also important to understand the flocculation-dispersion behaviour of the clay particles in FFT, especially, in light of the electrical double layer theory. An in-depth understanding of the colloid-electrolyte interactions as well as colloid-electrolyte-polymer interactions in FFT and their effects on the relevant engineering properties such as sedimentation and rheology is crucial to the improvement of flocculant-based dewatering technologies and for accelerating the settlement of the tailings.

2.2.2 Colloid-electrolyte interactions

By definition, clay minerals are hydrous aluminum phyllosilicates that comprise layers of tetrahedral sheets and octahedral sheets with the capacity for the substitution of one cation on the interlayer surface for another. Although such substitution does not alter the shape of the atomic structure, it leads to the development of a negative charge at the surface of the clay minerals (Kaminsky, 2011). The term ‘clay’ refers to a naturally occurring component of soil that consists mostly of the fine-grained aluminum phyllosilicates minerals. Because of their high specific surface area and ion exchange capacities, clay minerals play an important role in the surface chemistry of soils and sediments (Al-Ani and Sarapää, 2008).

The soil is a mixture of organic and inorganic constituents. The inorganic components, which typically constitute about 40% ~ 45% of the soil volume have been classified into two types of minerals: (a) primary minerals and (b) secondary minerals (Evangelou, 1998). The minerals that are formed during the crystallization process of the

molten magma are known as primary minerals. The secondary minerals are formed by the weathering of the pre-existing primary minerals (Evangelou, 1998). Among the most commonly found secondary minerals, the aluminosilicate minerals, which constitute a major portion of the clay-sized ($<2\ \mu\text{m}$) fraction of soils are composed of two basic structural units: (a) silicon-oxygen tetrahedrons and (b) aluminum-oxygen octahedrons (Evangelou, 1998).

Figure 2.7. Schematic of (a) silicone tetrahedron, where four oxygen atoms are surrounding a silicon atom, forming a tetrahedral structure; and, (b) aluminum octahedron, where six oxygen atoms are surrounding an aluminum atom, forming an octahedral structure (adapted from Al-Ani and Sarapää, 2008).

In a typical silicon-oxygen tetrahedron, one silica atom (Si^{4+}) is surrounded by four oxygen atoms (O^{2-}), and in an aluminum-oxygen octahedron, six oxygen atoms (O^{2-}) are bonded to an aluminum atom (Al^{3+}) at the center (Figure 2.7). In clay minerals, these tetrahedral and octahedral structures share oxygen atoms with their adjacent structural units and form tetrahedral and octahedral sheets respectively (Evangelou, 1998). The octahedral and tetrahedral sheets act as the basic building blocks for different types of clay minerals such as kaolins, smectites, etc. However, different arrangements of these sheets result in different structures and compositions for the clay minerals, which in turn, result in different physical and chemical properties for them (Al-Ani and Sarapää, 2008).

Very often, during the initial formation of clay minerals or during their subsequent alteration, an aluminum atom substitutes a silicon atom in the tetrahedral sheet, or divalent cations replace the aluminum atoms in the octahedral sheet. Such substitution of a Si^{4+} or an Al^{3+} cation by a cation with lower valence (such as Mg^{2+}) are known as isomorphous substitution and lead to the development of a permanent negative charge at the surface of the clay mineral, and subsequently, a negative electric potential at the surface (Evangelou, 1998). When the clay particle is immersed in an aqueous solution, the negatively charged particle surface attracts the positively charged cations available in the surrounding water in order to maintain the electrical neutrality in the clay-water system. Subsequently, because of the net surface charge, a layer of counter ions (ions with opposite charge of the colloid) is developed surrounding the particle (Evangelou, 1998). The non-uniform distribution of ions around the charged particle and the interaction of these charged species lead to the formation of an electrical double layer (EDL) (Evangelou, 1998).

Figure 2.8. Distribution of ions in the Stern model of EDL (adapted from Priesing, 1962).

Figure 2.8 shows the distribution of ions schematically in the Stern model of the EDL system. According to the Stern model of the electrical double layer, the electrical double layer comprises two parallel layers of charge surrounding the colloid particle: (a) an internal Fixed or Stern layer, and (b) an outer diffuse layer (Guo, 2012). In the Stern layer, the oppositely charged ions or counter ions (from the surrounding water) remain firmly attached to the surface of a negatively charged colloidal particle. In the outer diffuse layer, although the negatively charged colloid particle still attracts the oppositely charged ions, these positive ions are also repelled by the Stern layer, which prevents them from approaching the colloid. Hence, in the diffuse layer, most of the ions move freely in the

fluid under the influence of electric attraction rather than being firmly anchored to the particle surface and they create a charged atmosphere. Since the negative electric potential decreases exponentially with the increasing distance from the particle surface, the diffuse layer is characterized by a decrease in the concentration of counter ions with distance (Guo, 2012; Nik, 2013). However, at a distance farther away from the surface, there comes a region where the electrical forces of attraction try to pull the counter ions towards the surface, whereas a repulsive force of diffusion tries to pull them away from the surface. When these opposing forces become equal, a neutral charge starts to prevail, and the colloidal suspension becomes an electroneutral solution (Evangelou, 1998).

Since the clay mineral surfaces are not perfectly spherical or flat, which is the requirement of the electrical double layer (EDL) theory, it is difficult to apply the theory to the clay minerals at a quantitative level. Nevertheless, the electrical double layer theory is appropriate for qualitatively explaining the behaviour of clay minerals in a colloidal suspension including flocculation and dispersion behaviour (Evangelou, 1998).

2.2.3 Colloid-electrolyte-polymer interactions

Typically, the colloidal behaviour in the natural clay-water systems is governed by the dispersion-flocculation processes (Evangelou, 1998). When two clay particles come close to each other in the solution, the repulsive forces that result from the interaction of the negative charges distributed over each particle (electrical double layer forces) counteract the attractive forces between them (i.e. van der Waals forces). When these attraction and repulsion forces are in an equilibrium state, the fine clay particles remain dispersed and form a stable structure in a colloidal system (Tang, 1997).

The fine clay particles in the fluid fine tailings form a stable colloidal system. The highly dispersed state of the clay particles in tailings originates with the chemistry of the bitumen extraction process (Beier and Sego, 2008). Clay minerals, in the presence of chemical additives such as caustic NaOH, which is added during the oil sands extraction process attain an enhanced negative surface charge and the resulting repulsion between the negatively charged surfaces of the adjacent particles promotes dispersion of the clay particles (Beier and Sego, 2008). Although the dispersion of clays is necessary for efficient bitumen extraction by flotation, it also poses a challenge to the rapid dewatering of tailings since it hinders sedimentation and consolidation of the fine particles in tailings (BGC Engineering Inc., 2010). In FFT, the dispersed flatten clay particles form a card-house clay microstructure with large pore spaces entrapping the bulk of the process water. Such fabric of clay particles gives the tailings material a high water-retention capacity and reduces the self-weight consolidation rate (Scott and Dusseault, 1982; Tang, 1997; Wong et al., 2008).

If we want to improve the dewatering and settling characteristics of a colloidal system like tailings, it is essential to destabilize the equilibrium state and disrupt the stable dispersed structure in the colloidal suspension by bringing the particles together and forming larger aggregates (Nik, 2013). According to the DLVO theory, when these particles come close to each other, they require sufficient kinetic energy to overcome the repulsive energy barrier acting between them in order to agglomerate. Generally, there are several ways for eliminating the repulsive energy barrier and destabilizing the stable structure in the colloidal suspension including (a) adsorption and charge neutralization method (also known as charge patching), where the adsorption of the positively-charged

cationic polymers on the negatively-charged particle surfaces leads to the reduction of double layer repulsion forces, and eventually, to the destabilization of the colloidal particles (charge neutralization through the compression of double layer); and, (b) bridging mechanism, where the negatively-charged anionic polymers adsorb on the particle surfaces via hydrogen bonding. If the high molecular weight anionic polymer has a long polymer chain, which is extended away from the particle surface and into the solution, then, the long-chain polymer would adsorb onto the surface of several particles at the same time, thereby physically bringing them together and forming larger flocs through the flocculation process (Tripathy and De, 2006; Vedoy and Soares, 2015; Vajihinejad et al., 2019).

Figure 2.9. (a) A stable colloidal system with the particles in a dispersed state; and, (b) an unstable colloidal system with the particles aggregating to larger flocs.

When polymers are added to the colloidal suspensions such as clay-rich FFT and promote flocculation of the clay particles, the electrostatic repulsion forces between these fine particles are reduced. Because of this shift in the balance between the van der Waals attractive forces and electrostatic repulsion forces, the particles become attracted to each

other and start to coalesce, form flocs, and eventually, they settle out (Figure 2.9b). However, if the attractive forces fail to exceed the repulsive forces, the net force acts as a repulsive force, and the particles remain dispersed in the suspension (Figure 2.9a) (Mitchell and Soga, 2005).

The electrical double layer comprises counter ions firmly attached to the colloid surface in the Stern layer and a charged atmosphere surrounding the colloid in the diffuse layer. The thickness of this electrical double layer, a property that reflects the range of the electrostatic influence relative to the particle surface depends on the valence and the concentration of the ions in the colloidal system (Mitchell and Soga, 2005; Miller, 2010). After adding polymer to the clay suspensions, when the segment of the polymer chain is adsorbed on the colloids, the subsequent changes in the ionic valence and ion concentration reduces the double layer thickness. The double layer thickness governs the flocculation of the colloid particles as there is an inverse relationship between the double layer thickness and the tendency of the particles to flocculate (Mitchell and Soga, 2005; Miller, 2010). The suppression of the double layer allows the van der Waals attraction forces to dominate over the electrostatic repulsion forces, and subsequently, the net force acts as an attractive force and causes flocculation of the particles. Generally, as the electrical double layer gets thinner, the particles in suspension tend to flocculate more (Mitchell and Soga, 2005).

2.2.4 Zeta potential and flocculation

Within the electrical double layer of a clay particle in a clay-water suspension, a plane is thought to develop at the outer boundary of the Stern layer, which encompasses the colloidal particle and the ions that are closely associated to that particle and forms a

stable entity in such a way that those ions along with the colloidal particle move through the solution as a unit. This plane is known as the shear plane or slipping plane and the electric potential that exists between that unit and the surrounding dispersion medium at the surface of this shear plane is known as the zeta potential of the particle (Figure 2.8) (Hunter, 1988; Lyklema 2000; Hughes, 2000; Delgado et al., 2005). This electric potential, also known as electrokinetic potential, is typically expressed using the Greek letter zeta (ζ), that's why it is called zeta potential.

When polymers are adsorbed on the particles, the thickness of the adsorbed polymer layer on the particle surfaces also gradually increases. Such an increase in thickness changes the properties of the Stern layer and generates a hydrodynamic effect. Subsequently, the ions in the electrical double layer are displaced from the Stern layer prompting a displacement of the shear plane. Such a shift in the position of the shear plane further from the Stern layer alters the double layer thickness, which in turn decreases the zeta potential. Again, such a reduction in zeta potential prompts a reduction in surface charge, which results in the elimination of the repulsive energy barrier, and subsequently, leads to the flocculation of the particle (Mpofu et al., 2003). When an inadequate dosage of polymer is applied, an inadequate decrease in net negative charge on the particle surface results in the dispersion of the particles. By contrast, an excess dosage prompts the polymers to saturate the particle surfaces to such an extent that it prevents the colliding particles from getting firmly attached to the polymers as there is no free space available for polymer adsorption. Also, the free polymer molecules in the suspension start to repulse each other, which provides stability to the suspension rather than destabilizing it (Agarwal,

2002; Vedoy and Soares, 2015). Hence, when the polymer is applied to a colloidal suspension in such a concentration that it provides partial coverage of the particle surface and the unoccupied surface space is available for adsorption of other polymer molecules, such concentration promotes an effective flocculation of the particles by allowing the bridging flocculation to continue (Agarwal, 2002; Vedoy and Soares, 2015).

Although the zeta potential is an intermediate value and is not equal to the surface electric potential in the electrical double layer, it reflects the net surface charge of the colloidal particles in a colloidal suspension (Mitchell and Soga, 2005). Hence, the degree of electrostatic repulsion between the adjacent and similarly charged particles can be approximated by the magnitude of zeta potential. That's why the zeta potential is considered a key indicator of the stability of the colloidal suspensions. Generally, a higher absolute value of zeta potential refers to a stronger electrostatic repulsion between the particles, which also indicates that the particles are more dispersed, and the colloidal dispersion is more stable and will resist aggregation (Hanaor et al., 2012; Helmenstine, 2018). By contrast, lower absolute values of zeta potential indicate that the particles are overcoming the electrostatic repulsion forces and moving closer to each other to aggregate, which would lead to the flocculation of the particles (Hanaor et al., 2012; Helmenstine, 2018). For the clay particles in the clay-water suspensions, the values of zeta potential are typically measured in the range of 0 ~ 50 mV (Mitchell and Soga, 2005).

Since the behaviour of the colloids in the colloidal suspensions primarily depend on the properties of the particle-liquid interface and the zeta potential indicates the potential difference across the phase boundaries between solids and liquids, the measurement of zeta

potential is important to the practical study and control of colloidal stability and dispersion-flocculation processes in various types of colloidal dispersions including clay-water systems (Helmenstine, 2018). Especially, the increase in the thickness of the adsorbed polymer layer and the subsequent decrease in the zeta potential are considered important indicators of the effectiveness of polymers in flocculating a colloidal dispersion (Mpofu et al., 2003). Although it is not possible to measure the zeta potential directly, it can be determined experimentally. The equipment that is used for determining the zeta potential typically relies on either electrophoresis or electroacoustics-based techniques. Electrophoresis refers to the movement of charged colloidal particles relative to the fluid under the influence of an electric field (Helmenstine, 2017). The use of electrophoresis technique for measuring the zeta potential of clay suspensions (mostly kaolinite) has been reported in the literature (Greenwood et al., 2007). The technique has also been used for measuring the zeta potential of oil sands fine tailings (Rima, 2013). Rima (2013) measured different physicochemical properties of tailings bleed water (obtained from both flocculated and non-flocculated tailings) including zeta potential in order to investigate the changes in physicochemical properties before and after polymer adsorption. The zeta potential was found to decrease after adding the polymer, which was due to the shear plane displacement. Such a decrease in zeta potential was also found to correlate well with the dewatering parameters measured from the centrifuge test (Rima, 2013).

2.2.5 Summary: Clay-water-electrolyte system

The clay minerals in oil sands not only make the recovery of oil from oil sands challenging but also influence the tractability of the tailings produced from the bitumen

extraction process. The ultra-fine component of the oil sands fine solids and its behaviour has been found responsible for the formation of fluid fine tailings from the bitumen extraction plants and these ultra-fine clay particles also affect the tailings management since they dominate the settling behaviour (Tu et al., 2005). Due to the gap-graded grain size distribution, the whole tailings stream shows a segregating behaviour when it is discharged into the tailings ponds. The coarse fraction settles closest to the discharge point and the fines flow away from the discharge point. These fine clay particles remain dispersed and form a stable structure in a colloidal system (Tang, 1997). The electrical double layer that forms around the small, individual colloid particles plays an important role in the mechanism of the electrostatic stabilization of these colloids (Park and Seo, 2011). Since such colloidal stability impedes the settlement of the fine particles in tailings, polymers are used for destabilizing the colloidal suspension in tailings. When the polymers are added to the tailings, as the particles collide with each other, their electrical double layers begin to interfere; eventually, they overlap and get thinner, and their zeta potential values decrease. Subsequently, the flocculation tendency of the particles gets stronger and they start to aggregate and form rapid settling flocs making the settlement process faster. That's why, the measurement of properties that reflect the double layer properties in colloidal dispersions (i.e. zeta potential) is important to the investigation of the stability of a colloidal system such as FFT (Hanaor et al., 2012; Helmenstine, 2018).

Better management of the oil sands tailings in commercial oil sands operations largely depends on the improvement of the settling characteristics of the fine clays in an economical and environmentally acceptable manner (Chalaturnyk et al., 2002). Since there

is strong evidence that the poor settling behaviour of tailings results from the characteristics of these clay particles and their activities, it is important to understand the nature of clay minerals present in the highly ionic environments of oil sands tailings streams and also the role played by them in overall tailings behaviour, especially, the behaviour of fluid fine tailings (Kaminsky et al., 2006; Hooshier et al., 2010; Boxill, 2011). The effectiveness of the technologies that involve dewatering and thickening of tailings through polymeric flocculation depends on the improvement of the geotechnical properties of FFT through an efficient modification of the clay behaviour (Omotoso and Mikula, 2004).

2.3 Short-term dewatering behaviour of flocculated tailings

2.3.1 Polymer optimization for tailings dewatering

When a polymer is added to the FFT, the flocculation and dewatering efficiency depends on several factors including polymer properties such as molecular weight, the nature of the functional groups, ionic content and ionization degree, dosage, charge density; the hydrodynamic conditions used during polymer treatment such as intensity of mixing and the rate of polymer addition; and, tailings properties such as level of dissolved solids, the mineralogical composition, particle size, particle surface charge, pore water chemistry, etc. (Sworska et al., 2000; Vedoy and Soares, 2015).

For assessing the performance of different polymers, selecting the potential polymer treatment candidates, and determining the optimal polymer dosage for the full-scale or field-scale implementation, usually, a series of laboratory-scale screening experiments are performed on the flocculated tailings, and the best performing polymer or

the optimum polymer dosage is selected based on the evaluation of some specific key performance indicators (KPIs). The commonly used key performance indicators (KPIs) include capillary suction test time (CST), settling rate (from the settling column test), yield stress, clay-to-water (CWR) ratio (in flocs), solids content and net water release (Wells et al., 2011; Demoz and Mikula, 2012; Mizani et al., 2013, 2017; Boxill et al., 2018). Since the fFFT typically releases a significant amount of water within the initial 24 ~ 48-hour period of polymer mixing, most of the laboratory-scale research on polymer optimization focus on maximizing short-term dewatering such as dewatering over one- or two-days post-deposition or post-treatment (Mizani et al., 2017; Boxill et al., 2018). However, it has always been a great challenge to replicate the same polymer mixing and flocculation conditions and dewatering performances in the fields where the operating conditions can vary considerably. In order to overcome this challenge and to satisfy the dewatering and consolidation performance criteria post-deposition under field conditions, currently, a wide spectrum of screening criteria or key performance indicators (KPIs) are used for evaluating the performances of the polymers under a wide range of operating conditions including polymer dosing and transportation of flocculated tailings through the pipelines prior to deposition, where a wide range of factors may influence the flocculation processes and dewatering performances (Boxill et al., 2018).

Wells et al. (2011) reported on the bench and pilot-scale experiments that were conducted to examine the dewatering potential of the fFFT material as a function of polymer dosage, polymer injection and mixing, total shear applied, and clay-to-water ratio. Several polymer dosages in the range of 500 ~ 1700 ppm and different polymers including

anionic polyacrylamides were trialled. Wells et al. (2011) measured the water released by decanting the initial water release and performing the capillary suction time (CST) tests. They observed that the tailings-polymer mixture was able to attain an optimal permeability and release maximum free water only at an optimal polymer dosage. However, in the under-dosed or the overdosed condition, the amount of free water released by the FFT was relatively smaller (Wells et al., 2011).

Figure 2.10. The correlation between the buildup, rearrangement, and breakdown of flocs and the release of water and the change in the yield stress in fFFT (from Wells et al., 2011).

Wells et al. (2011) investigated the effect of mixing time or shear duration on the yield stress using a rheometer and also shed light on the effect of the shear energy applied to the fFFT on its post-deposition dewaterability potential and strength performance. The

buildup, rearrangement, and breakdown of flocs and the release of water were found correlated to the change in yield stress or the strength developed within the fFFT material (Figure 2.10). Wells et al. (2011) observed that excess shearing could result in a decline in the strength of fFFT material and reduce the permeability and dewaterability potential. Although it is necessary to apply adequate mixing energy to the FFT-polymer mixture for a specific period of time in order to disperse the polymers into the FFT materials and produce a well-flocculated FFT with good dewaterability potential, if the mixing is continued at high intensity for a period longer than the necessary, the larger aggregates or flocs resulting from the flocculation process may disintegrate into smaller flocs and result in poor dewaterability and strength (Yao, 2016). Hence, the buildup of yield stress in the fFFT material was found to be a function of the shear energy applied to the FFT-polymer mixture (Wells et al., 2011).

Demoz and Mikula (2012) investigated the flocculation of oil sands tailings to determine the influence of mixing energy (or shear energy) on dewatering performances. They used two high molecular weight anionic polymers and four different mixing tools with different impeller speeds for flocculating different FFT with different solids contents. The amount of water released in the settling columns and the capillary suction time (CST) were used as the criteria for evaluating their flocculation and dewatering performances. The capillary suction time was found inversely proportional to the amount of water released. For all the combinations of polymers and mixing tools used, both the peak in the amount of water released and the minimum in the capillary suction time were found to be a function of mixing time. Such findings implied that there is an optimal mixing time or

mixing energy for a given FFT-polymer mixture under a given set of mixing conditions, which also corresponds to the optimum flocculation and dewatering of that FFT-polymer mixture (Demoz and Mikula, 2012).

Figure 2.11. Dewaterability of a 21% solid content FFT sample (dosed with 1000-ppm of 0.4% by weight A3338 polymer and mixed at 600 rpm using a Rushton turbine impeller), measured over a period of 3 days (from Demoz and Mikula, 2012).

Mizani et al. (2013) examined the short-term effect of polymer-assisted flocculation and reported that the formation of flocs in polymer amended tailings resulted in considerable water release within 24 hours of polymer mixing. Mizani et al. (2013) investigated the effects of different polymer dosages and mixing durations on the dewatering potential of flocculated tailings (Figures 2.12, 2.13, and 2.14) and observed that relatively lower dosage had more dewatering potential in thin lift deposition (0.50 m).

Figure 2.12. The measurements of water released from the FFT mixed with different polymer dosages (from Mizani et al., 2013).

Figure 2.13. The measurements of solids in released water obtained from the FFT mixed with different polymer dosages (from Mizani et al., 2013).

Figure 2.14. The measurements of water released from the FFT mixed for different mixing durations (from Mizani et al., 2013).

Mizani et al. (2013) also investigated the rheological behaviour of the fFFT materials after they emerged from the pipelines and were deposited into the deposition cell. Since the polymer amended tailings are especially sensitive to shearing, the mixing intensity and duration (during transportation of fFFT through the pipelines) can disintegrate the flocs, reduce the yield stress, and affect the dewaterability potential. Hence, Mizani et al. (2013) focused on achieving the right balance between deposition and dewaterability so that the lift heights and slopes used during deposition would not prevent the fFFT from attaining their maximum dewaterability potential under field conditions (Mizani et al., 2013). Based on the results from the slump test, stress growth test, and constant stress test (controlled decreasing shear stress test), and the measurements of water released from the

fFFT, the polymer dosage of 850 g/ton (850 ppm) was found to be the optimum dosage for the dewatering of given tailings material in the short-term (Table 2.2) (Mizani et al., 2013).

Table 2.2. Summary of the results from rheological tests performed on the tailings mixed with different polymer dosages (from Mizani et al., 2013).

Dosage (g/ton)	Method of Measurement						
	Slump (Pa)	From Lubrication Theory (Pa)	Stress growth		Decreasing shear stress		Stress Relaxation
			Shear rate (S ⁻¹)	Max stress (Pa)	Starting shear stress (Pa)	Interpreted yield stress (Pa)	Ave Stress (Pa)
FFT	-	-	0.1	28.8	100	10	5.52
			1	28.0			
600	92	60	0.1	169	250	50-100	
			1	207	200	50-100	
725	125	95	0.1	255	450	50-100	
			1	323			
850	154	104	0.1	333	700	50-100	16.7
			1	510			
1,000	163	110	0.1	988	1,000	250	
			1	1,020			
1,200	187	-	0.1	1,000	1,300	-*	
			1	1,180			

Mizani et al. (2017) further investigated the rheology of fFFT by performing amplitude sweep tests, stress growth tests, and controlled stress tests on the samples dosed with 850 ppm of an anionic polymer (A3338). The samples were found to produce shear history-dependent apparent yield stress values. An almost full recovery of the structure following high shear indicated that the destructure under high shear is reversible. The flocs were disintegrated during high shear, thereby reducing the benefits of polymer application; however, the rapid recovery of the structure was able to regain most of the benefits (Mizani et al., 2017).

Although process variables such as polymer and tailings properties, mixing conditions (shear energy and duration) influence the flocculation and dewatering performance of oil sands tailings significantly, there is still relatively little information in the literature on the impacts of such process variables (Motta et al., 2018). Further research is needed to develop and implement advanced and automated process control technologies for optimizing the flocculation processes and improving the dewatering performance so that they could match the industry standards and expectations (Gumfekar et al., 2019).

2.3.2 Dewatering optimization technologies in wastewater treatment

There are similarities between the sludge in wastewater treatment and the fluid fine tailings in oil sands in that they both have similar fine particle sizes, the presence of organic matter, and the relatively high solids contents (Zhu, Tan, and Liu, 2017). Similar to the oil sands industry's challenge with the dewatering and densification of tailings deposits, optimization of conditioning and dewatering of sludge has been a major challenge for the wastewater treatment plants. There are many factors that affect the dewatering performance of sludges, and therefore, it is always difficult to identify an optimum polymer dosage that works for all types of sludges or under all types of operating conditions. Moreover, continuous adjustments to the polymer dosage are required because of the everchanging sludge characteristics (Al Momani and Örmeci, 2014b; Aghamir-Baha, 2014). Nevertheless, the wastewater treatment industry has made considerable progress with the dewatering technologies that use flocculants for sludge thickening and dewatering. By contrast, current flocculation and dewatering technologies that are used for the dewatering of FFT need more improvement from the fundamental and industrial perspectives

(Gumfekar et al., 2019). Hence, it is worthwhile to study the progress that the wastewater treatment industry has made with the development of innovative technologies for the improvement of flocculation performance (Zhu et al., 2017). There is a substantial body of research on new and novel approaches to sludge dewatering, some of which can potentially be adapted to oil sands tailings. For example, the measurement of absorbance of light by the polymers in centrate (sludge supernatant) using the technique of UV-vis spectrophotometry, which has been used for in-line and real-time monitoring of residual polymer concentration in centrate and the determination of optimum polymer dosage based on the measurements of polymer concentration (Gibbons and Örmeci, 2013; Al Momani and Örmeci, 2014a, 2014b; DiMassimo and Örmeci, 2015); and, the measurement of rheological parameters (i.e. peak torque and totalized torque), which has been utilized for describing the effects of polymeric flocculation on the physical characteristics and dewaterability of sludge and used as predictive tools for optimizing the polymer dosage (Örmeci and Abu-Orf, 2004, 2005, 2006; Abu-Orf and Örmeci, 2005; Örmeci, 2007).

2.3.3 UV-vis spectrophotometry

The results from the UV-vis spectrophotometry studies in wastewater treatment demonstrated that the polyacrylamide polymers absorbed light in the 190 ~ 230 nm range of wavelength with the highest absorbance measured at 191.5 nm and the UV-vis spectrophotometer was able to detect the incremental increases in polymer concentrations, which was reflected in the incremental increases in absorbance measurements (Gibbons and Örmeci, 2013; Al Momani and Örmeci, 2014a). A strong linear relationship was established between the polymer concentrations and the measured absorbance values and

a calibration curve was obtained from the linear relationship. Thus, the polymer concentrations in the centrate samples were determined directly by measuring the sample absorbances at a specific wavelength using a UV-vis spectrophotometer (Gibbons and Örmeci, 2013; Al Momani and Örmeci, 2014a). The method was found capable of detecting and measuring polymers even at very low concentrations.

When the polymer dosage was incrementally increased, the absorbances measured from the residual polymer concentrations in the centrate samples exhibited a ‘U’ shaped curve, where the dip corresponded to the optimum polymer dosage (Al Momani and Örmeci, 2014b). In the underdose range, the absorbance measured from the residual polymer in the centrate decreased with the increasing polymer dosage as the polymer added to the sludge was adsorbed increasingly on the sludge particles and solids (Al Momani and Örmeci, 2014b). When the polymer dosage approached an optimum value, more polymers were adsorbed on the particle surfaces and they covered most of the particle surfaces. Subsequently, the rate of further adsorption decreased due to the lack of adequate adsorption sites on the surfaces (Gregory, 1988; Chang et al., 2002). Once polymer dosage exceeded the optimum dosage or the sample became overdosed, absorbance values started to increase again because of the increase in residual polymer concentration in the centrate. The additional polymer, which was not adsorbed on the sample surfaces remained in the sludge water, thereby increasing the residual polymer concentration (Al Momani and Örmeci, 2014b). The optimum polymer dosage determined by the UV-vis spectroscopy was found to be in good agreement with the optimum dosage determined by the conventional dewatering tests such as capillary suction time (CST) test and filtration test

(Figure 2.15) (Al Momani and Örmeci, 2014b). Full-scale testing at three wastewater treatment plants using an in-line UV-vis spectrophotometer confirmed that the optimum polymer dosage could be identified and adjusted as needed by monitoring the lowest centrate or filtrate absorbance at or around 190 nm (DiMassimo and Örmeci, 2015).

Figure 2.15. Relationship between the CST, filtrate volume, and filtrate absorbance at 191.5 nm in the underdose, optimum dosage, and overdose range for the polymer CB 4350 (from Al Momani and Örmeci, 2014b).

Although there are several analytical and chemical methods for the identification and quantification of polymers including gas chromatography (GC), high-performance liquid chromatography (HPLC), mass spectrometry (MS), size exclusion chromatography (SEC), nuclear magnetic resonance (NMR) spectroscopy, and spectrophotometric determination using cationic dyes, these methods are not suitable for the process

optimization as they are not capable of in-line or real-time measurements (Gibbons and Örmeci, 2013; Al Momani and Örmeci, 2014a, Aghamir-Baha, 2014). By contrast, the UV-vis spectroscopy is a relatively simple and sensitive technique that can be used in-line and can provide real-time information on the residual polymer concentrations in the centrate. Such information can be used for correcting or adjusting the concentration of polymer being added to the digested sludge (Gibbons and Örmeci, 2013; Aghamir-Baha, 2014; DiMassimo and Örmeci, 2015). The technique has also been found effective in preventing overdosing of polymer, thereby reducing the operational costs and minimizing the adverse effects on the environment (Murray and Örmeci, 2008; Gibbons and Örmeci, 2013).

2.3.4 Torque rheometry

The results from the laboratory-scale torque rheometry and rheological experiments conducted on flocculated wastewater sludges demonstrated that quantifying the rheological properties, i.e., the degree of deformation in flocculated sludge in response to the applied shearing forces or the ability of the sludge to resist deformation could provide useful information about the strength of the floc network in sludge, and therefore, on the degree of flocculation and sludge dewaterability (Tixier et al., 2003; Murray and Örmeci, 2009). Several studies were conducted to investigate whether specific rheological parameters of the wastewater sludges could be used as a reliable field control parameter for the assessment and optimization of polymer dosage. Based on the test results, two different torque rheology-based methods were proposed for selecting the best performing polymer or the optimum polymer dosage (Abu-Orf and Dentel, 1999; Örmeci, Cho, and Abu-Orf, 2004; Örmeci and Abu-Orf, 2005; Abu-Orf and Örmeci, 2005; Örmeci, 2007).

A torque rheometer named Floccy Tester ODA-10 (Koei Industry Co., Ltd Japan), which was specifically designed to study the flocculation-deflocculation phases of slurries was used to determine the fundamental rheological properties of the flocculated sludge samples. The torque rheometer measures the torque on the mixing impeller, which is produced by the resistance of the flocculated sludge sample to the applied shearing forces. The typical output of a torque rheometer is a torque-time rheogram that depicts the torque-time relationship for the period during which the sludge is sheared (Örmeci et al., 2004). The equipment allows the measurement of peak torque (which is formed in the torque-time rheograms after adding the polymer directly into the samples) (Figure 2.16) and totalized torque (the area under a torque-time rheogram up to a selected torque application time, which reflects the relative strength of the floc network in the sample) (Örmeci, 2007). When the wastewater sludges were dosed with the incrementally increasing polymer dosages and the resulting peak torques and totalized torques were compared, they were observed to increase with the increasing polymer dosages in the underdose range, which was then followed by a local decrease at the optimum polymer dosage (Figure 2.17). Such local decreases were found to correspond to the lowest capillary suction time obtained from the CST tests, which validated the results obtained from the torque rheometry.

Figure 2.16. The torque–time rheograms and the peak torques formed in the rheograms, from the sludge samples conditioned with Clarifloc C-311 polymer (from Örmeci, 2007).

Figure 2.17. The totalized torque values estimated from the rheograms (obtained from the sludge samples conditioned with different polymer dosages) showing a local decrease at the optimum dosage (identified by the CST tests) (from Örmeci and Abu-Orf, 2006).

Full-scale dose-response testing at both laboratory and wastewater treatment facilities demonstrated that both peak torque and totalized torque could be used to identify the optimum polymer dosage for the conditioning and dewatering of wastewater sludge (Abu-Orf and Örmeci, 2005; Örmeci, 2007), and such torque rheology-based methods were found to reduce the polymer consumption by 50%, which resulted in major savings for the treatment plants (Örmeci, 2007).

2.4 Viscoelastic properties of flocculated tailings and ageing effect in tailings

After the step of polymeric flocculation, a key process in the tailings management involves the transportation of flocculated tailings through the pipelines and the systematic deposition of flocculated tailings into some containment cells or dedicated disposal areas. In conformance with the characteristics of non-Newtonian fluid behaviour, when subjected to high shear stress that exceeds the yield stress, the tailings materials strain irreversibly and start to flow continuously; however, at low shear stress, they tend to stop. Polymer amended FFT materials are especially sensitive to shearing. The mixing intensity and the duration of mixing and the shear caused during transportation have the potential for disintegrating the flocs and influencing the flow behaviour. When flocculated tailings are continuously sheared at high shear rates, the floc structures are destroyed, and subsequently, the apparent viscosity and yield stress as well as dewatering potential are reduced (Mizani et al., 2013, 2017). Contrary to such a ‘shear thinning’ phenomenon, the ageing effect that occurs in fFFT (when they are sitting idle in the pipelines or subjected to relatively low shear stresses) helps the materials in recovering their structure. After a period of rest, the fFFT materials start to regain their original structure and stiffness through the

restructuring of the particle network system (Soleimani et al., 2013; Mizani et al., 2014, 2017). Because of such a time-dependent ageing effect, even highly sheared fFFT materials were found to demonstrate significant yield stress (Simms et al., 2013).

When the tailings are being pumped or transported through the pipelines, they need to possess the liquid-like flow behaviour. However, when they are deposited, the flow needs to come to a stop so that the stacking can occur. While the tailings go through this transition between the liquid-like behaviour and solid-like behaviour, they exhibit both elastic and viscous behaviour or the viscoelasticity. Exceeding the yield stress is the point where this transition from elastic to viscous behaviour occurs (Mizani, 2010). The viscoelastic properties of the tailings material (such as elastic modulus or G' and viscous modulus or G'') can be determined as a function of the oscillatory stress from the oscillatory rheometry tests (Nasser and James, 2008). The elastic modulus (G') can be described as:

$$G' = \frac{\tau_c}{\gamma_c} \dots \dots \dots (2.1)$$

Where τ_c = shear stress, and γ_c = shear strain.

Elastic or small strain shear modulus (G'), also known as rigidity modulus or storage modulus represents the elastic behaviour corresponding to a small, temporary, and reversible deformation. When a linearly elastic material is subjected to a small strain shear stress, elastic deformation changes the shape of the material within its elastic limit. However, when the externally applied force is removed, the material regains its original shape or configuration. The maximum G' measured in the small strain region is called elastic shear stiffness (G_{max}) (Sorensen, 2006).

Soils, especially the fine-grained soils, show an elastic behaviour at low stress levels as the strain increases linearly with the stress applied. However, once a certain stress level is exceeded, the soil reaches a plastic state, where the strain increases continuously and the deformation becomes irreversible (Sorensen, 2006; Das, 2008). Various factors including soil composition, soil fabric, soil density, soil stress-strain history, chemical alterations of soils, current state, drainage, and strain rates have been found to influence the magnitude of the elastic shear stiffness in soils (Jardine, 1992). It has been observed that naturally structured clays show a higher G_{max} in comparison to the reconstituted natural clays at the same void ratio due to an ageing effect in undisturbed natural clays (Shibuya, 2000). The range of the linear elastic zone, which reflects the degree of resistance of the soils to an externally applied force has also been found to vary in soils depending on their different fabrics and interparticle bonding. Relatively softer or less stiff soils such as reconstituted clays show a relatively smaller range of linear elastic zone in comparison to the naturally structured clays (Jardine, 1992).

Figure 2.18. The change in G' with time during creep (from Anderson and Stoke, 1978).

Anderson and Stoke (1978) investigated the change in elastic shear modulus of a kaolinite clay sample under a constant confining pressure during consolidation (Figure 2.18). During primary consolidation, the shear modulus increased rapidly. However, after the complete dissipation of excess pore water pressure (i.e. after the end of primary

consolidation), during the creep period, the elastic shear modulus was observed to increase linearly with the logarithm of time. The change in elastic shear modulus during creep compression solely due to a decrease in the void ratio was estimated theoretically using an empirical formula proposed by Hardin and Black (1968) (Soga, 2005):

$$G = A \frac{(2.97-e)^2}{1+e} p'^{0.5} \dots\dots\dots (2.2)$$

Where A = a material constant; e = void ratio; and, p' = mean effective stress.

When compared, Anderson and Stoke (1978) found the increase in experimentally determined elastic shear modulus considerably greater than the increase in theoretically estimated elastic shear modulus (which was solely due to a reduction in the void ratio) and they attributed the additional increase in elastic shear stiffness during creep compression (the part of the experimentally determined elastic shear stiffness that could not be explained by a decrease in void ratio) to a stronger physical-chemical bonding between the particles.

When Lohani et al. (2001) conducted one-dimensional compression tests on reconstituted clays and subjected them to a sustained loading at different stress levels for different periods under a drained condition, the elastic shear modulus was found to increase with time and such a disproportionate increase in elastic shear modulus relative to the reduction in void ratio during creep compression was attributed to the inherent ageing effects such as bonding (Lohani et al., 2001).

Recently, Mizani (2016) used an Anton Paar Physica MCR301 model rheometer with a vane fixture for conducting oscillatory rheometry tests including amplitude sweep

tests and stress growth tests on the flocculated FFT samples prepared with different polymer dosages and also on the flocculated FFT samples of different ages for studying their deformation/destruction behaviour under externally applied loads or shears and reflocculation behaviour under the resting condition (which is an ageing effect).

Amplitude sweep tests are typically conducted on high viscosity materials such as soils and non-Newtonian fluids for determining the viscoelastic properties and also for examining the role of structure and water content on the viscoelastic behaviour, especially in the linear viscoelastic zone, where the material shows a predominantly linear elastic behaviour (i.e. reversible deformation) (Markgraf et al. 2006). During the amplitude sweep test, the sample is sheared at a constant oscillation frequency with the shear stress or shear strain increasing and the material shows a transgression from the elastic to plastic or viscous state as the material deforms irreversibly (Markgraf, 2006). A typical amplitude sweep test result indicates the range of linear viscoelastic behaviour of tailings (Figure 2.19). Within this range, typically, both G' and G'' remain almost unchanged with the G' having a larger value than the G'' , which implies that the sample structure is relatively undisturbed, and the behaviour of the material is largely elastic. This region is called linear viscoelastic range (LVE) and the plateau value of G' in the LVE region denotes the rigidity of the sample. Once the linear viscoelastic range is exceeded, both G' and G'' values start to change, and subsequently, the structure starts to degrade. Once the applied shear stress exceeds the yield stress, the value of G' drops sharply and becomes smaller than the G'' . The intersection of G' and G'' reflects the yielding point. Once that point is reached, the material starts to display viscous behaviour (Mizani et al., 2017).

Figure 2.19. A typical amplitude sweep test result showing variable deformation at a constant oscillation frequency with shear stress increasing with each oscillation.

Mizani et al. (2017) performed the amplitude sweep tests on the FFT samples dosed with 850 ppm of anionic polymer using three different oscillation frequency (10 Hz, 5 Hz, and 1.60 Hz) (Figure 2.20). The linear viscoelastic limit of the material was measured 50 Pa based on the oscillatory measurements. Irrespective of the oscillation frequency applied, at smaller stress values such as below 50 Pa, G' values remained unchanged as the oscillations of the vane were not able to deform the structure of the material beyond its elastic limit. However, beyond 50 Pa, the structure of the material started to degrade. Depending on the oscillation frequency applied, the material yielded completely at around 300 ~ 500 Pa. Such yielding behaviour was also evident in the greater value of G'' in comparison to the G' in that stress range and also in the predominantly viscous behaviour

displayed by the material. Higher oscillation frequency resulted in lower yield stress for the fFFT materials (Mizani et al., 2017).

Figure 2.20. The results obtained from the amplitude sweep tests performed on fFFT showing the linear viscoelastic regions at 3 different frequencies (from Mizani et al., 2017).

Mizani (2016) conducted three interval thixotropy tests (3ITT) on the fFFT samples for investigating the recovery of the original structure of polymer amended tailings after shearing. A combination of amplitude sweep tests (where the maximum strain/stress was increased at each cycle while maintaining a constant oscillation frequency) and stress growth tests (where the sample was sheared at a constant shear rate) was used to measure the ‘structure breakdown’ (under shear) and ‘structure recovery’ (at rest) in terms of increase/decrease in the elastic modulus before, during, and after shearing (Figure 2.21).

Figure 2.21. A typical three interval thixotropy test (3ITT) results showing the recovery of the original structure of the fFFT after shearing.

The baseline rheology (in terms of elastic modulus G') of the fFFT material was determined by applying an oscillating stress (10 Pa for 5 minutes) that was well below the linear viscoelastic range so that the material would undergo elastic deformation only. Then, the material was sheared at constant stress (500 Pa) for a short period (5 minutes). Then, an oscillating stress was applied again to the material (using the same frequency and the same amplitude as the first step: 10 Pa for 5 minutes). The thixotropic recovery of the sample structure was determined as the ratio of finally measured elastic modulus G' (measured after a resting period) to the initially measured elastic modulus G' (measured as baseline rheology) (Mizani, 2016).

Since the elastic modulus (G') of the FFT material is correlated to its fabric or structure, Mizani et al. (2017) found the measurements of elastic modulus from the oscillatory rheometry tests useful in investigating the decrease in material stiffness in response to the small strain shear stresses (destruction) or the increase in material stiffness under the resting condition due to the ageing effect (recovery of structure or reconstruction). The measurements of elastic modulus from the oscillatory rheometry tests also showed that the FFT materials were able to reflocculate at lower shear rates (after the structure had been destroyed at a high shear rate). Hence, the fFFT materials were observed to develop a shear history-dependent apparent yield stress (Mizani et al., 2017).

The materials were also found to develop time-dependent yield stress at the low stress levels. At low stresses, the material behaviour was dependent on the material structure and the material initially showed resistance to the shear. However, when the material was sheared at the same rate for a sustained period, the material eventually reached a time-dependent yield stress, where it started to flow. Hence, the behaviour of the fFFT material was a function of both ageing and destruction due to shearing (Mizani and Simms, 2016).

2.5 Geotechnical perspectives on clay behaviour

2.5.1 Creep in natural clay deposits

Creep or secondary compression refers to the time-dependent deformation in soils, which continues even after the excess pore pressure has been substantially dissipated. Creep compression occurs at practically constant effective stress (Robinson, 1999). In high

void ratio materials such as tailings, hydraulic conductivity-void ratio relationship is the property that dictates the rate and magnitude of settlement during primary consolidation (Suthaker and Scott, 1994; Gholami, 2014). While the excess pore water pressure dissipates, the tailings increase in effective stress and subsequently go through a deformation, and therefore, the void ratio changes. During creep or secondary compression, the hydraulic conductivity of the material still governs the rate of flow of water out of a deposit, however, the rate of change of effective stress is also affected by the viscous deformation of the soil structure (Soga, 2005; Sorensen, 2006). The resulting settlement or the change in volume is called volumetric creep compression or secondary consolidation (Soga, 1994). Although creep compression or secondary consolidation is thought to be a small fraction of primary consolidation, in those soft soils or clays that demonstrate considerable primary consolidation, viscous creep should be taken into account as an important deformation mechanism for determining the long-term settlement (Fatahi et al., 2013).

When Crawford and Bozozuk (1990) examined the long-term settlement behaviour of a sensitive, highly structured, normally consolidated Champlain clay in Ottawa under full-scale loading, they found evidence of secondary consolidation. In 1954, the ‘Accommodation Building’, a relatively light structure was built on a very compressible clay layer and the resulting settlement behaviour was recorded over the next 33 years. When the clay layer was subjected to a surface load (during construction and post-construction), the vertical effective stresses in the ground (obtained from in-situ measurements at different locations) increased to the preconsolidation stress level (59 ~ 60

kPa, measured in the laboratory) within a relatively short period (<2 years). Afterwards, the effective stresses remained at the same level as the laboratory-measured preconsolidation stress for a long period (>30 years); however, the consolidation continued throughout the period (Figure 2.22). The settlements measured in the field were found to differ considerably from the predictions of consolidation settlement from the laboratory tests (Figure 2.23) (Crawford and Bozozuk, 1990). Thus, the long-term study of the settlement behaviour of Champlain clays demonstrated that such sensitive clays could undergo consolidation for many decades under constant effective stresses, and such settlement behaviour could make predicting the consolidation settlements from the laboratory tests challenging.

Figure 2.22. The measurements of settlements at two different locations at the building site (from Crawford and Bozozuk, 1990).

Figure 2.23. Comparison between the compression curve measured in the field (that shows the relationship between the maximum vertical effective stresses and percent compression) and the average curve of compression under incremental increases of stress obtained from the laboratory tests (from Crawford and Bozozuk, 1990).

In an experimental study, Robinson (1999) investigated the compression behaviour of 4 different clays using a fixed-ring-type consolidometer, where the bottom porous stone was connected to a pressure transducer for measuring the excess pore water pressure during consolidation. Figures 2.24a and 2.24b show the typical time-compression curves obtained from the clays in the pressure range of 100 ~ 200 kPa and the corresponding time-pore water pressure data respectively. The pore water pressure measurements indicated the end of primary consolidation after a consolidation period of 600 minutes (approximately). However, the consolidation continued even after the complete dissipation of the excess pore water pressure, which was the secondary consolidation or creep portion of consolidation (Robinson, 1999).

Figure 2.24. (a) Typical time-compression curves for the clays; and, (b) the corresponding time-pore water pressure curves (from Robinson, 1999).

Although there have been many types of research on the creep behaviour of clays, the creep mechanisms and the factors influencing the creep in clays are not well understood yet owing to the complex structure of clay matrix (Varatharajan, 2011). There are many possible explanations for the creep mechanisms (Le, Fatahi, and Khabbaz, 2012). The mechanism of creep compression could be a combination of different processes, both at the microstructural and macrostructural levels (Le et al., 2012). Yin (2003) attributed the creep behaviour of soil to a combination of two processes including viscous flow of adsorbed water in the double layers on the clay particles (Bjerrum, 1967; Graham and Yin, 2001) and the viscous, irrecoverable rearrangement or adjustment of the clay particles or the soil structure (Yin, 2003; Fatahi et al., 2013; Le et al., 2015). From a macroscopic point of view, the creep compression occurs when a structural rearrangement or adjustment takes place in clays through the breakdown of the interparticle bonds and/or relative movement or rearrangement of the particles (in order to reach a new equilibrium state under an applied stress or to settle into a more stable condition) (Taylor, 1942; Soga, 1994; Le et al., 2012). On the other hand, the deformation of clay microstructure, which occurs due to the structural viscosity of the water within the clay microstructure explains the creep deformation from the microstructural point of view (Le et al., 2012).

Basically, it is the electrochemical properties of clay mineralogy that lead to the viscous flow of water within the clay microstructure, which results in a deformation of the clay microstructure units. Such rearrangement of the clay particles also results in a plastic structural resistance, which is known as structural viscosity and is thought to resist the relative movement between the clay particles and provide more stability to the clay

structure against further compression owing to an increase in the number of contacts between the clay particles (Bjerrum, 1967; Le et al., 2012; Le et al., 2015). That's why Mitchell (1993) defined creep as the time-dependent volumetric strains and/or shear strains that develop at a rate controlled by the viscous resistance of the soil structure. Such a viscous effect can be quantified through the magnitude of creep strain at fixed stress or the increase in shear stress due to the increase in strain rate. The secondary consolidation is a special case of creep compression that is also governed by the viscous resistance (Soga, 1994; Suthaker, 1997).

The theory and the equations for the analysis of one-dimensional consolidation of soils were presented by Terzaghi (1943). However, many of the assumptions, upon which Terzaghi's one-dimensional consolidation theory was based do not reflect the actual material behaviour observed in the field. Terzaghi (1943) assumed that hydraulic conductivity (k) is constant and the relationship between the void ratio and effective stress is linear and independent of time. However, practically, the relationship between the void ratio and effective stress in fine-grained soils or clays is time-dependent and strain-rate dependent. Hence, Terzaghi's one-dimensional consolidation theory is not applicable to the consolidation settlement calculation of clayey soils with creep (Yin and Feng, 2016).

So far, several improved methods have been proposed for the calculation of consolidation settlement of clays by incorporating creep. These methods can be divided into two broad categories based on the assumptions made while formulating them: (a) Hypothesis A; and, (b) Hypothesis B (Yin and Feng, 2016). The method based on Hypothesis A assumes that no creep compression occurs during the primary consolidation

period and it occurs only when the excess pore water pressure is completely dissipated and the secondary compression period is started (Yin and Feng, 2016). The creep compression that occurs during the secondary compression period can be expressed in terms of secondary compression coefficient or creep coefficient (Rowe, 2012; Yin and Feng, 2016):

$$\Delta e_{creep} = C_{ae}(\Delta \log t) \dots\dots\dots (2.3)$$

Where C_{ae} = creep coefficient obtained from the slope of the straight-line portion of the compressibility curve representing the secondary compression period; Δe_{creep} = creep compression that occurs during the secondary compression period; and, t = loading period.

The secondary compression behaviour of clays is typically explained and predicted by the C_{ae}/C_c law of compressibility, where C_c is the compression index (Mesri and Castro, 1987). The law assumes C_{ae}/C_c to be a constant for any soil during the secondary compression (Funk, 2013):

“There is a unique interrelationship between the time compressibility and the effective stress compressibility that holds true throughout the secondary consolidation stage.”

The typical values of C_{ae}/C_c for most of the inorganic soft clays are 0.04 ± 0.01 , and 0.05 ± 0.01 for the highly organic plastic clays (Mesri and Castro, 1987). When the C_{ae}/C_c law of compressibility is coupled with the unique end-of-primary void ratio-effective stress relationship (Mesri and Choi, 1985) (Figure 2.25), they present a useful approach for the determination of the consolidation settlement of clayey soils with creep (Watabe and Leroueil, 2015).

Figure 2.25. The secondary compression coefficient determined from the void ratio - log (time) curve (from Yin and Feng, 2016).

The method based on Hypothesis B involves a coupled consolidation analysis that uses the constitutive relationship for the time-dependent stress-strain behaviour of clays including creep compression during the primary consolidation period (Yin and Feng, 2016). Hypothesis B assumes that the void ratio is non-unique and is affected by the thickness of the soil layer and drainage conditions (Yin and Feng, 2016). A wide range of laboratory measurements and field observations indicated that excess pore water pressure dissipation and creep compression occur simultaneously (Figure 2.26) (Bjerrum, 1967; Degago et al., 2009; Watabe et al., 2012). The creep properties of soil are also influenced by the effective stress developed in the soil; as effective stress increases with depth and time, the creep related parameters such as creep coefficients and creep strain rates decrease (Le and Fatahi, 2014). The method based on Hypothesis B requires a numerical method for solving a set of non-linear partial differential equations (Yin and Feng, 2016).

Figure 2.26. The concept of ‘instant’ and ‘delayed’ compression by Bjerrum (1967) (from Bjerrum, 1967).

Figure 2.26 presents the concept of ‘instant’ and ‘delayed’ compression by Bjerrum (1967) (which was based upon the behaviour of clay structure in response to an increase in the effective stresses) as compared to the conventional concept of ‘primary’ and ‘secondary’ consolidation (which divides the compression into two components occurring before and after the dissipation of excess pore pressures) and shows that the creep or secondary compression could occur during the period of excess pore water pressure dissipation under constant effective stress (Bjerrum, 1967).

For characterizing the creep compression in clays under one-dimensional straining condition, Bjerrum (1967) suggested that the compression of clay sediment could be divided into two components: (a) an instantaneous compression; and, (b) a delayed compression or creep. When the clay is subjected to a loading, such an increase in effective stress is instantaneously transferred to the clay skeleton, even before the onset of drainage, and results in a volume change. Such volume change is called an instantaneous compression, and when plotted, such compression behaviour deviates from the compression behaviour displayed by the experimental curve theoretically constructed in the early stages of consolidation. The delayed compression is solely caused by the effect of creep, where soil volume changes over time under constant vertical effective stress (Bjerrum, 1967; Robinson, 1999; Jeeravipoolvarn et al., 2009). Recently, Carrier (2017) suggested that a substantial amount of the creep observed in a soft soil or clay sample can be attributed to a delayed consolidation in parts of that test specimen, specifically for those samples with low hydraulic conductivity (Carrier, 2017).

2.5.2 Modelling creep compression

Various numerical models have been developed for reproducing complex soil behaviour including creep phenomena. Among them, advanced time-dependent specific constitutive models such as Soft Soil Creep (SSC) model or Elastic Visco-Plastic (EVP) model are suitable for the soft soils that display time-dependent stress-strain behaviour (Waterman and Broere, 2004; Kaczmarek and Dobak, 2017). The SSC model has been founded on the modified Cam-Clay model. The model also includes some aspects of the viscous behaviour of soil under compression as it considers the logarithmic creep law for

secondary compression. The total volumetric strain in the SSC model is estimated as the summation of the elastic and viscoplastic strains (Fatahi et al., 2013) and the model assumes that the elastic strains in the soil are instantaneous, whereas viscoplastic strains develop over time. Various time-dependent features of soil including time-dependent compression, preconsolidation stress, and stress-dependency of stiffness are considered by the SSC model (Waterman and Broere, 2004, 2005). However, one limitation with the SSC model is that it assumes a constant creep rate with time and stress level, which may result in an overestimation and unrealistic prediction of the long-term consolidation of the soil deposits. Creep rate is thought to decrease with time by several researchers (Yin, 1999; Fatahi et al., 2013).

The original Elastic Visco-Plastic (EVP) model, which was proposed by Yin and Graham (1989) was based on the framework of the modified Cam-clay model and the constitutive model considered the influence of effective stress and strain rates on the time-dependent compression behaviour of clays (Fatahi et al., 2013). The model was basically adopted from and expanded on the concepts of instant and delayed compression and the timeline model, which was proposed by Bjerrum (1967) for considering the creep compression under one-dimensional straining condition. Because of the time-dependent nature of creep, a single $e-\log \sigma_v'$ curve or compressibility curve would not be able to describe the compressibility characteristic of a clay that exhibits a delayed compression. Hence, for describing the compressibility characteristic of such clays, Bjerrum (1967) suggested using a set of compressibility curves where each curve would represent the settlement response to a specific time of sustained loading (Jeeravipoolvarn, 2005).

Yin and Graham (1989) introduced new concepts such as ‘equivalent timeline’, ‘instant timeline’, ‘limit timeline’ and ‘reference timeline’, and by using these concepts in combination with a linear logarithmic creep function, they formulated a one-dimensional EVP constitutive model for explaining the time-dependent stress-strain behaviour of clays (Yin and Graham, 1989). Since the application of a linear logarithmic creep function could lead to an infinite creep strain with the time approaching infinity, Yin (1999) presented a non-linear creep function that includes a creep strain limit and a creep coefficient (Le et al., 2015). Over the years, further modifications and improvements have been made to the EVP model for the application under different stress-strain and boundary conditions and also for the analysis under three-dimensional conditions (Yin, Zhu, and Graham, 2002; Fatahi et al., 2013; Le et al., 2015). However, determining the creep parameters for applying to the EVP model remains a challenging task (Le and Fatahi, 2016).

2.5.3 Case studies of creep in natural clay deposits/slurries

Because of the creep effect, many natural clays show deformation behaviour even under a constant load or effective stress. When the engineering applications involve determining the long-term settlement of the large structures on natural clays such as the construction of Kansai International Airport in Japan, they require careful consideration of the rate-dependent compression behaviour of natural clays (Qi et al., 2018). The Kansai International Airport was constructed 5 km off the coast of Senshu on two artificial islands built in 18~20-metre-deep seawater in Osaka Bay (Mesri and Funk, 2014). When the construction started, as a part of the original design considerations, the islands were predicted to settle evenly over a 50-year period before stabilizing at 4-metre above the sea

level. However, within six years, the islands were found to settle more than that threshold in many places. The actual settlements were found to exceed the laboratory estimates of the rate of consolidation, which were estimated based on the end-of-primary (EOP) void ratio-effective vertical stress relationship of clays (Mesri and Funk, 2014).

The discrepancies between the predicted and actual settlements in the Osaka Bay Pleistocene clay prompted the researchers to study the long-term consolidation behaviour of soft clays using different approaches and estimate the secondary consolidation with high accuracy for determining the long-term settlement of large-scale structures on natural clays. Several researchers attributed the unusual settlement behaviour of Osaka Bay clay to a continuing creep or secondary consolidation at a load smaller than the preconsolidation pressure (Imai et al., 2005; Kobayashi et al., 2005; Oda et al., 2005; Tanaka, 2005). For explaining the long-term consolidation characteristics of soft clays such as Osaka Bay clay, Watabe et al. (2008) proposed a simplified method based on the isotache concept (which involves viscosity and considers the effect of the strain rate on the compression characteristics) using a reference compression curve (obtained from the constant rate of strain one-dimensional consolidation tests) and the relationship between the strain rate and the preconsolidation pressure corresponding to that strain rate (obtained from the long-term consolidation tests under a constant applied stress). Watabe et al. (2008) presented a simple relationship to relate the apparent preconsolidation pressure and strain rate:

$$\ln \frac{p'_c - p'_{cL}}{p'_{cL}} = c_1 + c_2 \epsilon_{vp} \dots \dots \dots (2.4)$$

Where p'_{cL} = a reference preconsolidation pressure; p'_c = actual preconsolidation pressure (a function of strain rate); $\dot{\epsilon}_{vp}$ = strain rate; and, c_1 and c_2 are the fitting coefficients.

Later, Watabe et al. (2012) analyzed the data on clay compressibility available in the literature and applied the proposed method to worldwide natural clays with different characteristics for characterizing their long-term consolidation behaviour including creep compression. The shift in apparent compressibility of a natural clay because of its strain rate-dependent compression behaviour was found relatively well-constrained in natural clays, i.e., between 0.7 and 1.5 of a reference preconsolidation pressure.

Figure 2.27. Variation in apparent preconsolidation pressure with strain rate in large data set on clay compressibility (from Watabe et al., 2012).

2.5.4 Structuration/ageing effect in natural clays

Many natural clay deposits have been observed to demonstrate characteristics and behaviour different than that of the reconstituted or remoulded samples prepared from the same clays and such differences have been attributed to a bonded soil structure developed within the clay deposits over a long period of time (Leroueil and Vaughan, 1990). Both geological and stress histories of the clayey soils have contributed to the formation of such structure through a combination of extremely complex, time-dependent processes involving various chemical, thermal, mechanical, and environmental factors (Liu and Carter, 1999). Although it is difficult to describe the origin and evolution of soil structure with absolute accuracy, there exist many experimental data in the literature concerning structuration or ageing effect in natural clays (Burland, 1990, Leroueil and Vaughan, 1990).

For soils, the term ‘structure’ refers to the combination of ‘fabric’ (the arrangement of the soil particles) and ‘bonding’ (interparticle forces) (Mitchell, 1976) and they both play a role in the structuration effect in clays (Burland, 1990). Crawford (1968) referred the term ‘structure’ in a clay to “*a complex space frame of solid particles held together by plastic and rigid bonds*” and the term ‘fabric’ to “*the size, shape and geometric arrangement of particles, ranging from a parallel to a random or card house orientation*”. The effect of structuration on the soil is typically manifested in a change in the arrangement of the particles and interparticle bonding, and subsequently, in an increase in apparent preconsolidation pressure (yield stress) (Liu and Carter, 1999). Hence, the structuration is the macroscopic effect of rearrangement of the microstructural elements in soils such as particles and aggregates and the subsequent bonding between them (Delage et al., 2010).

Locat and Lefebvre (1986) have defined the structuration in the soil as the amount of strength or stiffness that the soil has gained through a combination of time-dependent processes in the soil such as thixotropy or cementation. Bjerrum (1967) attributed the structuration effect in normally consolidated clays to the delayed or secondary consolidation during sustained loading, which is typically manifested in the time-dependency of the stress-compressibility relationships. In such clays, the yield stress often increases to a point that is past beyond the maximum past effective overburden pressure (p_c) (Bjerrum, 1967). Based on the observations of creep in soft clays that occurs during one-dimensional compression tests, Bjerrum (1967) proposed the concept of isochrones or timelines:

“if a normally consolidated sample is allowed to creep for a period of time and then subsequently loaded, the sample will be able to sustain a load in excess of the preconsolidation pressure without causing significant volumetric changes”.

Bjerrum (1967) proposed the ‘timeline’ concept to explain the delayed compression at constant effective stress in natural clay deposits, model the reduced creep rates resulting from the increased duration of loading, and describe the apparent preconsolidation pressure and overconsolidation ratio (OCR) that originate from the geological ageing effect (Yin and Graham, 1989; Suhonen, 2010). A plastic clay sample was consolidated in an oedometer by loading it in steps and then the load was sustained for 28 days (Figure 2.28). When the sample was loaded again with additional loads in steps, it was found to demonstrate a 25% increase in critical pressure or apparent preconsolidation pressure. The increase in apparent preconsolidation pressure, which was caused by the delayed

compression or secondary consolidation was an ageing effect and such an ageing effect influenced the overall compressibility of the sample (Rajot, 1992).

Figure 2.28. The results from the oedometer test on the plastic clay in Drammen (Bjerrum, 1967) showing the development of reserve resistance against further compression during 28 days of delayed compression under sustained loading conditions (from Rajot, 1992).

Leonards and Deschamps (1995) attributed the structuration/ageing effect in clays to a number of factors including (a) alterations of clay minerals; (b) change in pore water chemistry; (c) cementation; (d) mineral leaching or internal erosion; and, (e) a combination of time effect/volumetric creep strains at a constant effective stress. Depending on the conditions, the above-mentioned factors may have different degrees of influence on the structuration effect in clays; however, the combined effect of time and volumetric creep

compression has been considered to be the principal mechanism in the stiffer arrangement of the clay particles and the development of an apparent or quasi-preconsolidation pressure (Graham, Crooks, and Bell, 1983; Leonards and Deschamps, 1995). The structuration behaviour observed in natural clay deposits has mostly been attributed to the inherent ageing effects such as thixotropy, interparticle bonding, and cementation, whereas the natural creep process that arises from the viscous effect in clays has been considered as a minor contributing factor (Locat et al., 2003; Sorensen, 2006).

When a 'structured' soil is strained beyond a certain limit either by subjecting it to a vertical loading or by shearing it, it experiences a structural degradation and becomes 'destructured'. The same effect can be achieved through the process of remoulding, where the soil is disturbed to an extent that its natural internal structure becomes altered, shear strength decreases, and the soil gains compressibility (Leroueil and Vaughan, 1990). The influence of structuration on clays can be clearly seen when the properties of a structured clay are compared with the properties measured or inferred for the same clay in its disturbed state. Hence, Burland (1990) proposed using the compressibility and strength characteristics of reconstituted clays (the clays which has been thoroughly mixed at a water content equal to or greater than the original liquid limit) as a framework for interpreting the corresponding properties of natural clays so that the influence of time-dependent effects such as structuration/ageing could be determined (Burland, 1990).

A comparison of the compression tests performed on the naturally structured clay and the reconstituted clay prepared from same natural clay showed that compressibility curves obtained from the natural clay samples had a higher void ratio than that of the

reconstituted clay samples at the same effective stress, which was due to the structuration/ageing effect (Figure 2.29) (Locat and Lefebvre, 1985; Leroueil and Vaughan, 1990; Burland, 1990; Liu and Carter, 1999; Delage, 2010). Naturally structured clays also develop an undrained shear strength greater than the strength of the corresponding reconstituted/remoulded samples at the same void ratio or density (Bishop, Webb, and Lewin, 1965).

Figure 2.29. A comparison of compression behaviour showed by the structured soil and reconstituted/remoulded soil at the same effective stress (from Leroueil & Vaughan, 1990).

In Figure 2.29, the void ratio-stress space shown in stripes (outside the line demarcating the void ratio-effective stress area or the compression behaviour of reconstituted/remoulded soil) is denoted as 'structure permitted space' (Leroueil and Vaughan, 1990). Such an additional void ratio area (the striped area) indicates the limit to which the structured soil can demonstrate an additional stiffness in comparison to the

reconstituted/remoulded soil at the same stress level (which in turn results in less compressibility). This additional void ratio area is also an indicator of the extent to which the soil is structured. Such a degree of structuration also reflects the strength of the soil, which in turn determines how much stress that soil will endure before it starts to yield (Leroueil and Vaughan, 1990). Once the apparent preconsolidation pressure (yield stress) is exceeded, the structured soil starts showing relatively large compressibility and compression index (C_c) due to the structural breakdown. Also, the soil shows a relatively larger secondary compression for the undisturbed, naturally structured soil samples (Soga, 1994).

Liu and Carter (1999) identified three distinctive behaviours of structured soils where they clearly differ from the behaviour of reconstituted/remoulded samples prepared from the same soils:

- (a) The structured soil has a higher preconsolidation pressure in comparison to the corresponding reconstituted or remoulded soil, which makes the structured soil material stiffer in comparison to the corresponding reconstituted/remoulded soil, especially at relatively low stress levels. Thus, a comparison between the compression behaviour of the structured soil and the reconstituted/remoulded soil clearly shows the influence of structuration on the soil.
- (b) The structured soil can sustain an additional void ratio in comparison to the corresponding reconstituted/remoulded soil, which is also evident in the comparison between the compression behaviour of the structured soil and the corresponding reconstituted/remoulded soil.

(c) When the structured soil starts to yield during the consolidation test, it becomes more compressible in comparison to the corresponding reconstituted/remoulded soil. However, as destructuring of an initially structured sample progresses under the influence of increasing compression pressure, its compression behaviour gradually becomes similar to the compression behaviour of the reconstituted/remoulded soil. When destructuring is completed and the structure is destroyed, the behaviour of the two samples becomes identical (Liu and Carter, 1999).

During the destructuring process, once the vertical effective stress exceeded the preconsolidation pressure, plastic deformation is induced into the sample, and subsequently, the additional void ratio that has been sustained initially by the structured soil (before destructuring has started) starts to diminish. Although such plastic deformation is typically irrecoverable, the destructured soil may again recover its structure over time because of the chemical changes such as interparticle bonding (Liu and Carter, 1999). The recovery of the structure in reconstituted/remoulded clays with time is another interesting feature of the structuration/ageing effect in clays (Leroueil and Vaughan, 1990). Even when the soil structure is destroyed by loading and straining, the initial structure of the soil may recover to a good extent over time due to the structuration/ageing effect.

2.5.5 Structuration effect observed in clays in laboratory-scale tests

When Leonards and Ramiah (1959) investigated the influence of ageing on the one-dimensional compression behaviour of a reconstituted residual clay by testing the samples in standard and Lucite oedometers and by allowing the consolidated samples to rest for a period under different creep conditions (creep permitted/prevented), the results showed that

the samples developed resistance to the compression force while they were ageing owing to the time-dependent changes in the fabric or microstructure of the clay and such resistance was independent of the change in volume due to the creep compression (Leonards and Ramiah, 1959). When the aged samples were loaded again to a critical pressure such that the structural resistance was broken, Leonards and Ramiah (1959) termed that critical pressure as the apparent or quasi-preconsolidation pressure. Such an increase in preconsolidation pressure, which was greater than what was expected from a change in volume due to the secondary or creep compression showed the effect of structuration separately from the effect of creep compression (Leroueil and Vaughan, 1990).

Leonards and Altschaeffl (1964) loaded a reconstituted soil sample to 49 kPa in an oedometer and allowed the sample to rest at constant pressure for 90 days. When the sample was loaded again after 90 days of secondary compression, the preconsolidation pressure was increased to 61 kPa. Leonards and Altschaeffl (1964) postulated that it would not be possible to explain such an increase in preconsolidation pressure solely in terms of a decrease in the void ratio that occurred during the sustained compression period of 90 days. Rather, it was the structuration behaviour of soil, which is typically associated with the development of bonds between the particles produced such a high preconsolidation pressure (Leonards and Altschaeffl, 1964; Augustesen, Liingaard, and Lade, 2004).

Leroueil (1996) subjected a reconstituted Jonquiere clay sample to a vertical effective stress of 10 kPa and allowed the sample to consolidate under a constant compression pressure of 10 kPa for 120 days. After 120 days, when the sample was subjected to further compression pressure under increasing vertical effective stress, the

preconsolidation pressure was measured 18.5 kPa (Figure 2.30). If only the change in void ratio during creep were considered, the preconsolidation pressure would have been 11.5 kPa after 120 days. So, there were indications that the reconstituted soil sample developed a structure during the 120-day period of constant effective stress and such structuration effect caused an increase in apparent preconsolidation pressure (Leroueil, 1996).

Figure 2.30. The compression curve from Leroueil (1996) showing the structuration behaviour demonstrated by a reconstituted Jonquiere clay sample during the secondary compression period (from Augustesen, Liingaard, and Lade, 2004).

Similar to natural clays, the structuration effect has also been observed and reported in dredged clays. In fact, the dredged sediments are more similar to oil sands tailings deposits than the clay sediments since they also can stabilize at water contents much larger than their liquid limits. The dredged clays, which are typically removed from the riverbed and deposited into the disposal areas or used for the construction of reclaimed lands have

high water content and compressibility and very low shear strength (Shahriar et al., 2018). Zeng et al. (2016) studied the influence of ageing effect on the compression curves in dredged sediments from China. One-dimensional consolidation tests were conducted on three dredged clays with and without ageing and the corresponding changes in time-dependent vertical yield stresses were examined. The dredged clays with ageing behaved differently from those without ageing as they developed significant vertical yield stresses (or apparent preconsolidation pressures) under fixed consolidation loads and showed changes in the compression curve, and subsequently, structural resistance to the compression force (Figure 2.31). However, for the dredged clays, the magnitude of the change in preconsolidation pressure was relatively smaller in comparison to the natural clays (Zeng et al., 2016).

Figure 2.31. The example of ageing in dredged sediment (from Zeng et al., 2016).

Hence, there is ample evidence in the literature that shows if a clay sample is allowed to rest for a certain period in a laboratory under constant stress, the sample will develop structural strength, and when it is again subjected to a small load increment it will show resistance against further deformation (Moretto, 1946; Leonards and Ramiah, 1959; Bjerrum and Lo, 1963). Such an ageing effect has been attributed to the thixotropic behaviour of clays that involves the development of cohesive bonds between the particles. Similar ageing effect has been observed in field conditions (manifested in an increase in shear strength), where the creep compression has also been observed and found associated with a change in volume; however, the strength gain has been found independent of the reduction in volume due to creep (Terzaghi, 1943; Bjerrum and Wu, 1960).

2.5.6 Rate dependency of structured soils

Several experimental studies demonstrated the time and rate-dependent nature of soft clays and the test results also indicated that the degree of structuration in clays depends on the combined effect of time and strain rate (Sorensen, 2006). Clays were found to show rate-dependent stress-strain relationships; the loading sequence and the loading rate affected the preconsolidation pressure as well as undrained shear strength (Soga, 2005; Yin, Yin, and Huang, 2015). When Leroueil et al. (1985) conducted 18 different constant rates of consolidation (CRC) tests on Batiscan clay by applying different strain rates ($1.7 \times 10^{-8} \sim 4 \times 10^{-5} \text{ s}^{-1}$), the resulting compression curves were evidently rate-dependent as there was a shift in the location of the compressibility curves with the strain rate and the magnitudes of preconsolidation pressures (σ'_p) were also found dependent on the strain rate or compression rate (Figures 2.32a and 2.32b).

When Leroueil et al. (1985) conducted different types of oedometer tests including constant rate of strain (CRS) tests and controlled gradient tests (CGT) on natural clay samples collected from different sites, they observed a unique relationship between the preconsolidation pressure and strain rate as the change in preconsolidation pressure with strain rate was found to be very similar independent of the clay type (Leroueil et al., 1985).

Figure 2.32a. Results from the CRS oedometer tests conducted on Batiscan clay using different strain rates (from Leroueil et al., 1985).

Figure 2.32b. The change in preconsolidation pressure with the strain rates showing the rate dependency of one-dimensional compression curves of Batiscaan clay (from Leroueil, et al., 1985).

When Leroueil (1996) compared the compressibility curves obtained from two different types of compression tests (i.e. conventional oedometer test and constant rate of strain oedometer or CRS test, which were performed at different strain rates such as $1 \times 10^{-7} \text{ s}^{-1}$ and 3.3×10^{-6} respectively), the curves were found different. The preconsolidation pressure estimated from the CRS tests was found 25% higher than the preconsolidation pressure estimated from the conventional oedometer tests. Such difference could only be explained by the strain rate effects as relatively slower CRS tests allowed progressive structuration (ageing effect) at lower strain rate ($< 1 \times 10^{-7} \text{ s}^{-1}$) and resulted in much stiffer compression behaviour (Figure 2.33) (Leroueil, 1996; Augustesen, Liingaard, and Lade, 2004; Sorensen, 2006).

Figure 2.33. Compression curves obtained from the conventional oedometer test and the constant rate of strain oedometer or CRS test (from Leroueil, 1996).

2.5.7 Thixotropic behaviour of clays

Although the thixotropy has typically been classified as a rheological process, it is a common phenomenon in most of the clay-water systems. Hence, the thixotropy process has also been described from a geotechnical point of view. Accordingly, thixotropy is the process of softening that is caused by remoulding and followed by a time-dependent recovery of the original harder state at a constant water content and porosity (Mitchell, 1960). Mitchell and Soga (2005) described thixotropy as “*an isothermal reversible time-dependent process occurring under conditions of constant composition and volume whereby material stiffens while at rest*”. Although the phenomenon of thixotropy in

remoulded soils is not completely understood yet, it is generally believed that the thixotropy develops in structured systems and is typically manifested in a thixotropic strength loss and regain associated with the microstructural changes (Osipov et al., 1984; Markgraf et al., 2006).

Usually, the thixotropic strength gain in clays is estimated as a ratio of the undrained shear strength measured after an elapsed time to the undrained shear strength measured immediately after the remoulding/reconstitution at the same water content; such ratio is also known as the thixotropic strength ratio (in the case of compacted clays) or sensitivity (for saturated clays) (Skempton and Northey, 1952; Suthaker, 1997). The thixotropic strength ratio or sensitivity is used for quantifying the thixotropic effects in clays instead of using the actual value of the strength increase (Mitchell, 1960).

When Morretto (1948) investigated the effect of structuration on the strength development in remoulded clays, he observed increases in undrained shear strength over time with very negligible change in water content (Morretto, 1948). The undrained shear strength in natural clays is typically a function of the in situ effective stresses and the soil fabric and bonding, but not the water content or void ratio (Burland, 1990). For normally consolidated soils, there is an approximately linear relationship between the undrained shear strength (s_u) and the vertical effective stress (σ'_v). In the case of overconsolidated soils such as natural deposits of sedimentary clays, an empirical relationship between the undrained shear strength (s_u) and the preconsolidation pressure (σ'_p) (instead of vertical effective stress) has been proposed (Terzaghi et al., 1996):

$$\frac{s_u}{\sigma'_p} = 0.22 \text{ (for undisturbed samples)}$$

$$\frac{s_u}{\sigma'_p} = 0.27 \text{ (for remoulded samples)}$$

Where s_u = undrained shear strength, and σ'_p = preconsolidation pressure.

Such a proportional relationship implies that the increase in undrained shear strength (because of the thixotropic strength regain) and the increase in preconsolidation pressure (due to the structuration effect) are just the different manifestations of the same physical phenomenon as both thixotropy and structuration originate from the same process of stronger particle-particle interactions and bonding and the particle rearrangements (Nagaraj et al., 1990). A time-dependent thixotropic behaviour in remoulded or reconstituted clays, which results in an increase in undrained shear strength is also typically accompanied by an increase in preconsolidation pressure (due to the structuration effect) (Mesri, 1975). The only difference between them is that the structuration effect leads to a gradual recovery of stiffness in a remoulded or reconstituted clay sample, whereas thixotropy results in recovery of strength with time (Qi et al., 2018).

In literature, the thixotropic behaviour of clays has been analyzed and explained from both macroscopic and microscopic points of view (Banas, 1991). Among the early researchers, Moretto (1948) and Skempton and Northey (1952) investigated the thixotropic strength gain in remoulded clays and measured the sensitivity of clays, and Seed and Chan (1957) examined the time-dependent strength gain in compacted clays. The data presented by them clearly demonstrated that there was an increase in strength of the clayey soils at

constant water content because of the thixotropic effect and the processes of thixotropic strength loss and gain were found to be reversible in compacted silty clays (Mitchell, 1960). Mitchell (1960) described the thixotropic phenomena in fine-grained soils in detail and summarized the previous investigations on the behaviour of the thixotropic systems. Later, many other researchers explored the thixotropic behaviour of clays and shed light on various topics related to time-dependent thixotropic strength gain (Banas, 1991).

Microstructural investigations of thixotropic processes in clay soils using scanning electron microscopy (Osipov et al., 1984) revealed that microstructural changes in clays are directly related to the value of shear strengths and the increase in thixotropic strength with time is also correlated to the recovery of the initial microstructure in clays. Recently, Zhang et al. (2017) studied the change in the microstructure of Zhanjiang clay along with the evolution of strength in the clay sample during 500 days of thixotropic development; the goal was to explain the thixotropic mechanisms for clays from the microstructural point of view. Zhang et al. (2017) measured thixotropic strength development in remoulded Zhanjiang clay. Although the strength decreased substantially because of the disturbance during the remoulding process (from 175 kPa to 23.5 kPa), the strength increased to 33.5 kPa within 10 days. The strength was increased by almost 2 times and 2.5 times that of the initial disturbance value after 100 days and 500 days respectively (increased to 47 kPa and 57.9 kPa respectively from 23.5 kPa). The rate of strength increase in the remoulded clays was significantly higher in the initial stage of thixotropy; however, the strength increase rate gradually decreased with the thixotropic time (Figure 2.34) (Zhang et al., 2017).

Figure 2.34. The changes in thixotropic strength ratio of Zhanjiang clay and other clays and mineral clays including Detroit clay by Morretto (1948), Horten clay by Hansen (1950), Beauharnois clay by Morretto (1948), Shell haven clay by Skempton (1948), London clay by Skempton and Northey (1952), and bentonite, kaolin and illite clay by Skempton and Northey (1952) (from Zhang et al., 2017).

The SEM images and the MIP analyses shed light on the changes in microstructure and pore size distribution of clays during the thixotropic strength development. The loss of strength in clays due to the disturbance during remoulding and the subsequent strength regain due to thixotropy was reflected in the stages through which the clay microstructure changed from the initial structure to the dispersed structure, and again from the dispersed structure to the flocculated structure (Figure 2.35). Such changes in microstructure and pore sizes over time were thought to play a crucial role in the thixotropic strength development in Zhanjiang clay (Zhang et al., 2017).

Figure 2.35. The SEM images of (a) natural Zhanjiang clay and remoulded clay at different thixotropic times: (b) 1 day, (c) 3 days, (d) 7 days, (e) 30 days, (f) 100 days, (g) 300 days, and (h) 500 days (from Zhang et al., 2017).

Studies on thixotropy in clays identified several factors that influence the thixotropy in clays directly. They include the clay mineralogy, water content, rate of loading, axial strain, and most importantly, time (Banas, 1991). Different clay minerals were observed to

regain thixotropic strength at a different rate; for example, bentonite showed a prominent thixotropic characteristic as it regained strength rapidly, whereas kaolinite showed a nominal strength regain with almost no thixotropy (Skempton and Northey, 1952).

For clays or clay slurries, although it has never been possible to establish any unique relationship between the thixotropic strength gain and time, the samples have always been observed to gain strength over time irrespective of the test duration indicating that thixotropy is primarily a time-dependent phenomenon (Mitchell, 1960). Especially, when the clay slurries were tested at low stress levels and high water contents, a significant increase in shear strength was observed with time even after correcting for the reduction in void ratio/density (due to the dewatering and self-weight consolidation) (Sorensen, 2006).

Although water content is one of the most important factors influencing the thixotropic effect in clays, there has been conflicting evidence in the literature regarding the relationship between the water content and the degree of thixotropy. When the water content exceeds the liquid limit, clayey soils demonstrate a wide range of thixotropic behaviour starting from less thixotropic to significantly thixotropic (Skempton and Northey, 1952). Again, when water content approaches the plastic limit, some soils exhibit very little or no thixotropy (Seed and Chan, 1957), whereas some soils show significant thixotropy (Mitchell, 1960).

Regarding the effect of rate of loading on thixotropy, Banas (1991) postulated that the outcome of an increase in the duration of loading would be the combination of two simultaneously occurring factors: (a) the availability of a longer period for creep deformation to occur could work against the tendency to gain thixotropic strength, and (b)

at the same time, such an extended period of time could allow the thixotropic effects to develop and gain strength in clays, which could work against further creep compression.

2.5.8 Structuration effect in Leda clay

Leda clay is the name given to extremely sensitive fine-grained sediment commonly found in the valleys of St. Lawrence and Ottawa rivers (Eden and Crawford, 1957; Crawford, 1968). In literature, the clay has also been called as ‘Champlain’ clay, ‘Laurentian’ clay and ‘Rideau’ clay, however, the name ‘Leda clay’ has been the most acceptable and commonly used name (Eden and Crawford, 1957). Leda clay is called sensitive because the clay experiences a considerable loss of shear strength when it is remoulded (Alshawmar, 2014). Leda clay is also known as a quick clay. Owing to a structure that is ‘sensitive’ to distortion, the structural properties of Leda clays can change rapidly when they are strained (Crawford, 1968).

Leda clays are known to display a structuration effect in their natural state. When Walker and Raymond (1968) compared the consolidation behaviour of undisturbed Leda clay determined from the laboratory experiments to the theoretical predictions of average rates of consolidation, they observed a discrepancy. They attributed such discrepancy to the development of a structural bonding (due to cementation), which held the soil particles together and formed a structure within the clays, and subsequently, resulted in an apparent or quasi-preconsolidation pressure (Crawford, 1968). The effect of such bonding was manifested in the early stage of the compressibility curves. The clays demonstrated an elastic deformation with the load increasing. At this stage, there was a minimum disturbance to the structure, rather the clays demonstrated a structural resistance against

the compression force applied. Therefore, the rate of consolidation was impeded. However, once the preconsolidation pressure was exceeded, the bonds started to break down and the structure started to collapse causing a sharp slope in the compressibility curve. The effect of structure became increasingly less significant as the consolidation stress increased. The effect of time was the most important factor during this stage (Jarret, 1967). Hence, the structural bonding or the formation of a structure in Leda clay was found to influence the rate of its consolidation (Walker and Raymond, 1968).

Later, Yong and Nagaraj (1977) also demonstrated that Leda clay undergoes a change in its compression behaviour with time because of the formation of a soil structure within the deposits; however, when it is remoulded, it shows an increase in compressibility and decrease in yield strength (Figure 2.36) (Yong and Nagaraj, 1977).

Figure 2.36. Compressibility curves obtained from one-dimensional compression tests performed on the undisturbed and remoulded Leda clay (from Yong and Nagaraj, 1977).

Figure 2.37. The compression curves obtained from the Leda clay as tested by Quigley and Thompson (1966) (from Soga, 1994).

Quigley and Thompson (1966) studied the relationship between the soil structure (fabric or particle arrangement and interparticle bonding) and compression behaviour of Leda clay using the X-ray diffraction (XRD) technique and conventional consolidation tests. When an undisturbed sample was subjected to a pressure below the preconsolidation pressure, the sample showed stiffness and resisted compression (Figure 2.37). At the microstructural level, little change was observed in the particle arrangement. However, when the preconsolidation pressure was exceeded, the interparticle bonds were also found ruptured and there was a reorientation of the clay particles. In the case of compression of a remoulded sample, the interparticle bonding was already broken due to the process of remoulding; hence, the reorientation of the clay particles under the compression force was easier and the sample yielded at smaller effective stress producing a smoother compression

curve. Owing to a stronger interparticle bonding and more stable particle arrangement, structured natural clays were able to sustain greater vertical effective stress than the remoulded samples at the same void ratio. The study showed that the clay fabric is directly correlated to its compressibility characteristics (Quigley and Thompson, 1966).

Delage (2010) investigated the effects of changes in soil fabric and interparticle bonding on the compressibility of the Champlain clay based on the study of compressibility curves obtained from the one-dimensional consolidation tests and the SEM and MIP observations. He made a comparison between the compression curves obtained from three intact clay samples and a freshly remoulded clay sample (Figure 2.38). He correlated the changes in compressibility to the changes in soil fabric and attributed the increased apparent preconsolidation pressure to the stronger interparticle bonding (Delage, 2010).

Figure 2.38. The correlation between the changes in compressibility and the changes in soil structure (fabric and interparticle bonding) (from Delage, 2010).

2.5.9 Estimating the structuration effect in natural clays

In fine-grained soils, due to the creep and structuration effect, the yield stress often exceeds the maximum past effective overburden pressure (p_c) (Rowe, 2012). The increased yield stress due to the time-dependent effects in clays has been named as the apparent or quasi-preconsolidation pressure and such development has been found to result in a discrepancy between the observed and predicted settlements of clays (Ma et al., 2014).

When laboratory-scale tests were performed on natural clays and the samples were allowed to rest under constant effective stress for a period, a time-dependent volume change behaviour was observed, and the samples were also found to develop an apparent preconsolidation pressure. The creep compression (the time-dependent volume change behaviour of clays at constant stress) has been thought to be a contributing factor to such structural strength development in clays (Leroueil and Vaughan, 1990). In natural clay deposits, a similar increase in preconsolidation pressure has been observed, which has been attributed to the thixotropic effects in clays (Leroueil and Vaughan, 1990).

Burland (1990) characterized the soil structure in terms of compressibility and presented the concept of ‘intrinsic compression line (ICL)’ to describe the influence of structuration in natural sedimentary clays on their compressibility (Soga, 1994). Burland (1990) considered the properties of reconstituted clays as their ‘intrinsic’ properties since they are inherent to the clays irrespective of their natural states. He compared the properties of natural clays to their intrinsic properties and attributed the differences to the changes in the arrangement of the clay particles (soil fabric) and interparticle bonding (Burland, 1990). Burland used the term ‘intrinsic’ to describe the properties of clays when the clays are

reconstituted at a water content equal to or greater than the liquid limit (preferably at the range of $1.0 W_L \sim 1.5 W_L$) and consolidated under one-dimensional conditions. When a given soil is prepared (reconstituted) in such a specified manner, the measured properties reflect the basic properties of that soil, which are independent of its natural state or devoid of soil structure (Burland, 1990). The compression curve that represents the compression behaviour of a clay reconstituted at a specified water content is the ‘intrinsic compression curve’ and is used as a frame of reference for the compression behaviour of the clay at its natural state. For converting the intrinsic compression curve to a normalized curve, Burland (1990) introduced a new normalizing parameter called the void index (I_v):

$$I_v = \frac{e - e_{100}^*}{e_{100}^* - e_{1000}^*} = \frac{e - e_{100}^*}{C_c^*} \dots\dots\dots (2.5)$$

Where e_{100}^* and e_{1000}^* denote the intrinsic void ratios at the 100 kPa and 1000 kPa vertical effective stresses respectively and C_c^* is the intrinsic compressibility. The asterisk (*) is used to denote the intrinsic property.

A void index is a relative quantity that indicates the position of the current void ratio of clay in relation to the void ratios of the same clay at two specific vertical effective stresses (100 kPa and 1000 kPa respectively) (Liu, Zhuang, and Horpibulsuk, 2013). Burland (1990) assumed the void index (I_v) to be an indirect measure of the intrinsic compactness of a soil deposit; therefore, when $I_v < 0$, the soil is compact, whereas when $I_v > 0$, it is loose (Burland, 1990).

Figure 2.39. The use of void index I_v to normalize the intrinsic compression curve (from Burland, 1990).

When Burland (1990) plotted the normalized void index (I_v) against the effective stress based on the compressibility data obtained from a multitude of reconstituted natural clays, he noticed that for many reconstituted natural clays I_v fell on a unique line and named it an intrinsic compression line (ICL) (Figure 2.40). For a clay, the intrinsic compression line (ICL) can be measured experimentally or can be constructed using the following equation provided e_{100}^* and C_c^* are known:

$$I_v = 2.45 - 1.285x + 0.015x^3 \dots\dots\dots (2.6)$$

Where $x = \log \sigma'_v$ (in kPa).

Burland (1990) suggested another indirect method of constructing the intrinsic compression line (ICL) that is based on the empirical correlations between the constants of intrinsic compressibility (e_{100}^* and C_c^*) and the Atterberg limits:

$$e_{100}^* = 0.109 + 0.679e_L - 0.089e_L^2 + 0.016e_L^3 \dots\dots\dots (2.7)$$

$$C_c^* = 0.256e_L - 0.04 \dots\dots\dots (2.8)$$

Where e_L = Void ratio at the liquid limit.

Figure 2.40. The difference between the compressibility of in-situ or intact natural clays and the compressibility of reconstituted specimens (from Burland, 1990).

When Burland (1990) plotted the consolidation data from the intact field specimens (oedometer tests data from a large number of sites and the data from the sites considered

by Skempton, 1970), the line that passed through the data for the intact clays fell above the intrinsic compression line (ICL). This line was named ‘sedimentation compression line (SCL)’. A comparison between the intrinsic compression line (ICL) and the sedimentation compression line (SCL) demonstrated that naturally structured soils have different compression behaviour than the corresponding reconstituted soils. Over the range of $\sigma'_v = 10 \sim 1000 \text{ kPa}$, at any given void ratio or void index, a natural clay was found to sustain an effective overburden pressure which is approximately five times that sustained by the corresponding reconstituted clay sample at the same void ratio (Figure 2.40). The change in the arrangement of the particles and interparticle bonding in the naturally deposited clays was thought to be a contributing factor to such an increased resistance to the compression (Burland, 1990).

The intrinsic compression line (ICL) proposed by Burland (1990) could be used as a useful reference line for comparing the sedimentation compression curves of different natural clays and studying their compression characteristics. Although the proposed sedimentation compression line (SCL) is not a fundamental line, it is still useful and relevant since it represents the compression behaviour of most of the natural clay deposits. Especially, when the yield stress is not well-defined for a naturally overconsolidated soil, the sedimentation compression line (SCL) in connection to the intrinsic compression line (ICL) gives a useful means of determining the degree of overconsolidation in the soil (Grozic, Lunne, and Pande, 2003). When the vertical effective stress on a soil exceeds its yield stress or preconsolidation pressure, and subsequently, the soil fabric and interparticle bonding are destroyed, the resulting compression curve converges towards the intrinsic

compression line (ICL). However, in the case of reconstituted soils, the compression curves obtained from the oedometer would lie close to the ICL (Burland, 1990).

Based on the results obtained from the oedometer tests performed on four clay samples prepared at different initial water contents ($1.00 W_L \sim 1.75 W_L$), Cerato and Lutenegeger (2004) showed that for a given reconstituted soil sample the value of e_{100}^* is dependent on the initial water content. The initial water content was found to influence the compression curve of the reconstituted clay sample; the clay mineralogy and the corresponding soil properties such as liquid limit and cation exchange capacity were thought to be the factors that govern the influence of initial water content on the intrinsic compressibility. Cerato and Lutenegeger (2004) suggested using a standard range of initial water content during the sample preparation for determining the intrinsic compressibility. Otherwise, the estimated intrinsic parameter of the void index (I_v) might not be a truly intrinsic soil parameter and comparing test results from different sources would not be possible (Cerato and Lutenegeger, 2004).

Hong et al. (2010) investigated the effects of different initial water contents on the virgin compression of reconstituted clays, and based on the findings, they redefined the ICL by extending the range of initial water content to $W_0 = 0.7 W_L \sim 2.0 W_L$ with the effective vertical stress as low as 1.5 kPa. The resulting intrinsic compression line was called extended intrinsic compression line (EICL):

$$I_v = 3.0 - 1.87x + 0.079x^2 \dots\dots\dots (2.9)$$

Where $x = \log \sigma'_v$ (in kPa).

EICL was found to lie above ICL in the range of $\sigma'_v = 1.5 \sim 10 \text{ kPa}$; however, when σ'_v exceeds 25 kPa, theEICL converged to the ICL (Hong et al., 2010). Although Burland (1990) recommended the use of ICL for $\sigma'_v = 10 \sim 4000 \text{ kPa}$ and $W_0 = 1.25 W_L$, the introduction of EICL made it possible to use the ICL over a greater range of effective vertical stresses and initial water contents (Hong et al., 2010).

Based on the same assumption that the properties of reconstituted soils could be adopted as intrinsic properties (Burland, 1990), Liu and Carter (1999) investigated the compression behaviour of both reconstituted and natural soils to determine the influence of soil structure on the compression behaviour. They suggested the use of an additional void ratio (that is sustained by the soil structure in natural soils) as a direct indicator of soil structure since such void ratio reflects the difference in the behaviour of a structured soil and the corresponding reconstituted soil.

Liu and Carter (1999) presented a material idealization of the compression behaviour of reconstituted and structured soils (Figure 2.41). They expressed the void ratio of a structured soil (e) in terms of the corresponding void ratio of the reconstituted soil (e^*) using the following relationship, where e^* represents an intrinsic property of the reconstituted soil:

$$e = e^* + \Delta e$$

$$\text{So, } \Delta e = e - e^* \dots\dots\dots (2.10)$$

Where Δe is the additional voids ratio sustained by soil structure.

Figure 2.41. The scheme for analyzing the differences in reconstituted and natural clays proposed by Liu and Carter (1999) (from Liu and Carter, 1999).

Since the behaviour of reconstituted soil was considered as the reference behaviour and the associated properties to be the reference properties, the difference in the void ratio between the reconstituted and natural clays (Δe) was thought to be the effect of structuration. Liu and Carter (1999) proposed a basic principle for the compression behaviour of structured soils by introducing an inversely proportional relationship between the additional void ratio sustained by the soil structure and the current mean effective stress:

$$\Delta e = \frac{A}{p'} \dots\dots\dots (2.11)$$

$$\text{for } p' \geq p'_{y,i}$$

Where A is the 'structural compression factor', which was considered to be a constant for a given soil under a given type of compression; p' is the current mean effective stress; and $p'_{y,i}$ is the mean effective stress at which the structured soil starts to yield under the loading.

Since a linear relationship exists between the void ratio (e) and $\ln p'$ over a considerable range of effective stress (except at very low and very high stress levels) (Burland, 1990), Liu and Carter (1999) also used the following relationship for describing the compression behaviour of a structured soil:

$$e = e_{IC}^* + \frac{A}{p} - \lambda^* \ln p' \dots\dots\dots (2.12)$$

Where e_{IC}^* and λ^* are the intrinsic compression parameters of the reconstituted clays.

Since A was not a fundamental soil structure parameter and its value could vary depending on the initial virgin yield stress as well as deformation constraints imposed during compression, Liu and Carter (1999) introduced a new soil parameter, the structure index (S), which helped to replace A in the above equation by using the following relationship:

$$A = S p'_{y,i} \ln p'_{y,i} \dots\dots\dots (2.13)$$

Where the structure index (S) was given by the increase in the void ratio above the reconstituted compressibility curve divided by the natural logarithm of the vertical effective stress. The value of S depends on the type of soil as well as the degree of soil structure; however, theoretically, it would be a constant for the same soil with similar geological and stress histories (Liu and Carter, 1999).

Hence, the relationship between the additional void ratio and the current mean effective stress was rewritten as (by combining equation 2.11 and equation 2.13):

$$\Delta e = S \left(\frac{p_{y,i}'}{p'} \right) \ln p_{y,i}' \dots \dots \dots (2.14)$$

Also, the following hyperbolic relationship was derived to express the compression behaviour of a structured soil (by combining equation 2.12 and equation 2.14):

$$e = e_{IC}^* + S \left(\frac{p_{y,i}'}{p'} \right) \ln p_{y,i}' - \lambda^* \ln p' \dots \dots \dots (2.15)$$

$$\text{for } p' \geq p_{y,i}'$$

Liu and Carter (1999) reviewed twenty (20) case studies to examine whether the proposed hyperbolic relationship describes the compression behaviour of structured soils. The selected studies involved 20 different types of soils with 27 compression tests performed on them, covering a wide range of effective stresses (10 kPa ~ 3000 kPa) and void ratios (0.5 ~ 14.0). Although the tests involved one-dimensional compression, the mean effective stresses were not considered or estimated in most of the cases. Hence, the following relationships were used in those cases:

$$\Delta e = \frac{A_v}{\sigma_v'} \dots \dots \dots (2.16)$$

and

$$\Delta e = S_v \left(\frac{\sigma_{vy,i}'}{\sigma_v'} \right) \ln \sigma_{vy,i}' \dots \dots \dots (2.17)$$

Where σ_v' is the vertical effective stress and $\sigma_{vy,i}'$ is the vertical stress at which the sample starts to yield. A_v is the structural compression factor that corresponds to the one-

dimensional test conditions and S_v is the structure index that also corresponds to the one-dimensional conditions. A_v depends on the ratio of p' to σ'_v , whereas S_v depends on the ratio of p' to σ'_v and also on the value of $p'_{y,i}$. Provided that the ratio of p' to σ'_v is kept constant during the compression of the structured soil, such substitution does not affect the validity of the proposed hyperbolic relationship (Liu and Carter, 1999).

During the validation of the proposed hyperbolic relationship based on the examination of the available tests data, the values of the yield stress ($\sigma'_{v y,i}$) in the above equation were determined directly from the compression curves plotted in the e vs. $\ln \sigma'_v$ graphs. A_v was determined by finding the best match of equation 2.16 with the test data for the virgin yielding of the sample. Δe was estimated from the compression test conducted on the structured soil and the reconstituted clays. The values of $\sigma'_{v y,i}$ and A_v were determined independently; however, the values of S_v were determined from the values already estimated for the $\sigma'_{v y,i}$ and A_v . Using the values determined for A_v , the theoretical compression curves corresponding to equation (2.16) were superimposed on the experimental data. A comparison between the theoretical and experimental compression curves allowed for an assessment of the proposed hyperbolic relationship between the additional voids ratio and the current mean effective stress.

Figure 2.42 shows the comparison between the theoretical and experimental compression curves for the Champlain clay and Leda clay. The theoretical compression curves were in good agreement with the experimental compression curves, which indicated that the proposed hyperbolic relationship could describe the additional void ratio sustained

by the structured soil and predict its compression behaviour in terms of yield stress (or preconsolidation pressure) and S , a structure index [$\Delta e = S \ln \sigma_v'$] (Liu and Carter, 1999).

Figure 2.42. The theoretical compression curves and experimental compression curves obtained from the one-dimensional compression tests conducted on Champlain clay and Leda clay by Tavenas et al., 1974 and Yong and Nagaraj, 1977 respectively (adapted from Liu and Carter, 1999).

Recently, based on their data set of dredged sediments, Zeng et al. (2016) proposed an empirical equation, which predicts the yield stress to develop linearly with log time in the high water content dredged clays (due to the structuration/ageing effect). The proposed equation strongly depends on the ratio between the initial water content and the liquid limit, and weakly on the value of vertical effective stress:

$$(YSR - 1) = m \times \exp \left(n \times \frac{w_0}{w_L} \right) \dots \dots \dots (2.18)$$

Where YSR = the ratio of vertical yield stress to effective vertical stress; w_0 and w_L are the initial water content and liquid limit respectively; and, m and n are the parameters for determining YSR , which can be expressed as follows:

$$n = 1.55 \dots \dots \dots (2.19)$$

$$m = 0.005 - 0.031 \log(\sigma'_v) + 0.056 \log(t) \dots \dots \dots (2.20)$$

Where σ'_v = effective vertical stress; and, t = consolidation time, including primary consolidation time and ageing time.

When Zeng et al. (2016) compared the predicted YSR values with the experimentally obtained YSR values, predicted results were found in good agreement with the experimentally obtained values (Figure 2.43).

Figure 2.43. Comparisons of predicted and measured YSR values (from Zeng et al., 2016).

Since Burland (1990), several researchers have proposed different concepts for determining the compression behaviour of structured soils or for estimating the degree of structuration in structured soils based on their compression behaviour. Although most of the studies have been based on the intrinsic compression concept proposed by Burland (1990), they have also introduced new parameters such as structure index (S) (Liu and Carter, 1999), strength sensitivity (St) (Cotecchia and Chandler, 2000), and YSR (Zeng et al., 2016) and suggested empirical equations for quantifying or predicting the degree of structuration/ageing in clays. Nevertheless, yet, there exists no consolidation model that can describe the full range of consolidation behaviour in clays including the time-dependent structuration/ageing effect.

2.6 Time-dependent behaviour of oil sands tailings

2.6.1 Sedimentation and consolidation

The main processes that are considered in the tailings deposition mechanism are sedimentation and consolidation. Sedimentation is the prior stage of the settlement of tailings, which involves the downward movement of the fine solid particles under the force of gravity and the formation of sediment or a layer of tailings at the base of the pond, where effective stress does not exist (Kaminsky et al., 2015). Consolidation of tailings involves the slow settlement of fine-grained materials over time through the dissipation of excess pore water pressure and the subsequent gain in effective stress in response to either self-weight or vertical surcharge from a capping layer (BGC Engineering Inc., 2010; Jeeravipoolvarn, 2010). It is a time-dependent process that helps the tailings deposits to

dewater and strengthen through compression or densification and a reduction in void space (Kaminsky et al., 2015). Although sedimentation and consolidation, these two fundamental phenomena are conventionally treated as two separate processes, in practice they are interconnected. The boundary between sedimentation and consolidation is often unpredictable as the void ratio at which the effective stress initiates is not uniquely defined and varies with the initial water content of the slurry (Jeeravipoolvarn, 2010). An improved fundamental understanding of the liquid-to-solid transition and the associated strength gain, and the development of a unified theoretical formulation capable of characterizing the transformation from sedimentation to consolidation could lead to better design and management of the tailings facilities (BGC Engineering Inc., 2010).

Typically, sedimentation uses the force of gravity to separate the suspended solid particles from the tailings stream. The settling rate of the particles is influenced by the particle size and shape, density, and surface properties, which are again governed by the change in pH and salinity of the suspension, and the type and the amount of the flocculant used (Cabrera et al., 2009). The heavier particles such as sand settle quickly, whereas the fine particles such as clay remain in the suspension for a longer period and settle at a much slower rate. Such an environment promotes the segregation behaviour of the tailings particles, which leads to the preferential settlement of the particles of certain sizes in one or another physical zone (Mihiretu et al., 2009).

During the sedimentation process, often, there are mutual collisions between the particles interrupting their free settling. Subsequently, the soil particles settle together, and this type of settling is called ‘hindered settling’ or ‘hindered sedimentation’. A continuous

‘hindered settling’ of the solid particles results in the formation of a ‘sediment bed’, where effective stress does not exist. However, when the hindered sedimentation approaches an end, the particles in the sediment bed starts to exhibit grain-to-grain interactions (Bajwa, 2015). At the initial stage, the pore water carries the self-weight, which gives rise to the development of excess pore water pressure. It is called excess pore water pressure since it is a pressure within the pore water that is more than the hydrostatic equilibrium. After some time, this excess pore water pressure starts to dissipate via the expulsion of pore water. Generally, the primary consolidation of the newly formed sediment bed begins under the influence of the sediment self-weight and it is marked by the development of effective stress as the load from the tailings self-weight is transferred from the pore water to the soil skeleton (while the excess pore water pressure dissipates). The expulsion of air/water from the pores is followed by some rearrangements of the particles. The rearrangement of the particles prompts a volume change and this volume change leads to the compression, and subsequently, to the settlement of the fine-grained particles. The rate of volume change reflects the rate at which pore water moves out, which again depends on the hydraulic conductivity or permeability of the material (Gholami, 2014; Bajwa, 2015).

The poor dewaterability and extremely slow consolidation behaviour of FFT are often attributed to the low permeability (hydraulic conductivity) of the tailings material (that is associated with the highly dispersed state of the clay particles) and also to the buildup of a structure through thixotropy that impedes consolidation (Scott et al., 2013). Hydraulic conductivity governs the rate at which excess pore water pressure can dissipate. For low solids content or high void ratio materials such as tailings, hydraulic conductivity-

void ratio relationship is the property that dictates the rate and magnitude of settlement and consolidation (Suthaker and Scott, 1994). Fine tailings have low hydraulic conductivities (in the range of 10^{-6} to 10^{-9} m/s) even at a high void ratio of 5.4. Consequently, the excess pore pressure dissipates very slowly (Chalaturnyk et al., 2004). The primary consolidation in clay/FFT deposits ends with a complete dissipation of excess pore water pressure (when the excess pore water pressure returns to a hydrostatic equilibrium condition) and the formation of a consolidated bed (Imai, 1981; Leshchinsky, 2004; Dimitrova and Yanful, 2011). However, the creep compression or secondary consolidation, which originates from the plastic deformation of the bed under a constant effective overburden pressure continues long after the primary consolidation ends.

The conventional tailings disposal method involves hydraulic deposition as a slurry in a dammed impoundment, and subsequently, consolidation under self-weight. However, in the case of thickened tailings, where tailings are placed in different lifts, the tailings deposited earlier consolidate under their own weight, and also due to the weight of the subsequently placed tailings (Caldwell et al., 2014). When a new layer is deposited on top of an existing layer, initially, there is an increase in pore water pressure and this excess pore water pressure carries the new load. After some time, some water drains out of the system due to the increase in pore water pressure, which eventually causes the excess pore water pressure to dissipate, and then, the pore water transfers part of the load to the solids that are strong enough to support that load. This additional load pushes the particles in the solids to a denser, stronger position. Such densification raises the pore water pressure again. This cycle of developing excess pore water pressure and then dissipation through drainage

continues until the new surface load is supported entirely by the solids (Devenny et. al., 1993).

2.6.2 Consolidation theories and models

Generally, the rate of consolidation in a deposit depends on the magnitude of the applied load, the permeability, and the length of the drainage path. However, it is difficult to predict the time required for FFT to consolidate because of the limited amount of field consolidation data accumulated so far, and also due to the presence of a number of complicating factors including complex material properties and behaviour such as hydraulic conductivity that varies by several orders of magnitude as the deposit becomes denser; effective length of the drainage path as long drainage path does not favour consolidation in reasonable time; the presence of bitumen in pores and in discrete layers that hinders the rate of hydraulic conductivity; repulsive forces that oppose the development of a solids structure necessary to support the applied loads; variations in material composition and stratigraphy; and, the influence of time-dependent phenomena on the compressibility behaviour of the FFT (Devenny, 2010; Jeeravipoolvarn et al., 2014).

Karl Terzaghi proposed the classical one-dimensional small strain consolidation theory for the estimation of the settlements of soils. The theory was developed based on the consideration of fluid flow relationship, continuity equation, and the principle of effective stress. Mathematically, one-dimensional consolidation theory can be expressed as (Terzaghi, 1943):

$$c_v \frac{\partial^2 u}{\partial a^2} = \frac{\partial u}{\partial t} \dots \dots \dots (2.21)$$

Where c_v = coefficient of consolidation; a = a one-dimensional vertical coordinate; u = excess pore water pressure; and, t = time.

The theory was based on the assumption that hydraulic conductivity (k) is constant and the relationship between void ratio and effective stress is linear. Self-weight of soils was neglected, and an infinitesimal strain was considered. The movement of the solid particles was neglected and the boundary movement during the consolidation process was also ignored. It did not consider secondary consolidation or time-dependent effects such as ageing (Bharat and Sharma, 2011; Priestley et al., 2011). In conventional geotechnical engineering, the small strain consolidation theory is commonly used to predict the rate of excess pore pressure dissipation or consolidation. However, when considering extremely soft soils with very large deformations, the application of conventional consolidation analysis does not produce satisfactory results (Priestley, 2011).

Fluid fine tailings are generally deposited at high initial water content and high void ratio. Afterwards, the fine tailings go through a very large deformation and a change in the void ratio, which also changes the saturated hydraulic conductivity as well as compressibility of the tailings significantly in a non-linear manner (Priestley, 2011). Since fine tailings undergo such large strain consolidation, only a large strain consolidation theory that can explain the relationship between the void ratio and hydraulic conductivity in tailings can analyze the tailings consolidation behaviour rather than any conventional small strain consolidation analysis that assumes a constant hydraulic conductivity (Suthaker and Scott, 1997).

Mikasa (1965) and Gibson et al. (1967) were the first persons to formulate the non-linear consolidation equations independently and to develop one-dimensional finite strain consolidation theory for modelling the large deformation consolidation behaviour of soft fine-grained soils or slurries. The theory was introduced by refining the Terzaghi formulation and eliminating many of the assumptions that do not reflect the actual material behaviour observed in the field. The new theory removed the restrictions of the small strain and linearity and allowed more complex problems to be analyzed (Jeeravipoolvarn, 2010; Geier et al. 2011; Priestley, 2011). Gibson et al. (1967) introduced the one-dimensional finite strain consolidation theory by proposing a second-order partial differential equation in terms of void ratio and hydraulic conductivity:

$$\pm \left(\frac{\gamma_s}{\gamma_w} - 1 \right) \cdot \frac{d}{de} \left[\frac{k(e)}{(1+e)} \right] \frac{\partial e}{\partial z} + \frac{\partial}{\partial z} \left[\frac{k(e)}{\gamma_w(1+e)} \cdot \frac{d\sigma'}{de} \cdot \frac{\partial e}{\partial z} \right] + \frac{\partial e}{\partial t} = 0 \dots\dots\dots (2.22)$$

Where γ_s = solids density; γ_w = fluid density; e = void ratio; k = hydraulic conductivity; σ' = vertical effective stress; t = time; and, z = the reduced material coordinate.

The governing equation for the finite strain consolidation theory proposed by Gibson et al. (1967) includes the hydraulic conductivity and compressibility of tailings, which are two fundamental properties of void ratio function. The governing equation is expressed in terms of excess pore water pressure (u_e). Two important constitutive relationships are used in finite strain consolidation modelling: (a) a void ratio-effective stress relationship; and, (b) a void ratio-hydraulic conductivity. They are used to solve the governing equation, where the derivatives of effective stress and hydraulic conductivity need constitutive relationships to reduce the unknowns and solve for void ratios

(Jeeravipoolvarn, 2010). Void ratio-hydraulic conductivity relationship affects the completion of tailings densification over time and void ratio-effective stress relationship influences the densification progress within the consolidation process (Gholami, 2014).

Since the large strain consolidation model developed by Gibson et al. (1967) accounts for the large strain consolidation and the effect of self-weight, allows for the non-linearity of the material properties, and is based on the hydraulic conductivity-void ratio relationship and effective stress-void ratio relationship, the model can be applied to estimate the void ratio profiles for the one-dimensional tailings deposition scenarios and to predict their consolidation behaviour as they experience very large deformations. The model has been used in mine waste engineering extensively to solve the problems related to predicting the capacity of the tailings impoundments for different production schedules or deposition rates, different stages of deposition, and different impoundment geometries, and also to solve the problems related to predicting strength implications and pore pressure dissipation in the slurry (Jeeravipoolvarn, 2010; Gjerapic et al., 2008).

Over time, many researchers studied the large strain consolidation behaviour of the soft deposits, investigated the principal processes that influence their settlement after deposition such as flocculation, sedimentation, and self-weight consolidation, and they incorporated these concepts into the large strain consolidation theory. The non-linear constitutive relationships were obtained based on the laboratory experimental data and the numerical solutions were applied to solve the governing equation of finite strain consolidation. Later, the arrival of computers with their exceptional computing power as

well as finite element software made the solution of large deformation consolidation problems relatively easier (Priestley, 2011; Ahmed and Siddiqua, 2014).

Finite element analysis came as a superior choice for model development because of its strength in capturing the changes in material coordinates during large strain consolidation (Ito and Azam, 2013). The advantage of incorporating a wide array of flux and deformation boundary conditions in finite element analysis helped the consideration of more complex scenarios (Priestley et al., 2011). As technology advanced, two-dimensional and three-dimensional analysis of sedimentation and consolidation were also introduced. Although one-dimensional modelling is adequate for predicting the consolidation behaviour, further insight into the consolidation behaviour can be gained by using two and three-dimensional analyses (Priestley et al., 2011). Most commonly used models include FSCON 1-I and FSCON 2-I (Cargill and Schiffman, 1980), ACCUMV (Schiffman et al., 1992), NFSCONSOL (Wu, 1994), 1D CONDESO (Yao and Znidarcic, 1997), CONSOL 2D (Jakubick, McKenna, and Robertson, 2003), CC1 (Fox et al., 2005), and 3D CONDESO (Coffin, 2010) (Ahmed and Siddiqua, 2014).

Despite significant advancements with the consolidation theories and models, the numerical modelling of the consolidation of oil sand tailings remains challenging since several fundamental consolidation related phenomena or processes are yet to be understood completely (Chaudhary et al., 2015). On some occasions, even a large strain consolidation analysis has been proved to be inadequate in predicting the dewatering or volume change behaviour in soft deposits (Qi et al., 2017). Qi et al. (2017) attributed such short-comings of the large strain consolidation analysis to the fact that the analysis typically misestimates

or ignores the following processes or phenomena: (a) the water flow through the deposits due to evaporation or freeze-thaw; (b) the self-weight consolidation of very soft soils at the extremely low effective stress condition (Sills, 1999; Jeeravipoolvarn et al., 2009); and most significantly, (c) the time-dependent creep, thixotropy, and structuration behaviour, which affect the volume change and strength gain behaviour in the long-term (Jeeravipoolvarn et al. 2009). Soft soil deposits including tailings deposits demonstrate creep, thixotropy, and structuration behaviour during both initial settling stage and settlement that occurs in the long-term and such behaviour has significant implications for the dewatering, consolidation, and strength gain of tailings deposits (Qi et al., 2017).

Recently, Qi, Simms, and Vanapalli (2017) developed a program, named UNSAT-CON, which is capable of modelling large strain consolidation of saturated and unsaturated soft soil deposits including oil sands tailings. In 1997, Fox and Berles proposed a piecewise-linear finite-difference model called CS2 for the one-dimensional large strain consolidation. Qi, Simms, and Vanapalli (2017) expanded the formulation of CS2 to unsaturated conditions and developed a one-dimensional mass-conservative numerical formulation for coupled large-strain consolidation and saturated and unsaturated flow using a piecewise linear framework. The formulation was implemented in a novel algorithm named UNSAT-CON, using a finite-difference scheme (Qi, Simms, and Vanapalli, 2017). The proposed UNSAT-CON program was verified using numerical examples and physical tests including a column test on fine-grained oil sands tailings in the laboratory (Soleimani et al., 2014) and a field test on oil sands tailings (Dunmola et al., 2013a, b). The program was found capable of predicting a range of parameters including overall settlement, void

ratio, pore water pressure, and water content and required minimum calibration for making realistic predictions (Qi, Simms, Vanapalli, and Soleimani, 2017).

Recently, three commonly used creep models (Leoni et al. 2008; Yin and Graham, 1994, Rowe and Hinchberger, 1998) have been incorporated into the UNSAT-CON deposition simulation platform (Qi et al., 2018). When applied to the tailings deposits, the program was found capable of simulating simultaneous consolidation-creep effects in the flocculated tailings and the creep models generated similar results. However, no satisfactory approach has yet been formulated for incorporating the structuration effect into the large strain consolidation simulations; so, the uncertainty in final settlement predictions still remains (Qi et al., 2018).

2.6.3 Case studies of creep compression in unamended oil sands tailings

Since the fluid fine tailings are a composite material with unique features, a single factor or a single relationship does not govern the dewatering or consolidation behaviour of tailings. Rather, a wide array of physical and chemical properties and time-dependent effects such as creep, thixotropy, and structuration play important roles (Jeeravipoolvarn, 2005). Jeeravipoolvarn (2005) and Jeeravipoolvarn et al. (2009) reported on the compression behaviour of untreated oil sands tailings based on the results from the long-term, large-scale, self-weight consolidation tests conducted at the University of Alberta. A large strain consolidation model that incorporates creep was used to simulate the large-scale consolidation behaviour of oil sands tailings and the experimental data sets obtained from three 10-metre high, 0.9-metre diameter large standpipes were used for the theoretical verification (Figure 2.44). In 1982, one standpipe was filled with oil sands tailings and the

other two standpipes were filled with a mix of fine tailings and tailings sand. The first standpipe (which was filled with oil sands tailings) was monitored for more than 25 years for studying the self-weight consolidation behaviour of tailings and the second and third standpipes were monitored for studying the effects of entrained sand on the tailings consolidation behaviour (Jeeravipoolvarn, 2005; Jeeravipoolvarn et al., 2009).

Figure 2.44. (Left) 10-metre standpipe used at the University of Alberta for investigating the long-term dewatering and consolidation behaviour of FFT; and, (right) a cross-section of the 10-metre standpipe (from Jeeravipoolvarn, 2005; Jeeravipoolvarn et al., 2009).

Water-tailings interface settlement in the 10-metre standpipe was measured at intervals to keep track of the amount of compression of fluid fine tailings with time. The average solids content was measured to estimate the solids-water composition of tailings. The excess pore water pressures were measured at different heights of the standpipes for

monitoring the progress in consolidation since excess pore water pressure dissipation is an indication of the progress in consolidation (Jeeravipoolvarn, 2005).

After deposition, within a short period, the thixotropic gel strength started to develop, and a stronger soil structure was formed that started to behave like an overconsolidated structure. The pore water pressure tension (that was developed due to such a thixotropic effect) caused the excess pore water pressure to decline rapidly at the beginning with almost no settlement at the bottom. Later, as the creep or slow bond yielding of the fine matrix occurred, the excess pore water pressure increased slightly. However, soon, the excess pore water pressure started to decrease again with a decrease in total stress as the water-tailings interface settled. The measurements of pore water pressure and stresses in the 10-metre standpipe filled with tailings showed that the pore water pressure did not decline significantly over a period of 20.6 years, rather it remained very close to the total stress throughout the height of the standpipe (Figure 2.45a). Such a slow rate of pore water pressure dissipation was controlled by the hydraulic conductivity and the length of the drainage path. Overall, the excess pore water pressure behaviour in tailings was thought to be the resultant of creep-inducing pore water pressure and the dissipation of pore water pressure governed by hydraulic conductivity (Jeeravipoolvarn, 2005).

After 25.7 years, the FFT in the 10-metre standpipe settled more than 32% of its original height and there was a uniform settlement throughout the last 15 years (Figure 2.45b). However, there was very little to no effective stress development within the FFT material over time (Jeeravipoolvarn et al., 2009). After 10 years, only a small effective stress of 5 kPa was developed at the bottom 1 metre of the standpipe. Throughout the rest

of the period, there was no additional effective stress developed; however, the interface was settled by an amount of approximately 80 cm. The average solids content increased from 30.6% to 41.8% and the average void ratio decreased from 5.17 to 3.18 though only the bottom part of the standpipe appeared to be consolidated.

Figure 2.45. (a) Stress profile for the 10-metre standpipe after 20.6 years; and, (b) tailings-water interface settlement in the standpipe (from Jeeravipoolvarn et al., 2009).

Since the fluid fine tailings showed such a reduction in void ratio or a change in volume at very little to no effective stress, it was an indication of the creep behaviour of the tailings materials. If it were consolidation, such settlement would have been accompanied by a decrease in pore water pressure and effective stress development. Also, the overall volume change or compressibility behaviour would have been governed by the void ratio-effective stress relationship (or compressibility) relationship determined from the large strain consolidation test (Jeeravipoolvarn, 2005; Jeeravipoolvarn et al., 2009). Rather, it was the creep compression that occurred within the card-house floc structure of

the fine tailings (where the particles were rearranged under constant effective stress of zero) and contributed to such a uniform settlement. Not only the observation of FFT settlement in the 10-metre standpipe at the University of Alberta showed creep compression at constant effective stress, the effective stresses measured for the oil sands tailings that were settling in the tailings ponds were also very small (Jeeravipoolvarn, 2005).

When a numerical model was developed and used to examine the applicability of finite strain consolidation theory on the tailings material in the 10-metre standpipe and predict the interface settlement, the theory was found to overestimate the compression behaviour of tailings by 10% of the total height (Jeeravipoolvarn et al., 2009). Jeeravipoolvarn et al. (2009) discussed the time-dependent phenomena in untreated oil sands tailings that might have contributed to such overestimation. Based on the observation that the tailings in the standpipe settled over time at constant effective stress with a value very close to zero, they suggested that the tailings material demonstrated a creep behaviour, a phenomenon common in natural clays. Since creep is typically manifested in a time-dependent deformation behaviour under constant vertical effective stress, a single relationship between effective stress and void ratio that only corresponds to a specific time of loading would not be able to capture such time-dependent effect and describe the resulting compressibility characteristics of the tailings material (Jeeravipoolvarn et al., 2009). This is also the reason the conventional finite strain consolidation theory as well as conventional consolidation modelling fail to capture the volume deformation behaviour at constant or very low effective stresses and predict the final settlement of natural clay deposits or slurries with high accuracy. Especially, when the theory is applied to predict

the settlement behaviour of oil sands fine tailings, the result does not match the experimental data (Jeeravipoolvarn et al., 2008, 2009). Since the process of consolidation is an extremely time-consuming process in the oil sands tailings deposits and involves time-dependent effects such as creep and thixotropy, the compressibility relationship used in the finite strain consolidation model for predicting the final settlement is very important and should be representative of the material as much as possible (Jeeravipoolvarn, 2005).

2.6.4 Thixotropic behaviour of unamended oil sands tailings

Typically, sedimentation and self-weight consolidation are the principal dewatering mechanisms in tailings ponds. Although the process of sedimentation is completed within a short time of deposition, self-weight consolidation in FFT is a very complex and extremely time-consuming process. There are many factors that influence the consolidation mechanism; however, the rapid and high thixotropic strength gain of the FFT material warrants the most attention among them as such strength gain affects the rate and magnitude of consolidation of FFT in the long-term (Suthaker and Scott, 1997).

Generally, the physiochemical effect produced by the repulsive-attractive forces acting between the clay particles in a clay-water system is responsible for the development of thixotropy in tailings (Chatterji and Morgenstern, 1990; Miller, 2010). Miller (2010) suggested a relationship between the thixotropic behaviour exhibited by the clay-rich FFT and the physiochemical effect produced by the repulsive-attractive forces. These interparticle repulsive forces (double-layer repulsive forces) and attractive forces (long-range van der Waals attraction forces) are governed by the pore water chemistry and clay mineralogy and denoted by R and A respectively (Chatterji and Morgenstern, 1990; Miller,

2010). Assuming that either R or A or both have a significantly large magnitude in a clay-water system, Mitchell and Soga (2005) expressed the intergranular stress for the particles using a simplified equation:

$$\sigma'_i = \sigma' + (A - R) \dots\dots\dots (2.23)$$

Where σ'_i = true intergranular stress or true effective stress; σ' = skeletal forces (apparent effective stress); A = long-range van der Waals attraction force; and, R = interparticle double-layer repulsive forces. However, when R and A are negligible, the intergranular stress in the above equation becomes equal to the effective stress (Mitchell and Soga, 2005):

$$\text{True intergranular stress } (\sigma'_i) = \text{apparent effective stress } (\sigma') = \sigma - u \dots\dots\dots (2.24)$$

The combined effect of A and R has been defined as the net interparticle stress ($A - R$), which is basically an electrochemical force produced due to the physicochemical contributions from both repulsive and attraction forces (Mitchell and Soga, 2005).

Generally, the repulsive forces acting between the adjacent clay particles in a clay-water system are strongly influenced by the interaction between the adjacent double layers. When there is an increase in electrolyte concentration or cation valence in the system, the ‘thickness’ of the double layer decreases, thereby reducing the range of electrostatic influence relative to the particle surface. Subsequently, the repulsive forces (R) between the clay particles decrease, which allows the attractive forces (A) (mainly van der Waals forces) to become the dominant force, thereby promoting more bonding between the clay

particles. Hence, the interparticle spacings decrease and the intergranular stresses increase, resulting in a higher thixotropic strength for the system, which is typically manifested in higher undrained shear strength (Miller, 2010).

Although the pore water chemistry of FFT has a minor influence on the geotechnical properties, the development of thixotropic strength or gel strength in FFT is governed by the pore water chemistry (Miller, 2010). Since the undrained shear strength in FFT results from the chemical effects, the development of undrained shear strength is most noticeable at the lower effective stress, where the electrochemical interactions between the particles govern the FFT behaviour. However, at effective stresses above 10 kPa, the physical forces dominate the tailings behaviour (Jeeravipoolvarn, 2010).

Although both thixotropic behaviour and consolidation help the FFT materials to gain strength with time, they have adverse effects on each other (Mizani, 2016). Usually, due to the very slow dissipation of excess pore water pressures, effective stresses that develop in the tailings ponds are of very low magnitudes. At such low effective stresses, the thixotropic behaviour of the clay particles in FFT helps in building up bond or gel strength and developing a fines matrix with an overconsolidated structure that affects the rate and magnitude of initial self-weight consolidation (Miller, 2010). Such thixotropic behaviour has also been considered as a disadvantage to the dewatering (Mitchell, 1960). By holding the fine clay particles together, the thixotropic effect prevents them from further compression and release of additional water. That's why, the poor dewatering behaviour and the extremely low rate of consolidation of the FFT are often, in part, attributed to the thixotropic behaviour of tailings (Scott et al., 2013).

The thixotropic behaviour of oil sands tailings was observed both in laboratory and field conditions and an improved interparticle bonding that results from the chemical effects or changes was found to contribute to the development of an overconsolidated structure in tailings (Miller, 2010). In order to promote the rupture of these interparticle bonds in that overconsolidated structure and destroy the gel strength so that a significant consolidation settlement may occur, the tailings deposit must be subjected to vertical effective stress greater than the overconsolidation stress acting upon the deposit (Jeeravipoolvarn, 2005). Typically, the higher the rate of initial self-weight consolidation, the smaller the thixotropic strength gain; however, with time, as the rate of consolidation decreases, the thixotropic strength increases (Banas, 1991; Miller, 2010).

2.6.5 Studies on thixotropy in unamended oil sands tailings

There have been several oil sands tailings studies that shed light on the development of thixotropic gel strength in tailings materials and acknowledged the effect of thixotropic strength buildup on the different aspects of tailings behaviour, especially the compressibility behaviour in long-term (Banas, 1991; Suthaker, 1995; Jeeravipoolvarn, 2005, 2010; Miller, 2010). Suthaker (1995) discussed the change in consolidation behaviour of FFT over time and attributed this change to an ageing effect in FFT that changes the tailings fabric and causes an increase in preconsolidation pressure, which in turn reduces the compressibility of the FFT material. Hence, a single void ratio-effective stress relationship fails to capture the complete consolidation behaviour of FFT and results in an inaccurate interface settlement prediction (Suthaker, 1995). Suthaker and Scott (1997) conducted the cavity expansion tests on oil sands tailings and measured the thixotropic

strength development and showed that the oil sands fine tailings are in fact a highly thixotropic soil. When compared to the typical clays and clay minerals, oil sands tailings were found to develop relatively higher thixotropic strength. Suthaker and Scott (1997) tested five different tailings with different water contents over time. They observed that the thixotropic strength increased with the age of fine tailings and was increasing even after 450 days. They also found a strong inverse correlation between water content and thixotropic strength development. As water content decreased with depth in the tailings ponds, the magnitude of thixotropic strength varied as well, which in turn affected the consolidation behaviour of tailings with depth (Suthaker and Scott, 1997).

When Jeeravipoolvarn et al. (2009) investigated the compression behaviour of untreated oil sands tailings based on the results from the long-term, large-scale, self-weight consolidation tests conducted at the University of Alberta, they developed and used a numerical model to examine the applicability of finite strain consolidation theory on the tailings material in the standpipe and to predict the interface settlement. The theory was found to overestimate the compression behaviour of the tailings by 10% of the total height. Jeeravipoolvarn et al. (2009) theorized that the high thixotropic gain in strength led to an overconsolidation behaviour of the tailings material in the standpipe since the compressibility of this tailings material showed a preconsolidation behaviour at low effective stress of around 0.2 kPa. Earlier, Suthaker (1995) also showed that oil sands tailings could show a preconsolidation behaviour at low effective stress of 1 kPa and behave like an overconsolidated soil; however, when the effective stress was increased to 10 kPa, the overconsolidation effect diminished. The conventional finite strain

consolidation theory was not able to capture such an overconsolidation effect caused by a thixotropic gain in strength in the long-term standpipe test, and therefore, overestimated the consolidation behaviour of FFT (Jeeravipoolvarn et al., 2008, 2009).

When Miller (2010) investigated the effect of extraction water chemistry on the self-weight consolidation of caustic and non-caustic oil sands fine tailings, a thixotropic gain in strength was observed in the caustic fine tailings and such strength development was evident in the flattening of the compressibility curve at low effective stresses (Figure 2.46). Such flattening indicated an enhanced stiffness exhibited by the tailings in response to the compression force applied. The thixotropic strength development in caustic tailings was attributed to its finer particle size distribution and high electrolyte concentration in the pore water. However, such a thixotropic overconsolidation effect was also found to vary depending on the age of the tailings, drainage condition, and ore type (Miller, 2010).

Figure 2.46. Change in compressibility due to the thixotropic effect (from Miller, 2010).

In oil sands tailings, the long-term strength gain can be attributed to either thixotropic effect or a combination of thixotropic and consolidation effect (due to the decrease in water content associated with the self-weight consolidation and the subsequent increase in void ratio or density) (Banas, 1991; Suthaker, 1997; Miller, 2010). Miller (2010) performed shear strength tests on the untreated FFT samples to determine the overall or absolute shear strength and the thixotropic gain in strength with time (relative gain in strength). Miller (2010) used the interpolation techniques developed by Suthaker and Scott (1997) for correcting the influence of self-weight consolidation on the estimation of strength. A typical example of the gain in strength with time including both thixotropic and consolidation effect is shown in Figure 2.47 (Miller, 2010).

Figure 2.47. Shear strength of FFT and correction for consolidation (from Miller, 2010).

The thixotropy of FFT depends on the water content. When the water content is above 150%, thixotropic and consolidation strength development occur simultaneously. However, with the water content decreasing ($<150\%$), the thixotropic strength gain becomes prominent. With water content further decreasing ($<100\%$), the effect of consolidation strength development becomes negligible and the material shows a considerable increase in thixotropic strength (Figure 2.48) (Banas, 1991; Suthaker, 1997).

Figure 2.48. The thixotropic strength development in oil sands tailings with different water contents as measured from the cavity expansion tests (from Suthaker and Scott, 1997).

In addition to the water content, a few other factors including clay mineralogy, rate of loading, self-weight consolidation, and time were found to influence the thixotropy in the FFT directly. Although kaolinite is the most dominant clay mineral in FFT and it is not thixotropic at all, because of the addition of sodium hydroxide (NaOH) as a dispersing agent during the bitumen extraction processes (CHWE processes) and also due to the

presence of bicarbonates and bitumen, FFT becomes thixotropic in nature (Suthaker, 1997; Jeeravipoolvarn, 2005).

For thixotropic materials such as FFT, an increase in loading time could lead to an increase in strength through thixotropic strength gain because of the additional time available for the strength to develop (Jeeravipoolvarn, 2005). However, thixotropic strength gain could vary depending on different axial strains as such strength gain becomes increasingly significant at smaller strains because of the smaller restoration time required and the minimum degree of changes in the microstructure (Seed and Chan, 1957; Jeeravipoolvarn, 2005).

Figure 2.49. Thixotropic strength of FFT (from Suthaker and Scott, 1997).

Since thixotropy is a time-dependent effect, time plays an important role in the thixotropic strength gain in clays and FFT alike. Thixotropic strength was found to develop in FFT within a short time of shearing and increased continuously with age (Figure 2.49)

(Suthaker and Scott, 1997). While performing the lab-scale experiments, the aged fine tailings showed significant overconsolidation behaviour, whereas the younger samples showed minor overconsolidation behaviour (Miller, 2010). Geotechnical tests conducted on the FFT and clay samples at the University of Alberta (i.e. 10-metre standpipe test) also showed that there was a significant increase in shear strength due to the gradual establishment of thermodynamic equilibrium and restoration of microstructure over the course of time. However, when the effects of both creep deformation and thixotropy are considered together, the total effect of an increase in time could be a combination of the following: (a) a decrease in strength since there is adequate time for creep deformation to take place; and, (b) an increase in strength because of the thixotropic strength gain as there is available time for thixotropic strength development (Jeeravipoolvarn, 2005).

2.6.6 Structuration effect observed in fFFT and the methods proposed for quantifying the effect

The large strain consolidation modelling packages have presented ways of approaching the problems of modelling consolidation behaviour of slurries or mine wastes as such modelling packages allow for the inclusion of different material types and different flux and stress boundary conditions, drainage restrictions, etc. (Priestley et al., 2011). Since polymer amended FFT materials show similar behaviour, some forms of large strain consolidation theory have been used to analyze the complex mechanisms that control the permeability and consolidation of polymer amended FFT (Caldwell, 2014). Although polymer-aided initial dewatering and short-term sedimentation/consolidation have been studied extensively and modelled theoretically for the industrial dewatering processes, a

comprehensive understanding of the long-term settling, consolidation, and stabilization of oil sands tailings in the presence of polymers are yet to be established (Das et al., 2009). Field trials have already demonstrated that polymers can increase the solids content (by mass) considerably within a very short span of time following the deposition. However, whether flocculated tailings would show the same dewatering potential in the long-term is yet to be investigated (da Silva, 2011). For adequate densification and shear strength gain, in addition to adding polymer into the pipeline, deposited FFT also depends on further consolidation and environmental dewatering mechanisms (OSTC, 2012).

When the long-term dewatering and consolidation behaviour of the FFT samples flocculated with a high molecular weight anionic polymer were studied in a lab-scale experiment, a change in the compressibility behaviour was observed over time, which was thought to be due to the time-dependent structuration effect and found to influence the volume change behaviour (Salam et al., 2017, 2018a, 2018b). The experimental data generated from the column dewatering tests (using 10 cm tall columns) demonstrated that the time-dependent phenomena observed in fFFT play a significant role in the dewatering and consolidation performance of the polymer amended tailings deposits in longer-term, at least in some types of oil sands tailings (Salam et al., 2017, 2018a, 2018b).

Recently, the data reported from a pilot study of the deposition of FFT (in-line flocculation with an anionic polymer) showed evidence of time-dependent compressibility and structuration (Narges et al., 2019). The FFT deposit had an initial height of 11 metres (approximately). The in-situ effective stresses were estimated based on the in-situ measurements of pore water pressure and density. When these effective stresses were

plotted against the corresponding in-situ void ratios, the results suggested time-dependent compressibility effects in the FFT deposit (Figure 2.50) (Narges et al., 2019), which was similar to the findings from the longer-term laboratory-scale studies of fFFT samples in the 10 cm tall columns (Salam et al., 2017, 2018a, 2018b).

A field vane was used for measuring the peak and remoulded shear strength as well as the development of sensitivity (the ratio of peak to remoulded shear strength) (Figure 2.51). The sensitivity is used as an indicator of structuration/thixotropy in clays (Skempton and Northey 1952; Leroueil and Vaughan, 1990). Typically, the higher the sensitivity, the larger the structuration/thixotropic effect. The 11-metre deep fFFT showed an indication of the structuration effect as the sensitivity values increased with time (Narges et al., 2019).

Figure 2.50. In-situ measurements of the compressibility curve in an 11-metre deep pilot study of consolidation of polymer amended FFT (from Narges et al., 2019).

Figure 2.51. The ratio of peak to remoulded shear strength (sensitivity) in an 11-metre deep pilot study of consolidation of polymer amended FFT (from Narges et al., 2019).

Structuration effect has been observed in natural clays, dredged sediments, and in at least some oil sands tailings deposits. A careful review of the experimental data concerning structuration behaviour in natural clays (i.e. Burland, 1990), the results from the experimental investigation of the long-term dewatering and consolidation behaviour of fFFT (Salam et al., 2017, 2018a, 2018b), and the data reported from the pilot study (Narges et al., 2019) indicated that there are similarities in the time-dependent effects observed in fFFT and natural clays. However, the study of the structuration effect in the FFT deposits is especially important since such time-dependent behaviour may change the compressibility of the deposited materials over time, and subsequently, reduce the settlement and residual strength gain (in comparison to the values obtained from a conventional large strain consolidation analysis). At the same time, the peak undrained

strength of the tailings for a given density may increase. Thus, the structuration effect may have positive as well as negative implications for tailings management (Narges et al., 2019). Therefore, it is crucial to develop some models or methods for predicting the degree of structuration for a particular tailings-polymer mixture or an fFFT deposit; such knowledge may allow the operators to undertake appropriate strategies for minimizing the negative effects of structuration (Narges et al., 2019).

Burland (1990) generalized the compression behaviour of reconstituted clays and proposed the concept of intrinsic properties of reconstituted soils (or intrinsic compression concept) for evaluating the influence of soil structure on its compressibility. Later, Liu and Carter (1999) investigated the compression behaviour of both reconstituted and natural clays. Based on the intrinsic compression concept (Burland, 1990), Liu and Carter (1999) recommended the use of additional void ratios sustained by the natural clays as a direct indicator of soil structure since such additional void ratios reflect the difference in compression behaviour between a structured soil and the corresponding reconstituted soil.

Liu and Carter (1999) introduced a hyperbolic relationship for describing the additional void ratio sustained by the naturally structured soil and for predicting its compression behaviour in terms of preconsolidation pressure and structure index (S). They found that the increase in the void ratio of undisturbed, structured soil at the preconsolidation pressure relative to the void ratio of the corresponding reconstituted soil at the same effective stress could be linearly related to the preconsolidation pressure, as shown in Figure 2.41, where $\Delta e = S \ln \sigma'_p$ at the preconsolidation pressure. The structure index (S), a soil parameter introduced by Liu and Carter (1999) was given by the increase

in the void ratio above the reconstituted compressibility curve divided by the natural logarithm of the vertical effective stress (Liu and Carter, 1999).

Liu and Carter (1999) reviewed twenty case studies of the compressibility of reconstituted and natural clays. They compared the experimentally determined compression curves of structured clays to the theoretical compression curves that were constructed based on their proposed relationship. The theoretical compression curves were found in good agreement with the experimental compression curves (Liu and Carter, 1999).

Recently, Qi et al (2018) applied the concept of Liu and Carter (1999) to the compression curves obtained from the compression tests that were performed on the fFFT samples (Salam et al., 2017, 2018a, 2018b). These fFFT samples were thought to be structured based on the findings from other experiments conducted on them. From the structured sample's compressibility curve, Qi et al (2018) estimated a preconsolidation pressure of 100 kPa. When Liu and Carter (1999) estimated the preconsolidation pressures in naturally structured clays from the available compression tests data (for constructing theoretical compression curves), almost 90% of the preconsolidation pressures measured were less than 200 kPa, which was not significantly different from the preconsolidation pressures measured from the compression curves obtained from the compression tests performed on fFFT. Qi et al. (2018) considered the compressibility curve obtained from an independent large strain consolidation (LSC) test as the representative compressibility curve for the reconstituted or remoulded tailings samples since the properties determined from an LSC test would be more representative of an intrinsic state of tailings in comparison to the properties measured from the column test compressibility curve. After

comparing the curves (Figure 2.52), Qi et al. (2018) found a difference of 0.6 in the void ratio between the reconstituted and structured compressibility curves, which implied that an additional void ratio of 0.6 was sustained by the structured tailings (Qi et al., 2018).

Figure 2.52. Compressibility curves of differently aged fFFT samples (from Qi et al., 2018).

When Liu and Carter (1999) estimated the S values based on the preconsolidation pressures determined from the experimental compression curves, 90% of S values were below 0.2, while the average value was 0.12. For a structured flocculated tailings sample with a preconsolidation pressure in the range of 50 ~ 100 kPa, Qi et al. (2018) estimated the S to be 0.13 and 0.10 (for 50 kPa and 100 kPa respectively) using the difference in void ratio (Δe) between the intact and remoulded samples at 112 days ($\Delta e = S \ln \sigma'_v$ at the preconsolidation pressure) (Figure 2.53). Therefore, the degree of structuration in polymer-

amended fines dominated oil sands tailings appeared to be in the same order as the structuration measured in natural clay deposits.

Figure 2.53. Undisturbed/remoulded compression curves from 112-day old fFFT samples.

Zeng et al. (2016) proposed an empirical equation (equation 2.18) to determine the yield stress developed in the high-water content dredged clays due to ageing. When Zeng et al. (2016) compared the predicted YSR values (the ratio of vertical yield stress to effective vertical stress, estimated using equation 2.18) with the experimentally obtained YSR values for dredged clays, predicted results were found in good agreement with the experimentally obtained values. Recently, Narges et al. (2019) suggested using a simple rate law in combination with the empirical model proposed by Zeng et al. (2016). The viscosity bifurcation models in the rheology of clay suspensions (i.e. Coussot et al., 2002), which simulate the buildup of thixotropic strength at small time scales in the clay

suspensions are governed by the simple rate laws, which typically balance the breakdown of the network structure under the shearing action with an ageing term. An example of such a rate law is:

$$\frac{d\lambda}{dt} = a(\lambda_{max} - \lambda) - b\gamma \dots\dots\dots (2.25)$$

Where λ is the structure parameter; γ is the strain rate; t is time; and, a and b are the parameters (Narges et al., 2019).

Assuming that the preconsolidation pressure is linearly related to the structure parameter in an ageing sample, equation 2.25 can be used to predict a decreasing rate of ageing up to some maximum value, but only below a critical strain rate, which is a function of the ratio of a and b . When the thixotropic shear strengths developed in the replicate 10 cm tall columns were measured using a fall cone and plotted against the time, the onset of ageing was also observed to occur below a similar critical strain rate. Hence, the increase in preconsolidation pressure at a constant but low strain rate was predicted using the available fFFT data with equation 2.25 and was also compared to the prediction made based on equation 2.18 and the measured data (Figure 2.54). Both equations were found to replicate the measured data. Although equation 2.18 predicted an unbounded increase in preconsolidation pressure (Figure 2.55), such equation and other databases were thought to be beneficial in guiding parameter selection for equation 2.25 (Narges et al., 2019).

Figure 2.54. Zheng et al. (2016) model (equation 2.18) and strain rate limited model (equation 2.25) for predicting the rate of structuration compared to the measured data (from Narges et al., 2019).

Figure 2.55. Comparison of two different models in log time (from Narges et al., 2019).

2.7 Summary of the literature review and knowledge gaps

The slow settling of fine clay particles in oil sands residuals takes decades in tailings ponds and results in the accumulation of large volumes of fluid fine tailings over time, which poses the biggest challenge in tailings management. Adding polymers to the tailings can induce the flocculation of clay particles, and therefore, can improve the dewaterability of tailings, especially, over a short time frame. However, for a given FFT, the efficiency as well as the economy of the dewatering operations mainly depend on operating at the optimum polymer dosage and mixing conditions. Nevertheless, there is not much information in the literature on the impacts of such process variables on the flocculation and dewatering performance of tailings. Since polymer costs constitute a substantial portion of the tailings dewatering operation costs, further research is needed to develop an advanced and automated process control technology to quantify the dewatering efficacy immediately after the mixing of polymer with the tailings, identify the optimum polymer dosage, and correct or adjust the polymer dosage in real-time or near real-time. There are similarities between the sludge in wastewater treatment and the fluid fine tailings in oil sands. There is a substantial body of research on new and novel approaches to sludge dewatering, and some of this research can potentially be adapted to oil sands tailings.

Since the polymer amended tailings are especially sensitive to shearing, the mixing intensity and duration and the shear caused during pipeline transportation can disintegrate the flocs that are formed during the flocculation, and therefore, reduce the yield stress as well as dewatering potential of the flocculated tailings. However, when the same tailings are sitting idle in the pipelines or subjected to relatively low shear stresses, they start to

regain their original structure and stiffness through the restructuring of the particle network system, which is a time-dependent ageing effect (Soleimani et al., 2013; Mizani et al., 2017). A similar increase in stiffness has been observed in naturally structured clays during creep compression and attributed to the inherent ageing effects such as stronger physical-chemical bonding between the particles (Anderson and Stoke, 1978; Lohani et al., 2001).

Since the tailings go through a transition between the liquid-like behaviour and solid-like behaviour during their transportation through the pipelines and the post-deposition stacking, they exhibit both elastic and viscous behaviour or the viscoelasticity. Although the disintegration of flocs (destruction) under high shear during pipeline transportation of polymer amended tailings reduces the benefits of polymer application, an almost full recovery of the structure following high shear helps to regain most of the benefits (Mizani et al., 2017). For achieving the right balance between the dewaterability/strength and the mixing intensity/duration so that the flocculated tailings could attain an optimum dewaterability potential and strength under field conditions, it is necessary to conduct advanced rheological tests to simulate the pipeline transportation of tailings flocculated at different polymer dosages and investigate the effects of pipeline transportation (i.e. pipeline length and diameter) and mixing conditions (i.e. intensity and duration) on post-deposition dewatering and strength gain behaviour and the ageing effect.

Field trials have shown that polymers allow the dewatering technologies to increase the solids content to at least 45 ~ 55% within 48 hours following deposition (Matthews et al., 2011; Wells, 2011). However, for adequate densification and shear strength gain of the tailings deposits, the oil sands operators rely on the long-term dewatering mechanisms such

as consolidation, evaporation, and freeze-thaw, which typically work over a longer time frame. The consolidation of tailings is a time-dependent process that helps the tailings deposits to dewater and strengthen through compression or densification and a reduction in void space (Kaminsky et al., 2015). Although void ratio-effective stress relationship and hydraulic conductivity-void ratio relationship are the two most important properties that govern the consolidation behaviour of tailings, unamended tailings have been found to demonstrate time-dependent creep and thixotropic behaviour, which could affect the consolidation behaviour of tailings significantly (Jeeravipoolvarn, 2005; Jeeravipoolvarn et al., 2009; Miller, 2010).

Creep is any change in volume that occurs over time independent of the changes in effective stress. Creep has also been observed in many natural clays. Such viscous deformation typically manifests as a time-dependent compressibility curve, where the apparent preconsolidation pressure shifts with the strain rate. Thixotropy is a time-dependent reorganization of the fine clay particles due to the electrochemical forces acting between the particles in a clay-water system (Mitchell, 1960). The thixotropic effect in clays is most evident in the time-dependent strength recovery of clays after remoulding at constant water content and density (Skempton and Northey, 1952). Thixotropic strength gain over time has also been reported in unamended tailings (Jeeravipoolvarn, 2005; Jeeravipoolvarn et al., 2009; Miller, 2010). The thixotropic behaviour of the fine clay particles was thought to be the driving force in building up a bond or gel strength and developing a fines matrix with an overconsolidated structure that affects the rate and magnitude of initial self-weight consolidation in tailings, especially, at low effective

stresses (Miller, 2010). That's why, the poor dewatering behaviour and the very slow consolidation rate of the tailings deposits are often, in part, attributed to the thixotropic behaviour of the clay particles in tailings (Scott et al., 2013). However, when the effects of both creep compression and thixotropic strength gain are considered together for a tailings deposit, the total effect of an increase in time could be a combination of the following: (a) a decrease in strength since there is adequate time for creep deformation to take place; and, (b) an increase in strength because of the thixotropic strength gain as there is available time for thixotropic strength development (Jeeravipoolvarn, 2005).

Many natural clays exhibit a time-dependent behaviour called structuration, which is typically manifested in an increase in apparent preconsolidation pressure independent of the changes in density (Locat and Lefebvre, 1986; Leroueil and Vaughan, 1990; Burland, 1990; Liu and Carter, 1999; Delage, 2010; Sorensen, 2006). Locat and Lefebvre (1986) have defined the structuration in the soil as the amount of strength or stiffness that the soil has gained through a combination of time-dependent processes in soil. There exist many experimental data in the literature concerning the structuration effect in clays (Burland, 1990; Leroueil and Vaughan, 1990). Several researchers reported structuration in clays based on their findings from the long-term oedometer tests (Leonards and Ramiah, 1960; Leonards and Altshaeffl, 1964). When the samples were allowed to rest under constant effective stress for a period, a time-dependent volume change behaviour (a creep compression) was observed. During the same period, the samples also showed a significant increase in yield stress and developed a structural resistance against further compression. Such an increase in yield stress was found greater than the increase that was expected from

a change in volume due to creep compression, which distinguished the effect of structuration on structural strength development in clays from the effect of creep (Leroueil and Vaughan, 1990). Such a structuration effect was attributed to the thixotropic behaviour of clays that involved the development of cohesive bonds between the particles under creep conditions (Leonards and Ramiah, 1960; Leonards and Altshaeffl, 1964).

Because of structuration, most natural clays in their undisturbed or intact state show higher preconsolidation pressure and lower compressibility relative to the reconstituted or remoulded samples prepared from the same clays, which also makes the undisturbed or intact samples stiffer in comparison to the corresponding reconstituted or remoulded samples, especially at low stress levels (Locat and Lefebvre, 1986; Burland, 1990; Leroueil and Vaughan, 1990; Delage, 2010). Quigley and Thompson (1966) studied the structuration in Leda clay and showed that naturally structured clays are able to sustain greater vertical effective stress than the reconstituted or remoulded samples at the same void ratio owing to a stronger interparticle bonding and more stable particle arrangement. Naturally structured clays also develop an undrained shear strength greater than the strength of the corresponding reconstituted or remoulded samples at the same void ratio (Bishop et al., 1965).

Although little quantitative information is available in the literature on the time-dependent behaviour of polymer amended tailings, a careful review of the experimental data concerning the structuration behaviour in natural clays (i.e. Burland, 1990; Leroueil and Vaughan, 1990), the results from the experimental investigation of the long-term dewatering and consolidation behaviour of fFFT (Salam et al., 2017, 2018a, 2018b), and

the field data reported from a pilot study of the consolidation of fFFT in 11-meter deep deposits (Narges et al., 2019) indicated that there are similarities in the time-dependent effects observed in fFFT and natural clays. The polymer amended tailings showed evidence of time-dependent compressibility and structuration.

Since the performance of the tailings deposits depends on their consolidation and structuration effect may change the compressibility of deposited tailings over time, and subsequently, reduce the settlement, it is crucially important to be able to predict the degree of structuration in flocculated tailings. Although the differences in naturally structured clays and remoulded clays are widely studied, relatively little research has been conducted on the rate of structuration (or restructuring) in literature. Although creep is commonly modelled through the concept of rate-dependent compressibility, the conventional large strain consolidation models do not incorporate the effects of thixotropy or structuration in soft soils or deep deposits such as tailings deposits. It is unknown whether such time-dependent behaviour influences the long-term dewatering and consolidation performance of polymer amended tailings deposits, and we also do not know whether different polymer dosages and different polymer mixing methods have different effects on the structuration. Nevertheless, the study of the structuration effect in polymer amended tailings deposits is especially important since such knowledge bears on the optimization of tailings treatment, deposition, and management plans. It warrants an extensive investigation and assessment of the time-dependent phenomena in flocculated FFT to incorporate the effects of time-dependent phenomena into the large strain consolidation theory, reformulate the governing equations, and modify the numerical models.

Chapter 3: Material and methods

3.1 Oil sands tailings

Oil sands tailings samples were collected from a tailings pond in Northern Alberta, Canada, and shipped to Carleton University in Ottawa, Canada. They were received in large containers. Prior to the collection of fluid fine tailings from the container for conducting the experiments, to restore the tailings to their original composition and regain their initial solids content, the tailings inside the container were mixed again using a drum mixer (Model Number: DLM150VGD, Mixer Direct). Tailings were mixed continuously for 24 hours at an average speed of 129 rpm. For optimal mixing, the direction of the rotating blades was reversed every 2 hours by alternating between the forward and the backward mode. Tailings were then transferred into 20-litre buckets for conducting laboratory tests and investigations.

3.2 The properties of fluid fine tailings

Different laboratory tests and analyses were performed on the tailings to determine their physical, mineralogical, and chemical characteristics. Another objective of performing the characterization tests was to establish a baseline characteristic of the unamended FFT so that the changes in their properties (after adding polymers) and the changes occurring over a longer time frame could be detected and compared. The test results also helped to compare the properties of the FFT used in this study with the properties of the FFT used in other studies. The following table shows the difference in the properties of the FFT used and the FFT properties available in recent literature.

Table 3.1: Geotechnical properties of unamended FFT (compared with other FFTs).

Property	Unit	ASTM standard	Current study (2020)	Mizani (2016)	Yao (2016)	Beier (2015)	Bajwa (2015)	Miller (2010)
Solids content	%	D2216-19	31.00	33.37	32.00		42.00	
Water content	%	D2216-19	222.58	199.60	213.0		140.0	
Liquid limit	%	D4318-10	60.0	62.4	48~61	50.0	45.0	49.5~59.6
Plastic limit	%	D4318-10	28.0	26.7	26~29	21.0	19.0	25.8~30.5
Fines content	%	D7928-16	71.0	78.7	90~93	96.0		92~99
Clay content	%	D7928-16	39.0	30.0	45~50	52.0	40.0	41~49
Bitumen content	%	D95-05	1.41	2.90	1.30	3.00	3 ~ 4	0.35~0.50
Total bulk density	g/cm ³	D7263-09	1.20	1.20	1.21		1.20	1.05~1.06
Specific gravity		D854-10	2.12	2.20	2.30	2.44	2.22	2.45~2.55
Void ratio		D7263-09	5.10		4.90		3.17	

FFT properties such as solids content (C_s), gravimetric water content (GWC), specific gravity (G_s), liquid limit (LL), plastic limit (PL), void ratio, particle size distribution, and total bulk density were measured in the laboratory.

3.2.1 Solids content (C_s)

Solids content (C_s) is the ratio of the mass of solids in tailings (the non-water portion of tailings, which also includes bitumen and other minerals) to the total mass of tailings. A measurement of solids content helps to determine the solids-water composition of FFT (Miller, 2010). For the solids content measurement, samples were collected from the FFT in replicates and they were dried in an oven at a temperature of 105°C for 24 hours to remove the water part. The solids content was then calculated (for each sample) using the following equation (Mizani, 2016):

$$C_s = \frac{M_s}{M_T} \times 100 \dots\dots\dots (3.1)$$

Where C_s = solids content, expressed in %; M_s = mass of solids in tailings, measured after oven-drying; and, M_T = total mass that included both water and non-water portions, measured before oven-drying.

The average of the solids contents of the replicate samples was taken as final solids content.

3.2.2 Gravimetric water content (GWC)

Gravimetric water content (GWC) is the ratio of the mass of water in tailings to the mass of solids (the non-water portion of tailings, which also includes bitumen and other minerals). It is measured by drying the FFT sample in an oven at a temperature of 105°C for 24 hours (for removing the water portion of FFT), and then, by using the following equation (Mizani, 2016):

$$C_w = \frac{M_w}{M_s} \times 100 \dots\dots\dots (3.2)$$

Where C_w = gravimetric water content, expressed in %; M_s = mass of solids in tailings, measured after oven-drying; and, M_w = mass of water in tailings, estimated from the difference between the total mass of tailings (M_T) (which included both water and non-water portions of tailings and was measured before the oven-drying) and the mass of solids in tailings (M_s) (which was measured after oven-drying, after the removal of water).

3.2.3 Specific gravity (G_s)

For soils, the specific gravity (G_s) is the ratio of the mass of a given volume of solids in the soil to the mass of an equivalent volume of standard liquid. Typically, distilled water at standard temperature (4°C) and pressure are used as the standard liquid. Hence, specific gravity (G_s) is expressed by the following equation:

$$G_s = \frac{\rho_s}{\rho_w} \dots\dots\dots (3.3)$$

Where ρ_s = unit weight of soil solids; and, ρ_w = unit weight of distilled water at 4°C.

The specific gravity of the unamended FFT was determined according to the ASTM standard of ASTM D 854 – 10.

3.2.4 Liquid limit (LL)

It is a common practice to use the Atterberg limits of cohesive soils to identify, describe, or classify them or to determine their mechanical properties (Mitchell and Soga, 2005). According to Casagrande (1932b), for a fine-grained soil, the liquid limit is the water content at which the soil has an undrained shear strength of about 2.5 kPa. However,

later studies suggested that the liquid limit corresponds to an undrained shear strength in the range of 1.7~2.0 kPa (Wroth and Wood, 1978; Wood, 1985; Mitchell and Soga, 2005).

Generally, two types of devices, namely, Casagrande liquid limit device and fall cone device are used for determining the liquid limit values. The fall cone device is often preferred over the Casagrande device because it provides better repeatability and a less significant variation even when different devices are used by different operators. The method is also simple and economic (Houlsby, 1982; Leroueil and Le Bihan, 1996; Feng, 2000). Moreover, several studies (Keedwell, 1984; Koumoto and Houlsby, 2001) suggested that the method is suitable for the determination of the liquid limit of slurry type materials (Bajwa, 2015). Bajwa (2015) and Mizani (2016) used the fall cone method to determine the liquid limit of oil sands fine tailings.

Hansbo (1957) was the first one to develop the theoretical background of the fall cone test; later, Houlsby (1982) and Koumoto and Houlsby (2001) discussed the theory and the practice of fall cone test in detail. In this method, initially, a metal cone of a known weight and a known apex angle (which is fixed to a sliding shaft suspended vertically) is positioned in such a way that the tip of the cone just touches the surface of a sample but does not penetrate it. Then, the cone (along with the vertically sliding shaft) is released and allowed to drop freely into the sample under its own weight and the resulting penetration depth is measured. The measured penetration depth can be used to determine the liquid limit and plastic limit of the sample as well as its undrained shear strength (Houlsby, 1982; Leroueil and Le Bihan, 1996). Depending on different Geotechnical Engineering Standard organizations, there are several fall cone methods for determining the Atterberg limits.

They basically differ in terms of apex angle and the mass of the cone used and the depth of penetration associated with the liquid limit (Leroueil and Le Bihan, 1996).

3.2.5 Plastic limit (PL)

Feng (2000, 2001) proposed a linear logarithmic penetration depth versus logarithmic water content model for the determination of the plastic limit from the fall cone test results (Figure 3.1). The model is based upon the assumption that the cone penetration is 20 mm at the liquid limit (British Standard) and the undrained shear strength at the plastic limit is about 100 times the undrained shear strength at the liquid limit (from the data of Skempton and Northey, 1953). Hence, the plastic limit (PL_{100}) is determined using the following equation (Feng, 2000, 2001, 2004):

$$PL_{100} = c(2)^m \dots\dots\dots (3.4)$$

Where c = water content at the penetration depth of 1 mm; and, m = the absolute slope of the best-fit straight line drawn.

For clays, both parameters c and m can be determined experimentally by performing a series of fall cone tests on several samples (at least four) with varying degrees of water content (which would result in cone penetration depths of 3 ~ 25 mm) and analyzing the results. In equation 3.4, the value of 2 refers to a cone penetration depth of 2 mm (Harrison, 1988). When cone penetration depths are between 3 and 25 mm, the log-log plot (the logarithmic of water contents versus the logarithmic of fall cone penetrations) displays a linear relationship for any soil. For a British Standard fall cone, if this linear relationship is extended backwards, the intercept of the best-fit straight line with the water

content axis (y-coordinate) at the penetration depth of 2 mm would represent the plastic limit of the soil (Feng, 2000, 2001, 2004).

Figure 3.1. A linear logarithmic penetration depth versus logarithmic water content model for the determination of plastic limit (from Feng, 2001).

The numerical difference between the liquid limit and the plastic limit refers to the plasticity index (PI), which in turn refers to the range of water contents where the material shows plastic properties while transitioning from a semisolid state to a liquid state. According to the Burmister (1949) Classification of plasticity index, the plasticity index in the range of 10 ~ 20 indicates medium plasticity, whereas plasticity in the range of 20 ~ 40 refers to high plasticity. Generally, for clays, a high plasticity index indicates that the clays have high activity, and therefore, high cation exchange capacity (Kaminsky, 2008), and also, a higher undrained shear strength (Bjerrum, 1967).

3.2.6 Bitumen content

The bitumen content of the unamended FFT was determined by the Dean-Stark distillation extraction method (ASTM D95 – 05), and was expressed in terms of total mass (which includes water, solids, bitumen, and other minerals) using the following equation:

$$\text{Bitumen content} = \frac{M_b}{M_T} \times 100 \dots\dots\dots (3.5)$$

Where M_b = mass of bitumen in the FFT sample (in grams); and, M_T = total mass of FFT (in grams).

3.2.7 Void ratio

The void ratio (e) is a dimensionless quantity in soil mechanics that denotes the ratio of the volume of voids in soil to the volume of solids or soil particles. It is estimated using the following equation:

$$e = \frac{V_v}{V_s} \dots\dots\dots (3.6)$$

Where V_v = volume of voids in soil (empty/filled with water); and, V_s = volume of solids.

In saturated soil, all the voids are filled with water; however, in unsaturated soil, the voids are partially filled with air (Mizani, 2016).

3.2.8 Particle size distribution (fines content)

The particle size distribution of unamended FFT was determined through a combination of wet sieving and sedimentation (hydrometer) analysis (ASTM D7928 – 16).

Typically, the results from the hydrometer analysis are combined with the results from a sieve analysis for evaluating the fine-grained portion of a soil that has a wide range of particle sizes. The results are presented as the mass of percent finer versus the log of the particle diameter (Appendix B.1).

3.2.9 Total bulk density

The total bulk density or the wet bulk density of a soil is the ratio of the total mass of soil (that includes both solids and liquids) to the total volume of soil. It can be expressed by the following equation:

$$\rho = \frac{M_s + M_w}{V_T} \dots\dots\dots (3.7)$$

Where M_s = mass of the solids in FFT; M_w = mass of the liquids in FFT; and, V_T = volume of FFT.

3.2.10 Clay content (Methylene blue index test)

The methylene blue index test was performed to measure the clay content in unamended FFT sample, 7-day old flocculated FFT sample, and 42-day old flocculated FFT sample (following the method developed by Sethi, 1995, as a modification from the ASTM C837 – 99 standard test method for methylene blue index of clay). Methylene blue is a cationic dye with a strong affinity for the clay surfaces. It absorbs strongly to the clay surfaces and displaces the cations already present on the surfaces (such as Na^+ , Ca^{2+} , K^+ , and Mg^{2+}) (Kaminsky, 2014). The methylene blue index (MBI) of FFT was calculated based on the number of millilitres of methylene blue solution that was added to the FFT

sample during the test and absorbed a given mass of sample, and it was expressed in meq/100g (milliequivalents of methylene blue per 100 g of FFT sample), which was estimated by multiplying the number of millilitres of methylene blue used to titrate 100 g of sample by the molarity of methylene blue used (Kaminsky, 2014). Then, the percentage of clay in FFT was determined using the following empirical relationship, which was proposed by Sethi (1995):

$$\% \text{ Clay} = \frac{MBI \frac{meq}{100g} + 0.04}{0.14} \dots\dots\dots (3.8)$$

3.3 Preparation of polymer stock solution

An anionic polyacrylamide (PAM) flocculant (A3338) was used for the rapid dewatering of FFT samples. A3338 is a branched polymer with an average molecular weight of 18×10^6 g/mol (commercially available from SNF). In a plastic weighing dish, 4g of A3338 polymer was weighed using an analytical balance (Fisher Scientific, Sartorius AG Germany, LE225D) and decanted into a 1000 mL glass beaker and completed to 1000 mL with deionized water to prepare a polymer stock solution with a final concentration of 0.4%. The polymer solutions were stirred using a jar tester (Phipps and Bird, USA) at 200 rpm for 5 minutes and at 125 rpm for the following 55 minutes. Then the polymer solution was mixed with a hand blender for 10 seconds and left for maturation for 1 hour.

3.4 Preparation of flocculated FFT

To prepare the flocculated tailings, initially, a four-blade impeller (with a radius of 8.5 cm) attached to an overhead mixer (IKA EUROSTAR 60 control) was immersed in 2000 mL of FFT in a 5-litre pail. The mixing was initiated at a constant speed of 250 rpm

and continued for 15 minutes to make the tailings homogenous. Then, the required amount of 0.4% polymer stock solution was added to the FFT by injecting the solution close to the rotating impeller during mixing (the required amount was estimated based on the solids content of the unamended FFT). All the polymer stock solution was injected as fast as possible and in one shot into the FFT sample. Starting from the moment the polymer stock solution was injected into the FFT, the mixing was continued for 20 seconds.

The mixing method was originally adapted from a baseline procedure developed by Shell Canada in 2010 for replicating the in-line flocculation and producing field representative samples while conducting the laboratory-scale experiments. Later, Mizani (2016) optimized the procedure based on the measurements of short-term dewaterability (obtained from the capillary suction time tests and settlement tests) and yield stress. The FFT and the polymer stock solution were mixed at different speeds and the mixing was continued for different durations; subsequently, the polymer-FFT mixtures were tested for the dewaterability potential (capillary suction time test) and settleability (graduated cylinder column test). The test results indicated that flocculated tailings products that were produced by mixing at a speed of 250 rpm for 10 seconds would result in optimum dewatering behaviour and settling performance (Mizani, 2016). The rheology tests performed on the freshly mixed FFT-polymer mixtures and the yield stress measurements confirmed 250 rpm to be the optimum mixing speed. Mixing speeds above 250 rpm were found to cause over-shearing of the material, whereas mixing speeds lower than 250 rpm resulted in inadequate mixing (Mizani, 2016).

While the current research project was in progress, the mixing method was optimized again based on the sensitivity of the fFFT to the duration of shearing. The settleability of the FFT-polymer mixtures were determined from the short-term settlement tests (48 ~ 72 hours) conducted in graduated columns (K. Vandara, personal communication, February 21, 2018). The results indicated that mixing at a speed of 250 rpm for 20 ~ 30 seconds would result in optimum settling performance (Figure 3.2).

Figure 3.2. The results from the settlement tests using different mixing times (from Vandara, 2018).

Recently, Aldaeef and Simms (2019) proposed a quality control technique for improved flocculation of FFT, which was based on the relationship observed between the torque force developed during flocculation in a Couette rheometer and the quality of the flocculation (measured in terms of capillary suction time and settlement rate). Torque forces reached the peak after 15 ~ 25 seconds of polymer injection (Figure 3.3) and peak

torque values were thought to be associated with the formation of larger flocs as the rheometer was required to exert relatively greater energy for shearing the larger flocs. When such peak torque force readings measured and recorded during the process of flocculation were plotted against the capillary suction time values and the percentages of settlement (in the graduated columns, after 7 days) obtained from different flocculated samples, the capillary suction time values and the percentages of settlement were found to decrease and increase respectively with the peak torque forces increasing, and therefore, the peak torque forces indicated a better dewatering and settlement behaviour in the short-term (Figures 3.4 and 3.5). Additional mixing time resulted in a sharp reduction in the torque force, which was attributed to the over-shearing and the breakdown of the flocs (Aldaeef and Simms, 2019). Hence, based on the above observations and findings, mixing at a speed of 250 rpm for 20 seconds was adopted as the regular mixing method.

Figure 3.3. Torque development during flocculation (from Aldaeef and Simms, 2019).

Figure 3.4. The correlation between the dewaterability (CST values) and the peak torque forces (from Aldaeef and Simms, 2019).

Figure 3.5. The correlation between the settlement (%) and the peak torque forces (from Aldaeef and Simms, 2019).

Additionally, a second polymer mixing method was trialled, which was adapted from the Northern Alberta Institute of Technology (NAIT). In this method, a predetermined volume of the polymer stock solution was mixed with the FFT at a mixing speed of 320 rpm and the mixing was continued for 10 seconds after adding the polymer stock solution. Throughout this mixing period (10 seconds), the FFT was conditioned by holding a metallic spatula inside the pail. After 10 seconds of mixing, the mixing speed was reduced to 20 rpm. For the next 10 seconds, the conditioning by holding a metallic spatula was continued at a speed of 20 rpm. This additional step of conditioning was designed to ensure proper mixing and optimum flocculation of the FFT material.

Figure 3.6. Preparation of fFFT by mixing unamended FFT with a polymer solution.

3.5 Methods

This study investigated the influence of polymeric flocculation on the properties of FFT over time and also evaluated how the time-dependent phenomena observed in fFFT influenced its short-term as well as long-term dewatering and consolidation behaviour. In addition, methods were developed for the real-time or near real-time determination of optimum polymer dosage for the dewatering of tailings. A combination of tests and analyses including column dewatering test (using columns instrumented with pore water pressure sensors at different depths), conventional and modified consolidation test, fall cone test, oscillatory rheometry, low and high vacuum scanning electron microscopy, optical microscopy, and mercury intrusion porosimetry were employed to track the fundamental changes in the flocculated FFT materials. Several dose-response tests including capillary suction time (CST) test and graduated column settling test were performed for the determination of the optimum polymer dosage and for evaluating the flocculation performance.

Figure 3.7. A schematic diagram of the test methodologies used for the research.

3.5.1 Pore water pressure measurement

Column dewatering tests (using 10 cm tall columns instrumented with pore water pressure sensors at different depths) were conducted to investigate both consolidation and non-consolidation volume change behaviour and strength development properties in flocculated FFT in longer-term. The pore water pressure refers to the pressure of water held within the gaps between the particles or pores in soils (it is also known as soil matric suction or soil matric potential). The development of pore water pressure in the clay-water systems has been attributed to the combined action of the hydrostatic pressures arising from the mechanical effects, applied loads, and the interparticle forces that originate from the surface activity of the clay particles and produce physicochemical effects (Mitchell, 1962).

The pore water pressure sensors used for this study were the electric transducers capable of measuring the soil matric potential in the range of less than 100 kPa. A pore water pressure sensor comprises a porous, permeable ceramic cup connected to a vacuum gauge or a pressure measuring device through a water-filled plastic tube. When this ceramic cup is buried in the soil, the pore water comes into equilibrium with the water inside the plastic tube and the vacuum gauge (which acts as a reference pressure indicator), or the suction created inside the tube and vacuum gauge becomes equivalent to the soil matric potential as water flows through the porous ceramic cup. Thus, the pore water pressure is measured (Kirkham, 2014). The measurement of pore water pressure using pore water pressure sensors in combination with a data logger could help in monitoring the dissipation of excess pore water pressure in a soft soils or tailings deposit, which is also an indication of the progress in consolidation (Soleimani et al., 2014; Proskin et al., 2010).

Fast and direct measurement, continuous reading, and insensitivity to the soil salinity are some of the advantages of using pore water pressure sensors (Muñoz-Carpena et al., 2004). However, often, the phenomenon of cavitation, where vapour cavities are formed within the liquid in the porous ceramic cup or at its boundaries hampers the operation of pore water pressure sensors. Once the matric suction values are around 70 kPa, it is common to encounter problems related to the cavitation of water in the pore water pressure sensors (Vanapalli et al., 2004). To avoid cavitation and measure high soil matric suction values reliably, several advanced versions of pore water pressure sensors such as T5 and High Capacity Tensiometers (HCT) have been introduced. T5 measures suction in a range of 200 kPa in the field as well as in the laboratory (Ridley and Wray, 1995).

Figure 3.8. (a) Pore water pressure sensor (model T5 tensiometer from UMS) (https://twitter.com/ums_ag/status/433957941209620480); and, (b) datalogger (Delta DL2e datalogger from AT Delta-T devices) (<https://www.delta-t.co.uk/product/dl2e/>).

3.5.2 Oscillatory rheometry

Advanced rheological tests including amplitude sweep test and three interval thixotropy test (3ITT) were performed on the fFFT samples to examine the deformation of the samples in response to the small strain shear and to track the changes in their viscoelastic properties. An Anton Paar Physica MCR301 model rheometer with a vane fixture was used for performing the rheological tests. The vane had four thin blades arranged around a central shaft, and a diameter of 22 mm and a depth of 40 mm. For performing the rheological tests, the vane fixture was lowered into the sample inside a cylindrical sample holder (part number CC27) in the rheometer. The cylindrical sample holder had a diameter of 28.92 mm and a height of approximately 80 mm. When the sample was sheared using the vane, the resulting deformation in the sample was estimated and expressed in terms of elastic modulus (G') and viscous modulus (G''). G' was calculated from the recoverable strain during each oscillation, whereas G'' was calculated from the measured irrecoverable strain.

Figure 3.9. The Anton Paar Physica MCR301 model rheometer with a vane fixture.

3.5.3 Low/high vacuum scanning electron microscopy

Scanning electron microscopy (SEM) is a useful qualitative method for studying the arrangements of particles/flocs and pores/voids in soils as well as in tailings by generating electron micrographs of the samples (Romero and Simms, 2008). The major components of a scanning electron microscope include an electron gun within an electron column that creates a beam of electrons; a sample chamber, where the electron beam interacts with the sample; detectors, which monitor a variety of signals resulting from the beam-sample interaction; and, a viewing system that constructs an image from the signals (Kimseng and Meissel, 2001).

Figure 3.10. Schematic diagram of a scanning electron microscope (from Egerton, 2005).

Using a series of electromagnetic lenses, a conventional SEM device focuses and directs the electron beam emitted from the electron gun on the surface of a sample that is

mounted on a stage in the chamber area under a vacuum condition. When the energetic, accelerated electrons hit the sample surface and are decelerated, the electron-sample interactions lead to the dissipation of the kinetic energy possessed by the electrons as different types of signals including secondary electrons, backscattered electrons, diffracted backscattered electrons, photons, visible light, and heat (Swapp, 2017). A conventional scanning electron microscope is equipped with different types of electron detectors such as backscattered electron detectors (BSED), gaseous secondary electron detectors (GSED), and diffracted backscattered electron detectors. Backscattered electron detectors (BSED) and gaseous secondary electron detectors (GSED) detect the secondary electrons and form surface images based on the differences in signal intensity of the secondary electrons emitted from different locations on the sample (Romero and Simms, 2008; Bajwa, 2015; Mizani, 2016). Often, a secondary electron detector (SED) is located at the side of the electron chamber at such an angle that it helps to detect the secondary electrons more efficiently, and therefore, provides more detailed information on the morphology and topography of the sample surface (Khattak, 2018). Backscattered electrons, that are detected by the backscattered electron detectors placed near the surface of the sample are mainly used for displaying the contrasts in composition in multiphase samples (Swapp, 2017; Khattak, 2018). Diffracted backscattered electron detectors detect the diffracted backscattered electrons, which are mainly used for determining the crystalline structures as well as the orientation of minerals and micro-fabrics (Swapp, 2017; Khattak, 2018).

Typically, the scanning electron microscope scans a focused electron beam over a selected area on the surface of the sample and measures the intensity of the signals

generated by the interactions between the beam electron and the sample atoms at each point in the scanning pattern in order to generate a two-dimensional high-resolution black and white image. The image shows the spatial variations in sample properties such as external morphology and chemical composition (Greiser, 2009; Swapp, 2017). While generating the SEM micrographs, an SEM device can enlarge a sample in the range of 20 to 30,000 times of the original size with a spatial resolution of 50 to 100 nm (Bajwa, 2015). Also, an area as small as 5 μm can be imaged by the device (Bajwa, 2015). Selected point locations on the sample surface can be analyzed for determining the chemical compositions, crystalline structure, and crystal orientations in a qualitative or semi-quantitative manner (Swapp, 2017).

In the conventional SEM technique, typically, the sample requires some preparation or pre-treatment before it is imaged. The sample is made dehydrated and electrically conductive (Khattak, 2018). However, there is a variety of SEM called Environmental scanning electron microscopy (ESEM), also known as low vacuum SEM, which requires no pre-treatment or no conductive coating on the sample and allows to examine the wet samples at different temperature and pressure conditions while preserving their natural characteristics (Romero and Simms, 2008).

Although traditional SEM equipment operates at high vacuum condition, modern SEM equipment can operate at both high vacuum and low vacuum condition. A low vacuum condition inside the sample chamber minimizes the scattering of electrons by gas molecules, and thus ensures an environment that helps an effective operation of the electron beam emitted from the electron gun. However, such a vacuum condition may also affect

the quality of the image. If water vapour exists inside the chamber, then, the emitted secondary electrons may strike the water vapour and generate positive gaseous ions, which in turn may cause overcharging of the sample that affects the quality of the secondary electron images (Bajwa, 2015; Mizani, 2016). Nevertheless, the biggest advantage of low vacuum SEM is in that such vacuum condition allows investigation of specimens without coating, whereas a high vacuum SEM needs the specimens to be conductive or conductively coated (Goldstein et al., 2003)

The accuracy or usefulness of the SEM analysis largely depends on the sample preparation technique followed during the test. For the conventional scanning electron microscopy, often, the sample is dehydrated, and a conductive coating is applied to the sample to minimize the electron scattering. There are established methods of dehydrating the samples prior to examination in the scanning electron microscope such as the critical point drying method and freeze-drying (Bajwa, 2015). However, such methods have the potential for compromising the quality of SEM imaging and analysis (Greiser, 2009); especially, for those samples, which are wet or have a high void ratio such as expansive clays, there is a probability of sample disturbance (Mitchel and Soga, 2005).

Often, a sample preparation technique known as Cryo, which uses low temperatures to prevent the evaporation of water is used for scanning electron microscopy. It is crucial for many samples to retain their original composition in their fully hydrated state, including the oil sands tailings samples. During cryo sample preparation, samples are frozen rapidly under the vacuum conditions, which prevents the dehydration of the samples and preserves their natural structures (Greiser, 2009). Modern SEM device is equipped with the capacity

of adjusting the vacuum conditions over a wide range and functioning under controlled environmental conditions, which removes the need for applying conductive coatings to the samples, and hence, helps to preserve the fully hydrated condition and original microstructure of the samples (Bajwa, 2015).

The technique of scanning electron microscopy has been utilized successfully in different fields including the studies on clays or fine-grained soils. Several researchers have used the SEM technique as a microstructural investigation tool to examine the relationship between the clay microstructure and the mechanical behaviour of clays including the time-dependent effects. Delage et al. (2006) investigated the time-dependent ageing effects in compacted bentonite clay samples at constant water content and dry density from the microstructural perspectives. To monitor the changes in clay microstructure over time, Delage et al. (2006) used the pore size distribution measurement data produced by the mercury intrusion porosimetry (MIP) technique and the scanning electron microscopy (SEM) images generated from the freeze-dried samples and provided an interpretation of the ageing effects in terms of microstructural changes. When they compared two SEM micrographs generated from a freeze-dried clay sample (after 1 day and 90 days of sample preparation respectively), they observed significant changes in the clay microstructure. The appearance changed from porous to fewer interaggregate pores over time, and overall, there was a more homogeneous aspect (Delage et al., 2006).

The SEM technique has also been successfully applied to various mine tailings including oil sands tailings. Scanning electron microscopy was performed on the unamended FFT samples collected from the 10-metre standpipe test conducted at the

University of Alberta (Jeeravipoolvarn, 2005). Image analysis was conducted based on a total of 60 SEM micrographs (three samples were collected from three different depths and 20 images were generated from each sample) in order to measure the void ratio values and compare these with the void ratios obtained from the sample measurements. The results showed that there was a close agreement between them, which validated the accuracy of the SEM micrograph analysis in observing and measuring the changes in tailings fabric over time (Jeeravipoolvarn, 2005). Jeeravipoolvarn (2005) used the scanning electron micrographs to examine the changes in tailings fabric in connection with a thixotropic gain in strength and recapitulated the key findings from the thixotropic tests conducted by Banas (1991). Banas (1991) conducted shear strength tests including cavity expansion test and vane shear test on the fluid fine tailings to determine the absolute and relative gain in strength of the FFT material with time and the test results demonstrated a gradual recovery of the shear strength after remoulding, which continued even after 350 days.

Bajwa (2015) used SEM micrographs for studying the evolution of microstructure of polymer amended FFT for short-term dewatering. The tailings were mixed with different polymer dosages (500 ~ 4000-ppm) and the tailings fabric data obtained from the SEM image analyses (1 ~ 144 hours after polymer mixing) was used to explain the variability in dewatering and other related properties (i.e. void ratio, hydraulic conductivity, yield stress). The SEM micrographs showed that the tailings fabrics were different for different polymer dosages. Lower polymer dosages (i.e. 500 ~ 700-ppm) produced relatively smaller and regular-sized flocs (because of smooth, uniform aggregation of the fine particles), which also resulted in better dewaterability potential (due to the formation of porous or

channelized structure). However, with time, the structure became more packed and compacted, which resulted in a lower void ratio, lower hydraulic conductivity, and higher yield stress. By contrast, higher polymer dosages (i.e. 1500-ppm) produced larger and irregular-sized flocs, which resulted in poor dewaterability (Bajwa, 2015). The formation of different sized flocs and different tailings fabrics due to the application of different polymer dosages was found to affect the dewaterability potential in the short-term with varying degrees of void ratio, hydraulic conductivity, and yield stress (Bajwa, 2015).

Mizani (2016) used the scanning electron microscopy (SEM) to investigate the changes in the fabric of fFFT and correlate the changes in stiffness (estimated in terms of the elastic modulus) of the fFFT samples to the microstructural changes under both shearing and resting conditions (Mizani, 2016). When the fFFT materials were sheared under lower levels of stresses, the SEM micrographs generated before shearing, immediately after shearing, and 50 minutes after the completion of shearing showed similarities in microstructure, which implied that the fFFT can regain its elastic properties (Mizani, 2016). When the fFFT materials were sheared at relatively high shearing rates, the SEM micrographs generated before shearing, immediately after shearing, and 45 minutes after the completion of shearing showed that the microstructure was restored again 45 minutes after the shearing in spite of the complete breakdown of the FFT microstructure under the action of the shearing action. Such recovery of microstructure over time characterized the thixotropic behaviour of fFFT (Mizani, 2016).

3.5.4 Optical microscopy

Optical microscopy was performed on the FFT samples immediately after their mixing with the polymer for tracking the development of the flocs. Optical microscopy is a microstructural investigation technique used for examining a sample at the microstructural level through the magnification of a single lens or a series of lenses with visible light (Smith, 2018). Over the years, the optical microscopy technique has improved considerably from the simple microscope with a very basic design, which uses a single lens to magnify the image of the sample to the inverted microscope, where the sample is viewed from underneath and a series of lenses are used to magnify the sample image to a higher resolution (Smith, 2018). The optical microscopes that are commonly used in modern research use a combination of a standard charge-coupled device (CCD) camera system and a computer for generating high-resolution optical microscope micrographs and displaying them on the computer screen in real-time in order to visualize the images without the need for an eyepiece to view the sample. Because of the advancements in digital imaging and image processing technologies, modern optical microscopes are now capable of performing a multitude of quantitative measurements on a wide range of samples in their natural states (Khattak, 2018).

During the operation of an inverted optical microscope, a microscope slide with the sample is placed on the stage of the microscope. Since the stage is usually fixed, the focus is adjusted by bringing the objective lens closer to the sample or by moving it further away from the sample. The fine-tuning of the focus allows the light inside the tube of the microscope, which generates an enlarged, inverted image of the sample. The image can be

viewed through the eyepiece of the microscope as well as on the computer screen (Smith, 2018). The biggest advantage of optical microscopy is that it is suitable for almost every type of sample system including gas, liquid, and solid sample systems. However, it also has a disadvantage in that the optical microscope micrographs are typically low resolution (Herman and Lemasters, 2012).

Figure 3.11. (Left) the Nikon Eclipse *Ti* Optical microscope used for the optical microscopy, which is an inverted microscope; and, (right) optical pathways in an inverted microscope (modified from www.microscopegenius.com).

A wide spectrum of research areas including microbiology, microelectronics, nanophysics, biotechnology, pharmaceutical research, and histopathology use optical microscopy for viewing samples in greater detail (Smith, 2018). The technique of optical microscopy and the digital image analysis of optical microscope micrographs have been used for the study of soil porosity and pore size distribution (Heck and Elliot, 2007; Pires

et al., 2008, 2009; Passoni et al., 2014). Heck and Elliot (2007) used the optical microscopy technique for soil pore structure characterization and they calculated the voids in the soil by analyzing the optical microscope images using the software ImageJ. Pires et al. (2009) used a combination of a colour CCD camera and a petrographic microscope to generate 2-D digital images of soil samples to obtain information related to soil porosity and pore size distribution. The images were processed and analyzed using the software Noesis Visilog 5.4. Although the optical microscopy has been used in soil science, to the best of author's knowledge, optical microscopy has never been used to examine the fabric or to quantify the changes in size, shape, and numbers of the flocs/pores in oil sands tailings.

3.5.5 Mercury intrusion porosimetry (MIP)

Mercury intrusion porosimetry (MIP) is an analytical technique commonly used for obtaining quantitative information on the distribution of pores (pore size and volume) within the porous geomaterial. Pores in the range of $0.006 \sim 350 \mu\text{m}$ can be investigated using the MIP technique (Simms and Yanful, 2004). In this technique, a non-wetting liquid, in most cases which is mercury, is intruded progressively into the pores of a dehydrated porous sample at an increasingly high pressure using equipment known as porosimeter. The porosimeter uses a pressurized chamber to force mercury to intrude into the empty pores. When the pressure is applied, mercury fills the larger pores first; however, as the pressure gradually increases, the filling progresses to the relatively smaller pores (Diamond, 1970; Delage et al., 2006). The pressure required for filling the pores with mercury depends on the contact angle between the mercury and the porous sample, the shape of the pores, and the surface tension of the mercury that works against the pressure

at which the mercury is forced into the pores. The Washburn Equation (1921), which is the basis of the mercury intrusion method correlates the applied pressure to the pore diameter (Diamond, 1970):

$$d = - \frac{4\gamma \cos \theta}{P} \dots\dots\dots (3.9)$$

Where d = entrance or pore throat diameter; γ = surface tension of the mercury (its value is generally taken as 0.484 N/m at 25°C); θ = contact angle between the mercury and the pore wall (taken as 139° ~ 147° for the clay minerals); and, P = Pressure (required) = $P_G - P_L$, where P_L = absolute pressure causing intrusion and P_G = pressure of gas (Delage and Lefebvre, 1984; Delage et al., 2006; Romero and Simms, 2008). Several studies demonstrated that the high-pressure intrusion process used in the MIP technique does not affect the structure of the sample (Delage and Lefebvre, 1984; Penumadu and Dean, 2000).

MIP does not provide a direct measurement of pore sizes in the sample, but rather of pore entrance sizes (Delage and Lefebvre, 1984). A typical MIP technique result includes a presentation of the log differential intrusion curve vs. pore entrance size (known as pore size density function or PSD), which helps in the visual detection of the dominant pore modes in a dehydrated porous sample (Romero and Simms, 2008).

Sample preparation for the MIP tests requires special attention. Before mercury is intruded into the MIP test sample, it is required to remove the water that occupies the small pores inside the sample and prevents the entry of mercury. There are various drying techniques that can be used for dehydrating the samples such as air-drying, oven-drying, freeze-drying, and critical-point-drying. Several researchers investigated the application of

these drying techniques on the clay samples to determine whether they have any adverse influence on the initial fabric of the samples, which may compromise the reliability the MIP test results, especially, in the case of the soft and slurried clays (Gillott, 1970; Ahmed et al., 1974; Delage et al., 1982; Delage and Pellerin, 1984; Delage and Lefebvre, 1984; Mitchell and Soga, 2005). Since such clays are saturated and have a high water content, they also have a very porous and vulnerable structure, which may collapse easily unless an appropriate and effective drying method is used (Sasanian and Newson, 2013). Among the drying techniques, the freeze-drying technique has been acknowledged as the most reliable method since it causes minimum disturbance to the sample fabric. Especially, when the sample is heat and dry sensitive and has a high water content, freeze-drying is the best choice since it has minimum influence on the soil structure due to the rapid rate of freezing (Ahmed et al., 1974; Delage and Pellerin, 1984; Romero et al., 1999; Mitchell and Soga, 2005; Zeng et al., 2016).

Freeze-drying is the process of drying a frozen product by creating conditions for sublimation. Sublimation is the conversion of water from the solid state (ice) directly to the gaseous state (water vapour) without going through the liquid state. During freeze-drying, the samples are flash-frozen using liquid nitrogen which is already vacuum-cooled at -210°C below its boiling temperature (-196°C) (Zeng et al., 2016). A temperature below the boiling temperature ensures that there would be no boiling during the immersion of the sample into the liquid nitrogen and the heat transfer would be very fast (Delage and Pellerin, 1984; Delage, 2010). Samples are usually prepared in small sizes for accelerating the freezing process (Delage et al., 2006). While the sample is being flash-frozen, the pore

water inside the sample freezes and forms a very small ice crystal. Hence, the 9% volume expansion, which usually occurs in normal freezing conditions is avoided, thereby preserving the sample microstructure during dehydration (Delage et al., 2006). Afterwards, the frozen samples are placed inside a vacuum chamber in a freezer for sublimation. By running the sublimation process continuously for 24 hours under a controlled environment (temperature and pressure), the small ice crystals formed inside the sample are removed without passing through an intermediate liquid phase, and therefore, the microstructure disturbance during dehydration is minimized (Delage et al., 2006; Zeng et al., 2016).

Although the technique of mercury intrusion porosimetry is a relatively new technique, many researchers have already utilized this technique to study the structure developed in clays under different conditions including natural, reconstituted, remoulded, compacted, or consolidated states (Sasanian and Newson, 2013). Delage and Lefebvre (1984) studied the microstructure of a sensitive Champlain clay in both undisturbed and remoulded state and examined the progressive collapse of the clay structure at various levels of one-dimensional compression using a combination of scanning electron microscopy and mercury intrusion porosimetry. The investigation revealed the presence of an aggregated microstructure inside the natural clay samples with a pore network comprising both interaggregate and intra-aggregate pores. Although the remoulding process modified the organization of interaggregate links, the aggregates were still present after the remoulding. Delage and Lefebvre (1984) proposed an analytical technique for quantifying the changes in the microstructure of natural clays during compression in terms of changes in the pore size distribution. They compared the microstructure of the samples

consolidated at different stress levels of one-dimensional compression. They observed that the volume decrease during compression was prompted by a progressive and ordered collapse of interaggregate pores. When the sample was subjected to a compression pressure, the largest available pores in the sample were the first ones to collapse; hence, the changes in the volume of the largest existing pores led to a volume decrease during consolidation (Delage and Lefebvre, 1984; Delage, 2010). Such changes were reflected in a reduction in total porosity and a shift towards smaller pores. When all the large pores were collapsed, the smaller pores also started to collapse (Delage and Lefebvre, 1984; Griffiths and Joshi, 1989; Delage, 2010). When Griffiths and Joshi (1989) investigated the microstructural changes in four different clays due to consolidation by applying the MIP technique on the freeze-dried samples prepared from the consolidated clays, the test results agreed with the findings of Delage and Lefebvre (1984).

Simms (2003) studied the correlations between the pore size distribution measured from the mercury intrusion porosimetry (MIP) and the SEM images generated from the Reginal clay, Sarnia clay, and Halton till samples compacted at different degrees. The results showed that the particle sizes in the SEM micrographs were in the same order as the average pore diameters measured by MIP. There were few other studies that utilized the MIP technique to study the changes in microstructure of soils and explored the correlations between the pore size distribution and the permeability, consolidation characteristics, and water-retention behaviour; such correlations were found helpful in predicting the water-retention behaviour of soils as well as volume change behaviour (Simms and Yanful, 2004, 2005; Romero and Simms 2008).

Delage (2010) studied the effects of change in particle arrangement and interparticle bonding on the compressibility of Champlain clay and presented the results in a qualitative way. He made a comparison between three different one-dimensional compression curves obtained from three intact clay samples and a compression curve obtained from a freshly remoulded clay sample (Figure 3.12). He correlated the change in compressibility to the change in clay fabric (arrangement of particles) and attributed the increased apparent preconsolidation pressure (yield strength) to the stronger interparticle bonding. Delage (2010) described the change in clay fabric in terms of change in pore size distribution (PSD) curve obtained from the MIP tests. Poorly graded PSD curve was found to be correlated to an increase in compressibility, whereas well-graded PSD curves were found to be associated with reduced compressibility (Figure 3.12) (Delage, 2010).

Figure 3.12. The effect of changes in clay microstructure on its compressibility behaviour (from Delage, 2010).

Although the studies of clay microstructure using mercury intrusion porosimetry have shown the effectiveness of the technique in characterizing a clay with interconnected porosity through the quantitative evaluation of pore size distribution, mercury intrusion porosimetry has rarely been used to investigate the microstructure developed in slurried clay deposits or deep deposits such as tailings (Sasanian, 2011; Sasanian and Newson, 2013). Bajwa and Simms (2013) used the MIP technique for the quantitative measurements of tailings fabric as they determined the cumulative pore size distributions (CPSD) of polymer amended oil sands tailings to track the changes in pore size and shape. Bajwa and Simms (2013) determined the CPSD for the tailings treated with three different dosages of polymer (700, 1000, and 1500-ppm) and untreated tailings (no polymer) at the same initial water content (140%) and found that the treated FFT samples showed a considerably higher cumulative intrusion pore volume and also a greater porosity in comparison to the untreated tailings.

When Bajwa (2015) compared the CPSD of tailings dosed with different polymer dosages after 1 hour and 144 hours of deposition, the cumulative intrusion pore volume was found to decrease over time, which was attributed to the release of water (high initial dewatering) (Figure 3.13). The application of polymer in the optimum dosage range (500 ~ 700-ppm) resulted in a relatively higher cumulative intrusion at the beginning (after 1 hour); however, after 144 hours, the cumulative intrusion was considerably lower. By contrast, when the tailings were treated with a polymer dosage outside the optimum range (1000 ~ 1500-ppm), the change in cumulative pore volume after 144 hours was nominal (relative to the cumulative pore volume after 1 hour). Hence, it was demonstrated that the

MIP technique could detect the changes in tailings fabric due to different polymer dosages (Bajwa, 2015). The test results also indicated that tailings treated with optimum polymer dosage had a greater dewatering potential, which resulted in a lower pore volume with time. Such a change in pore size distribution was also thought to play an important role in the increase in yield stress of fFFT (Bajwa, 2015).

Figure 3.13. Change in the cumulative pore size distribution of fFFT (from Bajwa, 2015).

3.5.6 UV-vis spectrophotometry

Ultraviolet-visible (UV-vis) spectroscopy or ultraviolet-visible spectrophotometry refers to absorption spectroscopy in the ultraviolet-visible spectral region, which is frequently used for the quantitative analysis in analytical chemistry. The method was employed to determine the concentration of a polyacrylamide-based anionic polymer in the bleed water collected from fFFT samples using a UV-vis spectrophotometer (Figure 3.14).

The method was also used to determine the optimum polymer dosage required to achieve the best thickening and dewatering performance by monitoring the residual polymer concentrations in tailings water.

The UV-vis spectrophotometer measures the intensity of light passing through a sample (I) and compares it to the intensity of light before it passes through the sample (I_0). The ratio ($\frac{I}{I_0}$) is called the transmittance and is typically expressed as a percentage (%T):

$$\%T = \frac{I}{I_0} \times 100 \dots\dots\dots (3.10)$$

Where I = the intensity of light passing through a sample; I_0 = the intensity of light before it passes through the sample, and, %T = transmittance (the percentage of light that goes through the sample).

The absorbance A is estimated based on the transmittance (%T) using the following equation:

$$A = -\log_{10} \left(\frac{\%T}{100\%} \right) = \log_{10} \left(\frac{I_0}{I} \right) \dots\dots\dots (3.11)$$

Where A = absorbance (the amount of light absorbed by the sample).

The Ultraviolet-visible spectroscopy method is based on the theory that the colour of a substance has a close association with its electronic structure. A UV-vis spectrophotometer uses light in the ultra-violet and visible ranges on the sample to be analyzed, and subsequently, the sample exhibits absorption of light in the above ranges as the radiation prompts a change in the electronic state of the molecules or ions in the sample.

In UV-vis spectroscopy, such absorptions in the ultraviolet and visible ranges are measured in terms of wavelength expressed in nanometers (Thermo Spectronic, 2011).

Figure 3.14. Cary 100 Bio UV-vis Spectrophotometer used for UV-vis spectrophotometry.

3.5.7 Torque rheometry

Torque rheometry measures the viscosity-related torque generated by the resistance of a slurry to the shearing action in a laboratory device called torque rheometer. Floccky Tester ODA-10 (Koei Industry Co., Ltd Japan) is a torque rheometer specifically designed to study the flocculation-deflocculation phases of slurries (Figure 3.15). After a sample is placed in the rheometer beaker ($V = 200 \text{ mL}$), a three-vane impeller, which is driven by a motor is used to apply a range of rotational shear forces to the sample. The resistance of the sample to the applied shear is recorded as a strain in a transducer, which is then converted into an electronic signal, and subsequently, to the units of torque (mNm). After a prespecified time interval, the polymer stock solution is injected directly into the sample

through a couple of ports on the sides of the beaker while the sample is being sheared simultaneously. The typical output of the torque rheometer is a torque-time rheogram, where the y-axis in the torque-time plot indicates the torque developed during shearing action in mili-Newton-meter (mNm), and the x-axis indicates the duration of the rotational shear force applied to the sample in seconds. The equipment is operated by a computer equipped with a data-acquisition software. The software helps to select the operational parameters such as the rotational speed (rotation per minute or rpm), duration of the rotation (second), and the sensitivity of the detector, and to generate the results in terms of a torque-time relationship (torque-time rheogram).

Figure 3.15. (Left) The Floccky Tester torque rheometer; and (right) the cylindrical beaker with ports on the sides for injecting the polymer solution directly into the sample.

3.5.8 Undrained shear strength (fall cone test)

Shear strength of a soil refers to the maximum or ultimate shear stress that the soil can sustain without undergoing large deformations (Kaminsky et al., 2015). Undrained shear strength is the shear strength of soil with in-situ moisture. It is measured in an

undrained condition, where there is no flow of water into or out of the soil while the soil is subjected to a change in load. Since the water is unable to move into or out in response to the tendency of volume change, such a change in load also causes a change in pore water pressure as pore water carries most of the external loading. Kaminsky et al. (2015) have defined the undrained shear strength in FFT as:

“When the shear stress is applied relatively quickly or the drainage boundary conditions are such that the shear-induced excess pore pressure does not dissipate as the material is strained, then it is said that failure has taken place under undrained conditions, or that the undrained shear strength has been mobilized.”

In FFT, the undrained shear strength starts to develop only after the tailings have been sufficiently dewatered and improved in solids content or density (Beier et al., 2013). In addition to the solids content (or density), pore water chemistry, particle size distribution, and clay content and mineralogy affect the shear strength of tailings (Mitchell and Soga 2005; Sobkowicz and Morgenstern, 2009). For the successful remediation of tailings deposits, it requires a shear strength of about 20 kPa (McKenna et al., 2016).

Although it has been a common practice in geotechnical engineering to use a range of tests including triaxial and direct shear tests to determine the shear strength of soils, such conventional geotechnical techniques have been found unsuitable for the tailings since they have a very low shear strength, often as low as 0.5 kPa (Qiu and Sego, 2001). For these tailings with extremely low shear strength, a fall cone device (Figure 3.16) is considered to be the best tool for measuring the undrained shear strength since the device has the capacity to measure shear strengths as small as 1.5 kPa with reasonable accuracy and repeatability

(Dimitrova and Yanful, 2011). Lab vane shear test is another suitable method for measuring relatively low shear strengths in fully saturated samples, however, the sample disturbance caused by the rotation of the vane causes a reduction in the interparticle bonds and a rearrangement of the particles, which makes the assessment of structuration effect difficult.

Although the fall cone test is primarily a method used for determining the liquid limit of clay, the test is also considered as a strength test because of the well-established empirical correlations between the liquid limit determined and the undrained shear strength of the soil at that liquid limit (Houlsby, 1982). In fact, the fall cone test is a suitable method for determining the consistency of clay in both undisturbed and remoulded conditions (Leroueil and Le Bihan, 1996); consistency of soil refers to the strength with which soil materials are firmly held together and show resistance to deformation and rupture (FAO, 2018). Since the strength of the cohesive soil controls the penetration by fall cone, the measurement of cone penetration depth also provides a measurement of the soil strength (Kumar and Wood, 1999). Based on his extensive experimental work on the fall cone tests, Hansbo (1957) proposed the following equation to express the relationship between the undrained shear strength (s_u) and cone penetration (d):

$$s_u = \frac{kmg}{d^2} \dots \dots \dots (3.12)$$

Where s_u = undrained shear strength (in kPa); k = cone factor, an empirical constant that depends on the apex angle of the cone and whether the sample is undisturbed or remoulded; m = mass of the cone; g = gravitational acceleration = 9.81 m/s²; and, d = depth of cone penetration (in mm) (Leroueil and Le Bihan, 1996; Dimitrova and Yanful, 2011).

Figure 3.16. Fall cone used for the measurement of undrained shear strength.

Theoretical and experimental work on the fall cone test method (Hansbo, 1957; Houlsby, 1982; Wood, 1985; Koumoto and Houlsby, 2001) showed that a combination of factors including cone geometry (such as cone apex angle), cone surface roughness, soil heave formed around the cone during penetration, and dynamic effects of cone penetration could affect the cone factor (k) values (Mahajan and Budhu, 2009). For intact clays and an apex angle of 30° , Hansbo (1957) proposed a k value of 1. For remoulded clays with the same apex angle of 30° , Wood (1985) and Karlsson (1977) suggested a k value of 0.85 and 0.80 respectively (Leroueil and Le Bihan, 1996). Later, the k values were recalculated considering the smoothness or roughness of the cone surface. For an apex angle of 30° , Koumoto and Houlsby (2001) suggested using three different k values: 2.00 for the smooth surfaces, 1.33 for the semi-rough surfaces, and 1.03 for the rough surfaces.

There are several ways of assessing the degree of soil structure in clays; however, the sensitivity measurement is the most used method among them (Sorenson, 2006). When the natural clays are reconstituted or remoulded, they lose a portion of their undrained shear strength due to the destruction of soil structure. The sensitivity of a natural clay can be determined by comparing its undrained shear strength in the undisturbed state to its undrained shear strength in the reconstituted state at the same water content and thus the sensitivity quantifies the proportion of the intact strength that is lost due to the reconstitution/remoulding processes (Sorenson, 2006). Typically, a high sensitivity refers to a strong structuration/thixotropic effect. According to Terzaghi (1944), the sensitivity (S_t) of a natural clay can be estimated using the following relationship:

$$S_t = \frac{S_u}{S_{ur}} \dots \dots \dots (3.13)$$

Where S_t = sensitivity; S_u = undrained shear strength (undisturbed); and, S_{ur} = undrained shear strength of the reconstituted clay at the same water content (Sorenson, 2006).

3.5.9 Conventional one-dimensional consolidation test (oedometer test)

The conventional one-dimensional compression test or consolidation test, also known as the oedometer test simulates the one-dimensional compression and drainage conditions that the soil experiences in the field and the test result presents the soil stress-strain behaviour demonstrated during such one-dimensional compression. The test is typically conducted by applying vertical static loads to a cylindrical sample of saturated soil and measuring the corresponding settlements (Figure 3.17). The sample, which is typically 36 ~ 75 mm in diameter and 14 ~ 20 mm in thickness is enclosed in a metal ring

and placed on the porous stone. A loading cap, which also has a porous stone at the bottom is then placed on the top of the sample. Thus, the sample is placed between two porous stones. The sample within the metal ring and between the porous stones is mounted in a consolidation cell, which is then placed on the loading unit of an oedometer test frame. Water is poured into the consolidation cell around the sample so that the sample remains saturated. The presence of porous stones both at the top and bottom facilitates two-way drainage for the sample.

Vertical static loads are applied to the sample using dead loads and a static loading system, and the load is increased every 24 hours by doubling it relative to the last load applied. The corresponding changes in the sample thickness (settlement) are recorded during each loading increment. The range of vertical static loads used is selected based on the range of effective stress needed for the consolidation analysis of the sample under investigation. After the maximum load is applied for 24 hours, the sample is unloaded gradually by reducing the vertical loads by half every 24 hours. The swelling of the sample is recorded during each unloading. After unloading is completed, the sample is carefully removed from the consolidation cell, and the thickness and water content are measured.

Figure 3.17. (a) Typical oedometer test frame; and, (b) oedometer test setup at the lab.

From the measurements of change in thickness at the end of each loading step, the void ratio at the end of each loading step is estimated. When these void ratios are plotted against the corresponding effective stresses in a semi-logarithmic scale (e vs. $\log \sigma'$), the resulting curve, or more specifically, the slope of the linear portion of the resulting curve represents the compressibility behaviour of the saturated soil sample (where the soil is subjected to a vertical loading expressed in terms of effective stress, and the rate of consolidation is expressed in terms of void ratio). From the compression curve, one can also determine the soil parameters such as compression index (C_c), coefficient of volume compressibility (m_v), coefficient of consolidation (c_v), and swelling index (C_s). For a normally consolidated clay, compression index (C_c) is the parameter that is determined from the slope of the straight part of the compression curve and describes the change in void ratio as a function of the change in effective stress (required for computing the total settlement) (Figure 3.18) (Das, 2008).

Preconsolidation stress or pressure (σ'_p) is another parameter that is typically estimated from the consolidation test data (for an overconsolidated soil). Preconsolidation pressure is the maximum effective stress that a soil sample has experienced in the past (Solanki and Desai, 2008). In other words, preconsolidation pressure is the largest overburden stress beneath which the soil had once been consolidated (Casagrande, 1936). In general, previous loads, overburden pressure, and the groundwater level and its fluctuations are the primary factors that control the preconsolidation pressure in the soil. However, there are some cases where the preconsolidation pressure can be greater than the highest experienced overburden pressure, especially, if the soil has been subjected to the

time-dependent effects such as ageing, cementation, creep or secondary compression, and other soil structure related phenomena (Bjerrum, 1967).

Preconsolidation pressure is the single most important parameter that governs the overall clay behaviour, especially, in sensitive clays (Tavenas and Leroueil, 1979). Since preconsolidation pressure is the vertical effective stress beyond which large deformation takes place in soil (Leroueil et al., 1983), how much soil will settle depends on the magnitude of present overburden pressure relative to the magnitude of its preconsolidation pressure. Therefore, the strength and deformation properties of soil depend on its preconsolidation pressure (Persson, 2017).

Although many methods have been developed for determining the magnitude of preconsolidation pressure from the consolidation test data, Casagrande method (1936), which estimates the preconsolidation pressure from the void ratio-effective stress relationship (or compressibility curve) based on the point of greatest curvature has been the most standard and commonly used method for the determination of preconsolidation pressure (Figure 3.18) (Das, 2008; Solanki and Desai, 2008). Although the Casagrande (1936) method is easy to use, the accuracy of the method may vary depending on the presence and identification of a well-defined break in the e vs. $\log \sigma'$ plot (Grozic, Lunne, and Pande, 2003).

Figure 3.18. The schematic plot of a typical compression curve in the ' $e - \log \sigma'$ ' space for a given load increment that also shows the graphical procedure for the determination of compression index (C_c) and preconsolidation pressure (σ'_p) (from Das, 2008).

The steps generally followed in the Casagrande method (1936) (Das, 2008):

1. P , a point on the upper curved portion of the e vs. $\log \sigma'$ plot, which has the maximum curvature is selected based on visual observation.
2. A horizontal line PQ is drawn from the point P .
3. A tangent PR is drawn at P .
4. PS , a line bisecting the angle QPR is drawn.
5. The straight-line portion of the e vs. $\log \sigma'$ plot is extended backward to intersect the line PS at point T .
6. The effective stress corresponding to point T is the preconsolidation pressure (σ'_p).

3.5.10 Dewaterability tests

Several dewaterability tests including settling tests and capillary suction time (CST) tests were conducted to assess the performance of the polymers after they were added to the FFT at different dosages or using different mixing methods.

Settling test

Settling tests are typically conducted to determine the settling rate or to how rapidly the fine particles are settling in a suspension after they have been treated with a polymer (Figure 3.19). During the settling test, the change in the solid-liquid interface (mudline) is monitored and recorded as a function of time, and thus, a settling profile is generated.

Figure 3.19. The settling test performed on the FFT dosed with different polymer dosages.

Capillary suction time (CST) test

Capillary suction time (CST) test is a type of static filtration test that measures the filtration rate of a sludge/slurry sample using a filter paper as the medium, and therefore, quantifies the dewatering properties of a sample in terms of how rapidly that sample can be dewatered (Gumfekar et al., 2019). The CST apparatus consists of a cylinder/funnel placed on a platform equipped with two sensors and a digital timer connected to the sensors (Figure 3.20). Before conducting the test, a CST filter paper is placed between the platform and the funnel. When a flocculated sample is poured into the funnel, the filter paper generates a capillary suction pressure to draw the filtrate from the sample. Capillary suction time (CST) refers to the time taken by the filter paper to draw a certain volume of filtrate so that the filtered water could travel from the first sensor to the second sensor (Figure 3.21) (Sawalha, 2011).

Figure 3.20. The CST device with the funnels placed on a platform equipped with sensors.

Figure 3.21. The schematic diagram of the funnel and the sensors (from Sawalha, 2011).

When the CST tests are performed on the samples mixed with different dosages of a chemical conditioning agent such as polymer, the lowest value of capillary suction time indicates an optimum filterability and a high permeability and dewaterability, whereas a higher capillary suction time refers to a lower dewaterability or high water retention capacity (Jin et al., 2004; Wells et al., 2011; Gumfekar et al., 2019).

CST test is a simple test as no external source of pressure or suction is required. However, the accuracy and repeatability of the CST test depend on how homogenous and well-mixed the sample is (Sawalha, 2011; Sadighian et al., 2018).

3.6 Experimental methodology

3.6.1 Column dewatering test

10 cm tall column experiments were conducted using two different boundary conditions: (a) single-drainage and (b) double-drainage. The short height was chosen to maximize the influence of creep and minimize the role of self-weight consolidation. Typically, a short column height leads to the development of low effective stress within the deposited tailings material and the creep is thought to be one of the main mechanisms behind the deformations that occur at lower effective stresses (Miller, 2010; Mizani, 2016).

The difference between the single-drainage and double-drainage column is that the non-porous bottom in the single-drainage column didn't allow for any drainage, whereas the tailings in the double-drainage column were rested on a filter paper that allowed for drainage. The columns were kept covered at the top with the help of a tight lid. The space between the lid and the surface of the deposited tailings was kept closed in order to prevent the escape of the water vapour.

Two types of columns were used: (a) plastic acrylic columns (transparent) and (b) polypropylene columns (non-transparent). A single lift of polymer amended fluid fine tailings were deposited in the columns with the material reaching a height of 10 cm. Two pore water pressure sensors (model T5 from UMS) were installed into the columns at two different heights (at the elevations of 1.5 cm and 4.5 cm respectively). Before installing the pore water pressure sensors into the column, a standard de-airing procedure (as suggested by the manufacturer) was followed by using the vacuum equipment repeatedly on the pore

water pressure sensors for removing air. A set of replicate columns (similar sized columns with no pore water pressure sensors installed into them) was used for measuring the water content at different depths, the changes in water-solids interface height and void ratio, and for providing samples for other tests and analyses.

3.6.2 Oscillatory rheometry

For the amplitude sweep test, when the vane fixture was lowered into the sample, the vane oscillated at a constant low frequency, however, the maximum shearing stress was increased successively with each oscillation. The resulting deformation in the fFFT material was estimated and expressed in terms of elastic modulus (G') and viscous modulus (G''). The results from the amplitude sweep test also demonstrated the changes in the linear elastic range of the fFFT material and the yield stress. Such changes had already been found correlated to the changes in material structure as the elastic modulus strongly depends on the material fabric (Mizani et al., 2017). The region where the elastic modulus was relatively constant represented the true elastic range of the fFFT material and also indicated the presence of a particulate network (Mewis and Wagner, 2009). The point where G' was reduced and G'' became dominant represented the yield stress (past which viscous behaviour began to dominate the mechanical response of the material). In the case of fFFT material, the elastic modulus had already been found to vary substantially at constant density (Mizani et al., 2017); hence, when a change was observed in elastic modulus with time at constant density or void ratio, it was thought to indicate a change in the strength/stiffness of the fFFT sample, which originated from the changes in its microstructure, i.e., stronger interparticle bonding (Lohani et al., 2001).

Three interval thixotropy test (3ITT), which is a combination of amplitude sweep test (where the maximum stress was increased at each cycle while maintaining a constant oscillation frequency) and stress growth test (where the material was sheared at a constant shear rate) was performed to measure the ‘structure breakdown’ (under shear) and ‘structure recovery’ (at rest) (measured in terms of increase/decrease in the elastic modulus before, during, and after shearing). After placing a sample in the cylindrical sample holder (part number CC27), the elastic modulus was measured in a non-destructive manner by oscillating the vane at a constant low frequency for a short period (amplitude sweep test). Then, the sample was sheared at a constant strain rate (0.1 s^{-1}) for a specific period (stress growth test). Afterwards, the elastic modulus was measured again at specific time intervals (after 15 minutes, after 30 minutes, after 60 minutes, etc.). While the sample was in a resting condition in between the elastic modulus measurements, it was kept covered so that there was no loss of water and the density remained unchanged. The ratio of the elastic modulus measured at specific intervals to the original elastic modulus (initial elastic modulus) was used to describe the recovery of the sample structure over time and such recovery was an indication of thixotropic strength recovery in fFFT.

3.6.3 Low/high vacuum scanning electron microscopy

For this study, a Tescan Vega-II XMU SEM device at the Carleton Nano Imaging Facility (NIF), Carleton University was used for scanning electron microscopy. The device can operate at a low vacuum for the direct imaging of wet fFFT samples at their natural state and original composition. It also has the capacity to operate at a high vacuum, which is suited for high-resolution imaging. For facilitating the cryogenic sample preparation

technique at low temperature, the device is equipped with a Cryo stage. SEM micrographs were generated using the fFFT samples obtained from replicate columns. Maximum care was taken in transferring the columns to the Nano Imaging Facility (NIF) and collect the undisturbed samples from the columns without compromising the integrity of the samples. The samples were collected from the surface of the solids using a spatula and directly placed in the SEM chamber facing upwards. There was an electronic gun in a position perpendicular to the sample (90^0), which was used for emitting the electrons, whose interactions with the sample surface helped in imaging the surface profiles and topographies of the samples at variable pressures.

Using the cryogenic sample preparation technique, a rapid freezing stage of -40^0C was applied to the hydrated samples in the vacuum chamber. Then, the samples were scanned and imaged at a vacuum pressure of 50 Pa. A scanning speed of $148\text{ }\mu\text{s/pixel}$ and a working distance of $6 \sim 10\text{ mm}$ was used for generating the SEM micrographs. A nominal working distance was maintained in order to minimize the distance that the electron beam had to travel before impacting the sample surface. Since one of the original research objectives was to correlate the microstructural characteristics of fFFT (size, shape, and arrangement of the flocs and pores) to the macrostructural behaviour (compressibility, undrained shear strength, elastic response), highest precaution was taken to prevent any change in the microstructure of the flocculated FFT samples during SEM testing.

Although the SEM device can operate in different vacuum conditions including low vacuum scanning electron microscopy (LVSEM) and high vacuum scanning electron microscopy (HVSEM), for this study, mostly the low vacuum condition was used for

analyzing the wet FFT samples. Although in terms of resolution quality, the capacity of LVSEM is limited in comparison to the HVSEM, a low vacuum condition allows the device to examine the microstructure of the samples in their undisturbed state (Bajwa, 2015). SEM micrographs were generated from the samples using a variety of magnification scales (200X, 500X, and 1000X) and a range of field of view (250 ~ 750 μm) to visualize and quantify the pores of all ranges. The field of view refers to the area that is visible through the microscope for a specific magnification. Typically, the field of view decreases with the magnification scale increasing. 5 (five) different point locations were selected on each sample surface to ensure higher chances of repeatability. In spite of the relatively low resolution, the generated SEM images displayed the effects of dewatering and self-weight consolidation and also characterized the structuration and thixotropic behaviour of the fFFT samples. Especially, the size and pattern of the pores appeared to change over time.

In addition to the low vacuum SEM analysis, a high vacuum SEM was performed on the dehydrated, freeze-dried samples coated with gold-palladium. SEM with a gold-palladium coating produced better quality images in comparison to the low vacuum SEM. A high vacuum condition typically helps in avoiding scattering of the electrons, and therefore, maximizes the resolution. However, since there is always a probability of a change in volume of the high water content FFT samples while they are being freeze-dried, wet FFT samples (low vacuum SEM) are considered more reliable for the SEM analysis.

Recently, Khattak (2018) developed a combination of image processing techniques for measuring the fabric evolution of fFFT samples quantitatively from the SEM micrographs and applied these techniques to the SEM micrographs generated at different

stages of this research. The primary objective was to improve the conceptual models of tailing dewatering by incorporating the effects of structuration and thixotropy, which are typically reflected in the changes in the compressibility of tailings as well as in tailings fabric over time independent of the changes in density or effective stress (Khattak, 2018).

3.6.4 Optical microscopy

For this study, during the optical microscopy, the fFFT samples were imaged in their original state. The wet samples were collected from the surface of the replicate columns using a needle and they were placed carefully on the microscope slides so that the sample disturbance was minimum. The samples were imaged using a Nikon Eclipse Ti optical microscope at Carleton University. A magnification scale of 200X and a field of view of 650 μm across the width was used. The imaging software NIS Elements AR3.2 was used for image visualization and acquisition. In order to track the flocs development with time, the samples were collected from the columns on an hourly basis (1, 2, 4, 6, 8, 12, 24, and 48-hour). During optical microscopy, the positions of the slides were changed frequently to obtain at least 5 different images from 5 different locations on the same sample. Since the wet FFT samples tended to dehydrate under atmospheric conditions, the time between the sampling and the completion of imaging was kept minimum, never more than 5 minutes, in order to ensure that the samples were in their natural state while they were being imaged. As the density of FFT in the replicate columns was increasing, the samples collected from the surface became relatively dense, especially, after the first 48 hours of deposition. Hence, it became difficult for the light (that was emitted by the light source used by the optical microscope) to pass through the densifying samples, and

subsequently, it became impossible to image the samples irrespective of the polymer dosages applied or the boundary conditions used. Nevertheless, the optical microscope images generated over the first 48 hours of deposition were crucially important for studying the development of flocs since the particle-particle as well as particle-polymer interactions and the settling of flocs and particles were most prevalent during this period.

3.6.5 Mercury intrusion porosimetry (MIP)

For this study, the method used for the MIP sample preparation was based on the method of freeze-drying as discussed in Simms (2003) for compacted clays and Bajwa (2015) for polymer amended oil sands tailings. The fundamental principle in freeze-drying is sublimation, which is a type of phase transition where a substance changes from a solid state to a gaseous state without passing through a liquid phase. The technique works by utilizing temperature and pressure conditions. Initially, the material is flash-frozen; then, the pressure is reduced; and subsequently, the temperature is raised, which allowed the frozen water in the material to sublimate. If properly implemented, the technique could help to keep the microstructural features of the fFFT samples as much undisturbed as possible with a minimal change in volume (Romero and Simms, 2008; Bajwa, 2015).

Since it was difficult to collect the samples from the very wet polymer amended oil sands tailings and handle them, specially prepared small steel moulds were used to contain and give shape to the tailings samples at higher water contents. These molds had an inside volume of 0.5 ~ 0.6 cubic centimetre volume (0.8 cm * 0.8 cm * 0.8 cm).

For freeze-drying, a cylindrical container was filled with n-pentane (C_5H_{12}), which is a clear colourless liquid with a low melting point ($-129.8\text{ }^{\circ}C$). The n-pentane container filled with the liquid n-pentane was cooled in a liquid nitrogen bath ($-210^0C \sim -196^0C$) for 30 seconds so that the n-pentane could reach its melting point. When there was no visible sign of vapour coming from the n-pentane container, it was an indication that the n-pentane had reached its melting point. Pentane was used to accelerate the ‘flash’ freezing of the water within the samples, thereby minimizing the volume expansion (or the formation of water crystals). Then, the sample was placed in a steel grated thin cylindrical sample holder attached to a long rod. The sample holder was dipped in the n-pentane container while keeping that n-pentane container submerged in the liquid nitrogen bath throughout the time. The steel grated cylindrical sample holder was moved up and down to ensure that it would not freeze and get stuck to the wall of the n-pentane container. The freezing process was continued for approximately 60 seconds or until the sample turned white. Then, both n-pentane container and steel grated cylindrical sample holder were removed from the liquid nitrogen bath. The frozen sample was placed in a modified set up of a vacuum pump, a desiccator, desiccant beads, and icepacks within a freezer for sublimation. Previously, the desiccant beads were placed in an oven at $100^{\circ}C$ for 24 hours to make them moisture-free and reactivated. The sample was placed under vacuum for 24 hours; the goal was to remove the pentane or any remaining frozen water from the pores through the sublimation process.

After applying the freeze-drying technique on the samples collected from the replicate columns at intervals, the dehydrated samples were sent to the Geotechnical Research Centre at the Western University, London, Ontario. An AutoPore IV 9500

porosimeter, a porosimeter manufactured by Micromeritics was used to perform the MIP tests. The contact angle between mercury and the pore wall was 140° and the surface tension of mercury was 0.480 N/m.

3.6.6 UV-vis spectrophotometry

For the current study, the absorbance measurements in the UV-vis range were performed with a UV-Vis spectrophotometer (Cary 100 Bio UV-Vis Spectrophotometer, Varian Inc./Agilent Technologies, Canada) using a 1-cm quartz glass cell (Hellma Canada Ltd, Canada). The absorbance data were recorded using Scan software (Cary WinUV, Varian Inc., Canada) in the wavelength range of 190 ~ 800 nm. However, the absorbance values at 190 nm were used for the calculation of the polymer concentration because the results from the previous UV-vis spectroscopy studies showed that the polyacrylamide polymers strongly absorb light in the 190 ~ 230 nm range and beyond 260 nm of wavelength the amount of absorbance is insignificant (Gibbons and Örmeci, 2013; Al Momani and Örmeci, 2014a). Each absorbance measurement was repeated three times and the reported absorbance measurements were the average of three replicates.

3.6.7 Torque rheometry

This study investigated the potential of two rheological methods that are based on torque rheology and involve the measurement of rheological parameters to identify the optimum polymer dosage. The first method measures the peak torques that are observed in the torque-time rheograms after a predetermined amount of polymer solution is injected directly into the untreated tailings and mixed with the tailings using an impeller rotating at

a preselected speed. The method involves the measurement of torque values at three different successive steps using three different rotational speeds and allows the observation of both flocculation and deflocculation phases (Örmeci, 2007). Figure 3.22 shows the steps followed and the results typically obtained from this method. The first half of the peak or the upward portion shows the flocculation phase (when the flocs are formed), whereas the second half of the peak or the downward portion shows the deflocculation phase (when the degradation of flocs occurs).

Figure 3.22. The measurement of peak torque from the torque-time rheogram.

The formation of peaks in the torque-time rheograms can be explained based on the concept of elastic floc network (Michaels and Bolger, 1962; Firth and Hunter, 1976; Otsuba and Umeya, 1984; Langer et al., 1994). When the polymer is added to an unconditioned

sample, the strength of the chemical and physical bonds between the flocs that forms such an elastic floc network increases, which in turn increases the resistance of the sample to the shearing action of the impeller. So, the apex of the peak refers to the point where such bonds between the flocs are broken and the floc network structure is disintegrated (Langer et al., 1994). Hence, the peak torque is an indirect reference to the floc network strength of the sample and also to the degree of flocculation (Murray and Örmeci, 2008). When more polymer is added to the sample, the flocs become more stable and the bonds between the flocs become stronger. Since more energy is needed to break up the bonds and the peak torque corresponds to the amount of energy dissipated in breaking up the bonds, subsequently, the height of the peak increases (Abu-Orf and Dentel, 1999).

The second method involves the measurement of totalized torque (TTQ) or the total area under the flocculation curve in a torque-time rheogram, which stretches over a length of shear application time (Abu-Orf and Örmeci, 2005; Örmeci and Abu-Orf, 2005). The time starts from the moment the polymer is injected into the sample (i.e. 50 seconds) and continues up to a shear application time that coincides with the end of the deflocculation phase (i.e. 100 seconds). Totalized torque (TTQ) reflects the strength of the floc network formed in the flocculated sample and the variation in measured relative network strengths reflects the fundamental changes in the properties and behaviour of the sample material during both flocculation and deflocculation phases (Örmeci et al., 2004; Örmeci and Abu-Orf, 2005). Figure 3.23 shows the rheological method of measuring the totalized torque from a torque-time rheogram.

Figure 3.23. The measurement of totalized torque (the area bounded by the Time slices and Input slices) from the torque-time rheogram.

Örmeci and Abu-Orf (2005) derived mathematical models for estimating the relative network strength using a torque rheometer, which showed that the measurement of the area under the torque-time rheogram is representative of the total energy dissipation rate in a sludge system. The unit work energy required for breaking up the structure of a certain volume of sludge or the total energy dissipated during that process, which also reflects the network strength of that suspension can be expressed using the following equation (Michaels and Bolger, 1962):

$$W_u = \frac{[2\pi N*TTQ]}{V_b} \dots\dots\dots (3.14)$$

Where W_u = unit work energy (Joule/cm³); N = rotational speed (rpm); TTQ = totalized torque area or the area under the rheogram (Joule-min); and, V_b = sample volume (cm³).

The Floccky torque rheometer employed in this study uses a sample size of 200 mL and the totalized torque area (TTQ) is expressed in m J s; hence, equation (3.14) becomes:

$$W_u = \frac{[0.524N*TTQ]}{X} \dots\dots\dots (3.15)$$

Where W_u = work energy (J/Kg of dry solids); N = rotational speed (rpm); TTQ = totalized torque area or the area under the rheogram (m J s); and, X = solids concentration of sludge sample in g/L.

Thus, the totalized torque (TTQ) or the area under the torque-time rheogram is converted to the work energy required for processing a unit volume or mass of a given suspension (Chung, 1986), which also represents the total energy dissipated into the suspension during the process, and therefore, indicates the network strength (Michaels and Bolger, 1962). According to equation (3.14), if the sample volume and the rotational speed remain the same, the network strength is linearly proportional to the totalized torque (Örmeci et al., 2004; Örmeci and Abu-Orf, 2005). Therefore, for a given tailings composition, if the samples are dosed with different polymer dosages using the same sample volume and mixing intensity, the totalized torques measured from the torque-time rheograms can be used to compare the corresponding network strengths (Örmeci and Abu-Orf, 2005).

3.6.8 Undrained shear strength measurements (fall cone test)

For this study, to investigate the undrained shear strength development in the fFFT samples (dosed with different polymer dosages) over time, the fall cone tests were performed directly on the surface of the FFT samples deposited in 7.5 cm diameter and 10 cm tall polypropylene columns, which helped to avoid the difficulties that are generally associated with the placement of samples into the specimen cup (Feng, 2000). Maximum care was taken to select a relatively undisturbed and plain surface area for the cone to penetrate. For each polymer dosage used, 3 replicate columns were used for performing the fall cone test on a weekly basis. For keeping the boundary condition unchanged over time, the bleed water accumulated on the surface of the FFT samples in each column was removed carefully using a syringe and a fine needle before each measurement, and the same bleed water was restored again after each measurement. The mass of each column was recorded before and after the removal of bleed water as well as after the measurement of undrained shear strength, and finally, after the restoration of bleed water. The same water removal/restoration technique was followed during each measurement of undrained shear strength. The undrained shear strength values obtained from the undisturbed samples were compared to the undrained shear strength values obtained from the remoulded samples at the same water contents to determine the sensitivity of the fFFT samples.

3.6.9 Oedometer test and the determination of preconsolidation pressure

For the current study, oedometer tests were performed on the samples collected from the replicate columns to determine their void ratio-effective stress relationships and to track the changes in compressibility over time. Both undisturbed and remoulded samples

were tested for each sampling period. As it was observed in the case of natural sedimentary clays (Burland, 1990; Delage, 2010), a comparison between the compressibility behaviour of undisturbed and remoulded samples was found helpful in evaluating the influence of time-dependent structuration behaviour on the compressibility and strength characteristics of undisturbed samples.

3.6.10 Dewaterability tests

Settling test

Settling tests were typically carried out in 100 mL graduated glass columns (in duplicates) and the changes in the solid-liquid interface were monitored for 48 ~ 72 hours. However, in some cases, settling behaviour was observed for a relatively long period, up to several weeks.

CST test

Capillary suction time (CST) tests were conducted on the fFFT samples immediately after the mixing of the polymer. In addition to determining the optimum polymer dosage, CST tests were also used as an initial screening tool. Tests were performed to explore the dose-response relationship between the polymer dosage applied and the solids content of the FFT used and also as a check on the quality of the FFT-polymer mixture.

A Triton Type 319 Multipurpose CST equipment with 5 single radius test heads was used for performing the CST tests. Two types of CST filter papers were used. One of

them was a relatively thinner and lighter Ofite special CST filter paper (0.340 mm thickness) from OFI Testing Equipment with higher porosity (specifically developed for the petroleum industry applications involving very viscous or slow filtering systems). The other CST filter paper was a Whatman Grade 17 CHR type filter paper, which is a relatively thicker and heavier filter paper (0.920 mm thickness and 413 g/m² weight).

The same sampling procedure of collecting a mixture of flocs and released water from the sample surface was followed throughout the research in a consistent manner. However, filter paper characteristics such as pore size, thickness, and weight were found to influence the measured capillary suction times. Whatman Grade 17 CHR type filter paper generated relatively higher CST values because of the thickness of the filter paper, whereas Ofite special CST paper produced lower CST estimates due to the thinness. Capillary suction times were also found to vary depending on the polymer dosage and tailings properties (such as solids content).

Chapter 4: Determination of optimum polymer dosage using UV-vis spectrophotometry

4.1 Dewatering behaviour of polymer amended FFT: Need for optimum dosage

The slow settling of fine clay particles in oil sands residuals results in the generation of very large volumes of fine tailings in tailings ponds over time, thereby posing the biggest challenge in tailings management. The recent enactment of tailings regulations has accelerated the development of technologies for improving the dewaterability of FFT. Several technologies including in-line flocculation, tank thickening, and centrifugation involve a dewatering step using a polymer to drive flocculation. The application of synthetic polymers induces flocculation of the fine clay particles and release of water from a thick suspension of tailings solids, thereby improving the tailings dewaterability and settling behaviour and minimizing the volume of tailings. Field trials have shown that polymers allow the dewatering technologies to increase the solids concentration (by mass) to at least 45% within 48 hours following deposition (for in-line flocculation) and up to 55% (for centrifugation) (Matthews et al., 2011; Wells, 2011).

Most research on polymer optimization is typically directed towards maximizing short-term dewatering, over one- or two-days post-deposition or post-treatment. However, the effectiveness of the tailings dewatering treatment over months to years may be equally or more important to the deposit performance. To achieve the regulatory standards and develop a tailings deposit safe and strong enough for reclamation, it is thought that a solids content of at least 70% (which is equivalent to a gravimetric water content of 43%) is required (McKenna et al., 2016), which must be achieved through post-deposition

dewatering phenomena, such as consolidation, evaporation, or freeze-thaw. The recent changes in tailings regulation (Directive 085) also encourage consideration of dewatering over longer timescales, up to decades. Flocculation conditions (polymer dosage, mixing time, mixing speed, etc.), flocculation characteristics (floc size, density, settling velocity, etc.), and pore water chemistry (ionic strength, pH, and cation exchange capacity) play a prominent role in the short-term settling and dewatering of tailings. However, other factors such as consolidation gain importance in the long-term dewatering of tailings (Salam et al., 2016). Hence, the best performing polymer or the optimum polymer dosage that is selected for a given tailings composition based on its short-term dewatering performance could be different from the polymer or the polymer dosage that might work better for the same tailings in longer-term, which is due to the differences in the short-term and the long-term thickening and dewatering behaviour and mechanisms of tailings (Mizani et al., 2013).

Theoretically, an ideal flocculant should increase the efficacy of the tailings treatment technologies and result in higher water recovery and settlement without affecting the water chemistry or increasing the operational costs (Vedoy and Soares, 2015). However, selecting the appropriate polymer and optimizing its parameters such as dosage for a given tailings composition has always been a challenge for the oil sands industry. Polymer selection and optimization are directly associated with the treatment efficiency as well as the economy. Using too little or too much of polymer relative to the requirement may negatively impact the treatment efficiency, the quality of the end products, and above all, the cost-effectiveness of the process. Also, optimum polymer dosage may vary widely based on the changes occurring in tailings composition and characteristics, and therefore,

requires continuous adjustment. Hence, there is a need for optimization technologies that can assess flocculation performance and quantify dewatering efficacy immediately after the mixing of polymer into the tailings stream and can also provide feedback information to adjust and correct the polymer dosage in real-time on a continuous basis.

4.2 Dewatering optimization technologies

In wastewater treatment, a new technology has been developed that utilizes a UV-vis absorbance-based analytical method for in-line and real-time monitoring of residual polymer concentration in filtrate/centrate and determination of optimum polymer dosage for the dewatering of wastewater sludges (Gibbons and Örmeci, 2013; Al Momani and Örmeci, 2014a, 2014b; DiMassimo and Örmeci, 2015). The UV-vis or ultraviolet-visible spectrophotometry (or spectroscopy) refers to a technique of absorption spectroscopy in the ultraviolet-visible spectral region, which is frequently used for the quantitative analysis in analytical chemistry. The technique is based on the theory that the colour of a substance has a close association with its electronic structure. The UV-vis spectrophotometer uses light in the ultra-violet and visible ranges on the sample to be analyzed, and subsequently, the sample exhibits absorption in the above ranges as the radiation prompts a change in the electronic state of the molecules or ions in the sample. The absorptions are measured in terms of wavelength expressed in nanometers (Thermo Spectronic, 2011).

The results from the UV-vis spectroscopy studies showed that the polyacrylamide polymers strongly absorb light in the 190 ~ 200 nm range and the polymer concentration can be determined directly by measuring sample absorbance at these wavelengths (Gibbons and Örmeci, 2013; Al Momani and Örmeci, 2014a). The highest absorbance was measured

at 190 nm and beyond 240 nm there was no significant absorbance (Al Momani and Örmeci, 2014a). At 190 nm, the sample absorbance was found linearly correlated with the polymer concentration. When the polymer dosage was incrementally increased, the filtrate absorbance exhibited a 'U' shaped curve where the dip corresponded to the lowest polymer concentration; the lowest residual polymer concentration in filtrate again corresponded to the optimum polymer dosage (Al Momani and Örmeci, 2014b). Full-scale testing at three wastewater treatment plants using an in-line UV-vis spectrophotometer confirmed that the optimum polymer dosage could be identified and adjusted as needed by monitoring the lowest filtrate absorbance at or around 190 nm (DiMassimo and Örmeci, 2015).

Recently, the potential of the UV-vis absorbance based analytical method was evaluated for determining the optimum polymer dosage for achieving the best thickening and dewatering performance from oil sands tailings (Salam et al., 2016). Similar to the filtrate/centrate from the wastewater sludges, at 190 nm wavelength, the sample absorbance was found linearly correlated with the residual polymer concentration in tailings bleed water or reclaimed water. The method was also used to determine the optimum polymer dosage by monitoring the residual polymer concentration in tailings water. The detection limit of the method for polymer A3338 (SNF) was established as 0.37 mg/L in reclaimed water, which showed that the method has the sensitivity to detect very low polymer concentrations. Any potential interferences from fines, salinity, and pH were also investigated and were shown not to have an impact on the detection of the polymers. The results of the research indicated that the optimum polymer dosage for the dewatering of oil sands tailings could be determined by using an in-line UV-vis spectrophotometer (that can

generate real-time information) and directly measuring the residual polymer concentration in tailings water (Salam et al., 2016).

Recently, another technique was trialled successfully in the wastewater treatment plants for optimizing the polymer dosage in-line and in real-time, where a torque rheometer was used for measuring the rheological properties of wastewater sludge (Örmeci et al., 2004; Örmeci and Abu-Orf, 2005, 2006; Örmeci, 2007; Murray and Örmeci, 2008, 2009). The typical output of the torque rheometer is a torque-time rheogram that depicts the torque-time relationship for the period during which the sludge is subjected to a range of shears and the polymer is injected directly into the sludge. The rheometer measures the viscosity-related torque on the mixing impellers and this viscosity-related torque is produced by the resistance of the sample to the shearing action (Örmeci et al., 2004).

When trialled on the wastewater sludge, the torque rheometry allowed the observation of both flocculation and deflocculation phases (after adding polymer directly into the sample) and the measurement of rheological properties including peak torque and totalized torque (the area under a torque-time rheogram at a given time that reflects the relative strength of the floc network) from the rheograms. A local decrease in the measured peak torque and totalized torque values were found to correspond to the optimum polymer dosage (Örmeci, 2007). When the potential of such torque-rheology based methods was evaluated for determining the optimum polymer dosage for dewatering of oil sands tailings in a laboratory-scale experiment, the results demonstrated that the methods could be used to describe the effects of polymeric flocculation on tailings and identify the optimum polymer dosage that yields the best short-term dewatering.

4.3 Determination of optimum polymer dosage using UV-vis spectrophotometry

4.3.1 Background

The UV-vis absorbance-based analytical method was initially developed for the dewatering optimization of wastewater sludges and was successfully tested at three full-scale treatment plants using an in-line UV-vis spectrophotometer. A study was undertaken for evaluating the potential of the UV-vis method for monitoring the residual polymer concentration in reclaimed tailings water or bleed water and determining the optimum polymer dosage for the dewatering of tailings by measuring the absorbance at 190 nm. Also, the detection limit and the sensitivity of the method in tailings bleed water were determined and the effects of fines, salinity, and pH on polymer detection were evaluated. The main objective of the study was to explore the possibility of designing a process control system that could identify the optimum polymer dosage based on the measurement of absorbance and adjust the polymer dosage in real-time using an in-line UV-vis spectrophotometer.

4.3.2 Materials and methods

Oil sands tailings

Oil sands tailings and reclaimed water samples were collected from a tailings pond in Northern Alberta, Canada, and shipped to Carleton University in Ottawa, Canada. The total solids content of the tailings was measured as $35 \pm 0.3\%$.

Polymer stock solution and conditioning of tailings

Polymer A3338 (SNF), an anionic polyamide-based flocculant, was used to condition the tailings samples. In a plastic weighing dish, 1g of A3338 polymer (for preparation of 0.2% polymer stock solution) or 2g of A3338 polymer (for preparation of 0.4% polymer stock solution) were weighed using an analytical balance (Fisher Scientific, Sartorius AG Germany, LE225D) and decanted into a 500 mL glass beaker and completed to 500 mL with deionized water. The polymer solutions were stirred using a jar tester (Phipps and Bird, USA) at 200 rpm for 5 minutes and at 125 rpm for the following 55 minutes. Then, they were mixed with a hand blender for 10 seconds and left for maturation for 1 hour.

For conditioning of tailings, the polymer dosage added to the tailings samples was increased incrementally to cover the underdose, optimum dosage, and overdose ranges in the experiments. A predetermined volume of the polymer solution was injected in 250 mL of the tailings sample to produce the mixture with desired final polymer dosage and the mixture was mixed at 60 rpm for 80 sec (Yao et al., 2012) using a jar tester (Phipps and Bird, USA). The polymer dosage was calculated based on the dry solids content of samples and reported as g polymer/kg dry solids (DS).

CST and settling tests

The capillary suction time (CST) test is a commonly used filtration test to quantify dewatering properties. The CST apparatus consists of a cylinder placed on a platform equipped with two sensors and a digital timer connected to the sensors. The instrument measures the time it takes for the filtered water to travel from the first sensor to the second sensor, and shorter times indicate better filtration properties. Triton Type 319 Multipurpose

CST with 5 single radius test heads was used in the experiments. A CST paper (OFI Testing Equipment, USA) with higher porosity specifically developed for the petroleum industry applications involving very viscous or slow filtering systems was used in the experiments. CST tests were carried out immediately after the conditioning of the tailings samples with the polymer. After conducting the CST tests, the tailings samples dosed with six different polymer dosages were left to settle in six 100 mL graduated glass cylinders. Settling tests were carried out in duplicates and the solid-liquid interface was monitored for 48 hours. In some experiments, settling time was increased to 7 days.

Determination of polymer concentration

The polymer concentration in samples was determined by measuring their absorbance with a UV-vis spectrophotometer (Cary 100 Bio UV-vis Spectrophotometer, Varian Inc./Agilent Technologies, Canada) using quartz cuvettes (Hellma Canada Ltd., Canada). The absorbance data were recorded using Scan software (Cary WinUV, Varian Inc., Canada) in the wavelength range of 190 ~ 800 nm, and the absorbance values at 190 nm were used for the calculation of polymer concentration. Each measurement was repeated three times, and the reported absorbance measurements were the average of three replicates.

Polymer concentrations were first measured in deionized water and then in tailings supernatant. The tailings supernatant was collected from the top of the solids-liquid interface after the tailings samples were settled for 48 hours and diluted with deionized water using dilution factors of 10, 25, 50, and 100. Deionized samples with polymer did not require to be diluted before absorbance measurements.

Effect of fines on detection of polymer

To evaluate the effect of fines on the detection of polymer, settling tests were repeated. After 48 hours of settling, tailings supernatant was collected from the top of the solid-liquid interface and was centrifuged (Sorvall Legend Micro 21 Micro-centrifuge) in 2 mL centrifuge tubes at 14,300 rpm in order to remove the suspended fines. The absorbance values were then measured after diluting the samples with deionized water using dilution factors of 25, 50, and 100.

Effect of salts on detection of polymer

In these experiments, the reclaimed water collected from an oil sands tailings pond in Northern Alberta was used. A stock solution of 40 g/L of sodium chloride (NaCl) was prepared in deionized water. In 5 centrifuge tubes, the required volume of polymer stock solution as well as the required volume of NaCl stock solution were added to the reclaimed water such that the final samples had polymer concentration equal to 10 mg/L, with NaCl concentration values equal to 0, 250, 500, 750, and 1000 mg/L. The final volume of the sample in each centrifuge tube was 50 mL. The absorbance of the samples was then measured in the range of 190 ~ 260 nm.

Effect of pH on detection of polymer

In these experiments, both deionized water and reclaimed water collected from an oil sands tailings pond in Northern Alberta were used. In seven 50 mL centrifuge tubes, the required volume of polymer stock solution as well as the required volume of HCl (0.1 N) or NaOH (0.4%) were added to reclaimed water such that the final samples had a polymer

concentration equal to 10 mg/L with pH values equal to 3, 4, 5, 6, 7, 8, and 9. In seven other centrifuge tubes, the required volume of HCl (0.1 N) or NaOH (0.4%) was added to deionized water such that the final samples had pH values equal to 3, 4, 5, 6, 7, 8, and 9. The final volume of all samples was 50 mL. The pH of samples was measured using a pH meter (ORION 5 star, Thermo Scientific, Canada). The absorbance of the samples was measured and recorded in the range of 190 ~ 800 nm.

4.3.3 Results and discussions

Determination of optimum polymer dosage with conventional tests

CST test was used to monitor the improvement in the filterability of the tailings samples upon polymer addition. Six tailings samples were mixed with 6 different dosages of 0.4% polymer stock solution to give final polymer concentrations of 0.2, 0.4, 0.6, 0.8, 1.0, and 1.2 g/kg DS. The shortest CST time (1500 seconds) was measured at 0.6 g/kg DS, which indicated the optimum polymer dosage (Figure 4.1). Increasing the polymer dosage from 0.2 g/kg DS to 0.6 g/kg DS decreased the CST values from 2600 seconds to 1500 seconds indicating an improvement in dewaterability with increasing polymer concentration in the underdose range. However, overdosing the samples beyond 0.6 g/kg DS resulted in a deterioration in dewaterability and increased the CST values to 2200 seconds.

Figure 4.1. Determination of the optimum polymer dosage with the CST test.

In addition to the CST, settling tests were carried out after conditioning the tailings samples at polymer dosages of 0.2, 0.4, 0.6, 0.8, 1.0, and 1.2 g/kg DS. Settling behaviour of the tailings samples was recorded after 48 hours and 7 days in order to study the effect of settling time on the settling and consolidation characteristics at different polymer dosages. As expected, increasing the settling time increased the volume of the supernatant collected from above the solid-liquid interface as well as the solids concentration of the tailings collected from below the solid-liquid interface (Figure 4.2a and 4.2b). After 48 hours of settling, the highest supernatant volume and the highest solids concentration were measured at 0.8 g/kg DS, which indicated the optimum polymer dosage (Figure 4.2a). Increasing the polymer concentration until 0.8 g/ kg DS increased the water volume and solids concentration, but higher dosages than the 0.8 g/ kg DS deteriorated the settling

characteristics and decreased the supernatant volume and the solids concentration, which are typical indicators of the overdose range. After 7 days of settling, the changes in volumes and solids concentrations with different polymer dosages did not appear to follow the same trend (Figure 4.2b). The volume of the supernatant and the solids concentration gradually increased with increasing polymer dosage until 0.8 g/kg DS was reached. Beyond 0.8 g/kg DS, a deterioration in the settlement was not observed and in fact, a slight increase in supernatant volume and solids concentration was measured at 1.2 g/ kg DS. This change was likely due to the difference in the short-term and long-term thickening and dewatering behaviour of tailings (Mizani et al., 2013).

Figure 4.2(a). Determination of the optimum dosage with the settling test after 48 hours of settling.

Figure 4.2(b). Determination of the optimum dosage with the settling test after 7 days of settling.

Measurement of polymer concentration with absorbance method

Measurement of polymer in deionized water

Deionized water is free of impurities and initial experiments were carried out with deionized water to establish the absorbance spectra of polymer A3338. Deionized water was mixed with increasing dosages of 0.2% polymer stock solution to prepare samples with final polymer concentrations of 0, 0.5, 1, 2, 3, 4, 5, 10, 15, 20, 50, 100, 150, and 200 mg/L. The absorbance of the samples was measured in the wavelength range of 190 nm to 800 nm. There was no significant absorbance beyond 240 nm, and therefore, the absorbance spectra of samples are presented until 260 nm in Figure 4.3a. The working range of the

UV-vis spectrophotometer used was 0 ~ 4 AU, however, the suggested range for absorbance measurements was between 0 and 1 AU. For samples with less than 20 mg/L polymer, the highest absorbance was measured at 190 nm. Samples with very high polymer concentrations (50, 100, 150, and 200 mg/L) reached absorbance values of 2 ~ 2.7 AU, and the absorbance peak moved towards 192 ~ 196 nm due to the high concentrations of the samples. Diluting these samples would bring the peak absorbance back to 190 nm.

Figure 4.3b shows the calibration curve established by plotting the absorbance values of samples (0 ~ 50 mg/L) measured at 190 nm. Using the peak absorbance wavelength increases the sensitivity of the measurements and allows measuring lower polymer concentrations (Al Momani and Örmeci, 2014b). The results show that at 190 nm a strong linear relationship ($R^2 = 0.99$) exists between the sample absorbance and concentration, and polymer concentrations in unknown samples can be determined by using such a calibration curve.

Measurement of polymer in reclaimed water

Experiments were also repeated using reclaimed water collected from a tailings pond. Unlike deionized water, reclaimed water contains many dissolved and suspended contaminants as well as hydrocarbons that can interfere with the measurement of the polymers. In addition, the detection limit of the method in reclaimed water was established. The method detection limit (MDL) is defined as the minimum concentration of a substance that can be measured with 99% confidence and that is greater than zero (Glaser et al., 1981). MDL shows the lowest concentration that can be confidently measured and differentiated from the background noise. Hubaux and Vos (1970) developed a statistical analysis for the

determination of MDL for linear calibration curves, which was implemented in an excel spreadsheet available at the Chemiasoft website (www.chemiasoft.com). The spreadsheet readily calculates the MDL, upon providing some required values, along with standard concentrations and their corresponding absorbance measurements using a UV-vis spectrophotometer. For establishing the MDL, 7 sample replicates were used with 3 measurement replicates for each sample.

For confirmation of the absorbance method in reclaimed water and determination of the MDL, reclaimed water samples at different polymer concentrations (0, 0.5, 1.0, 1.5, 2.0, 2.5, 2.5, and 3.0 mg/L) were prepared. These samples were then diluted with deionized water using a dilution factor of 50. The samples required dilution due to the initially high absorbance of the reclaimed water. The results illustrating the absorbance spectra of the samples and the resulting calibration curve at 190 nm are presented in Figure 4.4a and 4.4b. The contaminants in the reclaimed water did not have a significant impact on the absorbance spectra and increasing the polymer concentration moved the absorbance curves up as expected. A strong linear relationship ($R^2 = 0.996$) between sample absorbance and polymer concentration was also established. Following the Hubaux and Vos (1970) statistical method, the detection limit calculated for the polymer A3338 in reclaimed water was 0.37 mg/L and confirmed the capability of the method in detecting very low polymer concentrations even in a complex matrix such as tailings reclaimed water.

Figure 4.3(a). Absorbance spectra of deionized water samples with 0-200 mg/L polymer.

Figure 4.3(b). Establishment of a calibration curve at 190 nm (0-50 mg/L polymer).

Figure 4.4(a). Absorbance spectra of reclaimed tailings water samples (0-3 mg/L polymer).

Figure 4.4(b). Establishment of calibration curve at 190 nm (0-3 mg/L polymer).

Determination of optimum polymer dosage with the absorbance method

The above experiments showed that the absorbance method can successfully be used to measure the polymer concentration in reclaimed water. The objective of these experiments was to determine whether the absorbance method can also be used to identify the optimum polymer dosage required to condition the oil sands tailings to achieve the best thickening and dewatering performance. Settling tests were carried out with tailings samples conditioned at 0.2, 0.4, 0.6, 0.8, 1.0, and 1.2 g/kg DS polymer in 100 ml graduated cylinders. After 48 hours of settling, supernatant samples were collected, and their absorbance values were measured. The absorbance values were used to determine the residual polymer concentration and were correlated to the optimum polymer dosage. In addition, the optimum polymer dosage was determined using the CST tests for comparison to the optimum polymer dosage determined with the absorbance method.

Figure 4.5a-e shows the results from the absorbance measurements at 190 nm as well as the results from the CST tests. The absorbance of the collected supernatant samples was measured at dilution factors of 10, 25, 50 and 100. The lowest CST values corresponded to the polymer dosage of 0.6 g/kg DS, which was identified as the optimum polymer dosage. The lowest absorbance value of the supernatant samples also corresponded to the polymer dosage of 0.6 g/ kg DS. In the underdose range (< 0.6 g/kg DS), increasing the polymer dosage decreased the absorbance of the supernatant samples until the optimum dosage was reached due to the improvement in the quality of the settled water after polymer addition. Exceeding the optimum dosage of 0.6 g/kg DS, however, resulted in an increase in the absorbance of the samples indicating an increase in the

residual polymer concentration in the supernatant. Similar results from filtration and centrifugation based dewatering tests were previously reported for wastewater sludge (Al Momani and Örmeci, 2014a; Aghamir-Baha and Örmeci, 2014). When the polymer is gradually added to a suspension of solids, it reacts with all available surfaces until the optimum polymer dosage is reached. At the optimum dosage, all available surfaces are saturated with polymer and any additional polymer added to the suspension remains in the water phase. Thus, in the overdose range, a linear increase in the residual polymer concentration in the water phase is observed with increasing polymer concentration (Gregory, 1988; Chang et al., 2002). The results illustrated in Figure 4.7 show that the dip on the absorbance curve can be used to identify the optimum polymer dosage required for the oil sands tailings. Increasing the dilution factor decreased the absorbance values as expected, but the dip in the absorbance curves could be clearly seen at all dilution ratios. Among the dilution ratios tested, 1:50 worked best with this tailings sample and brought the absorbance values below 1 without over-diluting the sample (Figure 4.5d).

Figure 4.6 presents the sample absorbance data from Figure 4.5 (DF = 50) but at wavelengths 220 nm, 254 nm, and 300 nm in addition to 190 nm. The results show that the dip in the absorbance observed at the optimum polymer dosage at 190 nm decreases radically at 220 nm and cannot be distinguished anymore at 254 and 300 nm. This confirms that any absorbance or turbidity measurement would not be able to detect this dip, and it is necessary to use 190 nm to be able to measure the residual polymer concentration. Based on the polymer characteristics, the peak absorbance wavelength for water-soluble polymers may shift between 185 and 200 nm.

Figure 4.5. Determination of optimum dosage with CST and absorbance of the tailings supernatant at 190 nm. Dilution factors are 0, 10, 25, 50, and 100.

Figure 4.6. The absorbance of the tailings supernatant samples at 190 nm, 220 nm, 254 nm, and 300 nm. The dilution factor is 50.

Effect of fine solids on the determination of optimum dosage

In order to determine the effect of fine solids on the identification of the optimum dosage, supernatant samples collected after 48 hours of settling tests were centrifuged to remove the suspended fines as much as possible. The absorbance values at 190 nm before and after centrifugation are illustrated in Figure 4.7a and 4.7b. Centrifugation was

successful in reducing the fines, which decreased the absorbance values. The trend of the absorbance curves did not change after centrifugation and the optimum dosage could still be found by searching the dip or the lowest absorbance value. These results showed that the fines did not have a significant impact on the measurements or on the determination of the polymer dosage.

Effect of salts on the detection of polymers

Salts naturally occur in oil sands ore and are usually present in high concentrations in oil sands tailings and reclaimed water. Thus, it is important to investigate the potential interference of salts on the absorbance and detection of polymers. In these experiments, NaCl concentration of reclaimed water containing 10 mg/L polymer was increased in a stepwise manner between 0 ~ 1000 mg/L and changes in the absorbance characteristics of the 10 mg/L polymer sample were studied. Adding NaCl did not change the shape of the absorbance curve and there were no new peaks (Figure 4.8a). The incremental addition of NaCl to polymer solutions was followed by a linear increase in absorbance at 190 nm (Figure 4.8b). This increase is likely due to the effect of chloride ions which absorb light below 200 nm (Korshin et al., 1997). However, this would not affect the detection and quantification of polymers since any salts present in the tailings would be part of the background absorbance and would be reflected in the calibration curves of polymers.

Effect of pH on the detection of polymers

The effect of potential pH changes on the detection of polymers with the absorbance method was also investigated. Figure 4.9 shows the absorbance values of reclaimed and

deionized water samples at 190 nm under different pH values (3 ~ 9). The pH change was achieved by adding HCl or NaOH to the samples. The trend named 'baseline' is the absorbance of deionized water with no added acid or base. When the absorbance of the deionized water was decreased to below 7 or increased to above 7, a gradual increase in the absorbance values was observed. This increase was likely due to the addition of ions to the deionized water, which lacks ions that can absorb UV light. An increase in light absorbance around 200 nm was previously reported for Cl^- and OH^- ions (Zuman and Szafranski, 1976; Korshin et al., 1997). For the reclaimed water samples, there was a little variance in absorbance with the change in pH due to the high concentration of ions present in the reclaimed water, and the results indicated that there would not be a significant impact on absorbance measurements. It was previously reported that pH remains relatively consistent in the range of 8-8.5 in a tailings pond, and it does not vary with depth (Wells, 2011).

Figure 4.7. Effect of fine solids on absorbance measurements: (a) without centrifugation; and (b) with centrifugation.

Figure 4.8. (a) Absorbance spectra of 10 mg/L polymer samples containing 0 ~ 1000 mg/L NaCl; and (b) Calibration curve at 190 nm.

Figure 4.9. Effect of pH on deionized water and tailings reclaimed water samples at 190 nm.

Implications for dewatering of oil sands tailings

The results of this study show that UV-vis spectrophotometry can be used to monitor the polymer concentration in oil sands tailings, and more importantly, it can also be used to determine the optimum polymer dosage required to achieve the best dewatering performance possible. The optimum polymer dosage corresponds to the lowest absorbance of the tailings water at 190 nm and can be determined by identifying the minimum dip in the absorbance curve as illustrated in Figure 4.7. The method is quick and simple, and it can be employed in real-time using in-line UV-vis spectrophotometers and auto-dilution

systems that already exist. Centrifugation and filtration-based mechanical dewatering methods are increasingly used for initial dewatering of oil sands tailings, and a minimum search algorithm can be employed to search for the minimum absorbance value in tailings filtrate or centrate and adjust the polymer dosage accordingly in real-time. The method presented in this paper was initially developed for the dewatering optimization of wastewater sludges and has been successfully tested and confirmed at three full-scale treatment plants using an in-line UV-vis spectrophotometer (DiMassimo and Örmeci, 2015). This study shows that dewatering of oil sands tailings can be optimized by monitoring the residual polymer concentration in the tailings water, which would lead to major savings by improving the treatment efficiency and reducing the volume of tailings stored in tailings ponds and help to reduce the environmental risks caused by tailings ponds.

4.3.4 Conclusion

In this study, an absorbance-based method was employed to determine the concentration of a polyamide polymer in reclaimed water collected from a tailings pond. The method was found capable of accurate detection and measurement of polymer concentrations. The detection limit of the method for polymer A3338 (SNF) was established as 0.37 mg/L in reclaimed water, which showed the method was sensitive enough to detect very low polymer concentrations. At 190 nm wavelength, sample absorbance was found linearly correlated with the polymer concentration. Furthermore, the method was also used to determine the optimum polymer dosage required to achieve the best thickening and dewatering performance by monitoring the residual polymer concentration in tailings water. When the polymer dosage was incrementally increased, the

absorbance exhibited a 'U' shaped curve where the dip or the lowest point on the curve corresponded to the optimum polymer dosage. Any potential interferences from fines, salinity, and pH were also investigated and were shown not to have an impact on the detection of the polymers. The ultimate goal of this research was to lay the foundation for the development of an automation system for optimizing the polymer dosage in-line and real-time and to make the thickening and dewatering technologies for the oil sands tailings more efficient and cost-effective. The results of the research indicated that thickening and dewatering optimization of oil sands tailings can be achieved by directly measuring the residual polymer concentration in tailings water using an in-line UV-vis spectrophotometer that can generate real-time information.

Chapter 5: Determination of optimum polymer dosage for dewatering of oil sands tailings using torque rheology

5.1 Introduction

The slow settling of fine clay particles in oil sands residuals takes decades in tailings ponds and results in the accumulation of large volumes of fluid fine tailings in tailings ponds over time, which poses the biggest challenge in tailings management. A pressing need for overcoming the challenges associated with the slow dewatering and consolidation of oil sands tailings deposits as well as the enactment of new tailings regulation has accelerated the development of tailings treatment technologies (Canadian Association of Petroleum Producers, 2018). Some of the improved tailings treatment technologies (i.e., in-line flocculation, tank thickening, and centrifugation technique) use polymers to promote the flocculation of clay particles for faster dewatering. However, the efficiency as well as the economy of the dewatering operations mainly depend on operating at the optimum polymer dosage. Selecting an appropriate polymer dosage for a given tailings composition or adjusting the polymer dosage based on the changes in tailings composition has always been a challenge for the oil sands industry. Adding the right polymer at an optimum dosage for tailings during its transportation to the tailings ponds for deposition could improve dewaterability significantly and result in higher water recovery and settlement decreasing the operational costs. By contrast, underdosing or overdosing of the same polymer could negatively impact the treatment efficiency, thereby leading to a sharp decline in the dewaterability and limiting the water that could be released from the tailings solids both in short- and long-term substantially increasing the operational costs (Cabrera et al., 2009;

Vedoy and Soares, 2015; Salam et al., 2016). Also, the polymer dosage required for an optimal dewatering performance could vary depending on the changes occurring in tailings composition and characteristics, and therefore, may require a continuous adjustment in field conditions. Hence, there is a need for optimization technologies that can assess flocculation performance and quantify dewatering efficacy immediately after the mixing of polymer into the tailings stream and also provide feedback information to adjust and correct the polymer dosage on a continuous basis. Since polymer costs constitute a substantial portion of the tailings dewatering operation costs, an optimization technique or method that could optimize the polymer dosage in real-time (or near real-time) could provide the oil sands industry with considerable operational and economic advantages and minimize the adverse effects on the environment from the unnecessary and excessive use of polymers.

There are similarities between the sludge in wastewater treatment and the fluid fine tailings in oil sands in that they both have similar fine particle sizes, the presence of organic matter, and the relatively high solids contents (Zhu et al., 2017). Similar to the oil sands industry's challenge with the dewatering and densification of tailings deposits, optimization of conditioning and dewatering of sludge has been a major challenge for the wastewater treatment plants. There is a substantial body of research on new and novel approaches to sludge dewatering, some of which can potentially be adapted to oil sands tailings. For example, the measurement of rheological parameters (i.e., peak torque and totalized torque), which are indicative of material properties (Dentel et al., 2005) has been utilized for describing the effects of polymeric flocculation on the physical characteristics

and dewaterability of sludge and also used as predictive tools for optimizing the polymer dosage (Örmeci and Abu-Orf, 2005, 2006; Abu-Orf and Örmeci, 2005; Örmeci, 2007). Recently, the monitoring of residual polymer concentration using UV-vis spectrophotometry (Gibbons and Örmeci, 2013; Al Momani and Örmeci, 2014a, 2014b) has been shown to be effective in oil sands tailings as well (Salam et al., 2016).

When a wastewater sludge is subjected to a shearing action in a torque rheometer and mixed with the polymer simultaneously, the chemical and physical bonds between the flocs help them to form an elastic floc network, which provides the sample with the strength to withstand the shear forces applied (Örmeci and Abu-Orf, 2005; Örmeci, 2007). The results from the laboratory-scale experiments showed that quantifying the rheological properties (i.e., peak torque and totalized torque) could provide useful information about the floc network strength or the ability of the flocculated wastewater sludge to resist deformation, and therefore, on the degree of flocculation/deflocculation and the dewaterability of the sludge (Tixier et al., 2003; Murray and Örmeci, 2009).

A study was undertaken for investigating the potential of two rheological methods that are based on torque rheology and involve the measurement of rheological parameters (i.e., peak torque and totalized torque) to identify the optimum polymer dosage for the dewatering of oil sands tailings. When the tailings sample was subjected to a shearing action in a torque rheometer and a polymer solution was injected into that sample simultaneously, the rheometer measured the torque on the impeller. The peak torque and the totalized torque were determined from the torque-time rheogram and they provided useful information about the strength of the floc network in the sample, and also on the

degree of flocculation. When the polymer dosage was increased incrementally and the resulting peak torques and totalized torques were plotted against the corresponding polymer dosages (six polymer dosages in total: 0.2, 0.4, 0.6, 0.8, 1.0, and 1.2 g/kg of dry solids), a local decrease was observed in both plots at the dosage of 0.8 g/kg of dry solids, which also corresponded to the lowest capillary suction time obtained from the CST test at the same dosage.

The objectives of this study were: 1) to investigate how the polymer-tailings interactions affect the torque-time rheograms (i.e., the peak torques that are formed immediately after the polymer injection); 2) to determine whether the measurement of rheological parameters (i.e., peak torque and totalized torque) could be used as a reliable control parameter to describe the effects of polymeric flocculation; and, 3) identify the optimum polymer dosage that would yield the best short-term dewatering and also translate into major savings for the industry.

5.2 Background on torque rheology methods for studying conditioning and dewatering

Method 1

The first rheological method used for this study measured the peak torques that were observed in the torque-time rheograms after a predetermined amount of polymer solution was injected directly into the untreated tailings and mixed with the tailings using an impeller rotating at a preselected speed (expressed in rotation per minute or rpm). The method involves the measurement of torque values at three different successive steps using

three different rotational speeds and allows the observation of both flocculation and deflocculation phases (Figure 5.1) (Örmeci, 2007).

Figure 5.1. The measurement of peak torque from the torque-time rheogram.

In Step 1, the sample is mixed at 420 rpm for 10 seconds. The first step is crucial for mixing tailings uniformly. In Step 2, the sample is mixed at 300 rpm for 40 seconds. The second step provides adequate time for the torque measurement to stabilize after a period of initial turbulence. After 50 seconds (or at the beginning of Step 3), the polymer is injected into the tailings through the ports on the sides of the beaker. Since mixing conditions can affect the resulting rheological properties and should be picked according to the characteristics of the fluid, different impeller speeds are trialled before finally

selecting the rotational speed for the third step. The goal is to determine the impeller speed that is most sensitive to the changes in rheological parameters of the fluid.

Once the polymer solution is injected into a sample, a peak is observed in the torque-time rheogram. The formation and deformation of the peaks typically span less than 60 seconds. Therefore, a minimum mixing duration ($> 60 \sim 120$ seconds) is required in Step 3 in order to observe the peak torque as well as both flocculation and deflocculation phases. Figure 5.1 shows the steps followed and the results typically obtained from Method 1. The first half of the peak or the upward portion shows the flocculation phase when the flocs are formed, whereas the second half of the peak or the downward portion shows the deflocculation phase when the degradation of flocs occurs.

The formation of peaks in the torque-time rheograms can be explained based on the concept of elastic floc network (Michaels and Bolger, 1962; Firth and Hunter, 1976; Otsubo and Umeya, 1984; Langer et al., 1994). When the polymer is added to an unconditioned sample, the strength of the chemical and physical bonds between the flocs that forms such an elastic floc network increases, which in turn increases the resistance of the sample to the shearing action of the impeller. So, the apex of the peak refers to the point where such bonds between the flocs are broken and the floc network structure is disintegrated (Langer et al., 1994). Hence, the peak torque is an indirect reference to the floc network strength of the sample and also to the degree of flocculation (Murray and Örmeci, 2008).

In wastewater sludges, the initial peak torque in the torque-time rheogram was found to increase with the increasing polymer dosage (Campbell and Crescuolo, 1982). As

more polymer is added to the sludge, the flocs become more stable and the bonds between the flocs become stronger. Since more energy is needed to break up the bonds and the peak torque corresponds to the amount of energy dissipated in breaking up the bonds, subsequently, the height of the peak increases (Abu-Orf and Dentel, 1999).

Method 2

Method 2 involves the measurement of totalized torque (TTQ) or the total area under the flocculation curve in a torque-time rheogram, which stretches over a length of shear application time in Step 3. The time starts from the moment the polymer is injected into the sample (i.e., 50 seconds) and continues up to a shear application time that coincides with the end of the deflocculation phase (i.e., 100 seconds). Figure 5.2 shows the rheological method of measuring the totalized torque from a torque-time rheogram.

The totalized torque (TTQ) reflects the strength of the floc network formed in the flocculated sample and the variation in measured relative network strengths reflects the fundamental changes in the properties and behaviour of the sample material during both flocculation and deflocculation phases (Örmeci and Abu-Orf, 2005).

Figure 5.2. The measurement of totalized torque (the area bounded by the Time slices and Input slices) from the torque-time rheogram.

Örmeci and Abu-Orf (2005) derived mathematical models for estimating the relative network strength using a torque rheometer, which showed that the measurement of the area under the torque-time rheogram is representative of the total energy dissipation rate in a sludge system. The unit work energy required for breaking up the structure of a certain volume of sludge or the total energy dissipated during that process, which also reflects the network strength of that suspension can be expressed using the following equation (Michaels and Bolger, 1962):

$$W_u = \frac{[2\pi N * TTQ]}{V_b} \dots\dots\dots (5.1)$$

Where W_u = unit work energy (Joule/cm³); N = rotational speed (rpm); TTQ = totalized torque area or the area under the rheogram (Joule-min); and, V_b = sample volume (cm³).

The Floccky torque rheometer employed in this study uses a sample size of 200 mL and the totalized torque area (TTQ) is expressed in m J s; hence, equation (1) becomes:

$$W_u = \frac{[0.524N*TTQ]}{X} \dots\dots\dots (5.2)$$

Where W_u = work energy (J/Kg of dry solids); N = rotational speed (rpm); TTQ = totalized torque area or the area under the rheogram (m J s); and, X = solids concentration of sludge sample in g/L.

Hence, the totalized torque (TTQ) or the area under the torque-time rheogram is converted to the work energy required for processing a unit volume or mass of a given suspension (Chung, 1986), which also represents the total energy dissipated into the suspension during the process, and therefore, indicates the network strength (Michaels and Bolger, 1962). According to equation (5.1), if the sample volume and the rotational speed remain the same, the network strength is linearly proportional to the totalized torque (Örmeci and Abu-Orf, 2005). Therefore, for a given tailings composition, if the samples are dosed with different polymer dosages using the same sample volume and mixing intensity, the totalized torques measured from the torque-time rheograms can be used to compare the corresponding network strengths (Örmeci and Abu-Orf, 2005).

5.3 Materials and methods

Torque rheometer

Floccky Tester ODA-10 (Koei Industry Co., Ltd Japan) is a torque rheometer specifically designed to study the flocculation-deflocculation phases of slurries (Figure 5.3). After a sample is placed in the rheometer beaker ($V = 200 \text{ mL}$), a three-vane impeller, which is driven by a motor is used to apply a range of rotational shear forces to that sample. While the polymer solution is injected directly into the sample through ports on the sides of the beaker, the rheometer measures the viscosity-related torque on the impeller, which is produced by the resistance of the flocculated samples to the shearing action. The output of the torque rheometer is a torque-time rheogram, where the y-axis indicates the torque in mili-Newton-meter (mNm), and the x-axis indicates the duration during which the sample is subjected to a rotational shear force.

Figure 5.3. (Left) The Floccky Tester torque rheometer; and (right) the cylindrical beaker with ports on the sides for injecting the polymer solution directly into the sample.

Oil sands tailings samples

Oil sands tailings samples were collected from a tailings pond in Northern Alberta, Canada, and shipped to Carleton University in Ottawa, Canada. The total solids content of the tailings was measured as $35 \pm 0.5\%$. The electrical conductivity of the pore water was 1660 micro-siemens/cm, while the dominant ions were Na^+ (320 mg/L), Cl^- (130 mg/L), and CO_3^{2-} (377 mg/L). The properties of unamended tailings samples are summarized in the following table. For the reader's interest, detailed rheology of these tailings is presented in Mizani et al. (2017).

Table 5.1. The properties of unamended tailings samples.

Parameters	Average value
Initial solids content (%)	35.0 ~ 35.5
Initial water content (%)	181.7 ~ 185.7
Specific gravity	2.2
Liquid limit (%)	55.0
Plastic limit (%)	27.0
Clay content (MBI test) (%)	29.64
Clay mineralogy (XRD analysis)	Kaolinite (70%), Illite (30%)

Preparation of polymer solution and conditioning of tailings

Polymer A3338 (SNF), an anionic polyamide-based flocculant, was used to condition the tailings samples. In a plastic weighing dish, 2g of A3338 polymer was weighed using an analytical balance (Fisher Scientific, Sartorius AG Germany, LE225D) and decanted into a 500 mL glass beaker and completed to 500 mL with deionized water

to prepare a polymer solution with a final concentration of 0.4%. The polymer solutions were stirred using a jar tester (Phipps and Bird, USA) at 200 rpm for 5 minutes and at 125 rpm for the following 55 minutes. Then, it was mixed with a hand blender for 10 seconds and left for maturation for 1 hour.

A predetermined volume of the polymer solution was injected directly into the tailings sample in the rheometer to produce the tailings-polymer mixture with the desired final polymer dosage. The solution volume was increased incrementally to cover the under dosage, optimum dosage, and overdosage ranges. The polymer dosage was calculated based on the dry solids content of samples and reported as g polymer/kg dry solids (DS).

Capillary suction time (CST) and settling tests

The capillary suction time (CST) test is a type of static filtration test that measures the filtration rate of a slurry using a filter paper as the medium, and therefore, quantifies the dewatering properties of a sample in terms of how rapidly that sample can be dewatered (Gumfekar et al., 2019). The CST apparatus consists of a cylinder placed on a platform equipped with two sensors and a digital timer connected to the sensors. Before conducting the test, a CST filter paper is placed between the platform and the cylinder. When a flocculated sample is poured into the cylinder, the filter paper generates a capillary suction pressure to draw the filtrate from the sample. Capillary suction time (CST) refers to the time taken by the filter paper to draw a certain volume of filtrate so that the filtered water could travel from the first sensor to the second sensor. When CST tests are performed on the samples mixed with different dosages of a chemical conditioning agent (i.e., polymer), the lowest value of the capillary suction time indicates an optimum filterability and a high

permeability, whereas a higher capillary suction time refers to a lower filterability or high water retention capacity (Jin et al., 2004; Wells et al., 2011; Gumfekar et al., 2019).

In this study, Triton Type 319 Multipurpose CST equipment with 5 single radius test heads was used and a CST filter paper (OFI Testing Equipment, USA) with higher porosity that is specifically developed for the petroleum industry applications involving very viscous or slow filtering systems was used for conducting the CST tests.

In addition to CST tests, settling tests were carried out after conditioning the tailings samples. Tailings samples dosed with six different polymer dosages (0.2 - 1.2 g/kg DS) were left to settle in six 100 mL graduated glass cylinders. The thickness of the settled solids layer and the advancement of the solid-liquid interface were monitored for the subsequent 48 hours.

Selection of impeller speed

In order to determine the impeller speed that is most sensitive to changes in the rheological parameters in Step 3, a range of rotational speeds (100, 200, 300 and 400 rpm) were trialled on the tailings samples conditioned at four different polymer dosages (0, 0.3, 0.6, and 1.2 g/kg DS). The resulting peak torque and totalized torque values were compared to determine the optimum impeller speed.

Peak torque and totalized torque determination

For the measurement of rheological parameters, 200 mL of unconditioned tailings sample was placed in the cylindrical beaker. Then, the sample was sheared at a rotational

speed of 420 rpm for the first 10 seconds, and at a speed of 300 rpm for the next 40 seconds. After 50 seconds, a predetermined dosage of the polymer A3338 was injected directly into the tailings while the sample was sheared at a speed of 400 rpm. Immediately after the polymer injection, a steep peak was observed in the torque-time rheogram, which was due to the formation of the flocs and the increased resistance on the impeller surface in response to the shear. The torque values along with the peak were measured and reported by a data-acquisition software that was included with the Floccky Tester. The same rheological analysis was performed on six different tailings samples dosed with six different polymer dosages 0.2, 0.4, 0.6, 0.8, 1.0, and 1.2 g/kg DS. The resulting torque-time rheograms and the peak torque values provided information on the strength of the floc network and the degree of flocculation in the samples for different polymer dosages.

Using the ‘Time slice’ and ‘Input slice’ functions of the data-acquisition software, a total of four slice lines were set on the torque-time rheogram for determining the totalized torque (TTQ). The area under the flocculation curve selected for the totalized torque analysis included both upward (flocculation phase) and downward portions (deflocculation phase) of the peak and was bounded by the two horizontal slice lines (Time slices) and two vertical slice lines (Input slices) (Figure 5.2). While setting the Time slices, an appropriate shear application time (~ 100 seconds) was selected so that the adverse effect of the additional shearing during rheological measurements could be avoided. The network strength of the sample was determined using equation (5.2).

5.4 Results and discussions

Determination of optimum polymer dosage

Dewaterability of conditioned tailings samples were assessed using CST and settling tests. CST tests were performed immediately after the mixing of tailings with the polymer (Figure 5.4). All measurements were conducted in duplicates, with the mean value reported. The CST values ranged from 1144 to 1756 seconds. The polymer dosage corresponding to the shortest capillary suction time obtained (1144 seconds) was determined to be the optimum dosage (0.8 g/kg DS).

Figure 5.4. CST test results for the tailings flocculated at different polymer dosages.

In addition to CST tests, settling tests were carried out after conditioning the tailings samples at polymer dosages of 0.2, 0.4, 0.6, 0.8, 1.0, and 1.2 g/kg DS. Tailings samples dosed with six different polymer dosages were left to settle in six 100 mL graduated glass cylinders and the solid-liquid interface was monitored for the subsequent 48 hours. After

48 hours of settling, the highest supernatant volume as well as the highest solids settlement were measured at 0.8 g/kg DS, which indicated the optimum polymer dosage (Figure 5.5).

CST tests and settling tests were carried out in duplicates.

Figure 5.5. Settling test results for the tailings flocculated at different polymer dosages.

Determination of optimum impeller speed

Four different rotational speeds (100, 200, 300, and 400 rpm) were trialled at four different polymer dosages (0, 0.3, 0.6, and 1.2 g/kg DS) to determine the optimum impeller speed (Figures 5.6, 5.7, 5.8, and 5.9). When compared to the torque rheograms generated at 0 g/kg DS dosage (Figure 5.6), the torque rheograms generated at 0.3, 0.6, and 1.2 g/kg DS dosages showed that the rheograms obtained from the conditioned tailings were

different from the rheogram obtained from the unconditioned tailings, and also showed how the polymer-tailings interactions affected the formation of peak torques.

For each polymer dosage, when the peak torques and the totalized torques generated by different impeller speeds were compared, they were found to increase with the increasing impeller speed. 400 rpm produced the highest torque values and well-defined peaks. When the sample was sheared at 400 rpm after the polymer injection, the flocculated sample developed an elastic floc network with high strength and degree of flocculation. The torque rheometer also showed good reproducibility of measurements at 400 rpm impeller speed.

When the samples were sheared at 100 rpm, the changes in the torque-time rheograms after the polymer injection were minimal, especially at the lower polymer dosages (i.e., 0.3 g/kg DS), which was probably due to incomplete flocculation under a low shearing rate (Yao, 2016). The impeller speeds of 200 rpm and 300 rpm also failed to generate a significant output signal in a consistent manner. Hence, 100 rpm, 200 rpm, and 300 rpm speeds were not considered for further evaluation and discussion. 400 rpm was selected as the optimum impeller speed for this tailings sample, and the tailings were mixed with the polymer at 400 rpm for 360 seconds in Step 3. The selection of the right impeller speed that is most sensitive to changes in rheological parameters is important in understanding the behaviour of tailings during conditioning and determination of the optimum polymer dosage.

Experiments conducted for selecting the optimum impeller speed also shed light on the relationship among mixing, polymer dose, and formation of peaks. In the underdose

range (0.3 g/kg DS), all peaks occurred at the same time due to the rapid adsorption of available polymer on solids and consumption of free polymer in tailings (Figure 5.7). Around the optimum polymer dosage, the formation of peaks was delayed at lower impeller speeds due to the slower rates of polymer incorporation into the tailings matrix, and consequently increasing the time it takes for flocculation and deflocculation phases (Figure 5.8). This resulted in shifting of the peaks to the right at lower impeller speeds following the order of 400 rpm < 300 rpm < 200 rpm < 100 rpm in terms of the time it takes for the peaks to appear. In the overdosage range, the excess polymer in tailings led to the formation of multiple large peaks indicating the reoccurrence of flocculation and deflocculation phases due to the still available residual polymer in the tailings matrix (Figure 5.9).

Figure 5.6. Torque rheograms for 0 g/kg DS polymer dosage (100, 200, 300, and 400 rpm).

Figure 5.7. Torque rheograms for 0.3 g/kg DS polymer dosage (100, 200, 300, & 400 rpm).

Figure 5.8. Torque rheograms for 0.6 g/kg DS polymer dosage (100, 200, 300, & 400 rpm).

Figure 5.9. Torque rheograms for 1.2 g/kg DS polymer dosage (100, 200, 300, & 400 rpm).

Determination of optimum polymer dosage

After the selection of 400 rpm as the impeller speed, six different polymer dosages (0.2, 0.4, 0.6, 0.8, 1.0, and 1.2 g/kg DS) were mixed with the tailings. Figure 5.10 presents the torque-time rheograms produced at these polymer dosages.

Figure 5.10. Torque rheograms for increasing polymer dosages.

Based on rheological measurements illustrated in Figure 5.10, Figure 5.11 shows the magnitudes of the peak torques measured at increasing polymer dosages covering the underdose, optimum dosage and overdosage ranges. When the polymer dosage was increased incrementally in the underdose range, the magnitudes of the torques, as well as peaks, increased as the tailings showed more and more resistance to the shearing action. Around the optimum dosage, the peak torque of the highest magnitude was observed at 0.6 g/kg DS dosage, and at the next polymer dosage, the peak torque value declined; hence, there was a local decrease at 0.8 g/kg DS dosage. Afterwards, in the overdosage range, the peak torque value started to increase again.

Figure 5.11. Peak torque values measured for increasing polymer dosages.

When a polymer is added incrementally to a slurry sample in a torque rheometer and also slowly starts flocculating, the sample increases in viscosity, which in turn increases the resistance of the sample to the applied shear. Once the sample approaches an optimum flocculation level, there is a decrease in the total surface area of the particles, and subsequently, a decrease in the drag force. Such a decrease in drag force leads to a decrease in the resistance of the sample to the shear measured as lower torque values. When the optimum polymer dosage is exceeded, the excess polymer in the tailings supernatant increases the viscosity, which can be measured by an increase in torque values.

In this experiment, the peak torque was observed at 0.6 g/kg DS dosage, which implied that the sample dosed with 0.6 g/kg DS demonstrated the highest network strength

and resistance to the applied shear. There was a decrease in the peak torque value at 0.8 g/kg DS dosage due to the release of free water. Once the polymer dosage exceeded the optimum range, the peak torques started to increase in height. When the polymer reached the overdosage range, higher polymer dosages produced higher peaks as the flocs became more stable with stronger bonding between them, thereby requiring more energy to destroy them.

The decline in resistance to the applied shear at the optimum polymer dosage (0.8 g/kg DS) was also an indication of a reduced network strength, which was caused by the rupture of the network bonds. The reduced network strength allowed the flocculated tailings to release a major portion of the water that was associated with the flocs as free water. Such release of water lowered the drag force further, and subsequently, the peak torque values declined (Abu-Orf and Örmeci, 2005; Örmeci and Abu-Orf, 2006). However, when the polymer exceeded the optimum dosage range, the presence of residual, unmixed polymer in the sample reflocculated the ruptured network bonds again, and subsequently, the amount of energy necessary to break up the bonds reached a level even higher than the amount of energy needed for optimum dewatering. Hence, the peak torque value increased again in the overdosage region (at dosage > 0.8 g/kg DS).

Similar to the peak torques, the totalized torque (TTQ) values were also determined for the same polymer dosages using the data-acquisition software (Figure 5.12). Starting from the moment the polymer was injected into the sample (i.e., 50 seconds), the area under the flocculation curve in the torque-time rheogram was calculated up to an appropriate shear application time that coincides with the end of the deflocculation stage (i.e., 100

seconds). When the measured totalized torque values were plotted against the corresponding polymer dosages, the TTQ values increased with the increasing polymer dosage up to 0.6 g/kg DS; however, similar to the peak torques, a local decrease or dip was observed in the TTQ values at 0.8 g/kg DS dosage.

The network strengths of the flocculated tailings samples were also calculated by applying the already calculated totalized torque values to equation (5.2) (Figure 5.13). Similar to the totalized torque values, initially, the network strength values increased with the increasing polymer dosage; however, a drop was observed in the network strength values at 0.8 g/kg DS. At higher dosages, the network strength started increasing again due to excess polymer.

Figure 5.12. Totalized torque values measured for increasing polymer dosages.

Figure 5.13. Network strength values measured for increasing polymer dosages.

Validation with the dewaterability tests

A comparison between the peak torque and totalized torque values measured by the torque rheometer for different polymer dosages and the capillary suction times obtained from the CST and settling tests for the same polymer dosages showed that the local decrease in the peak torque values (Figure 5.11) and also the dip observed in the totalized torque values (Figure 5.12) corresponded to the lowest capillary suction time (Figure 5.3) and the highest solids settlement (Figure 5.4). Therefore, the optimum polymer dosage determined from the peak torque, totalized torque, and network strength values appeared to correlate well with the optimum dosage identified by the CST and settling test results (Figures 5.14 and 5.15).

Figure 5.14. CST, solids settlement, and peak torque for increasing polymer dosages.

Figure 5.15. CST, solids settlement, and totalized torque for increasing polymer dosages.

5.5 Conclusion

In this study, two methods that are based on torque-rheology were employed to determine the optimum polymer dosage for the dewatering of oil sands tailings. The first method measured the peak torques that were produced in the torque-time rheograms when the polymer was injected directly into the tailings in a torque rheometer. Peak torque provided useful information on the strength of the floc network in the flocculated samples, and also on the degree of flocculation. The second method measured the totalized torques or the areas under the flocculation curves in the torque-time rheograms, which reflected the relative strength of the floc network. When the peak torques and totalized torques were measured for different polymer dosages and compared, they were observed to increase with the increasing polymer dosage up to the optimum dosage, followed by a local decrease at the optimum polymer dosage. When compared to the results obtained from the dewaterability tests, the dip observed in the peak torque and the totalized torque values corresponded to the lowest capillary suction time and the highest solids settlement obtained from the dewaterability tests at the same dosage. The torque rheometer used in this study also has a model that can be used in-line and real-time and has the potential to achieve dewatering optimization at full-scale applications.

Chapter 6: Investigation of creep, thixotropy, and structuration in flocculated FFT

6.1 Introduction

Most research on polymer optimization is typically directed towards maximizing short-term dewatering, over one- or two-days post-deposition or post-treatment. However, the effectiveness of the tailings dewatering treatment over months to years may be equally or more important to the deposit performance. Since there are differences between the short-term and long-term thickening and dewatering behaviour and mechanisms of tailings, the best performing polymer or the optimum polymer dosage that is selected for a given tailings composition based on its short-term dewatering performance could be different from the polymer or the optimum polymer dosage that might work better for the same tailings in longer-term. The flocculation conditions (polymer microstructure and dosage, mixing time and speed, etc.), flocculation characteristics (floc size, density, settling velocity, etc.), and pore water chemistry (ionic strength, pH, and cation exchange capacity) play a prominent role in the short-term settling and dewatering of tailings. However, other factors such as consolidation gain more importance in the long-term dewatering in tailings deposits. Although the polymer-assisted flocculation technologies have accelerated the settling and dewatering performance of FFT, still it is not feasible to meet the regulatory requirements of reducing tailings volume and developing a certain shear strength by relying solely on the polymer addition; further dewatering through post-deposition mechanisms such as consolidation, desiccation, and freeze-thaw is essential (Charlebois, 2012). The recent changes in tailings regulation (Directive 085) also encourage consideration of dewatering over longer timescales, up to decades (AER, 2019).

The consolidation of tailings involves the slow settlement of fine-grained materials over time through the dissipation of excess pore water pressure and the subsequent gain in effective stress in response to either self-weight or vertical surcharge from a capping layer (BGC Engineering Inc., 2010; Jeeravipoolvarn, 2010). The consolidation of fines dominated FFT works slowly owing to the low hydraulic conductivity at relatively high void ratios. The hydraulic conductivity of FFT as well as fFFT are considerably below that of the typical clay deposits (Figures 6.1 and 6.2). The time required for consolidation of the fairly deep deposits (>50 m) currently used for the final disposal of fines dominated tailings is long, from years to decades. Over such time frame, the time-dependent creep, structuration, and thixotropic behaviour of tailings have been observed to influence its consolidation behaviour (i.e. 10-metre standpipe test on unamended oil sands tailings at the University of Alberta) (Jeeravipoolvarn, 2005; Jeeravipoolvarn et al., 2009; Miller, 2010). However, it is yet to be examined whether fFFT demonstrates similar time-dependent creep, structuration, and thixotropic effects or whether such effects have any implications for the long-term dewatering and consolidation performance of fFFT.

Figure 6.1. Hydraulic conductivity relationships for (top) sedimentary clays; (middle) residual soils; and, (bottom) unamended oil sands tailings (from Ito and Azam, 2013).

Figure 6.2. Hydraulic conductivity of unamended FFT and thickened fFFT (from Wilson et al., 2017).

The recent Alberta Energy Regulator (AER) regulation (Directive 085) requires the oil sands operators to develop a plan to manage both existing fluid tailings and future tailings production (AER, 2019). Under the Directive 085, the operators must set project-specific targets to ensure that the fluid fine tailings are in a ready-to-reclaim state within ten years of the end of the mine's life, and the operators need to work towards this target over the mine life (AER, 2019). The enactment of these regulations has accelerated the development of tailings treatment technologies (Canadian Association of Petroleum Producers, 2019). Some of the improved tailings treatment technologies such as in-line flocculation, tank thickening, and centrifugation technique use polymers to promote flocculation for faster dewatering. Field trials have shown that the application of polymers allows such technologies to increase solids concentration to at least 50% in the short-term,

usually within days (Matthews et al., 2011; Wells, 2011). However, to meet the regulatory standards and develop a deposit safe and strong enough for reclamation, it is thought that a solids concentration of at least 70% (equivalent to a gravimetric water content of 43%) is required, which is also close to the plastic limit of the material (McKenna et al., 2016). To take the tailings deposits to such a state, it is necessary to optimize the tailings deposition and management strategies, which again warrants a comprehensive understanding of the longer-term dewatering processes in FFT such as consolidation and potentially others such as creep.

An accurate prediction of consolidation behaviour is crucial to the sustainable management of tailings during operation, closure, and reclamation; however, it also needs an in-depth understanding of the large strain consolidation behaviour of the tailings deposits (Ito and Azam, 2013). Both creep and thixotropic behaviour have been observed to influence the final consolidation or settlement behaviour of unamended tailings deposits (Jeeravipoolvarn et al., 2009). Although creep is commonly modelled through the concept of rate-dependent compressibility, the conventional large strain consolidation models do not incorporate the effects of structuration and thixotropy. To incorporate the effects of creep, thixotropy, and structuration into the large strain consolidation theory, reformulate the governing equations, and modify the numerical models, it requires an extensive investigation and assessment of the time-dependent phenomena in FFT.

This research investigated the nature of creep, thixotropy, and structuration in oil sands tailings, specifically in polymer amended FFT, from an experimental perspective. Experiments were conducted in different batches using different polymer dosages, different

polymer mixing methods, different boundary conditions, and different scales of tests. The experiments included a combination of long-term column dewatering tests with pore water pressure measurements (to characterize both consolidation and non-consolidation volume change behaviour of fFFT under saturated conditions), oscillatory rheometry tests (to investigate the thixotropic strength buildup/recovery in the aged samples), conventional and modified oedometer tests (to track the changes in compressibility and preconsolidation pressure/yield stress of fFFT and estimate the creep compression), fall cone tests (to determine the undrained shear strength and sensitivity), low/high vacuum scanning electron microscopy (for qualitative/quantitative evaluation of the changes in fFFT fabric and to correlate these changes to the changes in compressibility and strength characteristics), mercury intrusion porosimetry (to determine the changes in pore size distribution and to correlate them to the macroscopic behaviour of fFFT), optical microscopy (for qualitative/quantitative evaluation of post-flocculation changes in grains/flocs size), and zeta potential (to track the changes in post-flocculation colloidal stability for the evaluation of flocculation efficiency). Also, the implications of creep and thixotropic and structuration effects for the longer-term dewatering and consolidation performance of fFFT were examined and discussed.

6.2 Materials

6.2.1 Oil sands tailings

Oil sands tailings samples were collected from a tailings pond in Northern Alberta, Canada, and shipped to Carleton University in Ottawa, Canada. Different laboratory tests and analyses were performed to determine the physical, mineralogical, and chemical

characteristics of unamended fluid fine tailings. The initial solids content was 30% ~ 31% and the liquid limit was 60%. The sands to fine ratio (SFR) was 0.25. The clay content obtained from the Methylene Blue Index (MBI) analysis was 28%. According to the X-ray diffraction (XRD) results, the composition of the clay fraction was 68% ~ 72%% Kaolinite and 28% ~ 32% Illite. Total Dissolved Solids (TDS) in the pore water collected from the unamended fluid fine tailings was 1050 mg/L, electrical conductivity was 1590 micro-S/cm, while the dominant cations were sodium at 340 mg/L. Typical material characteristics of the tailings used are listed in Table 6.1.

Table 6.1: Tailings properties

Parameters	Average value
Initial solids content (%)	30 ~ 31
Initial water content (%)	222 ~ 228
Hydrocarbons (%)	1.4
Initial wet density	1.20 ~ 1.21
Initial void ratio	5.1
Specific gravity	2.12
Liquid limit (%)	60.0 ~ 62.0
Plastic limit (%)	27.0 ~ 29.0
Plasticity index (%)	33.0
D ₉₀ , D ₆₀ , D ₅₀ , D ₁₀ (μm)	100, 11, 6, 0.8

During the course of the experiments, multiple samples of unamended FFT and fFFT (of different ages) were sent to an external laboratory for conducting different characterization tests including pore water chemistry, X-ray diffraction (XRD) analysis, Methylene blue index (MBI) test, and Dean-Stark analysis. Also, the particle size distribution and Atterberg's limits were determined at the laboratory for unamended FFT. The results are included in Appendix B.1.

6.2.2 Polymer stock solution

An anionic polyacrylamide flocculant (A3338) was used for the rapid dewatering of FFT samples. A3338 is a branched polymer, with an average molecular weight of 18×10^6 g/mol, which was commercially available from SNF. In a plastic weighing dish, 4g of A3338 polymer was weighed using an analytical balance (Fisher Scientific, Sartorius AG Germany, LE225D) and decanted into a 1000 mL glass beaker and completed to 1000 mL with deionized water to prepare a polymer stock solution with a final concentration of 0.4%. The polymer solutions were stirred using a jar tester (Phipps and Bird, USA) at 200 rpm for 5 minutes and at 125 rpm for the following 55 minutes. Then the polymer solution was mixed with a hand blender for 10 seconds and left for maturation for 1 hour.

6.2.3 Preparation of flocculated FFT

To prepare the flocculated tailings, initially, a four-blade impeller (with a radius of 8.5 cm) attached to an overhead mixer (IKA EUROSTAR 60 control) was immersed in 2000 mL of FFT in a 5-litre pail. The mixing was initiated at a constant speed of 250 rpm and continued for 15 minutes to make the tailings homogenous. Then, the required amount

of 0.4% polymer stock solution was added to the FFT by injecting the solution close to the rotating impeller during mixing (the required amount was estimated based on the solids content of the unamended FFT). All the polymer stock solution was injected as fast as possible and in one shot into the FFT sample. Starting from the moment the polymer stock solution was injected into the FFT, the mixing was continued for 20 seconds. The mixing method was originally adapted from a baseline procedure developed by Shell Canada in 2010 for replicating the in-line flocculation and producing field representative samples while conducting the laboratory-scale experiments. Mizani (2016) optimized the procedure based on the measurements of short-term dewaterability (obtained from the capillary suction time tests and settlement tests) and yield stress. The test results indicated that flocculated tailings products that were produced by mixing at a speed of 250 rpm for 10 seconds would result in optimum dewatering and settling behaviour (Mizani, 2016). Later, the mixing method was optimized again based on the sensitivity of fFFT to the duration of shearing. The result from the short-term settlement tests (48 ~ 72 hours) in graduated columns (K. Vandara, personal communication, February 21, 2018) and the relationship observed between the torque force developed during flocculation in a Couette rheometer and the quality of flocculation (measured in terms of capillary suction time and settlement rate) (Aldaef and Simms, 2019) indicated that mixing at a speed of 250 rpm for 20 seconds would result in an optimal dewatering performance.

6.3 Methods

Column dewatering experiments were conducted using two different boundary conditions: (a) single-drainage; and, (b) double-drainage (Figure 6.3). Typically, a short

column height leads to the development of low effective stress within the deposited tailings material and the creep is thought to be one of the main mechanisms behind the deformations that occur at lower effective stresses (Miller, 2010). Hence, a relatively short height of 10 cm was selected in order to maximize the influence of creep and minimize the role of self-weight consolidation for a single, thin lift of tailings deposition. Nevertheless, such short column height still allowed for the construction of profiles of density/water content with depth upon destructive testing.

Figure 6.3. A schematic diagram of the single-drainage and double-drainage columns.

Both single-drainage and double-drainage column experiments were conducted in different batches using two different dosages of anionic polymer, 600-ppm and 800-ppm respectively. When the single-drainage column experiment was conducted using a polymer dosage of 600-ppm, a single lift of polymer amended FFT was deposited in two transparent plastic acrylic columns with the material reaching a height of 10 cm inside the columns.

Acrylic columns were chosen based on the consideration that monitoring the settlement and dewatering behaviour of flocculated FFT material would be easier if the columns were built of transparent material. A pore water pressure sensor (model T5 from UMS) was installed into each of these columns at an elevation of 1.5 cm from the base of the columns to monitor both positive and negative pore water pressures (Figure 6.4).

Similar to the single-drainage columns dosed with the 600-ppm of anionic polymer, in the case of the 800-ppm single-drainage column experiment, a single lift of polymer amended FFT was deposited in two transparent plastic acrylic columns with the material reaching a height of 10 cm inside the columns. However, in this case (800-ppm), two pore water pressure sensors (model T5 from UMS) were installed into one of these two columns (at the elevations of 1.5 cm and 4.5 cm respectively) to monitor both positive and negative pore water pressures (Figure 6.6). The use of multiple pore water pressure sensors at different elevations allowed for the measurements of pore water pressure and effective stress development at different elevations. The other plastic acrylic column (that had no pore water pressure sensor) was used for measuring the dewatering and settlement rate. Both columns were kept covered at the top with the help of a tight lid. The space between the lid and the surface of the deposited tailings was also kept closed in order to prevent the escape of water vapour. The non-porous and sealed bottom in the single-drainage columns simulated the field drainage conditions where an impermeable layer underneath the fluid fine tailings layer in a tailings pond prevents the downward drainage (Miller, 2010).

Additionally, a transparent plastic acrylic column (17 cm length X 22 cm width X 50 cm height) was used for conducting the single-drainage dewatering test on a large scale

using a polymer dosage of 800-ppm (Figure 6.7). The column was equipped with three T5 pore water pressure sensors at three different elevations (5 cm, 15 cm, and 25 cm respectively) for the measurements of pore water pressure.

Similar to the single-drainage column dewatering experiments, a single lift of polymer amended FFT dosed with the same anionic polymer (A3338) was deposited in the columns for conducting the double-drainage column dewatering experiments. In the 600-ppm double-drainage column experiment, the column used was a 7.5 cm diameter polypropylene column with a pore water pressure sensor installed at an elevation of 1.5 cm from the base of the column (Figure 6.5). However, in the 800-ppm double-drainage column experiment, there were two transparent plastic acrylic columns; one of them was equipped with two pore water pressure sensors at different elevations (1.5 cm and 4.5 cm respectively) from the base of the column (Figure 6.6). The other plastic acrylic column (with no pore water pressure sensor) was used for measuring the dewatering and settling rate. In both 600-ppm and 800-ppm double-drainage columns, the columns were kept covered at the top in order to prevent the escape of water vapour. In both cases, during deposition, the columns were filled with flocculated tailings up to a height of 10 cm.

In the 600-ppm double-drainage column dewatering experiment, the tailings within the column were rested on a filter paper placed at the base of the column, which allowed for downward drainage. The filter paper was kept wet. In the 800-ppm double-drainage column experiment, the columns with the filter papers at their base were placed on a tray filled with water, and then, the flocculated tailings were deposited into the columns. The level of water in the tray was always kept at 1 cm. In the double-drainage columns, both

downward and upward drainage were allowed since they were designed to replicate the field drainage conditions where there is a porous sand layer lying just below the layer of FFT in a tailings pond (Miller, 2010). The experimental data obtained from the 800-ppm double-drainage column dewatering experiment are presented in the ‘Results and discussion’ section and the results from the 600-ppm double-drainage column dewatering experiment are included in Appendix A.2.

For both single-drainage and double-drainage column experiments, a set of replicate columns (7.5 cm diameter polypropylene columns) was used for measuring the changes in total height, water-tailings interface height, gravimetric water content, and void ratio over time (Figure 6.6). Samples were collected from the replicate columns periodically in order to perform other tests and analyses including oscillatory rheometry tests, conventional and modified oedometer tests, fall cone tests, low and high vacuum scanning electron microscopy, optical microscopy, zeta potential measurements, and mercury intrusion porosimetry (Figure 6.8). For both 800-ppm single-drainage and 800-ppm double-drainage replicate columns, water contents were measured at 1 cm intervals across the height of the settled solids. In spite of the difference in sidewall effect, the excess pore water pressure dissipated in both plastic acrylic columns and polypropylene columns at the more or less same time.

An investigation was conducted separately to assess the effects of different polymer dosages and deposition rates on the excess pore water pressure buildup and dissipation in fFFT and the results indicated that both polymer dosage and deposition rate have considerable influence on the excess pore water pressure buildup and dissipation rate. The

interparticle repulsive forces between the clay particles (that arise from the interaction of the electric double layers on the clay surfaces) and the adsorptive forces (i.e. London van der Waals forces) are important components of pore water pressure measurements (Mitchell, 1962). When the polymer is added to the FFT, since the attractive forces begin to dominate over the interparticle repulsive forces and the electric double layer thickness decreases, the pore water pressure measurement reflects the net effect of these changes. That's why the higher polymer dosages (i.e. 800-ppm ~ 1000-ppm), which tend to promote a greater flocculation tendency and produce larger flocs were observed to generate higher pore water pressure values in comparison to the lower polymer doses (i.e. 400-ppm ~ 600-ppm) and unamended FFT (Appendix A.1). Also, when a rapid deposition rate was used instead of a slow deposition rate, the pore water pressure values increased. A slower filling rate generated relatively lower values of load-induced excess pore water pressure and resulted in a relatively greater settlement rate, whereas a higher filling rate resulted in higher values of pore water pressure and lower settlement rate. Since the generation of excessively high excess pore water pressure could impede the rate of settlement, especially, in the tailings with low permeability (Zheng et al., 2018), a higher filling rate was avoided during the deposition of fFFT into the columns. When a slower deposition rate was used, the pore water pressures generated by different polymer dosages were in the same range. Since the deionized water sample was free from the particle surface forces and there were no active physicochemical forces in the deionized water, the peak pore water pressure values were comparatively lower for the deionized water, even when a higher deposition rate was used.

Capillary suction time tests were performed for both 600-ppm and 800-ppm polymer dosages (for both single-drainage and double-drainage) in order to explore the dose-response relationship between the polymer dosage applied and the solids content of the tailings and also as a check on the quality of the polymer-tailings mixture.

Figure 6.4. Single-drainage columns (600-ppm) with pore water pressure sensor at 1.5 cm.

Figure 6.5. Double-drainage column (wet filter paper at the base) (600-ppm).

Figure 6.6. (Top) Single and double-drainage columns (with pore water pressure sensors at 1.5 and 4.5 cm elevations); and, (bottom) replicate columns (polymer dosage: 800-ppm).

Figure 6.7. A large single-drainage column with 3 pore water pressure sensors (800-ppm).

Figure 6.8. Use of replicate columns on a weekly basis for conducting different tests.

Oscillatory rheometry test

Oscillatory rheometry tests including amplitude sweep tests and three interval thixotropy tests (3ITT) were performed on the samples collected from the 600-ppm and 800-ppm single-drainage replicate columns and 800-ppm double-drainage replicate columns at intervals to track the changes in the viscoelastic properties of fFFT, and therefore, to investigate the deformation/destruction behaviour of the samples in response to the small strain shear and also their re-flocculation behaviour under the resting condition. An Anton Paar Physica MCR301 model rheometer with a vane fixture was used for conducting the tests.

During the amplitude sweep test, the sample was placed in a cylindrical sample holder in the rheometer and the vane fixture was then lowered into the sample. Then, the sample was sheared at a constant oscillation frequency with the shear stress increasing at each cycle of oscillation. The resulting deformation in the tailings materials was estimated and expressed in terms of elastic modulus (G') and viscous modulus (G'') (Figure 6.9). G' was estimated from the recoverable strain during each oscillation, whereas G'' was estimated from the measured irrecoverable strain. Additionally, the amplitude sweep test results demonstrated the changes in the linear elastic range in response to the shear applied. The region where the elastic modulus was relatively constant represented the true elastic range of the fFFT material and also indicated the presence of a particulate network in the material (Mewis and Wagner, 2009). The point where the material showed a transgression from the elastic to plastic (or viscous) state and started to deform irreversibly or the point where G' dropped sharply and G'' became dominant represented the yield stress (past which

the viscous behaviour began to dominate the mechanical response of the material). Since the elastic modulus represents a material's resistance to being deformed elastically (or stiffness) and is strongly dependent on the material fabric, the changes observed in elastic modulus also reflected the changes in material structure over time (Mizani et al., 2017).

Figure 6.9. A typical amplitude sweep test result showing the changes in viscoelastic properties in response to the small strain shear.

Three interval thixotropy (3ITT) test, which is a combination of amplitude sweep test and stress growth test (where the material was sheared at a constant rate) was performed to measure the destruction of the structure during shearing and the post-shear structure recovery in resting condition (measured in terms of increase/decrease in elastic modulus before, during, and after shearing) (Figure 6.10). Initial elastic modulus (G') of the flocculated FFT material was determined by applying a stress that was well below the linear

viscoelastic range so that the material would undergo elastic deformation only. Then, the material was sheared at a constant rate for a short duration. After a resting period, the elastic modulus was measured again by applying oscillating stress with the same frequency and amplitude as in the first step. The thixotropic recovery of the sample structure was determined as the ratio of final elastic modulus G' (measured after a resting period) to the elastic modulus G' initially measured (Mizani, 2016).

Figure 6.10. A typical three interval thixotropy test result showing the recovery of structure.

Oedometer test

For both single-drainage and double-drainage and for both polymer dosages, the conventional one-dimensional consolidation test (also known as oedometer test) was

performed on the aged samples collected from the bottommost layer of the replicate columns (in an undisturbed condition). For the 800-ppm single-drainage and 800-ppm double-drainage column dewatering experiments, in addition to the undisturbed samples, remoulded samples were also tested at intervals. A close examination of the compressibility curves obtained from the undisturbed and remoulded samples helped in evaluating the effect of time-dependent behaviour of tailings on its compressibility and strength characteristics (Burland, 1990; Delage, 2010).

For the 800-ppm single-drainage and double-drainage column experiments, a 'modified' oedometer test was performed on the samples collected from the replicate columns. One of the oedometer test frames in the laboratory was modified by installing a pore water pressure sensor (T5 from UMS) at the base of the consolidation cell in a vertical position so that when the sample was mounted on the consolidation cell, the porous ceramic cup at the tip of the pore water pressure sensor penetrated the sample and measured the variations in pore water pressure (Figure 6.11). When vertical static loads were applied to the sample, the corresponding changes in the sample thickness (settlement) as well as the changes in pore water pressure were recorded. Instead of doubling the load after 24 hours, the load was doubled only when the excess pore pressure was completely dissipated. When both pore water pressure and settlement (due to primary consolidation and creep) were measured and plotted against the logarithm of time for each cycle of loading individually, it helped to determine whether there was any settlement after the complete dissipation of excess pore water pressure. Such a settlement would indicate a creep compression in the sample (a decrease in void ratio with time at constant effective stress).

Figure 6.11. Modified oedometer test (settlement with pore water pressure measurement).

Fall cone test

A fall cone device made by M & L Testing Equipment was used for measuring the changes in the undrained shear strength of the fFFT samples based on the measurements of cone penetration. The cone had an apex angle of 30° and the weight of the cone including the vertically sliding shift was 40 g. The device was equipped with a release button for facilitating the smooth release of the cone and a 40 mm diameter dial gauge with 0.01 mm subdivisions for the measurements of penetration made by the cone into the sample upon release. Although the fall cone test (Standard British fall cone method, British Standard BS 1377) is a commonly used alternative to the Casagrande method for measuring the Atterberg limits (British Standards Institution, 1990), the test has also been used for measuring the undrained shear strength of clays in both undisturbed and remoulded

conditions. The test was performed directly on the surface of the flocculated FFT materials deposited in 7.5 cm diameter and 10 cm tall polypropylene (PP) replicate column (Figure 6.12). A replicate column was used exclusively for performing the fall cone test on a weekly basis. Additionally, several replicate columns were used for preparing remoulded samples at intervals and their undrained shear strengths were measured at the same water contents as the undisturbed sample. The sensitivity of the fFFT material was determined by comparing the undrained shear strengths measured in its undisturbed state to the undrained shear strengths measured in its remoulded state.

Figure 6.12. (Left) A typical fall cone used for measuring the undrained shear strength; and, (right) measurement of undrained shear strength on the surface of the fFFT samples.

Fabric estimation techniques: MIP, optical microscopy, and SEM

For both single-drainage and double-drainage columns dosed with 800-ppm of polymer and single-drainage columns dosed with 600-ppm of polymer, the mercury intrusion porosimetry (MIP) technique was applied to the freeze-dried samples for the pore size distribution measurements. For freeze-drying, the samples were flash-frozen using liquid n-pentane, which was already cooled in a liquid nitrogen bath ($-210^{\circ}\text{C} \sim -196^{\circ}\text{C}$) for 30 seconds (so that the n-pentane could reach its melting point of -129.8°C). Then, the frozen samples were placed inside a vacuum chamber in a freezer for sublimation (Figure 6.13). The freeze-drying method helped in keeping the microstructural features of fFFT samples as much undisturbed as possible with a minimal change in volume.

Figure 6.13. The steps for the sample preparation for the mercury intrusion porosimetry (MIP) analysis using the techniques of freezing-drying and sublimation (clockwise).

Optical microscopy was performed on the samples collected from both single-drainage and double-drainage replicate columns dosed with 600-ppm and 800-ppm of polymer; the goal was to track the development in floc formation in the fFFT samples after the mixing of the polymer. In optical microscopy, the fFFT samples were imaged in their original state; however, the technique can only be used for the relatively wet samples (during the first 48 hours of mixing) because beyond 48 hours the light source can not penetrate the densifying tailings. The wet samples were collected from the surface of the columns using a needle and placed carefully on the microscope slides so that the sample disturbance was minimum (Figure 6.14). The samples were imaged using a Nikon Eclipse Ti optical microscope. A magnification scale of 200X and a field of view of 650 μm across the width was used. The imaging software NIS Elements AR3.2 was used for image visualization and acquisition.

Figure 6.14. (Left) The sample collected from the column using a needle; and, (right) the placement of the sample on a microscope slide for optical microscopy.

During the first 48 hours of polymer mixing, the zeta potentials of the tailings bleed water were also measured to determine the colloidal stability of the fFFT samples or the status of the electrochemical attractions between the charged clay particles in fFFT.

For both single-drainage and double-drainage and for both polymer dosages, the scanning electron microscopy (SEM) images were generated using the samples collected from the surface of the solids settled in the replicate columns. A Tescan Vega-II XMU SEM device at the Carleton Nano Imaging Facility (NIF) was used for conducting the low vacuum scanning electron microscopy (Figures 6.15 and 6.16). Using the cryogenic sample preparation technique, a rapid freezing stage of -40°C was applied to the hydrated fFFT samples in the vacuum chamber. Then, the samples were scanned and imaged at a vacuum pressure of 50 Pa. SEM images were generated using a variety of magnification scales (200X, 500X, and 1000X) and a range of field of view ($250 \sim 750 \mu\text{m}$) to visualize and quantify the grains and pores of all ranges. Five different point locations were selected for each sample to ensure higher chances of repeatability. SEM images helped to observe the changes in size, shape, and arrangement of grains/flocs and pores in fFFT over time.

In addition to the low vacuum SEM, a high vacuum SEM was performed on the freeze-dried samples. High vacuum SEM analysis with a gold-palladium coating on the freeze-dried samples produced better quality images in comparison to the low vacuum SEM analysis on the wet samples. Nevertheless, since there was a probability that high water content fFFT samples had a change in volume (i.e. a change in the original microstructure of the samples) when they were freeze-dried and sublimated, a high vacuum SEM was considered less reliable than the low vacuum SEM.

Figure 6.15. Tescan Vega-II XMU SEM device used for scanning electron microscopy.

Figure 6.16. (Left) The sample collected from the column; and, (right) a frozen sample.

6.4 Results and discussions

6.4.1 Dewaterability/settleability (column dewatering test)

Single-drainage column (600-ppm)

Pore water pressure declined rapidly to the hydrostatic pressure by Day-5; however, excess pore water pressure was completely dissipated around Day-49 and the pressure did not change appreciably thereafter (Figure 6.17). By contrast, the tailings in the replicate columns showed substantial volume change throughout the experiment duration, declining in water content from 225% to 78% after 91 days (Figure 6.18). Water content was measured at 3 different depths (top, middle, and bottom) (Figure 6.21). The dewaterability potential was estimated as the net water release (Figure 6.23). The changes in void ratio and solids settlement were estimated from the measurements of weight, total height, water-tailings interface height, water release, and water content (Figures 6.25 and 6.27).

Single-drainage column (800-ppm)

Pore water pressure declined rapidly after the deposition (Day-2). Afterwards, the pressure started to decline, but slowly (Figure 6.19). Pore water pressure declined to the hydrostatic pressure around Day-35 and remained close to the hydrostatic pressure for the rest of the experiment period. By contrast, the tailings in the replicate columns underwent volume change throughout the time with the water content declining from 245% to 133% after 91 days, and to 98% after 168 days (Figure 6.20). Water contents were measured at 1 cm intervals across the height of the solids (Figure 6.22). The net water release, void ratio, and solids settlement were determined as well (Figures 6.24, 6.26, and 6.28).

Figure 6.17. Pore water pressure measured from the 600-ppm single-drainage columns.

Figure 6.18. Water content at different depths (600-ppm single-drainage replicate column).

Figure 6.19. Pore water pressure measured from the 800-ppm single-drainage column.

Figure 6.20. Water content at different depths (800-ppm single-drainage replicate column).

Figure 6.21. Water content at different elevations over time (600-ppm single-drainage).

Figure 6.22. Water content at different elevations over time (800-ppm single-drainage).

Figure 6.23. Net water release measured from 600-ppm single-drainage replicate columns.

Figure 6.24. Net water release measured from 800-ppm single-drainage replicate columns.

Figure 6.25. Solids settlement over time in the 600-ppm single-drainage replicate columns.

Figure 6.26. Solids settlement over time in the 800-ppm single-drainage replicate columns.

Figure 6.27. Change in the void ratio in the 600-ppm single-drainage replicates columns.

Figure 6.28. Change in the void ratio in the 800-ppm single-drainage replicate columns.

Effect of sedimentation

In the literature, the settlement that occurs in the FFT deposits during the first few days or weeks of deposition has been attributed to both consolidation and non-consolidation volume change behaviour of tailings. Like creep and thixotropy, sedimentation has also been purported to explain the non-consolidation volume change in FFT (Chaudhary et al., 2015). A test was conducted to determine the water content at the water-tailings interface in the 800-ppm single-drainage replicate columns at which the influence of sedimentation diminishes and the self-weight consolidation starts to act as the key dewatering and settlement mechanism (Carrier and Freeman, 2018).

Three replicate columns were filled with fFFT to different heights and the changes in water-tailings interface height were measured and recorded for the first 2 days (or until there was no more settlement). Then, the average final solids content in each column was estimated based on the known values of initial solids content and the specific gravity of the material (Carrier and Freeman, 2018). The estimated average solids content value included the effects of both sedimentation and consolidation (and, creep and thixotropy). When these average solids content values for the three replicate columns were plotted against the corresponding tailings-water interface heights (after 2 days) and the data were extrapolated back to a final interface height of zero, the intercept value gave us an estimate of the solids content corresponding to the end of sedimentation (Carrier and Freeman, 2018). From the test conducted, the ‘end of sedimentation’ solids content at the water-tailings interface in the single-drainage replicate columns was estimated as 32.6%. (Figure 6.29). By combining the estimated ‘end of sedimentation’ solids content and the gravimetric water

content profile measured at the top of the solids (at the water-tailings interface) from the 800-ppm single-drainage column dewatering experiment, the process of sedimentation was found to be the dominant dewatering mechanism till Day-8 (Figure 6.30).

Figure 6.29. Replicate column test to determine the ‘end of sedimentation’ solids content.

Figure 6.30. The self-weight consolidation became the dominant dewatering mechanism in the 800-ppm single-drainage replicate columns starting from Day-8 (based on the ‘end of sedimentation’ solids content).

Effective stress development

Assuming that the pore water pressures developed in the polypropylene replicate columns at the elevations of 1.5 and 4.5 cm were same as the pore water pressures measured in the plastic acrylic column (using the pore water pressure sensors) at the same elevations and by estimating the total stresses developed in the replicate columns at the same height as the pore water pressure sensors, the changes in effective stress over time were determined using the following equation:

$$\sigma' = \sigma - u \dots\dots\dots (6.1)$$

Where σ' = effective stress; σ = total stress; and, u = pore water pressure.

The effective stresses were measured and plotted against the void ratios for both 600-ppm and 800-ppm single-drainage replicate columns (Figure 6.31). For the 600-ppm samples, after 56 days, the effective stress developed at an elevation of 1.5 cm was estimated to be 0.05 kPa (at a void ratio of 2.32) and after 84 days, the effective stress developed was 0.0525 kPa (at a void ratio of 1.84). So, there was very little increase in effective stress after Day-56 (i.e. after the end of primary consolidation). For the 800-ppm samples, the effective stress measured after Day-35 was 0.09 kPa at an elevation of 1.5 cm and 0.01 kPa at an elevation of 4.5 cm (at a void ratio of 3.52). After Day-84, the effective stress measured was 0.10 kPa and 0.06 kPa respectively (at a void ratio of 2.99). After 119 days, the effective stresses developed were 0.11 kPa and 0.065 kPa respectively (at a void ratio of 2.65). So, similar to the 600-ppm single-drainage samples, once the excess pore water was dissipated, the changes in effective stresses in 800-ppm were relatively small.

Figure 6.31. Effective stresses at 1.5 cm in the 600- and 800-ppm single-drainage columns.

Observation: Single-drainage columns (600-ppm vs. 800-ppm)

When both 600-ppm and 800-ppm pore water pressure profiles and gravimetric water content profiles were compared, the 800-ppm replicate columns generated a considerably higher magnitude of initial dewatering and consolidation (From Day-1 to Day-14). The rate of dewatering and settlement became relatively slower in the 800-ppm columns afterwards. Although the dissipation of excess pore water pressure took a relatively long period in the 600-ppm columns (Day-49 in comparison to Day-35 for the 800-ppm columns), by the end of primary consolidation, the 600-ppm columns demonstrated more dewaterability potential (water content declined to 118% after Day-49) and greater settlement (31% solids settlement) in comparison to the 800-ppm columns (water content declined to 168% after Day-35; solids settlement was 27% at Day-35). Once the excess pore water pressure was completely dissipated, both 600-ppm and 800-ppm columns showed a creep compression with a slow but uniform decline in water content. However, the creep compression rate declined as the samples aged. In the 800-ppm single-drainage columns, creep compression slowed down considerably around Day-133 (after attaining a solids content of 49%, an average void ratio of 2.38, and a 34% solids settlement). However, in the 600-ppm single-drainage columns, the water content was still declining at a uniform rate when the experiment was terminated (Day-91). Overall, 600-ppm single-drainage replicate columns demonstrated a better dewatering and consolidation performance in comparison to the 800-ppm single-drainage columns in the longer-term.

The difference in the dewaterability potential of the 600-ppm and 800-ppm samples was evident in the comparison of water contents across the height of the solids in the

columns (Figure 6.21 and 6.22). After Day-91, in the 800-ppm columns, there was an 85% drop in gravimetric water content at the top and a 125% drop at the bottom. By contrast, 600-ppm samples displayed a relatively larger drop in water content with 145% at the top and 155% at the bottom during the same period. 800-ppm samples showed a better dewaterability at the beginning in comparison to the 600-ppm samples. However, later, their progress with the dewaterability slowed down, and they showed less dewaterability and settleability relative to the 600-ppm samples. Both 600-ppm and 800-ppm columns showed a change in water content at different elevations throughout the time, even at the surface, even when there was no change in effective stress or pore water pressure, which clearly shows mechanisms other than consolidation such as creep played a role here.

Before commencing both single-drainage and double-drainage column experiments, CST tests were performed to ensure an optimum dewaterability with relation to the application of polymer. The results from the CST test indicated that a dosage of 800-ppm would perform better than the dosage of 600-ppm in the short-term (Figure 6.32). Additionally, settlement tests were conducted using a pair of graduated cylinders. The 48 hours results from the settlement test also indicated that a dosage of 800-ppm would result in a slightly greater settlement than the 600-ppm. Such findings matched what we observed during the initial stages of the 600-ppm and 800-ppm column experiments. However, when the settlements in the same pair of graduated cylinders were measured after a period of 140 days, there was almost no difference between them in terms of settlement (Figure 6.33). In this connection, if we compare the water content profiles and the settlements obtained from the 600-ppm and the 800-ppm single-drainage column experiments, we would see that the

800-ppm columns did not necessarily produce a better performance over a larger time-scale; rather, a dosage of 600-ppm was found to perform relatively better in longer-term.

Figure 6.32. CST test results from the samples dosed with different polymer dosages.

Figure 6.33. Settlement test (600 and 800-ppm): (left) after 48 hours, (right) after 140 days.

Double-drainage column (800-ppm)

Pore water pressures declined rapidly after the deposition; they remained almost at the same values until Day-14, and then, started to decline gradually. At the elevation of 4.5 cm, the pore water pressure was completely dissipated by Day-21, and then, continued to decline further. However, after Day-45, the decline in pore water pressure considerably slowed down and remained nearly steady for the rest of the period (till Day-168). At the elevation of 1.5 cm, the pore water pressure was completely dissipated after Day-53 and remained steady at a level just below the hydrostatic pressure for the rest of the period (Figure 6.34). By contrast, the tailings in the replicate columns displayed a substantial and uniform dewatering and volume change behaviour. Gravimetric water content declined from 245% to 160% after 21 days, to 111% after 56 days, and to 69% after 168 days (Figure 6.35). The void ratios, solids settlements, and the water contents at 1 cm intervals throughout the height of the solids were determined from the replicate columns.

Figure 6.34. Pore water pressure measured from the 800-ppm double-drainage column.

Figure 6.35. Water content at different depths (800-ppm double-drainage replicate column)

Figure 6.36. Changes in water content at different elevations (800-ppm double-drainage).

Figure 6.37. Solids settlement over time in the 800-ppm double-drainage replicate columns

Figure 6.38. Change in the void ratio in the 800-ppm double-drainage replicate columns.

Figure 6.39. Effective stresses at 1.5 and 4.5 cm in the 800-ppm double-drainage columns.

The effective stresses were measured and plotted against the void ratios for the 800-ppm double-drainage columns (Figure 6.39). After 119 days, effective stress increased to 0.18 kPa at 1.5 cm, and to 0.29 kPa at 4.5 cm.

Observation: Single-drainage vs. Double-drainage (800-ppm columns)

Apparently, the double-drainage boundary condition facilitated a significantly higher rate of dewatering as well as higher magnitudes of primary consolidation during the primary consolidation period (water content declined to 111% with 44% solids settlement after Day-56 when the pore water pressure was completely dissipated at 1.5 cm elevation) in comparison to the 800-ppm single-drainage (water content declined to 168% with 27% solids settlement after Day-35 when the pore water pressure was completely dissipated at 1.5 cm elevation). At the elevation of 4.5 cm, the primary consolidation was completed in the double-drainage columns by Day-21 (average water content was 160% and solids settlement was 34%), whereas in the single-drainage columns, the primary consolidation was completed at the same elevation around Day-42 (average water content was 163% and solids settlement was 26%). A relatively higher rate of dewatering and settlement in the double-drainage columns was probably due to the different boundary conditions.

Post-consolidation dewatering and settlement were evident in both 800-ppm single-drainage and 800-ppm double-drainage columns. If we consider the solids settlement that occurred after the end of primary consolidation, double-drainage columns experienced a greater creep compression (solids settlement increased to 55% from 39%) relative to the single-drainage columns (solids settlement increased to 34% from 27%). Apparently, in both single-drainage and double-drainage columns, the creep compression declined

significantly after a certain amount of dewatering and settlement have been accomplished. In the 800-ppm single-drainage columns, the creep compression slowed down considerably around Day-133 (after attaining a solids content of 49%, an average void ratio of 2.38, and a 34% solids settlement). In the 800-ppm double-drainage columns, the creep compression slowed down relatively earlier, around Day-91 (after attaining a solids content of 54%, an average void ratio of 1.84, and a 49% solids settlement). Therefore, the rate of dewatering and settlement in the double-drainage columns during the creep period (after the end of primary consolidation and before the creep compression slowed down significantly) was considerably greater than the rate of dewatering and settlement in the single-drainage columns. At least 10% solids settlement occurred in the double-drainage columns, whereas 7% solids settlement occurred in the single-drainage columns over a relatively longer creep period. Apparently, the double-drainage boundary condition favoured the creep compression as well.

The difference in the dewaterability potential of the 800-ppm single-drainage and 800-ppm double-drainage samples, especially, during the period of primary consolidation, was evident in the comparison of water contents across the height of the solids in the columns (Figure 6.22 and 6.36). 800-ppm single-drainage samples displayed a considerable initial dewatering and settlement performance, however, later, their progress with the dewaterability slowed down. By the time the excess pore water pressure was completely dissipated (Day-35), the average water content declined from 245% to 168% (37% solids content). By contrast, after demonstrating a similar rapid initial dewatering behaviour during the first 7 days, 800-ppm double-drainage columns continued dewatering

at a relatively higher rate (in comparison to the single-drainage columns) until the excess pore water pressure was completely dissipated. By the time the primary consolidation was completed (Day-56), the average water content declined from 245% to 111%. After Day-168, when the experiment was completed, there was a 170% drop in water content at the top and a 180% drop at the bottom in the double-drainage columns. By contrast, single-drainage columns displayed a relatively smaller drop in water content with 85% at the top and 125% at the bottom during the same period. In both cases, when there was no significant change in effective stress or pore water pressure (after Day-56), the changes observed in water content at different elevations over time indicated a creep compression in the flocculated FFT materials.

By the end of the primary consolidation period, the effective stresses developed at both elevations (1.5 cm and 4.5 cm respectively) were considerably higher in the double-drainage columns (0.14 kPa and 0.19 kPa respectively) in comparison to the effective stresses developed in the single-drainage columns (0.09 kPa and 0.01 kPa respectively), which contributed to a higher dewatering and settlement in the double-drainage columns. Also, after Day-119, the effective stresses developed in the double-drainage columns (0.18 kPa and 0.29 kPa at the elevations of 1.5 cm and 4.5 cm respectively) were greater than the effective stresses developed in the single-drainage columns (0.11 kPa and 0.065 kPa respectively). Although the effective stresses remained almost constant during the creep period in the single-drainage columns, the effective stresses increased significantly in the double-drainage columns, especially, at the elevation of 4.5 cm (although there was no change in pore water pressure during this period). Such an increase in effective stresses

might have been due to a downward hydraulic gradient imposed upon the flocculated FFT samples since the hydrostatic pressure of the bottom drainage was not raised to the hydrostatic pressure level of the tailings inside the double-drainage columns.

Large single-drainage column (800-ppm)

The pore water pressure measurements from the large column showed a steady decline in pore water pressure for the first five weeks since deposition. During the same period, there was a considerable settlement as the solids settled down to a height of 21 cm from the initial height of 35 cm. Once the excess pore water pressure was dissipated, the pressure declined very little, but the solids continued to settle indicating a non-consolidation volume change behaviour of fFFT in the large column, which might have been due to the creep compression. The initial void ratio and water content were 5.13 and 245% respectively and the final void ratio and water content were 2.07 and 91% respectively (at Day-133) (Figures 6.40 and 6.41).

Figure 6.40. Pore water pressure at 5, 15, & 25 cm elevations of a large column (800-ppm).

Figure 6.41. Change in large column solids height with time (polymer dosage: 800-ppm).

Summary (column dewatering test)

Before investigating the dewatering and settlement behaviour in the columns, it was anticipated that the gravimetric water content would stop declining with the complete dissipation of excess pore water. However, the water contents were found to decline and the tailings to settle even after the end of primary consolidation, though at a relatively slower rate. In both single-drainage and double-drainage columns, large and small, the initial dewatering and primary consolidation were followed by a substantial secondary compression and a uniform rate of dewatering even when there was no or very little change in pore water pressure (and in effective stress). Such dewatering and volume change behaviour could be attributed to some processes other than the consolidation such as creep. The following figures show the differences in the dewaterability potential and settlement behaviour of the 600-ppm single-drainage and 800-ppm single-drainage columns (and the 800-ppm double-drainage columns) as well as differences in the magnitudes of their initial dewatering, primary consolidation, and creep compression.

Figure 6.42. The changes in average gravimetric water content with time measured from the 600-ppm & 800-ppm single-drainage and 800-ppm double-drainage replicate columns.

Figure 6.43. Net water release measured from 600- and 800-ppm single-drainage columns.

Figure 6.44. The changes in void ratios in the 600-ppm and 800-ppm single-drainage and 800-ppm double-drainage replicate columns.

Figure 6.45. The changes in solids settlement in the 600-ppm and 800-ppm single-drainage and 800-ppm double-drainage replicate columns.

Overall, 600-ppm single-drainage columns showed a higher rate of dewatering and settlement in comparison to the 800-ppm single-drainage columns in the longer-term. Even, their dewatering and settlement performances were comparable to the 800-ppm double-drainage columns. The double-drainage columns showed a relatively rapid and greater settlement at the beginning in comparison to the single-drainage columns. Because of the imposed boundary conditions, the effective stresses developed in the double-drainage columns were larger. Also, the hydrostatic pressure of the bottom drainage was not raised to the hydrostatic pressure level of the flocculated FFT inside the double-drainage columns; hence, the flocculated FFT materials inside the columns might have been subjected to a downward hydraulic gradient, which might have contributed to a rapid and greater settlement (Miller, 2010). In the double-drainage columns, although the excess pore water pressure dissipated near the water-tailings interface (at 4.5 cm) rapidly (by Day-21), the rate of excess pore water pressure dissipation was relatively slower near the bottom (at 1.5 cm) and the excess pore water pressure was completely dissipated by Day-53. By the time the primary consolidation was completed in the double-drainage columns (Day-53), the 600-ppm single-drainage columns had almost the same dewatering and settlement rate as the 800-ppm double-drainage columns.

6.4.2 Thixotropic strength buildup/recovery (oscillatory rheometry)

Amplitude sweep test

Amplitude sweep test, an advanced oscillatory rheometry test was performed on the 600-ppm (single-drainage) and 800-ppm (both single-drainage and double-drainage) samples. Figures 6.46, 6.47, 6.48, 6.49, and 6.50 illustrate the results of the amplitude

sweep tests performed on the 30-minutes, Day-14, Day-35, Day-56, and Day-70 600-ppm single-drainage samples respectively. The oscillation frequency was kept constant at 1.6 Hz; however, the maximum stress was increased successively with each oscillation. X-axis represented the maximum stress (in Pa) during a given oscillation. The resulting deformation in tailings materials was estimated and expressed in terms of elastic modulus (G') (in Pa) and viscous modulus (G'') (in Pa). Y-axis represented the elastic modulus and viscous modulus. Amplitude sweep tests also demonstrated the changes in the linear elastic range over time and correlated these changes to the changes in material structure (since the elastic modulus is strongly dependent on material fabric) (Mizani et al., 2017).

600-ppm single-drainage sample

Figure 6.46. Change in viscoelastic properties (30-min. 600-ppm single-drainage sample).

Figure 6.47. Change in viscoelastic properties (Day-14 600-ppm single-drainage sample).

Figure 6.48. Change in viscoelastic properties (Day-35 600-ppm single-drainage sample).

Figure 6.49. Change in viscoelastic properties (Day-56 600-ppm single-drainage sample).

Figure 6.50. Change in viscoelastic properties (Day-70 600-ppm single-drainage sample).

If we consider the change in elastic shear stiffness ($G_{\max}$) (maximum elastic modulus) over time, the rate of increase in elastic modulus was greater during the creep period (after Day-49, when the excess pore water pressure was completely dissipated) than the rate of increase in elastic modulus during initial dewatering and primary consolidation period (Day-1 ~ Day-49) (Figure 6.51). Similar to the elastic shear stiffness ($G_{\max}$), the rate of increase in yield stress measured during the creep period was greater than the rate of increase observed during the initial dewatering and primary consolidation period. When Anderson and Stoke (1978) investigated the changes in elastic modulus of a kaolinite clay sample under a constant confining pressure during a consolidation test, they noticed a similar disproportionate increase in elastic modulus during creep compression relative to an increase in elastic modulus estimated for the same period assuming that a reduction in void ratio was the sole contributing factor to such an increase. Anderson and Stoke (1978) attributed the additional increase in elastic shear stiffness during creep compression (the part of the measured elastic shear stiffness that could not be explained by a decrease in the void ratio over time) to a stronger physical-chemical bonding between the particles (a thixotropic effect) (Anderson and Stoke, 1978).

When the elastic shear stiffness ($G_{\max}$) values and the gravimetric water contents measured over time were plotted together, the increase in elastic modulus was found correlated to the decline in water content (Figure 6.51). When the water content became relatively stable after undergoing a rapid decline at the initial stage and approached 100% (around Day-56), the elastic modulus started to increase considerably. Although the decrease in the void ratio (or the increase in density) during initial dewatering and primary

consolidation contributed to an increase in elastic shear stiffness of the flocculated FFT samples, the elastic modulus was found to increase at a significantly higher rate when there was a nominal change in water content (or void ratio). Apparently, the flocculated tailings gained a significant amount of stiffness only after a certain amount of water was removed, which allowed the fine particles to agglomerate and develop stronger interparticle bonding, and therefore, to gain strength.

Figure 6.51. Increase in $G_{\max}$ with the water content declining (600-ppm single-drainage).

800-ppm single-drainage sample

Figures 6.52, 6.53, 6.54, and 6.55 illustrate the results of the amplitude sweep tests performed on the Day-1, Day-14, Day-42, and Day-84 800-ppm single-drainage samples. The oscillation frequency was kept constant at 1.6 Hz.

Figure 6.52. Change in viscoelastic properties (Day-1 800-ppm single-drainage sample).

Figure 6.53. Change in viscoelastic properties (Day-14 800-ppm single-drainage sample).

Figure 6.54. Change in viscoelastic properties (Day-42 800-ppm single-drainage sample).

Figure 6.55. Change in viscoelastic properties (Day-84 800-ppm single-drainage sample).

Figure 6.56. Increase in $G_{\max}$ with the water content declining (800-ppm single-drainage).

800-ppm double-drainage sample

Figures 6.57, 6.58, 6.59, 6.60, and 6.61 illustrate the results of the amplitude sweep tests performed on the Day-1, Day-14, Day-28, Day-56, and Day-84 800-ppm double-drainage samples. The oscillation frequency was kept constant at 1.6 Hz.

Figure 6.57. Change in viscoelastic properties (Day-1 800-ppm double-drainage sample).

Figure 6.58. Change in viscoelastic properties (Day-14 800-ppm double-drainage sample).

Figure 6.59. Change in viscoelastic properties (Day-28 800-ppm double-drainage sample).

Figure 6.60. Change in viscoelastic properties (Day-56 800-ppm double-drainage sample).

Figure 6.61. Change in viscoelastic properties (Day-105 800-ppm double-drainage sample)

Figure 6.62. Increase in $G_{\max}$ with the water content declining (800-ppm double-drainage).

Observation: Single-drainage (600-ppm versus 800-ppm)

Similar to the 600-ppm single-drainage samples, the elastic shear stiffness ($G_{\max}$) (or maximum elastic modulus) was found to increase in the 800-ppm single-drainage samples with gravimetric water content declining (Figure 6.62). However, in the 800-ppm samples, the elastic modulus started increasing noticeably earlier than the 600-ppm samples (Figure 6.63). In the 600-ppm samples, the excess pore water pressure dissipated completely around Day-49, and 89% of the total bleed water had been released around the same time (by Day-56). However, in the 800-ppm samples, the excess pore water pressure was dissipated around Day-35 and 90% of the total bleed water had also been released by that time. Hence, it was evident that the increase in elastic modulus had an inverse relationship with the volume of water present in the fFFT sample. The elastic shear stiffness ($G_{\max}$) (or maximum elastic modulus), which reflects the material stiffness started to increase only after most of the water had been released and such an increase in material stiffness was more prominent in the 800-ppm single-drainage samples. Apparently, when dosed with 800-ppm of polymer, the tailings developed a stronger particle-particle interaction and bonding due to the ageing effect leading to a higher $G_{\max}$ for the older samples (Anderson and Stoke, 1978).

Figure 6.63. Increases in $G_{\max}$ over time with the water content declining (600-ppm and 800-ppm single-drainage samples).

Observation: Single-drainage (600-ppm/800-ppm) vs. Double-drainage (800-ppm)

In comparison to both 600-ppm and 800-ppm single-drainage samples, 800-ppm double-drainage samples showed an exceptionally high increase in elastic modulus (and therefore, in material stiffness). Similar to the 800-ppm single-drainage samples, the elastic modulus in the 800-ppm double-drainage samples started to increase earlier than the 600-ppm single-drainage samples; however, at a much higher rate. In the 800-ppm single-drainage samples, the rate of increase in elastic modulus slowed down with the gravimetric water content and void ratio declining; however, in the 800-ppm double-drainage samples, the rate of increase in elastic modulus increased almost linearly with the water content and

void ratio declining (Figure 6.64). Apparently, both stronger particle-particle interactions or bonding and a rapid decline in water content (higher rate of dewatering) contributed to an increase in stiffness in the 800-ppm double-drainage samples.

Figure 6.64. Increases in $G_{\max}$ over time with the water content declining (800-ppm single-drainage and 800-ppm double-drainage).

When Sorensen (2006) conducted one dimensional oedometer compression tests on the reconstituted kaolin clays along with bender element testing (for the measurement of $G'_{\max}$) using a nominal strain rate ($1.2 \times 10^{-6} \text{ s}^{-1}$), an exponential relationship was derived from the test results for describing the relationship between the elastic modulus ($G'_{\max}$) and the void ratio at a constant stress (Sorensen, 2006):

$$\frac{G'}{p_r} = A' \cdot \exp^{-n' \cdot e} \dots\dots\dots (6.2)$$

Where p_r = a reference pressure introduced to make the equation dimensionally consistent, typically taken as 1 kPa; A' and n' = positive dimensionless material parameters; and, e = void ratio.

If we compare the measured elastic modulus ($G_{\max}$) values (experimentally obtained) to the elastic modulus ($G_{\max}$) values estimated based on the changes in void ratio over time (using equation 6.2), we could see significant discrepancies between the experimentally obtained values and the theoretically estimated values, especially, in the 800-ppm samples (Figure 6.65 and 6.66). Both A' and n' were unknown for the flocculated tailings, but since both kaolin and fluid fine tailings consist primarily of the kaolinite, the same A' and n' values used by Sorensen (2006) (2572 and 3.54 respectively) were used for estimating the theoretical values of elastic modulus of fFFT.

Figure 6.65. Experimentally & theoretically obtained G' values (800-ppm single-drainage).

Figure 6.66. Experimentally & theoretically obtained G' values (800-ppm double-drainage)

Summary: Amplitude sweep test

In both single-drainage and double-drainage samples, as well as in both 600-ppm and 800-ppm samples, the elastic modulus ($G_{\max}$), linear elastic region, and the yield stress of the fFFT materials increased with the ageing of the sample (though at different rates). Since elastic modulus (G') is strongly dependent on the material fabric, such changes in elastic modulus or in the linear elastic range indicated a change in the tailings microstructure over time, which contributed to an increase in stiffness as well as yield stress. The rate of increase in elastic modulus was highest in the 800-ppm double-drainage samples (Figure 6.67). Apparently, irrespective of the polymer dosage used, the double-drainage boundary condition, which resulted in the rapid dewatering of the samples, played a more crucial role in such strength development in the flocculated FFT materials.

Figure 6.67. Different rates of increase in maximum elastic modulus ($G_{\max}$) for different samples (600-ppm & 800-ppm single-drainage and 800-ppm double-drainage samples).

If we combine the observations of dewaterability/settleability from the replicate columns with the results from the amplitude sweep tests, we would see that the rate of increase in elastic modulus was relatively slower in the 600-ppm single-drainage samples, and subsequently, the stiffness or the thixotropic strength developed over time was not adequate to impede the self-weight consolidation process in the 600-ppm replicate columns. Hence, the rate and magnitude of both dewatering and settlement were considerably greater in the 600-ppm samples, especially, in the longer-term. By contrast, 800-ppm single-drainage samples showed a relatively slower dewatering rate and less creep compression because of the significant thixotropic strength development.

Three interval thixotropy test

The three interval thixotropy test (3ITT), which is a combination of amplitude sweep test and stress growth test was performed to measure the ‘structure breakdown’ (under shear) and ‘structure recovery’ (at rest) (measured in terms of increase/decrease in the elastic modulus before, during, and after shearing). Initially, the baseline elastic modulus (G') was measured by conducting an amplitude sweep test on a sample at a low oscillation frequency for 5 minutes. Then, the same sample was sheared at a constant strain rate of 0.1s^{-1} for 5 minutes. Subsequently, the elastic modulus (G') of the sample was measured again by conducting another amplitude sweep test for 5 minutes. The elastic modulus (G') values were measured again after a resting period of 30 minutes and 60 minutes. In between the amplitude sweep tests, during the resting periods, the samples were kept completely covered to minimize the effect of evaporation. Also, the water that was accumulated on the surface was collected and measured to evaluate the effect of dewatering on the recovery of structure. Figures 6.68 and 6.69 show the recovery of G' after the shearing of a 1-day old 600-ppm single-drainage sample, whereas Figures 6.71 and 6.72 show the recovery of G' after the shearing of a 70-day old 600-ppm single-drainage sample.

When the fFFT was subjected to a shearing action at a constant rate, initially, the deformation was elastic. But, later, the deformation became viscoplastic with the continuous shearing. Subsequently, the bonding between the particles became weaker and there was a decrease in the strength of the material. However, once the shearing stress applied to the material was removed and the sample was allowed some resting period, the fFFT started to regain its original structure as well as strength (Jeeravipoolvarn, 2005).

Figure 6.68. Recovery of G' after shearing in the Day-1 600-ppm single-drainage sample.

Figure 6.69. Thixotropic recovery of structure in Day-1 600-ppm single-drainage sample.

Figure 6.70. Recovery of G' after shearing in the Day-70 600-ppm single-drainage sample.

Figure 6.71. Thixotropic recovery of structure in Day-70 600-ppm single-drainage sample.

Although 48% of the elastic modulus lost during shearing was recovered 60 minutes after shearing in the case of a 1-day old sample, the percentage of recovery decreased in the 70-day old sample (from 48% to 23%). However, the maximum elastic modulus (initial elastic modulus) was found to increase in the 70-day old sample significantly (from 405 Pa in the 1-day old sample to 801 Pa in the 70-day old sample), which indicated that the sample gained elastic modulus considerably over time; in fact, the elastic modulus almost doubled. Probably, such an increase in stiffness in the older sample (70-day old sample) contributed to the slower recovery of elastic modulus after shearing. Nevertheless, such recovery demonstrated a significant thixotropic strength regain in the disturbed/destructured sample, which also showed the reversible characteristics of structuration/thixotropy. Despite the difference in the percentage of recovery, the amount of elastic modulus recovered after 60 minutes in the 70-day old sample (76 Pa) was comparable to the amount of elastic modulus recovered in the 1-day old sample (84 Pa).

Figures 6.72 and 6.73 show the recovery of G' with time after the shearing of 800-ppm single-drainage samples of different ages and 800-ppm double-drainage samples of different ages respectively. In the 800-ppm single-drainage samples, although the percentage of recovery increased up to Day-42, the percentage of recovery decreased afterwards (however, the initial elastic modulus continued to increase). Similar to the 600-ppm single-drainage samples, such observations from the 800-ppm single-drainage samples indicated that once the single-drainage fFFT materials reached a certain level of stiffness/strength, the rate of thixotropic strength recovery started to decrease.

Figure 6.72. Thixotropic structure recovery in the 800-ppm single-drainage samples.

Figure 6.73. Thixotropic structure recovery in the 800-ppm double-drainage samples.

If compared to the 600-ppm or 800-ppm single-drainage samples, the recovery of the structure was more prominent in the 800-ppm double-drainage samples. Probably, a rapid and significant amount of initial dewatering in the replicate columns and the subsequent changes in void ratio, in addition to the thixotropic effect, contributed to a higher rate of structure recovery in the 800-ppm double-drainage samples. Similar to the percentage of recovery, the amount of elastic modulus recovered (60 minutes after shearing) increased as the samples got older. Unlike 600-ppm or 800-ppm single-drainage samples, the rate of thixotropic strength recovery did not start to decrease in the double-drainage samples after reaching a certain stiffness or strength level. In general, the thixotropic effect, which is typically manifested in a thixotropic recovery of strength in remoulded tailings materials was more prominent in the FFT dosed with the 800-ppm of polymer than in the FFT dosed with the 600-ppm. Apparently, the fFFT materials were highly thixotropic and able to recover their structure/strength considerably after the remoulding; however, the rate of such recovery appeared to slow down with time.

The water that was accumulated on the surface of the samples during shearing and also during the resting periods was collected and measured. The small volume of water indicated that dewatering had a very nominal effect on the recovery of the structure measured from the 3ITT tests. Especially, the older samples (> 28 days old) released a minimum (1 ~ 2 mL) or no water, whereas they showed maximum structure recovery.

Summary: Three interval thixotropy test

The results from the 3ITT tests demonstrated that the flocculated FFT materials are highly thixotropic in nature. Although the structure was destroyed under the influence of

shearing, it was also recovered considerably within a short period, which implied that the structure is recoverable (the recovery was measured in terms of G' values, which are correlated to the material fabric). The percentage of recovery increased with time (up to a certain level of stiffness/strength) and the samples recovered most of the elastic modulus they lost due to the shearing (the percentage of recovery may vary depending on the polymer dosage and the boundary condition used). Although the thixotropic recovery of structure/strength in the 600-ppm single-drainage sample was comparable to the 800-ppm single-drainage samples at the beginning (i.e. 1-day old sample), the older 600-ppm samples (i.e. 70-day old) were able to regain only a small fraction of the elastic modulus (that was lost due to the shearing). By contrast, the older 800-ppm samples recovered most of their structure/strength. Such a time-dependent recovery of G' in fFFT after shearing and structural breakdown was an indication of ageing effect (a thixotropic recovery of structure), which has also been observed to influence the rheological behaviour of flocculated tailings in a pipeline or during deposition (Mizani et al., 2017).

6.4.3 Change in compressibility and yield stress (oedometer test)

Conventional oedometer test

600-ppm single-drainage sample

Oedometer tests were performed on the samples collected from the replicate columns at specific time intervals for both single-drainage and double-drainage and both 600-ppm and 800-ppm column dewatering experiments. Tests were conducted on the 56-day and 120-day old 600-ppm single-drainage samples. Figure 6.74 represents the

compressibility curves obtained from the 600-ppm single-drainage samples. With time, as the samples got older (i.e. went through an ageing effect), there was a change in their compressibility behaviour. The older sample developed a stiffness, became less compressible, and therefore, showed resistance to the vertical compression force acting upon it. Because of the time-dependent structuration effect, an apparent preconsolidation pressure (yield stress) was developed in the 120-day old sample, which contributed to such stiffness.

Figure 6.74. Compressibility curves from 56 & 120-day 600-ppm single-drainage samples.

800-ppm single-drainage sample

Similar to the 600-ppm single-drainage column dewatering experiment, oedometer tests were performed on the samples collected from the 800-ppm single-drainage samples

(Day-56, Day-84, Day-112, and Day-168 samples) (Figure 6.75). The 56-day old 800-ppm single-drainage sample developed more stiffness and showed less compressibility in comparison to the 600-ppm single-drainage sample of the same age (56-days old). As the 800-ppm samples gradually got older (Day-56 → Day-84 → Day-112), they also increased in apparent preconsolidation pressure (yield stress) but decreased in compressibility. However, beyond a certain time range (>Day-112), the changes in the 800-ppm single-drainage compressibility curve (i.e. Day-168 sample) with time were not as much distinguishable as they were in the case of less-aged samples (i.e. Day-56, Day-84, and Day-112 samples). The increase in apparent preconsolidation pressure or the change in compressibility behaviour slowed down significantly. The samples stopped gaining stiffness and there was no more settlement (creep compression) as well. At this point of dewatering (after Day-168), apparently, the structuration/ageing effect in the flocculated tailings would not contribute further to the stiffness of the fFFT materials, even if they were allowed more time for ageing because the fine particles had already formed a closely-packed system with no or minimum voids (that would allow the water to flow) and reached an equilibrium state. The viscous flow of the free water within the clay microstructure also declined and an increased structural viscosity started to resist the relative movement between the fine particles and provided more stability to the structure against further compression. Therefore, it is likely that the amount of further deformation (creep compression) of the clay microstructure units would be very nominal, and thus, the creep compression of the tailings material was limited by the time-dependent structuration effect (Sorensen, 2006; Le, Fatahi, and Khabbaz, 2012; Le et al., 2015).

Figure 6.75. 56, 84, 112, and 168-day old 800-ppm single-drainage compressibility curves.

Oedometer tests were also performed on the remoulded samples prepared from the Day-84 and Day-112 800-ppm single-drainage columns. Figures 6.76 and 6.77 show the compressibility curves for the Day-84 800-ppm single-drainage undisturbed and remoulded samples and the Day-112 800-ppm single-drainage undisturbed and remoulded samples respectively. Figure 6.78 shows the compressibility curves from two single-drainage remoulded samples of different ages (Day-84 and Day-112 remoulded samples).

Figure 6.76. Undisturbed/remoulded Day-84 800-ppm single-drainage compression curves

Figure 6.77. Undisturbed/remoulded Day-112 800-ppm single-drainage compression curve

Figure 6.78. The compressibility curves obtained from the remoulded samples of different ages (84-day and 112-day old 800-ppm single-drainage remoulded samples).

800-ppm single-drainage (50 cm tall large column)

Figure 6.79 presents the compressibility curves from the 800-ppm single-drainage samples, which were obtained from the different layers (5 cm, 15 cm, and 30 cm) of the 50 cm tall column at the end of the experiment (after 180 days of self-weight consolidation). Apparently, all the three samples developed an apparent preconsolidation pressure, especially, the 15 cm and 30 cm samples, which resulted in reduced compressibility. An increase in yield strength in the fFFT material, especially at the bottom layers resulted in increased stiffness, which counteracted the compression force from the upper layers and slowed down the secondary compression (creep) of the fFFT material.

Figure 6.79. The compressibility curves obtained from the 800-ppm samples collected from different elevations of a large single-drainage column (50 cm tall column).

Figure 6.80. Comparison of compressibility curves from 10 cm and 50 cm column samples.

When compared to the 112-day old 800-ppm single-drainage undisturbed samples' compressibility curve (collected from the 10 cm tall replicate column), 180-day old 5 cm and 15 cm samples (collected from the 50 cm tall column) showed similar compressibility behaviour, which implied that different scales of column dewatering test (larger/smaller column) resulted in buildup of stiffness in the fFFT material at different rates (probably due to the different rates of structuration effect in different-sized columns) (Figure 6.80).

800-ppm double-drainage sample

Figure 6.81 presents the compressibility curves obtained from the 28, 42, 70, and 224-day old 800-ppm double-drainage samples. Day-42 sample developed an apparent preconsolidation pressure (yield strength) and the compressibility was also decreased relative to the Day-28 sample. The yield strength further increased in the Day-70 sample and the sample showed resistance to the compression force. However, afterwards, as the samples aged, there were minimal changes in compressibility and yield strength. Similar to the single-drainage samples, after a certain period ($> \text{Day-126}$), the double-drainage samples stopped gaining additional stiffness and the rate of settlement (creep compression) became very nominal. Oedometer tests were performed on the remoulded samples prepared from the Day-70 double-drainage column (Figure 6.82).

Figure 6.81. 28, 42, 70, and 224-day old 800-ppm double-drainage compressibility curves.

Figure 6.82. Undisturbed/remoulded Day-70 800-ppm double-drainage compression curve

Observation: Changes in compressibility because of the structuration/ageing effect

As we have seen it in natural clays in the past (Locat and Lefebvre, 1985; Burland, 1990; Delage, 2010), the development of a strong interparticle bonding between the fine clay particles and their subsequent rearrangement (due to the structuration/ageing effect) are correlated to an increased apparent preconsolidation pressure (yield strength) and the subsequent changes in compressibility behaviour in the undisturbed FFT samples. The application of scanning electron microscopy on the fFFT samples demonstrated that there was a decrease in total pore volume and a rearrangement of the fine particles with time (Bajwa, 2015). The influence of such time-dependent changes in the microstructure of fFFT samples on their compressibility and strength characteristics became evident when the compressibility curves from the undisturbed samples were compared to the remoulded samples' compressibility curves (Figures 6.76 and 6.77).

Because of the structuration/ageing effect in the undisturbed samples, the undisturbed samples showed relatively greater resistance to the vertical compression loading acting upon them (Burland, 1990; Delage, 2010), especially, during the first few 24-hour loading cycles, when the samples were subjected to the nominal effective stresses. By contrast, remoulded samples showed no or minimum resistance and high compressibility in response to the same vertical compression loading since the samples had already gone through a destructuring process during remoulding, which weakened the interparticle bonds in the remoulded samples and caused changes in their fabric. However, when subjected to a significantly higher loading, the undisturbed samples showed a compressibility behaviour similar to the remoulded samples.

When the compressibility curves from two remoulded samples of different ages were compared (Day-84 and Day-112 samples), they were observed to display the same compressibility behaviour (Figure 6.78). Due to the remoulding process, the structures within both samples were destroyed and restored to their similar pre-structuration structure; that's why they showed similar compression behaviour under vertical loading. Such similarities between the remoulded samples' compressibility behaviour was also a clear indication of the time-dependent structuration effect in the fFFT samples.

Observation: Changes in compressibility depending on the polymer dosage

Apparently, a relatively prominent structuration/ageing effect occurred in the 800-ppm samples, which led to a less porous and more compact microstructure with time, and subsequently, to an increase in the apparent preconsolidation pressure (yield strength) that resulted in more resistance to the vertical compression loading. However, the rate of such an increase in yield strength or the change in compressibility gradually slowed down with time. That's why the compressibility curves obtained from the relatively older (i.e. aged) samples showed very little change (i.e. 112-day and 168-day old 800-ppm single-drainage samples). In general, an 800-ppm dosage of polymer tends to promote a significant structuration/ageing effect in flocculated tailings in the longer-term, which could also potentially impede or slow down the progress of self-weight consolidation. By contrast, because of a relatively moderate structuration/ageing effect, a 600-ppm dosage could lead to a relatively greater creep compression and more dewatering in the longer-term. The findings from the column dewatering tests and the oscillatory rheometry tests were also found to be in good agreement with such observations.

Observation: Changes in compressibility depending on the boundary condition

When an 800-ppm polymer dosage was applied to the FFT samples, in comparison to the single-drainage boundary condition, the double-drainage boundary condition resulted in a relatively significant change in compressibility. Such a difference in the compressibility behaviour resulted in different magnitudes of creep compression and dewatering for the single-drainage and double-drainage boundary conditions.

Modified oedometer test (creep compression under static vertical loading)

Modified oedometer tests were performed on the 14-day and 168-day old 800-ppm single-drainage samples and 210-day old 800-ppm double-drainage samples. Figures 6.83 and 6.84 show the settlement and pore water pressure data for two different loading steps (from 3.3 to 6.6 kPa and from 53 to 106 kPa respectively) for the 168-day old 800-ppm single-drainage sample. In Figure 6.83, the load applied to the sample (6.6 kPa) was well below the apparent preconsolidation pressure. The corresponding secondary compression is very nominal, almost undetectable. Such observation is not atypical of the results obtained from the compression tests performed on clays where the observed rate of creep is low or almost undetectable for an overconsolidated soil (Qi et al., 2018).

Figure 6.83. Settlement & excess pore water pressure in a loading step from 3.3 to 6.6 kPa.

Figure 6.84. Settlement & excess pore water pressure in a loading step from 53 to 106 kPa.

In Figure 6.84, where the loading approached the apparent preconsolidation pressure or even exceeded, the magnitude of secondary compression is measurable (Qi et al., 2018). The inset figure shows that settlement continued even after the complete dissipation of excess pore water pressure. Based on the settlement and pore water pressure data presented in Figure 6.84, Qi et al. (2018) estimated that for a load of 106 kPa, the secondary compression would be equivalent to an additional change in the void ratio of 0.095 over 10 years and 0.105 in 100 years.

Similar to the 168-day old 800-ppm single-drainage sample, in a 210-day old 800-ppm double-drainage sample, the creep or the secondary compression was found negligible at low effective stresses. However, the creep became noticeable when the sample was subjected to a load close to its preconsolidation pressure (Figures 6.85 and 6.86).

Figure 6.85. Settlement & excess pore water pressure in a loading step from 1.8 to 3.6 kPa.

Figure 6.86. Settlement, excess pore water pressure in a loading step from 28.6 to 57.2 kPa.

6.4.4 Undrained shear strength and sensitivity (fall cone test)

A fall cone was used for measuring the changes in the undrained shear strength of the fFFT samples based on the measurements of cone penetration. Figure 6.87 shows the undrained shear strengths measured from the 800-ppm single-drainage sample. Although the strength developed was nominal initially, the material started gaining strength slowly by Day-28. Following rapid initial dewatering (water content declined to 171% at Day-28 from 245% at Day-1), the excess pore water pressure dissipated at or around Day-35 and 90% of the total bleed water was released by that time. By Day-49, the rate of decline in water content slowed down significantly. Around the same time, the fFFT started gaining strength at a significantly higher rate. Therefore, apparently, the increase in material strength had an inverse relationship with the dewatering in the fFFT sample (Figure 6.88).

Figure 6.87. Undrained shear strength development in 800-ppm single-drainage samples.

Figure 6.88. The inverse relationship between the water content and undrained shear strength in 800-ppm single-drainage samples.

The undrained shear strengths were also determined for the remoulded fFFT samples at the same water contents as the undisturbed samples. The sensitivity of the fFFT was estimated from the ratio of the undrained shear strengths of the undisturbed samples to the undrained shear strengths of the remoulded samples (Figure 6.89). Since high sensitivity refers to a stronger structuration effect or more thixotropic hardening, a significant increase in sensitivity values after Day-49 indicated a larger thixotropic hardening with the water content decreasing.

Usually, the thixotropic strength development in a sample is attributed to a combined effect of structuration/ageing effect and a change in the void ratio/density (due to the dewatering). Hence, when the undrained shear strengths of the undisturbed samples were compared to the undrained shear strengths of the corresponding remoulded samples (prepared at the same water contents as the undisturbed samples), the differences observed in undrained shear strength between the undisturbed and remoulded samples reflected the effect of the structuration/ageing solely (Figure 6.90).

Figure 6.89. Sensitivity measured from the undisturbed and remoulded shear strengths.

Figure 6.90. Structuration effect in the material contributing to the strength development.

6.4.5 Fabric estimation techniques

Mercury intrusion porosimetry (MIP)

The mercury intrusion porosimetry (MIP) technique was applied to the freeze-dried single-drainage samples (600-ppm and 800-ppm) and double-drainage samples (800-ppm) for the measurement of pore size distribution. The results showed that the technique could provide a characterization of the pore structure of polymer amended oil sands tailings including the pore size distribution curve (cumulative intrusion pore volume versus pore diameter plot), threshold pore diameter, and peak pore mode volume/diameter (log differential pore volume vs. pore diameter plot). The shape of the pore size distribution curve characterized the microstructure of the tailings samples; hence, the changes in a curve shape (i.e. the decrease in cumulative intrusion pore volume over time) provided an indication of the changes in tailings fabric (Cook and Hover, 1993). The threshold pore diameter, which is the pore diameter corresponding to the steepest slope of the pore size distribution curve also indicates the smallest pore diameter that is continuous within a sample and corresponds to the pressure at which mercury rapidly intrudes the most of a sample or the highest rate of mercury intrusion inside the pores (Cook and Hover, 1993; Panesar and Francis, 2014; Roshani, Fall, and Kennedy, 2018). Hence, a decrease in the threshold pore diameter is correlated to an increase in fine particles in the tailings samples and a decrease in hydraulic conductivity (Fall, Benzaazoua, and Ouellet, 2005; Roshani, Fall, and Kennedy, 2018). The threshold pore diameters determined from the individual pore size distribution curves served as an index to compare the pore structures of the fFFT samples of increasing ages (Panesar and Francis, 2014).

In this study, the pore size distribution data (cumulative intrusion pore volume versus pore diameter plots) showed that the cumulative intrusion pore volume reduced gradually with the Day-1 samples producing the highest cumulative intrusion. In the case of 800-ppm single-drainage samples, after the initial dewatering period (Day 1 ~ 14), the change in cumulative intrusion pore volume was nominal and the pore size distribution curves gradually converged. Similar was the observation with the 600-ppm single-drainage samples. However, relative to the 800-ppm samples, the 600-ppm samples had smaller cumulative intrusion pore volumes, which explained the relatively higher rate of initial dewatering observed in the 800-ppm single-drainage samples. The pore size distribution curves obtained from the double-drainage samples didn't converge over time, rather the cumulative intrusion pore volume decreased over time, which was in line with the relatively higher rate and a prolonged period of dewatering observed in the double-drainage replicate columns. Both single-drainage and double-drainage samples showed a decreasing proportion of the frequency of larger pores with time.

In the case of 800-ppm single-drainage samples, the threshold pore diameter was found to decrease with time (decreased to 0.70 μm from 0.90 μm over a period of 98 days). In comparison, 600-ppm single-drainage samples had a nominal change in the threshold pore diameter value (decreased to 0.65 μm from 0.70 μm over a period of 70 days). Although 800-ppm single-drainage sample had a greater threshold pore diameter at the beginning (i.e. 0.90 μm at Day-1), which apparently resulted in a higher rate of initial dewatering, both 600-ppm and 800-ppm single-drainage samples had comparable threshold diameters once the primary consolidation was completed (i.e. 0.65 μm and 0.72

μm respectively at Day-70). The threshold diameter decreased more rapidly in the 800-ppm double-drainage samples (decreased to $0.40\ \mu\text{m}$ from $0.80\ \mu\text{m}$ over a period of 56 days).

The log differential pore volume distribution data (log differential pore volume vs. pore diameter plots) generated from the fFFT samples of increasing ages showed a shift in the peak pore mode towards smaller pore diameter with time, and consequently, the pore diameters at which the pore size distributions of the samples were centred decreased. Initially (i.e. at Day-1), the pore size distributions of 600-ppm and 800-ppm single-drainage samples were centred at the pore diameters of $0.50\ \mu\text{m}$ and $0.65\ \mu\text{m}$ respectively, whereas, at Day-70, the pore size distributions were centred at the pore diameters of $0.40\ \mu\text{m}$ and $0.50\ \mu\text{m}$ respectively. The shift in the peak pore mode towards smaller pore diameter with time was more prominent in the 800-ppm double-drainage samples, which was found to be in good agreement with a relatively rapid increase in the solids content and the subsequent consolidation/densification and strength development in the double-drainage samples. At Day-1 and Day-56, the pore size distributions of the 800-ppm double-drainage samples were centred at $0.50\ \mu\text{m}$ and at $0.30\ \mu\text{m}$ respectively.

Overall, the results from the mercury intrusion porosimetry analyses showed that the pore size distributions of the fFFT samples changed with time and such changes were found to be in good agreement with the changes observed in the macroscopic behaviour of the samples in the replicate columns (i.e. dewatering, consolidation, strength development, etc.) and the changes observed in other microstructural investigations (i.e. scanning electron microscopy and optical microscopy). In general, the decrease in pore volume was

more prominent in the double-drainage samples relative to the single-drainage samples, and in the 600-ppm single-drainage samples when compared to the 800-ppm single-drainage samples. The pore size distribution data and the log differential pore volume distribution data for both single-drainage and double-drainage samples as well as individual cumulative intrusion pore volume vs. pore diameter plots and log differential pore volume vs. pore diameter plots are included in Appendix C.

Scanning electron microscopy and optical microscopy

For both single-drainage and double-drainage boundary conditions and both 600-ppm and 800-ppm polymer dosages, the scanning electron microscopy images were generated using the samples collected from the surface of the fFFT deposited and settled in the replicate columns. A Tescan Vega-II XMU SEM device at the Carleton Nano Imaging Facility (NIF) was used for conducting the low vacuum scanning electron microscopy. A Nikon Eclipse Ti optical microscope, in combination with the imaging software NIS Elements AR3.2 was used for generating the optical microscopy images from the samples during and immediately after the flocculation. Qualitative observations/visualizations of the SEM and optical microscopy images were used in combination with quantitative fabric analysis techniques (Khattak, 2018) to examine the changes in tailings microstructure over time. Using a combination of advanced digital image processing techniques, Khattak (2018) studied the changes in flocs sizes and distributions within 0 ~ 48 hours of polymer mixing and also the changes in pore and grain sizes and distributions in fFFT in longer-term, up to Day-70.

Low vacuum scanning electron microscopy

Figures 6.91, 6.95, 6.100, and 6.105 show the original SEM micrographs generated from the 600-ppm single-drainage, 800-ppm single-drainage, 600-ppm double-drainage, and 800-ppm double-drainage samples respectively. Qualitatively, the SEM micrographs reflected the changes in size, shape, and arrangement of the grains/flocs and pores in fFFT over time. For conducting the quantitative fabric analysis of fFFT, Khattak (2018) used the same sets of SEM images. The brighter areas in the images reflected the grains/flocs on the surface of the sample, whereas the darker areas reflected the pores/voids, both interparticle and intra-particles. An image processing software named Fiji ImageJ was used for the fabric investigation and quantitative analysis of the images and detailed information was extracted on the total number of pores and grains, total image porosity, pore diameter, and the changes in the cumulative average grains/flocs size over time (Khattak, 2018). When the images obtained from different batches of samples collected under the same experimental conditions were analyzed, the image processing techniques were able to generate repeatable trends in porosity and grain and pore size distribution (Khattak, 2018).

SEM: 600-ppm single-drainage sample

In the 600-ppm single-drainage column dewatering experiment, since the samples were collected from the surface and there was a relatively lower amount of dewatering at the surface from Day-7 to Day-28 (gravimetric water content declined from 213% at Day-7 to 186% at Day-28), Day-7, Day-14, and Day-28 SEM micrographs showed relatively less variance. It was difficult to make out any differences in the pattern of porosity, which seemed to remain relatively small pores dispersed throughout the image. Nevertheless, the images showed a decrease in the area of dark tint zones with time. After Day-28, the replicate columns underwent a substantial amount of dewatering at the surface (from where the samples were collected) and the water content declined to 96% (at Day-70) from 186% (at Day-28). Hence, the Day-72 SEM micrograph demonstrated a tailings fabric significantly different than the previous micrographs. The flocs/aggregates appeared to grow or coalesce with more interlocking between them. The pores, which were initially small but frequent became larger and less frequent and decreased in total volume.

Figure 6.91. SEM images of 7, 14, 28, and 72-day old 600-ppm single-drainage samples showing the changes in the size, shape, and arrangement of the grains and pores (clockwise).

When Khattak (2018) analyzed 3 different sets of the original 600-ppm single-drainage SEM images, a steady decline was observed in the total number of pores with time (Figure 6.92). In line with the porosity measured from the 600-ppm single-drainage replicate columns, the image porosity also displayed a declining trend (Figure 6.93). The

decline in pore number was attributed to the growth of flocs during flocculation. A detailed quantitative analysis of pore size distribution from the binary images (generated from the original SEM images during the process of digital image processing) indicated that the changes in pore number observed with time were most prevalent in the lowest range of sizes (<10 microns) (Figure 6.94). The total number of grains also decreased; however, the average size of grains and pores increased.

Figure 6.92. The number of pores measured from the 600-ppm single-drainage sample images (adapted from Khattak, 2018).

Figure 6.93. Image porosity in the 600-ppm single-drainage (adapted from Khattak, 2018).

Figure 6.94. The number of pores in the 0 ~ 40 μm size range in the 600-ppm single-drainage sample images (adapted from Khattak, 2018).

SEM: 800-ppm single-drainage sample

Figure 6.95. SEM images of Day-1, Day-7, Day-47, and Day-70 800-ppm single-drainage samples showing the changes in the size, shape, and arrangement of the grains and pores (clockwise).

In the 800-ppm single-drainage SEM micrographs, from Day-1 to Day-70, there were noticeable changes in tailings fabric, which were also in line with the rate of

dewatering observed in the 800-ppm single-drainage replicate columns. With time, as the water content declined, there were fewer pores, and overall, the appearance became more homogeneous. The flocs also appeared to grow.

The quantitative fabric analysis of the SEM images generated from the 800-ppm single-drainage samples produced a substantially higher pore count as compared to the SEM images generated from the 600-ppm single-drainage samples (Figure 6.96) (Khattak, 2018). Apparently, the 800-ppm single-drainage samples displayed a better dewaterability potential because of their different image porosities and pore counts (Figure 6.97 and Figure 6.98 respectively). However, the 600-ppm samples attained similar porosities by the end of the experiment (Figure 6.99) (Khattak, 2018).

Figure 6.96. The number of pores in the 800-ppm single-drainage sample images (adapted from Khattak, 2018).

Figure 6.97. Image porosity in the 800-ppm single-drainage (adapted from Khattak, 2018).

Figure 6.98. Pores in the 600- and 800-ppm single-drainage (adapted from Khattak, 2018).

Figure 6.99. Porosity in 600- & 800-ppm single-drainage (adapted from Khattak, 2018).

SEM: 600-ppm double-drainage sample

When the SEM images obtained from the 600-ppm double-drainage samples (Figure 6.100) were analyzed (Khattak, 2018), although the overall trend in pore count was the same for both single-drainage and double-drainage columns, the numbers decreased at a relatively rapid rate in the double-drainage columns (Figure 6.101). There was a sharp decline in image porosity, especially, in comparison to the single-drainage columns (Figure 6.102). Such a trend in image porosity agreed with the trend observed in the porosity measured from the double-drainage replicate columns. A detailed quantitative analysis of the pore size distribution showed a decreasing trend in pore count for the pores in the lowest size range ($0 \sim 10 \mu\text{m}$), which was similar to the trend observed in the 600-ppm single-drainage samples; but, the rate of decline was relatively rapid in the double-drainage

samples (Khattak, 2018). Figures 6.103 and 6.104 show the overlay binary images that were generated from the 600-ppm double-drainage SEM images (Day-21 and Day-35 respectively) using the software ImageJ for investigating the trends in pore and particle size distributions with time. The images showed a considerable increase in floc size and interlocking between the flocs/aggregates from Day-21 to Day-35 (Khattak, 2018).

Figure 6.100. SEM images of Day-1, Day-7, Day-21, and Day-35 600-ppm double-drainage samples showing the changes in the size, shape, and arrangement of the grains and pores (clockwise).

Figure 6.101. No. of pores in the 600-ppm double-drainage (adapted from Khattak, 2018).

Figure 6.102. Image porosity in 600-ppm double-drainage (adapted from Khattak, 2018).

Figure 6.103. Growth in floc size from a binary image (at Day-21) (from Khattak, 2018).

Figure 6.104. Growth in floc size from a binary image (at Day-35) (from Khattak, 2018).

SEM: 800-ppm double-drainage sample

Figure 6.105. SEM images of 1, 7, 47, and 60-day old 800-ppm double-drainage samples showing the changes in size, shape, and arrangement of the grains and pores (clockwise).

The 800-ppm double-drainage SEM images displayed the same trend as the 800-ppm single-drainage SEM images, however, the changes in tailings fabric with time were more pronounced (Figure 6.105). Similar to the trend observed in the SEM images, the results from the bench-scale experiments also showed that the double-drainage samples in

the replicate columns (both 600-ppm and 800-ppm) had better dewaterability in comparison to the respective single-drainage samples. Apparently, the boundary condition influenced the dewatering behaviour of the fFFT samples, and such influence was found repeatable from sample to sample (Khattak, 2018).

High vacuum scanning electron microscopy

Figure 6.106. SEM images (high vacuum SEM) from the 7, 28, 56, and 98-day freeze-dried 800-ppm single-drainage samples (200X magnified) (clockwise).

Figure 6.106 shows the SEM images generated from the freeze-dried 800-ppm single-drainage samples. The trend was similar to the SEM images generated from the wet samples; the freeze-dried samples also became less porous and more homogenous.

Optical microscopy

600-ppm single-drainage sample

Optical microscopy images were generated at intervals during the first 48 hours of polymer mixing (Figure 6.107). Beyond 48 hours, it was not possible to produce optical microscopy images because of the excess density or thickness of the fFFT samples placed on the microscope slides. Using the software ImageJ, Khattak (2018) processed the selected optical microscopy images digitally and performed the quantitative fabric analysis of the optical microscope images for tracking the changes in the total number of flocs and floc size distribution with time. Although the total floc area increased in general, there was a decrease in the total floc area between 8 to 24-hour (Figure 6.108). Such decline was attributed to the settlement and segregation of the larger flocs. In addition to an increasing trend in average floc size, an increase in the maximum floc size was also observed.

Although the flocs in the size range of $0 \sim 10 \mu\text{m}$ increased in number initially, up to 24 hours, afterwards, they decreased. Based on the analyses of the images, the initial increase in the count was thought to be due to the attachment of very small non-detectable particles to the flocs in the $0 \sim 10 \mu\text{m}$ range as they agglomerated. However, the subsequent decrease (24 ~ 48-hour) was attributed to the flocculation process, which resulted in aggregation of smaller flocs and subsequently, the formation of larger flocs (Khattak 2018).

Figure 6.107. Optical microscopy images from the 600-ppm single-drainage samples.

Figure 6.108. The changes in total flocs area over the first 48 hours (after polymer mixing) in the 600-ppm single-drainage samples (adapted from Khattak, 2018).

600-ppm double-drainage sample

When Khattak (2019) performed the quantitative fabric analysis of the selected optical microscope images generated from the 600-ppm double-drainage samples (Figure 6.109) and investigated the changes in the total number of flocs and floc size distribution, the double-drainage samples had higher numbers of flocs in different size ranges in comparison to the single-drainage samples. After 48 hours, the maximum sizes of the flocs were 230 ~ 250 microns and 310 ~ 320 microns for the single-drainage and double-drainage samples respectively (Figure 6.110) (M. Khattak, personal communication, June 5, 2019). Apparently, the double-drainage boundary condition resulted in a reduced distance between the dispersed particles in FFT and subsequently accelerated the rate of

aggregation and flocculation, which in turn resulted in a better dewaterability, which was also evident in the rapid initial dewatering in the replicate columns.

Figure 6.109. (Left) The images from the 600-ppm double-drainage sample; and, (right) the processed images (from the same images) for quantitative analysis (Khattak, 2019).

Figure 6.110. The changes in flocs number and size distribution in the 600-ppm single-drainage and 600-ppm double-drainage samples (from Khattak, 2019).

800-ppm single-drainage sample

When Khattak (2018) performed a similar quantitative analysis of the 800-ppm single-drainage optical microscopy images (Figure 6.111), there was an increase in the maximum floc size over time and such an increase was found to be more repeatable, especially, at the lower size ranges. At 1 hour, the maximum floc sizes were 70 ~ 100 microns and 100 ~ 120 microns for the 600-ppm and 800-ppm single-drainage samples respectively. After 24 hours, the maximum floc sizes increased to 150 microns and 230 microns respectively, and by the end of 48 hours, the maximum floc sizes increased to 330 microns and 370 microns respectively (Figure 6.112) (Khattak, 2018).

Figure 6.111. Optical microscopy images from the 800-ppm single-drainage samples.

Figure 6.112. The changes in flocs number and size distribution in the images obtained from the 600-ppm and 800-ppm single-drainage samples (from Khattak, 2018).

Figure 6.113. Flocs area for the 600- and 800-ppm samples (adapted from Khattak, 2018).

The change in the total area of the flocs followed a similar trend as the maximum floc size and was found to reflect the cumulative change in solids mass with time. The change in total solids mass was attributed to the flocculation process and the subsequent decrease in water content. In the first few hours of polymer mixing (1 ~ 8 hours), 800-ppm single-drainage samples showed a rapid increase in the solids relative to the 600-ppm single-drainage samples. However, after 48 hours, the total area of the flocs became approximately the same for the 600-ppm and 800-ppm samples (Figure 6.113) and the change in total solids mass also became the same. Such observation was in total agreement with the results from the graduated cylinder settlement tests. During the settlement tests, after 24 hours, the observed settlement of the flocculated tailings was greater in the 800-ppm samples, however, after 7 days, there was little difference in the height of the tailings-water interface (Khattak, 2018).

800-ppm double-drainage sample

When the 800-ppm double-drainage sample images were compared to the images obtained from the 800-ppm single-drainage samples, after 48 hours, the maximum floc sizes were estimated to be 300 ~ 310 microns and 320 ~ 330 microns for the single-drainage samples and double-drainage samples respectively (Figure 6.114) (M. Khattak, personal communication, June 5, 2019).

Figure 6.114. (Left) The images from the 800-ppm double-drainage sample; (right) the processed images (from the same images) for quantitative analysis (from Khattak, 2019).

Figure 6.115. The changes in flocs number and size distribution in the images obtained from the 800-ppm single-drainage and double-drainage samples (from Khattak, 2019).

Summary: Optical microscopy and scanning electron microscopy

The optical microscopy images showed an increase in floc sizes during the first 48 hours of polymer mixing. The SEM images also showed changes in tailings fabric over the long-term (Khattak, 2018). Since the analysis of the 800-ppm optical microscopy images produced relatively larger maximum floc sizes relative to the 600-ppm samples, based on the quantitative fabric analysis of optical microscopy images (<24 hours), the 800-ppm polymer dosage appeared to be the optimal choice. However, the quantitative fabric analysis of both optical microscopy and SEM images from the 600-ppm samples suggested that a considerable change occurred in tailings fabric both in the short-term and long-term,

which eventually resulted in the formation of same-sized flocs in the 600-ppm and 800-ppm samples (Khattak, 2018).

The SEM images from the double-drainage samples showed a significant increase in floc size and a sharp decline in image porosity and the number of pores relative to the single-drainage samples (both 600-ppm and 800-ppm). Similar to the trend observed in SEM images, the results from the bench-scale experiments also showed that the double-drainage samples in the replicate columns (both 600-ppm and 800-ppm) underwent rapid sedimentation and showed a better dewatering performance in comparison to the respective single-drainage samples. The double-drainage boundary condition was thought to result in lower water content at a relatively rapid rate, which in turn reduced the distance between the dispersed particles, and therefore, accelerated the rate of flocculation, and subsequently, sedimentation and consolidation (Khattak, 2018). Hence, Khattak (2018) postulated that it was the boundary condition, not the polymer dosage, which played a more significant role in increasing the size of the microstructural flocs (Khattak, 2018).

6.4.6 Zeta potential measurement

In order to understand the physicochemical interactions that occur in the FFT suspensions immediately after the addition of polymer and during the first 48 hours of mixing (when the particles show a tendency to flocculate), the zeta potential of the tailings bleed water was determined using the equipment Zeta Meter System 4.0. Zeta Meter System 4.0 measures zeta potential using a technique called microelectrophoresis. The equipment comprises a viewing chamber called electrophoresis cell (it has two electrodes at its two ends), a powerful microscope for the direct observation of the individual colloids

in the electrophoresis cell, and the main Zeta Meter unit that is connected to the electrophoresis cell (Figures 6.116 and 6.117). The Zeta Meter unit applies an electrical field across the test solution (bleed water) in the electrophoresis cell by energizing the electrodes at an appropriate voltage (which was determined based on the specific conductance of the sample). The unit estimates the electrophoretic mobility or the zeta potential directly from the electrophoretic velocity recorded during the colloid movement. The transparent electrophoresis cell is mounted on the microscope, which is equipped with a light source for projecting lights on the test solution inside the electrophoresis cell at a high intensity. Under the influence of the applied electric field, the charged colloid particles start moving toward the oppositely charged electrode through the solution at an electrophoretic velocity; since the clay particles are negatively charged, they migrate toward the positive electrode or anode. The distance travelled by any individual colloid inside the electrophoresis cell is tracked across a grid in the display of the microscope. As the particle completes its movement from one end to the other end of that grid, the main unit estimates the electrophoretic mobility from its electrophoretic velocity (the velocity of the particle per unit of the electric field is the electrophoretic mobility) and converts this electrophoretic mobility into the zeta potential. Since the colloids move toward the positive electrode, the electrophoretic mobility is counted negative, and so the zeta potential.

Figure 6.116. The Zeta Meter 4.0 system used for the measurement of zeta potential.

Figure 6.117. The microelectrophoresis technique for zeta potential measurement.

For this study, the test solution for the Zeta Meter testing was prepared using a dilution factor of 2000 (by adding 1 mL of bleed water to the 2000 mL of deionized water). Deionized water was used during the process of dilution in order to avoid disturbing the existing equilibrium state at the particle surfaces as much as possible. Before running the

test, the test solution was stirred and mixed well in order to make it homogenous. For each test solution, 20 randomly selected colloids were tracked using the microscope. The zeta potential value of each of these colloids was recorded and the median value estimated from these values was taken as the representative zeta potential value for the whole test solution. After adding polymer to the FFT, zeta potentials were measured at specific time intervals.

Pate and Safier (2016) created a chart showing the different states of colloidal stability corresponding to the different ranges of zeta potential (Table 6.2). Zeta potential is typically measured in millivolts (mV). The colloids with high zeta potential values (absolute values, negative or positive) refer to an electrically stabilized colloidal suspensions and colloids with low zeta potential values (absolute values, negative or positive) refer to a colloidal suspension with a tendency to flocculate.

Table 6.2. Colloidal stability for ranges of zeta potential (Pate and Safier, 2016).

Zeta potential (mV)	Colloidal stability
0 to ± 5	Rapid agglomeration
± 10 to ± 30	Incipient stability
± 30 to ± 40	Moderate stability
± 40 to ± 60	Good stability
$> \pm 60$	Excellent stability

Generally, a zeta potential value of greater than 30 or below -30 indicates a moderate to good stability for a colloidal suspension, where the particles remain in a dispersed state due to the mutual electrostatic repulsion (Pate and Safier, 2016). However, when the zeta potentials are measured in the range of ± 10 to ± 30 mV, they represent the beginning of aggregation (due to a reduction in net negative charges or electrostatic repulsion) (Pate and Safier, 2016). Net negative charge plays a central role in controlling the clay dispersion behaviour (Chorom and Rengasamy, 1995). If the zeta potential values approach ± 5 mV or close to zero, they refer to a state of rapid agglomeration, where flocculation occurs at a faster pace and leads to rapid sedimentation. When the zeta potential values equal zero, they refer to a completely destabilized colloidal dispersion, where the particles are flocculated (the negative surface charge is completely neutralized by the positive counter ions) (Pate and Safier, 2016).

Figure 6.118. Zeta potential values measured from the 600-ppm single-drainage samples.

Figure 6.119. Zeta potential values measured from the 800-ppm single-drainage samples.

The results from the 600-ppm single-drainage test solution showed that the colloidal dispersion of clay particles in the tailings bleed water was destabilized immediately after the addition of polymer and the dispersion showed a tendency towards flocculation (Figure 6.118). The initial downward trend in zeta potential value was due to the disruption of the equilibrium state and the subsequent particle aggregation. However, after 6 hours, the dispersion started to regain stability and reached the state of a ‘good colloidal stability’ within 24 hours. By contrast, when zeta potentials were measured for the 800-ppm single-drainage test solution, the solution showed a mild flocculation tendency during the first 12 hours; then, the solution started to regain stability, but slowly (Figure 6.119).

If we draw a comparison between these results, it will show that the 600-ppm single-drainage test solution regained colloidal stability faster than the 800-ppm single-drainage test solution and showed a ‘good stability’ within the first 24 hours. The 800-ppm single-drainage test solution was relatively slow with the recovery of the colloidal stability, and even after 48 hours, the measured zeta potential values indicated that the solution was still in the state of ‘incipient stability’. Such observation implied that although a 600-ppm dosage of polymer resulted in a greater flocculation tendency for the colloidal solution immediately after the mixing of polymer (reached the peak within the first 6 hours), the solution also reached the state of good colloidal stability within 24 hours (therefore, there was a shorter time span for the flocculation). By contrast, an 800-ppm dosage of polymer displayed a relatively lower flocculation tendency; however, it also showed a slow recovery of colloidal stability, thereby indicating a relatively longer flocculation period in the beginning. Such observation correlated well to the dewatering and settlement properties measured during the single-drainage and double-drainage column dewatering tests. In the 600-ppm single-drainage replicate columns, after 24 hours, the gravimetric water content declined to 215% from 225% (initial water content at 0-hour), whereas in the 800-ppm single-drainage replicate columns, the water content declined to 238% from 245%. However, by the end of the Day-2, the water content declined to 213% from 215% (at Day-1) in the 600-ppm single-drainage replicate columns, whereas in the 800-ppm single-drainage replicate columns, the water content declined to 224% from 238% (at Day-1). Therefore, although the 600-ppm samples showed a greater flocculation tendency in the beginning, by the end of the flocculation period, the 800-ppm samples showed a better

dewaterability, which was probably due to the tendency of the samples to flocculate over a longer period.

In another experiment, another set of zeta potential measurement was conducted at the Oil Sands and Coal Interfacial Engineering Facility at the University of Alberta (N. Yang, personal communication, March 9, 2018). During this experiment, fFFT (dosed with 800-ppm of A3338 polymer) was used as a test solution instead of bleed water. The instrument used was Model DT1200 (Figure 6.120), which works based on the principle of electroacoustic technique and can determine both particle size distribution and zeta potential in an aqueous system without any need for dilution. An electroacoustic probe tip was immersed in the test solution to measure the vibration current of the particles, and then, the zeta potential was calculated based on the measured current and the particle size distribution (Figure 6.121).

Figure 6.120. The measuring unit of a DT 1200 equipment (Dispersion Technology, 2018).

Figure 6.121. Zeta potential measurement using the electroacoustics method (DT 1200).

Figure 6.122. Zeta potential measured from the 800-ppm single-drainage (DT 1200).

The initial zeta potential values obtained from the DT 1200 (during the first few hours of polymer mixing) were relatively lower in comparison to the values obtained from the Zeta Meter 4.0 system during the same period, which indicated that the test solution (fFFT) in DT 1200 had a greater tendency to flocculate at the initial stage and quickly reached the state of ‘rapid agglomeration’ (Figure 6.122). The zeta potential values gradually decreased (went upward in the zeta potential versus time plot) and approached the state of ‘incipient stability’. The difference in the values obtained from the Zeta Meter 4.0 and DT 1200 can be attributed to the different measurement techniques as well as different sample preparation techniques. However, both equipment and techniques displayed a similar pattern with the zeta potential values. Overall, the zeta potential measurement, in combination with quantitative fabric analysis of optical microscopy images helped to track the formation of flocs in the fFFT samples along with the changes in colloidal stability (which occurred simultaneously as a direct result of the destabilization of the colloidal suspension because of the polymer activity).

6.4.7 Summary: Creep, thixotropy, and structuration effect in fFFT

When a combination of lab-scale experiments was conducted on the fFFT samples to investigate the long-term dewatering and consolidation behaviour including time-dependent creep and structuration effect, a change in volume was observed due to the creep and there was also a change in compression behaviour because of the structuration effect. In order to investigate how the changes in compressibility with time may potentially influence the dewatering and consolidation performance of the tailings deposits and affect the accuracy of final settlement prediction, a simple numerical analysis was performed based on the method suggested by Carrier and Freeman (2018). Carrier and Freeman (2018) proposed a spreadsheet-based method for estimating the final height of a deposit of oil sand fine tailings without using a large-strain, non-linear consolidation computer program. The analysis does not consider the sedimentation process and assumes that the deposited tailings are always submerged, and the boundary condition is single-drainage. When the maximum depth of the deposition area, the initial solids content, and the specific gravity of the tailings are known, the method divides the total depth of the deposit into small layers and estimates the final effective stress at the top of each layers and calculates the final void ratio using the compressibility relationship ($e = a\sigma_v'^b$). Finally, the final height of each layer is determined based on the estimated initial and final void ratios and initial height. Then, the final settlement of the deposit is calculated by adding the final heights of all the layers (Carrier and Freeman, 2018).

Recently, Qi and Simms (2019) proposed an analytical function for estimating the final height of a soft soil/tailings deposit:

$$\text{Final height} = z + \frac{az[\gamma_s - \gamma_w]^b}{b+1} \dots\dots\dots (6.3)$$

Where $z = \frac{\text{Initial height } (h_0)}{e_0+1}$; γ_s = unit weight of solids; and, γ_w = unit weight of water. The parameters a and b can be estimated by fitting the compressibility curve with a power function ($e = a\sigma_v'^b$, where e and σ_v' are void ratio and effective stress respectively).

Now, if we assume a 50-metre tailings deposit and determine the parameters a and b by fitting the compressibility curves obtained from the oedometer tests with a power function, we can calculate the final settlement by using the method suggested by Carrier and Freeman (2018) and also by using the analytical function suggested by Qi and Simms (2019) for the final height estimation.

Since the influence of time-dependent changes in tailings microstructure on its compressibility and strength characteristics became most evident when we compared the compressibility curves obtained from the undisturbed samples to the compressibility curve obtained from the remoulded samples, the final density profile as well as the final height of a 50-metre tailings deposit were also estimated based on the compressibility curves derived from the undisturbed and remoulded samples of same ages (i.e. 84-day old undisturbed and remoulded 800-ppm single-drainage samples, 112-day old undisturbed and remoulded 800-ppm single-drainage samples) (Figures 6.123 and 6.124).

The increase in apparent preconsolidation pressure (yield strength) and the subsequent change in compressibility in the undisturbed samples relative to the remoulded samples (due to the structuration/ageing effect) resulted in different final height estimations

for the same tailings deposits. The differences between the estimated final heights were in the range of 2.25 ~ 3.25 metres. Both the spreadsheet-based method suggested by Carrier and Freeman (2018) and the analytical functions proposed by Qi and Simms (2019) produced almost identical results for the final height estimations. Depending on the sample age (84-day old or 112-day old), the degrees of structuration in the undisturbed 800-ppm single-drainage samples were different, and subsequently, the corresponding final heights (24.3 m and 23.1 m respectively) and the density profiles were also different. However, irrespective of the sample age (84-day old or 112-day old), the final heights and the density profiles derived from the remoulded samples' compressibility curves were fairly close (21.0 m and 20.8 m for the 84-day old and 112-day old remoulded samples respectively) since there were no structuration effects in them (due to the 'structure breakdown' during the remoulding procedure). Figures 6.123 and 6.124 clearly show the effect of changes in compressibility (due to the structuration effect) on the dewatering and consolidation performance of a tailings deposit in the longer-term.

Overall, the fFFT samples developed a thixotropic strength/stiffness with time, and subsequently, there was an increase in apparent preconsolidation pressure (yield strength) and a decrease in compressibility, which eventually impeded the self-weight consolidation in fFFT in longer-term by limiting the creep compression. Hence, the time-dependent creep compression and structuration effect in fFFT were found to affect the dewatering and consolidation performance of the tailings deposits.

Figure 6.123. The final settlement estimated from the Day-84 800-ppm compressibility.

Figure 6.124. The final settlement estimated from the Day-112 800-ppm compressibility.

Chapter 7: Investigation of creep, thixotropy, and structuration effect in Leda clay

7.1 Introduction

This chapter presents a detailed description of the investigation of creep, thixotropy, and structuration in Leda clay. In order to compare the structuration effect observed in natural clays and polymer amended oil sands tailings, the same experiments were conducted on both Leda clay and fFFT samples. Leda clay was selected as a comparative material because the Leda clay deposits are known to display a structuration/ageing effect in their natural state. The data presented include the results from the column dewatering tests conducted on the clay samples, and also the results from the oscillatory rheometry tests, conventional one-dimensional consolidation tests, and fall cone tests, which were performed to examine the strength/stiffness development in the clay samples over time. The experimental data obtained were explained in light of the concept of time-dependent phenomena of creep, thixotropy, and structuration. Comparisons were drawn between the structuration effect observed in the clay-rich fFFT samples and the Leda clay samples.

7.2 Materials

The Leda clay used in this study was collected from the Navan landfill in Ottawa. Typical properties are summarized in the following table (Aldaeef and Rayhani, 2017).

Table 7.1: Leda clay properties

Parameters	Average value
Initial solids content (%)	56.7
Initial water content (%)	76.3
Dry density (g/cm ³)	1.41
Clay content (%)	71.0
Liquid limit (%)	50.8
Plastic limit (%)	27.5
Plasticity index (%)	23.3
D ₉₀ , D ₆₀ , D ₅₀ , D ₁₀ (μm)	10, 1, <1, <1
Specific gravity	2.7
Mineralogy	Illite (83%), Kaolinite (11%)

7.3 Methods

The original gravimetric water content of the clay was 76.3%, but it was increased to 100 ~ 110% (approximately twice the liquid limit of the original Leda clay) by adding the required amount of deionized water. Before adding the water, the clay sample was crushed into smaller pieces manually. The water was added in a way so that the clay sample became fully saturated and completely homogenous. Then, a single lift of clay sample was deposited in several identical polypropylene columns with the material reaching a height of 10 cm inside the columns.

The columns were kept covered at the top with the help of a tight lid. The space between the lid and the surface of the clay sample was also kept closed in order to prevent the escape of water vapour. The non-porous and sealed bottom in the single-drainage columns simulated the field drainage conditions where an impermeable layer underneath the clay deposits prevents the downward drainage.

The column experiment was conducted in two different batches. In both cases, one of the polypropylene columns was used on a weekly basis for measuring the changes in gravimetric water content, total height, water-solids interface height, and void ratio with time. Water contents were measured either at three different depths (top, middle, and bottom of the solids) or at 1 cm intervals across the height of the solids. Samples collected from the columns were also used for performing several tests and analyses including oscillatory rheometry test, conventional one-dimensional oedometer test, fall cone test, and low vacuum scanning electron microscopy.

Figure 7.1. Single-drainage column dewatering test (10 cm tall column) on the Leda clay.

7.4 Results and discussions

7.4.1 Dewaterability/settleability (column dewatering tests)

During the 1st batch of the experiment, over a period of 8 weeks, the gravimetric water content in the Leda clay samples declined to 83% from 110%. The water content gradually declined till Day-28, and the change in water content with time was minimal afterwards. The volume of the total water released (bleed water) was nominal throughout the period and the volume of the bleed water accumulated on the surface of the solids in any single sample never exceeded 6 mL. After Day-56, the solids settlement (%) in the clay sample was only 1.50%. Hence, the samples showed an insignificant volume change behaviour, which was also evident in the slow decline of the void ratio over time.

Figure 7.2. Change in gravimetric water content in Leda clay samples over time (1st batch).

Figure 7.3. Change in the void ratio in the Leda clay samples over time (1st batch).

Figure 7.4. Solids settlement in the Leda clay samples over time (1st batch).

For the replication purpose, the same 10 cm tall column dewatering test was performed again on another batch of Leda clay samples collected from the same Navan landfill in Ottawa. The initial water content was measured 76%, but it was increased to approximately 100% (twice the liquid limit of the original Leda clay) by adding the required amount of deionized water. Although there was a steady decline in the water content up to Day-49 (declined to 84.25% from the initial water content of 98.50%), there was little accumulation of water on the sample surfaces. Also, the degree of saturation estimated from the samples had always been 100% or very close to 100%. Hence, an investigation was conducted to determine whether the Leda clay has a strong affinity for water which might have allowed the clay particles to hold on to a significant amount of water and prevented the water from accumulating on the surface. In addition to 105⁰C, a higher temperature of 200⁰C was used while conducting the oven-dry test on the remaining samples. At Day-63, the water contents measured from the dry-oven test were found 88.95% (using a temperature of 200⁰C) and 83.97% (using a temperature of 105⁰C) respectively. Water content analyses performed at Day-77 and Day-91 also produced similar results with higher water contents obtained at higher temperatures. Hence, performing the dry-oven test at 200⁰C resulted in more water removal from the clay particles than at a temperature of 105⁰C, and on average, there was a difference of approximately 5% between the water contents measured at two different temperatures. Such discrepancies could be attributed to the presence of bound water in Leda clay at the particle level, which remained firmly held to the particles through molecular and electric cohesion, and heating at a temperature of 105⁰C for 24 hours was not adequate for their complete removal. Such characteristic of Leda clay has been attributed to an open fabric

structure, which allows the clay to hold high water content without flowing (Taha, 2010). Hence, there was a minimum accumulation of water on the sample surfaces. Figure 7.5 shows the water content profiles (Day-1 ~ Day-91, @105°C and Day-63 ~ Day-91, @200°C). Figure 7.6 shows the degree of saturation estimated from the Leda clay samples over time.

Figure 7.5. Water content measured in the clay samples (using 105°C/200°C) (2nd batch).

Figure 7.6. Degree of saturation measured in the clay samples (105°C/200°C) (2nd batch).

Figure 7.7. Change in water contents across the height of the solids in Leda clay (2nd batch).

Figure 7.8. Solids settlement in the Leda clay samples over time (2nd batch).

Figure 7.9. Change in the void ratio in the Leda clay samples over time (2nd batch).

7.4.2 Time-dependent thixotropic strength buildup/recovery in Leda clay

Amplitude sweep test

During the 1st batch of the experiment, oscillatory rheometry tests including amplitude sweep tests and three interval thixotropy (3ITT) tests were performed on the samples collected from the Leda clay columns to track the changes in the viscoelastic properties and investigate the deformation/destructuration behaviour of the samples in response to the small strain shear and also the restructuration behaviour under the resting condition. An Anton Paar Physica MCR301 model rheometer with a vane fixture was used for conducting the tests. Figures 7.10, 7.11, 7.12, and 7.13 illustrate the results of the amplitude sweep tests performed on Day-1, Day-3, Day-7, and Day-14 Leda clay samples respectively. The oscillation frequency was kept constant at 1.6 Hz (same as the frequency used for the fFFT samples); however, the maximum stress was increased successively with each oscillation. X-axis represented the maximum stress (in Pa) during a given oscillation. The resulting deformation in clays was estimated and expressed in terms of elastic modulus (G') and viscous modulus (G''). Y-axis represented the elastic modulus and viscous modulus (in Pa). Amplitude sweep tests also demonstrated the changes in the linear elastic range over time and correlated these changes to the changes in material structure (since the elastic modulus or G' is strongly dependent on material fabric) (Mizani et al., 2017).

Figure 7.10. Change in viscoelastic properties (Day-1 Leda clay sample).

Figure 7.11. Change in viscoelastic properties (Day-3 Leda clay sample).

Figure 7.12. Change in viscoelastic properties (Day-7 Leda clay sample).

Figure 7.13. Change in viscoelastic properties (Day-14 Leda clay sample).

Figure 7.14. Increase in $G_{\max}$ with a decrease in water content in the Leda clay samples.

When the elastic shear stiffness ($G_{\max}$) values and the gravimetric water contents were plotted together, the increase in maximum elastic modulus was found correlated to the decline in water content over time (Figure 7.14). Although the decrease in the void ratio (or the increase in density) during initial dewatering and primary consolidation period contributed to an increase in the elastic shear stiffness of clays, the elastic modulus was found to increase more when there was a substantial decline in the water content (i.e. at Day-14). Apparently, the dewatering allowed the fine particles to coalesce and develop stronger interparticle bonding, and subsequently, to gain strength. Hence, the elastic modulus, linear elastic range, and yield stress increased rapidly.

Generally, such a disproportionate increase in maximum elastic modulus or elastic shear stiffness ($G_{\max}$) can not be explained solely by a decrease in void ratio due to the

dewatering. Anderson and Stoke (1978) attributed such an additional increase in elastic shear stiffness to a stronger physical-chemical bonding between the particles (which is a thixotropic effect) (Anderson and Stoke, 1978).

Three interval thixotropy test

A combination of amplitude sweep test (where the maximum stress was increased at each cycle while maintaining a constant oscillation frequency) and stress growth test (where the materials were sheared at a constant shear rate), known as the three interval thixotropy test (3ITT) was performed to measure the ‘structure breakdown’ (under shear) and ‘structure recovery’ (at rest) (measured in terms of increase/decrease in the elastic modulus before, during, and after shearing). Figures 7.15, 7.16, 7.17, and 7.18 show the recovery of G' after the shearing of 1-day old, 3-day old, 7-day old, and 14-day old Leda clay samples respectively. In all four cases, the percentage of recovery increased with time and the samples recovered the elastic modulus that was lost due to shearing.

Figure 7.15. Thixotropic recovery of structure in the 1-day old Leda clay sample.

Figure 7.16. Thixotropic recovery of structure in the 3-day old Leda clay sample.

Figure 7.17. Thixotropic recovery of structure in the 7-day old Leda clay sample.

Figure 7.18. Thixotropic recovery of structure in the 14-day old Leda clay sample.

Figure 7.19. Comparison of thixotropic structure recovery in clay samples of different ages.

During the 3ITT test, the water that was accumulated on the surface of the samples during the shearing as well as resting periods were collected and measured. The nominal volume of water indicated that the dewatering had a minimal effect on the recovery of the structure measured from the 3ITT tests. The 1-day old sample recovered 115% of the elastic modulus that was lost due to shearing (after 60 minutes of shearing), whereas the 14-day old sample recovered 214% of the elastic modulus that was lost due to shearing (Figure 7.19). Such observation implied that the clay samples demonstrated a thixotropic effect, which was clearly manifested in a thixotropic recovery of structure/strength in clays (since they regained their elastic shear modulus with time). Although the structure was broken down under the influence of shearing, it was also recovered considerably within a short period, which shows that the thixotropic effect is reversible.

7.4.3 Change in compressibility and yield stress (oedometer test)

The conventional one-dimensional consolidation or oedometer tests were performed on the clay samples collected from the columns at specific intervals. In addition to the undisturbed samples, oedometer tests were also performed on the remoulded samples prepared from the Leda clay columns of the same ages at the same water contents. Figure 7.20 shows the compressibility curves for both undisturbed and remoulded 56-day old Leda clay samples (from the 1st batch of the experiment). Because of a structuration/ageing effect, which resulted in the development of apparent preconsolidation pressure (or yield strength), the undisturbed sample showed relatively greater resistance to the vertical compression forces, especially, during the first couple of 24-hour loading cycles (when the sample was subjected to the nominal effective stresses). Since the remoulded material had already gone through a destructuring process during remoulding, the interparticle bonds became weaker, and there were significant changes in its fabric. Subsequently, the remoulded sample showed minimal resistance and high compressibility. Nevertheless, when subjected to a significantly higher loading (which exceeded the yield strength), the undisturbed sample started showing a compressibility behaviour similar to the remoulded sample.

Figure 7.20. Day-56 undisturbed & remoulded Leda clay samples' compressibility curves.

The same phenomena were observed when the oedometer tests were performed on the samples obtained from 14, 35, 63, and 91-day old Leda clay columns in the 2nd batch of the experiment. The compressibility curves obtained from the undisturbed samples showed a change in compression behaviour over time. The samples developed stiffness; hence, there was an increase in yield strength and a decrease in compressibility (Figure 7.21). When the compressibility curves obtained from the undisturbed samples and the corresponding remoulded samples (prepared from the same Leda clay at the same water contents) were compared (i.e. Day-35 undisturbed sample and remoulded sample, Day-63 undisturbed sample and remoulded sample, etc.), the remoulded samples were found to show less stiffness (Figures 7.22 and 7.23). Apparently, the process of remoulding destroyed the microstructure of the 'structured' soil formation in Leda clays (which was developed within the clays over time while the sample was in an undisturbed state).

Figure 7.21. Change in compression behaviour in the clay samples over time (2nd batch).

Figure 7.22. Difference in the compressibility of Day-35 undisturbed/remoulded samples.

Figure 7.23. Difference in the compressibility of Day-63 undisturbed/remoulded samples.

Figures 7.22 and 7.23 demonstrate the effects of structuration in undisturbed samples and also how the effects of structuration in undisturbed samples (increase in stiffness or apparent preconsolidation pressure) are reduced by the process of remoulding.

When the compressibility curves from two remoulded Leda clay samples (i.e. Day-35 remoulded sample and Day-63 remoulded sample) were compared, both samples showed the almost same compressibility behaviour once they were remoulded (Figure 7.24). But when their corresponding undisturbed compressibility curves were plotted together and compared, both showed a sign of resistance to the external compression force, with the Day-63 sample showing a relatively greater stiffness (Figure 7.25). Hence, Figure 7.25 shows how the time-dependent structuration effect contributed to different degrees of stiffness in the undisturbed Leda clay samples of different ages.

Figure 7.24. Compressibility behaviour of the Day-35 and Day-63 remoulded clay samples.

Figure 7.25. Compressibility behaviour of the Day-35 & Day-63 undisturbed clay samples.

7.4.4 Change in undrained shear strength (fall cone test)

During the 2nd batch of the experiment, a fall cone device was used for measuring the undrained shear strength of the Leda clay samples based on the measurements of cone penetration. Although the fall cone test is a commonly used alternative to the Casagrande method for measuring the Atterberg limits, the test has also been used for measuring the undrained shear strength of clays in both undisturbed and remoulded conditions. The test was performed directly on the surface of the clay samples deposited in 7.5 cm diameter and 10 cm tall polypropylene replicate columns. 3 replicate columns were used exclusively for performing the fall cone test on a weekly basis.

Figure 7.26. Undrained shear strengths measured from the Leda clay samples over time.

Figure 7.26 shows the undrained shear strengths measured from the clay samples over time. Although the strength developed was nominal initially, the material started

gaining strength after Day-35. When the gravimetric water contents at the sample surfaces (where the fall cone tests were performed) and the undrained shear strengths were plotted together against the time, the increase in material strength appeared to have an inverse relationship with the dewatering (Figure 7.27).

Figure 7.27. The inverse relationship between undrained shear strength and water content.

7.4.5 Change in fabric (visual observations from the SEM images)

The scanning electron microscopy (SEM) images were generated from the samples collected from the sample surface in the polypropylene columns. A Tescan Vega-II XMU SEM device at the Carleton Nano Imaging Facility (NIF) was used for conducting the low vacuum scanning electron microscopy. Using the cryogenic sample preparation technique, a rapid freezing stage of -40°C was applied to the wet clay samples in the vacuum chamber,

and then, the samples were scanned and imaged at a vacuum pressure of 50 Pa. Figures 7.28 show the SEM micrographs produced from the Leda clay samples during the 1st batch of the experiment and Figure 7.29 shows the SEM micrographs produced during the 2nd batch of the experiment.

Figure 7.28. SEM images (1000X) from the 1, 7, 14, and 42-day old Leda clay samples.

During the 1st batch of the experiment, initially, the samples had a grainy texture with lots of different-sized fine particles and only a few pores visible on the surface (Day-1 image in Figure 7.28). After a week, the sample surface became less grainy and the difference between the individual particle started to disappear rapidly (Day-7 image in

Figure 7.28). Over time, the surface appeared to become more flat, smooth, homogenous and compact (Day-14 and Day-42 images in Figure 7.28). The SEM images obtained during the 2nd phase of the experiment showed the same trend (Figure 7.29).

Figure 7.29. SEM images (2000X) from the 3, 7, 35, and 91-day old Leda clay samples.

The changes at the microstructural level in Leda clays, which are typically manifested in the changes in pore and grain numbers and size distributions, have already

been found closely associated with the macroscopic behaviour changes such as a change in compressibility over time (Delage, 2010).

7.4.6 Summary: Creep, thixotropy, and structuration effect in Leda clay

During both batches of the experiment, following initial dewatering, the rate of dewatering and settlement in the Leda clay samples declined significantly, and afterwards, there was very nominal or almost no settlement in the samples for the rest of the period. With time, clays gained strength/stiffness and developed an apparent preconsolidation pressure due to a structuration/ageing effect. Due to enhanced yield strength, clays showed more resistance to the compression force during the consolidation test, and also impeded the self-weight consolidation in the samples in the polypropylene columns.

The preconsolidation pressures measured from the compressibility curves and the undrained shear strengths measured from the fall cone tests indicated that Leda clay developed strength/stiffness at a much greater rate than the fFFT. Subsequently, the clay samples displayed less dewaterability and settleability relative to the fFFT samples. Although in different degrees, both Leda clay and fFFT samples exhibited similar structuration behaviour, which resulted in a change in compression behaviour over time for both and bore implications for their dewatering and consolidation behaviour. Hence, the structuration/ageing effect observed in polymer amended oil sands tailings is not atypical of the structuration/ageing effect observed in the natural clay deposits.

Chapter 8: Influence of polymer dosages on time-dependent effects in FFT

8.1 Introduction

The poor dewaterability and the slow settling of fine clay particles in oil sands tailings, which is a by-product of the bitumen extraction process from the oil sands ores, have resulted in the formation of large volumes of fluid fine tailings (FFT) in tailings ponds over the years. The use of high molecular weight synthetic polymers, which promote flocculation of the fine particles and help them to aggregate has been found to improve the dewaterability and settling behaviour of FFT considerably, especially, in the short-term. However, to meet the regulatory standards and develop a deposit strong and safe enough for reclamation, it is necessary to optimize the dewatering performance of the fFFT deposits in the longer-term as well. The consideration of optimum dewatering over longer timescales again warrants a comprehensive understanding of the longer-term dewatering processes such as consolidation, and also other time-dependent effects such as creep, thixotropy, and structuration.

When the long-term dewatering and consolidation behaviour of the tailings samples flocculated with a high molecular weight anionic polymer were studied in a lab-scale experiment, we observed a change in volume over time due to creep and also a change in the compressibility behaviour because of the time-dependent structuration effect which is similar to what have been observed in many natural clay deposits (Salam et al., 2017, 2018a and 2018b). Although the difference in structured intact clays and remoulded clays are widely studied, there is little information on the rate of structuration (or restructuring) in

literature. Since the performance of the oil sands tailings deposits depends on their consolidation, and the time-dependent phenomena of creep and structuration could affect their compressibility over time, it is crucially important to determine the rate of structuration in the tailings materials. It is also important to examine whether modifying the properties of tailings at the pre-deposition treatment stage (i.e. treatment of FFT with chemical amendments such as polymers) could affect the magnitude of influence of structuration on the compressibility behaviour in longer-term. Although it has been a common practice to treat the FFT with polymers to make the dewatering process faster, it is yet to be investigated whether the creep and structuration in FFT could be influenced by using different polymers or polymer dosages or polymer mixing methods. Such knowledge could help us in selecting the best performing polymer or the optimum polymer dosage, and also the mixing method, which in turn could help in optimizing the long-term dewatering and consolidation performance of the tailings deposits. Hence, this research investigated the longer-term dewatering and consolidation behaviour of FFT flocculated with 5 different dosages of an anionic polymer from an experimental perspective. Two different mixing methods were used for mixing the polymers with the FFT. The experiments included column dewatering tests, conventional oedometer tests, fall cone tests, and low vacuum scanning electron microscopy. The implications of different polymer dosages and polymer mixing methods for the creep and structuration effect in polymer amended FFT deposits, and subsequently, for the longer-term dewatering and consolidation performances were discussed based on the findings from the tests.

8.2 Materials

Oil sands tailings

Oil sands tailings samples were collected from a tailings pond in Northern Alberta, Canada, and shipped to Carleton University in Ottawa, Canada. Different laboratory tests and analyses were performed to determine the physical, mineralogical, and chemical characteristics of the untreated or unamended fluid fine tailings. The initial solids content was 30% ~ 31% and the liquid limit was 60%. The sands to fine ratio (SFR) was 0.25. The clay content obtained from the Methylene Blue Index (MBI) analysis ranged from 28% to 32%. According to the X-ray diffraction (XRD) results, the composition of the clay fraction was 68 ~ 72% Kaolinite and 28 ~ 32% Illite. Total Dissolved Solids (TDS) in the pore water collected from the raw fluid fine tailings was 1050 mg/L, electrical conductivity was 1590 micro-S/cm, while the dominant cations were sodium at 340 mg/L. Typical material characteristics of the tailings used are listed in Table 8.1.

Table 8.1. Physical properties of the untreated fluid fine tailings.

Parameters	Average value
Initial solids content (%)	30 ~ 31
Initial water content (%)	222 ~ 228
Hydrocarbons (%)	1.4
Initial wet density	1.19 ~ 1.20
Initial void ratio	5.1
Specific gravity	2.12

Liquid limit (%)	60.0 ~ 62.0
Plastic limit (%)	27.0 ~ 29.0
Plasticity index (%)	33.0
D ₉₀ , D ₆₀ , D ₅₀ , D ₁₀ (μm)	100, 11, 6, 0.8

An anionic polyamide-based polymer (A3338) was used to prepare the polymer amended oil sands tailings samples. In a plastic weighing dish, 4g of A3338 polymer was weighed using an analytical balance (Fisher Scientific, Sartorius AG Germany, LE225D) and decanted into a 1000 mL glass beaker and completed to 1000 mL with deionized water to prepare a polymer stock solution with a final concentration of 0.4%. The polymer solutions were stirred using a jar tester (Phipps and Bird, USA) at 200 rpm for 5 minutes and at 125 rpm for the following 55 minutes. Then, the polymer solution was mixed with a hand blender for 10 seconds and left for maturation for 1 hour.

To prepare the polymer amended FFT samples, 2 different mixing methods were used. The first mixing method was originally adapted from a baseline procedure developed by Shell Canada in 2010 for replicating the in-line flocculation and producing field representative samples while conducting the laboratory-scale experiments. Initially, a four-blade impeller (with a radius of 8.5 cm) attached to an overhead mixer (IKA EUROSTAR 60 control) was immersed in 2000 mL of FFT in a 5-litre pail. The mixing was initiated at a constant speed of 250 rpm and continued for 15 minutes to make the tailings homogenous. Then, the required amount of 0.4% polymer stock solution was added to the FFT by injecting the solution close to the rotating impeller during mixing (the required amount was

estimated based on the solids content of the unamended FFT). All the polymer stock solution was injected as fast as possible and in one shot into the FFT sample. The mixing was continued for 20 seconds starting from the moment the polymer stock solution was injected into the FFT.

Mizani (2016) optimized the above-mentioned mixing procedure based on the measurements of short-term dewaterability (obtained from the capillary suction time and settlement tests) and yield stress. The test results indicated that the fFFT that was produced by mixing at a speed of 250 rpm for 10 seconds resulted in optimum dewatering and settling performance (Mizani, 2016). Later, the mixing method was optimized again based on the sensitivity of the fFFT to the duration of shearing. The result from the short-term settlement tests (48 ~ 72-hour) (K. Vandara, personal communication, February 21, 2018) and the relationship observed between the torque force developed during flocculation in a Couette rheometer and the quality of flocculation (measured in terms of capillary suction time and settlement rate) (Aldaeef and Simms, 2019) indicated that mixing at a speed of 250 rpm for 20 seconds would produce an optimal dewatering performance.

The second polymer mixing method was adopted from the Northern Alberta Institute of Technology (NAIT). In this method, the mixing was initiated at a constant speed of 320 rpm and continued for 15 minutes to make the tailings homogenous. Then, a predetermined volume of the polymer stock solution was added to the FFT by injecting the solution close to the rotating impeller during mixing. All the polymer stock solution was injected as fast as possible and in one shot into the FFT sample. Starting from the moment the polymer stock solution was injected into the FFT, the mixing was continued for 10

seconds at a mixing speed of 320 rpm. Throughout this mixing time, the FFT inside the pail was conditioned by holding a metallic spatula inside the pail. After 10 seconds, the mixing speed was reduced to 20 rpm. Then, the mixing along with the conditioning was continued at a speed of 20 rpm for another 10 seconds. This additional step of conditioning was designed to ensure proper mixing and optimum flocculation of the FFT material.

Five different dosages of polymer, including 0-ppm (no polymer or unamended FFT, which was used as a control sample) were used for this experiment. Three different dosages: 400-ppm, 600-ppm, and 1000-ppm were mixed with the FFT using the regular mixing method (250 rpm@20 seconds). Two different dosages: 600-ppm and 800-ppm were mixed using the method adopted from the NAIT (320 rpm@10 seconds + 20 rpm@10 seconds + conditioning using a spatula throughout the mixing duration).

8.3 Methods

The column dewatering experiment was conducted using a single-drainage boundary condition and a single lift of FFT dosed with the predetermined amount of anionic polymer A3338 was deposited in the polypropylene columns with the material reaching a height of 10 cm. Typically, a short column height leads to the development of low effective stress within the deposited tailings material and the creep is thought to be one of the main mechanisms behind the deformations that occur at lower effective stresses (Miller, 2010). Hence, a relatively short height of 10 cm was selected to maximize the influence of creep and minimize the role of self-weight consolidation for a single, thin lift of tailings deposition. Nevertheless, such short column height still allowed for the construction of profiles of density/water content with depth upon destructive testing. After

the deposition, the columns were kept covered at the top with the help of a tight lid. The space between the lid and the surface of the deposited tailings was also kept closed in order to prevent the escape of water vapour.

Figure 8.1. A schematic diagram of the PP column used for the column dewatering test.

There were equal numbers of columns (14) for each polymer dosage. Every week one column from each dosage was selected for destructive testing and samples were collected from each 1 cm layer of the solids for different tests and analyses. The changes in total fFFT height, water-solids interface height, weight, water content, solids content, and void ratio for the whole column and the changes in water content and solids content for each 1 cm layer of the solids were measured and recorded (Figure 8.2). Four different criteria were used for assessing the performance of the polymer dosages and mixing methods used and their connections to the creep, thixotropy, and structuration: (a) dewaterability; (b) settleability; (c) stiffness; and, (d) strength.

Dewaterability

Generally, when the FFT is mixed with a polymer, the dewaterability potential of the mixture, which also gives an impression of the flocculation efficiency can be estimated as the net water release (NWR) by taking both sources of water into the account: (a) the water that is used in the preparation of the polymer stock solution; and, (b) the water that is present in the original unamended FFT (Wilson et al., 2012). The following formula is used for the determination of net water release:

$$\text{Net water release (NWR)} = \frac{W_R - W_A}{W_O} \dots\dots\dots (8.1)$$

Where W_R = the volume of the water released; W_A = the water added to the polymer stock solution; and, W_O = the volume of initial water in the FFT.

If the estimated net water release (NWR) value turns out to be positive, then, the material is considered as flocculated. A higher value of NWR indicates a well-flocculated solution. Generally, as the flocculation/aggregation of particles progresses over time and the flocs/particles continue to settle, NWR increases in value. However, if the NWR value remains negative or close to zero even after a considerable time, it indicates a poorly flocculated mixture with poor dewaterability potential. In addition to net water release (NWR), the changes in average water content and the changes in water content at different solids heights provided us with an assessment of the dewaterability potential. A consistent decline in water content over time is expected to corroborate well with a steady increase in net water release (NWR), whereas a non-uniform or minimum change in water content indicates a poor dewaterability potential.

Settleability

For assessing the settleability, the change in the void ratio of the solids as well as the change in solids height with time was recorded. The change in solids height in the polypropylene columns was expressed as percentage settlement of the solids since they had different initial sample heights (± 10.0 cm) at the beginning (despite taking maximum care to ensure a uniform 10 cm height for all the columns). Typically, the settlement (the percentage settlement) is expected to increase with the decreasing void ratio.

Compressibility and preconsolidation pressure

Conventional one-dimensional oedometer tests were performed on the samples collected from the replicate columns to determine their void ratio-effective stress relationships and track the changes in compressibility behaviour with time. Preconsolidation pressures were estimated from the compression curves (obtained from the oedometer tests) using the Casagrande method. Oedometer tests were conducted on both undisturbed/intact and remoulded samples. Because of the structuration effect, most natural clays in their undisturbed or intact state show higher preconsolidation pressure and lower compressibility in comparison to the remoulded or reconstituted samples of the same clays. When a sample is subjected to the remoulding process, it goes through a ‘destructuration’ process and loses its strength/stiffness. However, since the structuration effect is reversible, reconstituted or remoulded samples regain a component of this preconsolidation pressure over time, which is typically manifested in an increase in stiffness that allows the samples to resist the compression from an external force (Qi et al., 2018). The preconsolidation pressures determined from the compression curves provided us with a measurement of the

sample stiffness, and therefore, a mean to quantify the degree of structuration effect in the fFFT samples over time.

Undrained shear strength

A fall cone was used for measuring the undrained shear strengths developed in the fFFT samples over time. Although the fall cone test is primarily a method used for determining the liquid limit of clays, the test is also considered and used as a strength test because of the well-established empirical correlations between the liquid limit determined and the undrained shear strength of the soil at that liquid limit (Houlsby, 1982). The device has a metal cone that falls under its own weight (when it is released using a lever) and penetrates the sample placed right underneath the cone. Since the strength of the cohesive soil controls the penetration by fall cone, the measurement of cone penetration depth provides a measurement of the soil strength (Kumar and Wood, 1999). Hansbo (1957) proposed the following equation to express the relationship between the undrained shear strength (s_u) and cone penetration (d):

$$s_u = \frac{kmg}{d^2} \dots\dots\dots (8.2)$$

Where s_u = undrained shear strength (in kPa); k = cone factor, an empirical constant that depends on the apex angle of the cone as well as surface roughness, and also whether the sample is undisturbed or remoulded; m = mass of the cone; g = gravitational acceleration = 9.81 m/s²; and, d = depth of cone penetration (in mm) (Leroueil and Le Bihan, 1996; Dimitrova and Yanful, 2011).

Microstructural investigation

Scanning electron microscopy (SEM) images were generated using the samples collected from the columns. A Tescan Vega-II XMU SEM device was used for the image analysis. Using the cryogenic sample preparation technique, a rapid freezing stage of -40°C was applied to the wet FFT samples in the vacuum chamber. Then, the samples were scanned and imaged at a vacuum pressure of 50 Pa. SEM images helped to monitor the changes in size, shape, and arrangement of the flocs with time.

8.4 Results and discussions

8.4.1 Dewaterability

Immediately after the mixing of polymers, the capillary suction time (CST) tests were performed to explore the dose-response relationships between the polymer dosages and the clay content of the tailings material (Figure 8.2). CST test is a type of static filtration test that measures the filtration rate using a filter paper as the medium, and therefore, quantifies the dewatering properties. When CST tests are performed on the samples mixed with different dosages of a chemical conditioning agent (i.e. polymer), the lowest value of the capillary suction time (CST) usually refers to an optimum filterability and dewaterability (Jin et al., 2004). The results from the CST tests showed that the capillary suction times recorded were almost similar for the 600, 800, and 1000-ppm samples, with the 800-ppm (NAIT) sample generating a relatively lower CST value, and thereby showing slightly better dewaterability in the short-term.

Figure 8.2. Results from the CST tests conducted on FFT mixed with different polymer dosages.

The unamended FFT had an initial water content of 224.23% (~ solids content of 30.84%), but since it was mixed with different volumes of polymer stock solutions that included different volumes of water (in order to prepare FFT samples dosed with different polymer dosages), the initial water contents (at Day-0) of the samples flocculated with different polymer dosages were different (in the range of 223 ~ 253%). Subsequently, the 10 cm tall fFFT samples in the columns had different initial solids contents and different initial void ratios.

Although the 1000-ppm samples had the highest initial water content, they exhibited a sharp decline in their water content profiles at the initial stage of the experiment

(Day-0 to Day-28). However, after Day-28, the 1000-ppm samples showed very little change in their water contents (Figure 8.3). By contrast, 600-ppm samples (both regular and NAIT) showed a relatively better dewaterability through a uniform drop in their water contents over time.

Figure 8.3. Change in water content in the FFT samples mixed with different polymer dosages.

The difference in dewaterability between the 600-ppm and 1000-ppm samples was also evident in the comparison of water contents across the height of the solids in the columns (Figure 8.4). After Day-91, there was only a 30% drop in water content throughout the solids within the 1000-ppm columns. There was also a relatively little change in water contents at the bottom layers from the water contents at the top layers. By contrast, 600-

ppm samples (both regular and NAIT) showed a relatively larger drop in water content with 80% at the top and 100% at the bottom (Figure 8.5). The water contents measured at the bottom layers were considerably lower than the water contents measured at the top layers throughout the experiment period, which is likely to reflect the self-weight consolidation in the fFFT material. 800-ppm (NAIT) samples displayed a better dewaterability at the beginning in comparison to the 600-ppm samples (up to Day-42). However, afterwards, their progress with the dewaterability slowed down, and finally, they showed less dewaterability and settleability relative to the 600-ppm samples. Following the same trend as the water content profiles, the solids content profiles also showed that despite attaining a higher solids content at the beginning, 1000-ppm samples had relatively slower growth in longer-term (Figure 8.6). By contrast, 600-ppm samples (both regular and NAIT) were found to produce greater solids content, and therefore, better dewaterability.

Figure 8.4. Change in water content across the height of the solids in 1000-ppm samples.

Figure 8.5. Change in water content across the height of the solids in the 600-ppm (regular).

Figure 8.6. Change in solids content in FFT samples mixed with different polymer dosages.

During the determination of gravimetric water contents, the wet soil samples are typically dried in an oven at a temperature of 105⁰C for 24 hours to remove the water, and subsequently, the water content and solids content are estimated using the equations 3.1 and 3.2. For this experiment, to determine whether drying at 105⁰C could remove the water completely from the wet fFFT samples, the 91-day old samples were dried at 4 different temperatures (105⁰C, 120⁰C, 200⁰C, and 550⁰C) for the same duration (24 hours). The solids contents measured from the samples dried at 105⁰C, 120⁰C, and 200⁰C showed little difference; however, when the samples were dried at 550⁰C, there was a significant difference as the volatile solids component of the total solids were removed at high temperature (Figures 8.7, 8.8, 8.9, 8.10, and 8.11). Since it has been the standard practice to dry the samples at 105⁰C and 105⁰C is the temperature that has been used consistently for drying the wet samples during this research project, the gravimetric water contents and solids contents measured at 105⁰C were considered for further analysis.

Figure 8.7. Solids content measured at 4 different temperatures for the 400-ppm samples.

Figure 8.8. Solids content measured at 4 different temperatures for the 600-ppm (regular).

Figure 8.9. Solids content measured at 4 different temperatures for the 600-ppm (NAIT).

Figure 8.10. Solids content measured at 4 different temperatures for the 800-ppm (NAIT).

Figure 8.11. Solids content measured at 4 different temperatures for the 1000-ppm samples.

When the volumes of the water released from the FFT samples dosed with different polymer dosages (water that was accumulated on the top of the solids settled in the columns) were plotted against the time, the 800-ppm (NAIT) and 1000-ppm samples were found to generate highest volumes at the beginning. However, after Day-28, the volume of water released by the 600-ppm samples (both regular and NAIT) became almost equal or comparable to the volume of water released by the 800-ppm (NAIT) and 1000-ppm samples (Figure 8.12). At the end of the experiment, by Day-91, 600-ppm samples (regular) had the highest release of water.

Figure 8.12. The volume of water released by FFT mixed with different polymer dosages.

When the net water release (NWR) values were estimated for the FFT samples mixed with different polymer dosages and plotted against the time, despite showing significant dewaterability potential at the beginning, the 1000-ppm sample resulted in a relatively lower net water release values in longer-term (Figure 8.13). In comparison, 600-ppm samples (both regular and NAIT) displayed a consistent increase in net water release throughout the period, and therefore, showed better dewaterability. Similar to the 1000-ppm samples, 800-ppm samples (NAIT) also had higher net water release values up to Day-28; however, afterwards, they had relatively slower growth in NWR, and finally, they showed less dewaterability potential relative to the 600-ppm samples (regular).

Figure 8.13. Net water release in the FFT samples mixed with different polymer dosages.

Overall, in terms of dewaterability, 600-ppm samples (both regular and NAIT) were relatively better performers. The samples that were mixed with higher dosages of polymer displayed better initial dewatering behaviour; however, in the long-term, their progress with the dewaterability became slower. Although 800-ppm (NAIT) samples displayed a dewaterability potential comparable to the 600-ppm samples (NAIT), 1000-ppm samples didn't improve in dewaterability over time.

8.4.2 Settleability

For all the different dosages, the degree of saturation was estimated, and the results indicated that the samples remained 100% saturated throughout the experiment period

(Figure 8.14). The void ratios were determined based on the changes in sample height. When the void ratios were plotted against the time, 600-ppm samples (both regular and NAIT) were found to generate the lowest void ratios (Figure 8.15). Their void ratios decreased in a consistent manner, which indicated a good settlement. 800-ppm samples (NAIT) experienced a sharp drop in the void ratio at the beginning (up to Day-14). Later, the void ratio decreased relatively slowly, and finally, 800-ppm (NAIT) samples had a relatively higher void ratio than the 600-ppm samples. In longer-term, 1000-ppm samples also had a higher void ratio. Although the 1000-ppm samples displayed a considerable drop in the void ratio in the first two weeks, after Day-14, there was a nominal change.

When the solids settlement was expressed in terms of decrease in solids height over time (% decrease in solids height) and plotted against the time, 600-ppm samples (both regular and NAIT) and 800-ppm samples (NAIT) demonstrated almost similar settleability in longer-term (Figure 8.16). By contrast, 1000-ppm samples had a relatively lower settleability. Both the 800-ppm sample (NAIT) and the 1000-ppm sample displayed a rapid settling behaviour at the beginning. But, with time, their settling rates decreased, especially, in the 1000-ppm samples. Similar to the void ratio, after the initial dewatering stage, 1000-ppm samples displayed very little progress in terms of their settling behaviour.

Overall, in terms of settling performance, 600-ppm samples (both regular and NAIT) were more consistent as the solids in the samples settled uniformly with time while the void ratio was also decreasing steadily.

Figure 8.14. The degree of saturation estimated from the FFT dosed with different dosages.

Figure 8.15. The change in the void ratio of the FFT mixed with different polymer dosages.

Figure 8.16. Solids settlement in the FFT samples mixed with different polymer dosages.

8.4.3 Preconsolidation pressure (yield stress)

At specific time intervals, samples were collected from the columns and the conventional consolidation tests (oedometer tests) were performed on them. 0-ppm and 400-ppm samples were found to be too yielding for conducting the oedometer tests. In the case of the 600-ppm samples, only after Day-35, it was possible to conduct the test. When the test was conducted on a Day-63 600-ppm sample (regular), the sample showed a sign of developing stiffness. Day-91 600-ppm sample (regular) showed relatively greater stiffness; however, there was still no distinct apparent preconsolidation pressure or yield strength development (Figure 8.17).

800-ppm samples (NAIT) showed a sign of developing stiffness from the beginning (Day-14 and Day-35 samples). By Day-63, the sample became relatively stiffer and showed resistance to the compression force applied. By the end of the experiment (Day-91), there was a visible increase in preconsolidation pressure (Figure 8.18). Such a change in compressibility behaviour with time and an increase in preconsolidation pressure (yield strength) were clear indications of the structuration effect in fFFT. By contrast, 1000-ppm samples showed considerable stiffness from the very beginning, starting from Day-14. By Day-63, there was a visible increase in yield strength and a decrease in compressibility (Figure 8.19). However, after Day-63, apparently, the change in compressibility behaviour became slower and there was no increase in preconsolidation pressure.

Figure 8.17. Change in compressibility behaviour in the 600-ppm samples (regular).

Figure 8.18. Change in compressibility behaviour in the 800-ppm samples (NAIT).

Figure 8.19. Change in compressibility behaviour in the 1000-ppm samples over time.

In addition to the undisturbed or intact samples, oedometer tests were also conducted on the remoulded samples. The remoulded samples were prepared by shearing the undisturbed samples to an extent that the clay particle structure that was developed earlier under the resting condition was destroyed. When the compressibility curve obtained from an undisturbed fFFT sample was compared with the compressibility curve obtained from the remoulded sample of the same fFFT material, there was a significant difference in their compressibility behaviour. Figure 8.20 and 8.21 show the compressibility curves obtained from the Day-35 1000-ppm undisturbed and remoulded samples and Day-63 800-ppm (NAIT) undisturbed and remoulded samples respectively. Since the remoulded samples experienced the ‘destructuring’ process during their preparation, they lost a part of their shear strength, and hence, showed relatively less stiffness or resistance to the external compression force applied. Such a difference in the compression behaviour between the undisturbed and remoulded samples, which was also reflected in the difference in void ratio at the preconsolidation pressure (of the undisturbed sample) was evidence of time-dependent structuration effect in flocculated FFT. The difference in the stiffness or the void ratio between the undisturbed and remoulded sample was relatively prominent in the Day-35 1000-ppm sample in comparison to the Day-63 800-ppm (NAIT) sample, which implied that the structuration effect was more prominent in the 1000-ppm sample at the beginning.

Figure 8.20. The compressibility behaviour of the Day-35 1000-ppm undisturbed and remoulded samples.

Figure 8.21. The compressibility behaviour of the Day-63 800-ppm (NAIT) undisturbed and remoulded samples.

When the compressibility curves obtained from two remoulded samples such as Day-35 1000-ppm remoulded sample and Day-63 800-ppm (NAIT) remoulded sample were compared, despite their different ageing period (Day-35 and Day-63), different polymer dosages, and different mixing methods (regular and NAIT), both displayed almost the same compressibility behaviour once they were remoulded (Figure 8.22). Apparently, after remoulding, both samples reverted to their similar pre-structuration state and displayed their similar intrinsic properties. Hence, the time-dependent effects of structuration in the undisturbed samples were reduced by the process of remoulding.

Figure 8.22. The compressibility curves obtained from the Day-35 1000-ppm remoulded sample and the Day-63 800-ppm (NAIT) remoulded sample.

However, when the compressibility curves from the corresponding undisturbed samples (Day-35 1000-ppm undisturbed sample and Day-63 800-ppm undisturbed sample) were compared, both samples showed an increase in stiffness with the Day-35 1000-ppm sample showing relatively greater stiffness despite being a relatively less-aged sample (Figure 8.23). Hence, the degrees/rates of structuration were different in the FFT samples dosed with different polymer dosages. However, such rates were also found to vary with time as the 800-ppm (NAIT) undisturbed sample developed a greater yield strength relative to the 1000-ppm undisturbed sample in the long-term (at Day-91).

Figure 8.23. The compressibility curves obtained from the Day-35 1000-ppm undisturbed sample and the Day-63 800-ppm (NAIT) undisturbed sample.

Provided that the FFT sample is mixed with the polymer at an optimum mixing speed for an optimum duration (so that over-shearing due to excess mixing speed or

insufficient mixing due to lower speed can be avoided), the increase in yield stress of the fFFT material typically depends on the ageing or structuration effect in the material (Mizani, 2016). Since the FFT samples that were mixed with the polymer dosages of 800-ppm (NAIT) and 1000-ppm were found to increase more in preconsolidation pressure (or stiffness) with time in comparison to the FFT samples mixed with other polymer dosages (i.e. 600-ppm), the structuration effect was apparently more prominent in the 800-ppm (NAIT) and 1000-ppm samples than the others.

8.4.4 Undrained shear strength

A fall cone was used for measuring the undrained shear strengths developed in the fFFT samples. The undrained shear strength has been found to increase in most natural clay deposits over time, which has been attributed to the thixotropic strength gain within the materials (Bjerrum, 1967). When the undrained shear strengths measured from the FFT samples flocculated with different polymer dosages were plotted against the time, 800-ppm (NAIT) and 1000-ppm samples were found to develop the highest strength. By contrast, 600-ppm samples (both regular and NAIT) were found to develop a relatively smaller amount of strength (Figure 8.24). The results also showed how the thixotropic strength buildup in the fFFT samples could vary depending on the polymer dosages applied.

When the undrained shear strengths and water content profiles were plotted together, the results were found to shed some light on how the thixotropic strength buildup in the samples is correlated to the rate of change in their water contents (Figure 8.25). In the case of 800-ppm (NAIT) and 1000-ppm samples, the strength started to increase noticeably only after the initial rapid decline in water content slowed down towards the

end of primary consolidation around Day-28. Although the decrease in the void ratio (or the increase in density) as a result of initial dewatering and self-weight consolidation contributed to an increase in the strength of fFFT at the initial stage, shear strength was found to increase at a much greater rate after the rate of change in water content became relatively nominal with time. Apparently, the tailings gained strength only after a sufficient amount of water had been removed from the samples and when the samples appeared to reach an equilibrium in terms of water content. Although some additional water was released from the samples with the development of structuration afterwards, a relatively stable condition allowed the fine particles to coalesce and develop stronger interparticle bonding. Hence, the change in void ratio or density (due to the dewatering and self-weight consolidation) was thought to be a minor factor in the strength gain of the 800-ppm and 1000-ppm samples; rather, it was the thixotropic effect that contributed the most.

In the 600-ppm samples (both regular and NAIT), the water contents decreased in a consistent manner over a relatively larger time frame (up to Day-63) and the samples didn't develop any noticeable strength throughout the time. However, after Day-63, when the water contents started to decrease at a relatively slower rate, the samples also started to buildup strength at a greater rate. Nevertheless, by Day-91 or by the end of the experiment, the strengths developed in the 600-ppm samples (both regular and NAIT) were relatively smaller than the strengths developed in the 800-ppm (NAIT) and 1000-ppm samples (Figure 8.26).

When the undrained shear strength development in the ageing Leda clay samples was compared to strength development in the FFT samples of same ages but dosed with

different polymer dosages, all the samples including the Leda clay samples appeared to develop a fairly low shear strength at the beginning and remained at the almost same level up to 30 ~ 40 days. However, after Day-30 ~ Day-40, the fall cone shear strength began to increase considerably (Figure 8.27). Hence, both Leda clay and fFFT samples were observed to develop significant thixotropic strength as the samples appeared to reach an equilibrium in terms of water content though the rate of strength development varied depending on the material type (FFT/Leda clay) as well as the polymer dosages used (FFT).

Figure 8.24. Undrained shear strengths measured from the fall cone tests.

Figure 8.25. Correlation between water content and thixotropic strength (800/1000-ppm).

Figure 8.26. Correlation between water content and thixotropic strength (600-ppm).

Figure 8.27. Undrained shear strength development in the Leda clay and the fFFT samples.

Structuration effect and thixotropic behaviour

Preconsolidation pressures were estimated from the compressibility curves obtained from the Day-14, Day-35, Day-63, and Day-91 1000-ppm undisturbed samples using the Casagrande method of determining preconsolidation pressure. When the estimated preconsolidation pressures and the undrained shear strengths measured from the 1000-ppm samples were plotted together against the time, both preconsolidation pressure and undrained shear strength showed a similar increasing trend and were also found to be proportional (Figure 8.28). Based on the estimated values, an empirical relationship was observed between the preconsolidation pressure and the undrained shear strength:

$$\frac{\text{Undrained shear strength}}{\text{Preconsolidation pressure}} = 0.18 \dots\dots\dots (8.3)$$

A similar proportional relationship between the preconsolidation pressure and the undrained shear strength has been observed in natural deposits of sedimentary clays; Terzaghi et al. (1996) proposed the following empirical relationship for clays:

$$\frac{\tau_f}{\sigma'_p} = 0.22 \dots\dots\dots (8.4)$$

Where τ_f = undrained shear strength; and, σ'_p = preconsolidation pressure.

Such similarities in the relationships between the preconsolidation pressure and the undrained shear strength implied that both natural clays and fFFT samples show similar time-dependent strength and stiffness development behaviour. Apparently, the increase in undrained shear strength (due to the thixotropic strength regain) and the increase in apparent preconsolidation pressure or the recovery of stiffness (due to the structuration effect) are just the different manifestations of the same physical phenomenon since both thixotropy and structuration originate from the same process of stronger particle-particle interactions and bonding and the subsequent particle rearrangements (Nagaraj et al., 1990). The only difference between them is that the structuration effect leads to a gradual recovery of stiffness in a reconstituted or remoulded clay sample, whereas thixotropy results in recovery of strength with time (Qi et al., 2018).

Figure 8.28. The empirical relationship between the preconsolidation pressure and undrained shear strength measured in 1000-ppm samples over time.

Microstructural analysis (scanning electron microscopy)

The technique of scanning electron microscopy (SEM) was used for monitoring the changes in size, shape, and arrangement of the particles or flocs in the samples. Samples were collected from the surface of the solids. Figure 8.29 shows the changes in the fabric of the 600-ppm (NAIT), 800-ppm (NAIT), and 1000-ppm samples over time (Day-3 vs. Day-91). The samples became less porous and more compact and homogenous in their appearances with time. The corresponding changes in particle-particle interaction and bonding were thought to be the main causes of the structuration effect.

Figure 8.29. SEM images generated from the (top) 600-ppm (NAIT) samples, (middle) 800-ppm (NAIT) samples, and (bottom) 1000-ppm samples (500X magnified).

8.4.5 Implications of structuration effect for compressibility behaviour

Typically, the large strain consolidation modelling is used to predict the compression behaviour of the tailings deposits. For performing the large strain consolidation analysis, key consolidation properties such as compressibility and hydraulic conductivity are determined from the large strain consolidation tests (Jeeravipoolvarn et al., 2015). Large strain consolidation analysis provides us with the final settlement of the deposits. However, the settlement predicted by the model could vary from the settlement in the real field conditions. Because, the time-dependent phenomena of creep and structuration, which the original finite strain consolidation theory does not consider may change the compressibility and hydraulic conductivity relationships in FFT over time, which in turn, may produce a final settlement different than the settlement predicted by the model (Jeeravipoolvarn et al., 2008).

To examine the implications of the structuration effect for the compression behaviour of polymer amended tailings, the final density profile of a 50-metre tailings deposit was generated using a simple numerical analysis method (Carrier and Freeman, 2018). Two different density profiles were produced based on two different compressibility curves: (a) compressibility curve obtained from the Day-91 1000-ppm sample in its undisturbed state; and, (b) compressibility curve obtained from the Day-91 600-ppm sample in its remoulded state (Figure 8.30). The Day-91 1000-ppm undisturbed sample showed significantly greater stiffness during the oedometer test (which was evident in its higher preconsolidation pressure). By contrast, the Day-91 600-ppm remoulded sample produced relatively less stiffness as well as no preconsolidation pressure. A comparison

between the density profiles generated by these two dissimilar samples manifested the effects of structuration on the compressibility behaviour clearly. The Day-91 1000-ppm undisturbed sample resulted in a higher void ratio as well as higher final elevation (23.7 m), which was due to its stiffness. By contrast, the Day-91 600-ppm remoulded sample produced a considerably lower void ratio as well as lower final elevation (19.1 m) because of its more compressible nature. Final density profiles showed that the time-dependent structuration effect in fFFT could change its compressibility behaviour over time, and therefore, the final settlement in the real field conditions could differ from the final settlement predicted by the conventional consolidation models.

Figure 8.30. Final density profiles of a 50-metre tailings deposit (based on the compressibility curves obtained from the undisturbed and remoulded fFFT samples).

8.4.6 The link between the polymer dosage and structuration effect

The table below categorizes the polymer dosages used in this experiment based on the assessment of their different properties over time: (a) dewaterability; (b) settleability; (c) preconsolidation pressure (or recovery of stiffness); and, (d) undrained shear strength (or thixotropic strength recovery).

Table 8.2. Assessment of performances of different polymer dosages.

Criteria	High	Medium	Low
Dewaterability	600 (Regular), 600 (NAIT)	800 (NAIT)	1000
Settleability	600 (Regular), 600 (NAIT)	800 (NAIT)	1000
Preconsolidation pressure/stiffness	800 (NAIT)	1000	600 (Regular)
Undrained shear strength	800 (NAIT)	1000	600 (Regular), 600 (NAIT)

In general, the higher dosages (i.e. 800-ppm and 1000-ppm) of the polymer A3338 resulted in greater stiffness and strength (manifested in a higher preconsolidation pressure and undrained shear strength respectively) and also in less dewaterability potential and settleability relative to lower dosages (i.e. 600-ppm). 600-ppm samples (both regular and

NAIT) were found to develop relatively smaller or no preconsolidation pressure and nominal undrained shear strength; however, they resulted in relatively greater dewaterability potential and settlement. Apparently, there was an inverse relationship between the polymer dosage and the dewaterability and settleability, and a direct relationship between the polymer dosage and the strength and stiffness.

Figure 8.31. Turbidity of the water released from the flocculated samples after Day-7.

At the initial stage of the experiment (at Day-7), the water released by the lower polymer dosages (i.e. 0-ppm and 400-ppm) were relatively turbid and visibly rich in total suspended solids (TSS), which was probably due to the inadequate amount of polymer in the FFT samples (Figure 8.31). There was not enough polymer for a complete particle neutralization. By contrast, the higher polymer dosages were found to produce less turbid water, water released from the 800-ppm (NAIT) samples and the 1000-ppm samples being the clearest among them. Generally, clear water (no turbidity) is an indication of good flocculation. Water released by the 600-ppm samples (both regular and NAIT) was slightly turbid with traces of suspended solids. Again, the water released by the 600-ppm (NAIT)

samples was relatively clear among them, which was probably due to the additional step of conditioning performed during the application of the NAIT mixing method.

In the case of higher polymer dosages (i.e. 800-ppm and 1000-ppm), after adding the polymer stock solution, because of more efficient bridging of the fine clay particles in the FFT samples (through the adsorption of polymer chains on the particle surfaces and the collision of the particles), which resulted in a more efficient flocculation, and subsequently, in the formation of larger flocs/aggregates (which was confirmed based on visual observations as well as zeta potential measurements and quantitative analysis of the SEM and optical microscopy images), the fFFT samples, in addition to generating relatively clear bleed water also showed considerable dewaterability and settleability at the beginning as the larger flocs settled rapidly under gravity. However, in longer-term, higher polymer dosages, despite their superior initial flocculation performances and high water release appeared to be an impediment to the optimum dewatering or settling behaviour. The larger flocs also resulted in a greater stiffness for the samples and the buildup of such stiffness within the fFFT samples hindered the self-weight consolidation at very low stresses, which also reduced the dewaterability potential. Apparently, the formation of larger flocs, and subsequently, rapid initial dewatering, which also contributed to a relatively shorter primary consolidation period acted as the prerequisite for the structuration effect in the fFFT samples.

The effects of different polymer dosages on the thixotropic strength buildup in the fFFT samples were also evident in Figure 8.32. At the onset of developing strength, when there was no or minimal effect of change in density on the strength buildup, the higher

polymer dosages (i.e. 800-ppm and 1000-ppm) developed a significantly higher undrained shear strength in comparison to the lower polymer dosages (i.e. 600-ppm regular, 600-ppm NAIT, and 400-ppm).

Figure 8.32. The shear strength developed in FFT dosed with different polymer dosages.

The 600-ppm samples (both regular and NAIT), which did not display a flocculation efficiency as good as the 800-ppm (NAIT) or 1000-ppm samples showed a considerable and consistent improvement in dewaterability and settleability with time. The 600-samples also did not develop as much stiffness or strength as the 800-ppm (NAIT) or 1000-ppm samples, which eventually helped them in achieving a greater dewaterability potential and settling performance in longer-term. 800-ppm of polymer dosage (800-ppm

NAIT) resulted in considerable flocculation at the beginning, and subsequently, rapid initial dewatering. The rate of strength development in the 800-ppm (NAIT) samples was considerably and consistently greater than the 600-ppm samples throughout the duration, and in the long-term, the 800-ppm (NAIT) samples demonstrated less dewaterability and settleability relative to the 600-ppm samples.

In the case of unamended FFT or 0-ppm samples (control sample), the initial dewatering, which was comparable to the initial dewatering of the samples mixed with polymers was due to the rapid sedimentation or gravity-driven settling of the particles (there was no polymer or no flocculation process for promoting the aggregation of the fine clay particles or disrupting the stable colloidal system in the 0-ppm samples). There was no strength or stiffness development in the 0-ppm samples over time; however, it still did not help the dewaterability or settlement process beyond a certain limit. In fact, after a certain time (Day-42), very little dewaterability or settlement was observed in the 0-ppm columns. Since Day-42, till the end of the experiment, the settlement of the solids (%) was in the range of 18 ~ 21% consistently and the volume of water released never exceeded 84 mL. Similar was the observation with the 400-ppm samples (since there was not enough polymer to drive adequate flocculation/aggregation of the fine clay particles). After Day-42, there was almost no change in the amount of water released (92 mL) and the settlement of the solids (%) was consistently in the range of 21 ~ 22%. By contrast, at Day-91, the decrease in solids height (or the increase in solids settlement, expressed in percentage) and the increase in the volume of water released (dewaterability) were still continuing in the 600-ppm (regular and NAIT), 800-ppm (NAIT), and 1000-ppm samples.

8.4.7 KPI used by the industry for evaluating the performance of polymers in the short-term and the optimization of the long-term dewatering performance

For assessing the performance of different polymers, selecting the potential polymer treatment candidates, and determining the optimal polymer dosage for the full-scale or field-scale implementation, typically, a series of laboratory-scale screening experiments are performed on the flocculated tailings, and the best performing polymer or the optimum dosage is selected based on the consideration of specific key performance indicators (KPIs). The commonly used key performance indicators (KPIs) include capillary suction test time (CST), settling rate (from the settling column test), yield stress, clay-to-water (CWR) ratio (in flocs), solids content and net water release at 24 hours (Wells et al., 2011; Demoz and Mikula, 2012; Mizani et al., 2013, 2017; Boxill et al., 2018). Since the amount of water released from the fFFT within the initial 24 ~ 48-hour period of polymer mixing constitutes most of the total free-draining water in fFFT, for practical reason, most laboratory-scale research on polymer optimization is directed towards maximizing short-term dewatering over one- or two-days post-deposition or post-treatment (Mizani et al., 2017; Boxill et al., 2018).

Although the polymer-aided flocculation was thought to be a minor influence on the consolidation characteristics of tailings (Hogg et al., 1993), this study showed that polymer dosages have significant influences on the time-dependent thixotropic and structuration behaviour of fFFT; especially, the degree of structuration in fFFT was found correlated to the polymer dosages used. Structuration appeared to manifest only when the water content became stable after a sharp decline in water content (due to the rapid initial

dewatering at the beginning) or after the end of primary consolidation, and the structuration effect appeared to become prominent once the rate of dewatering or the rate of self-weight consolidation or the density of the sample reached a certain level. Apparently, the fFFT samples that exhibited the highest short-term dewatering potential (i.e. 800-ppm and 1000-ppm) also exhibited the highest degree of structuration and such time-dependent effect influenced their dewaterability and settlement behaviour in longer-term. Hence, any specific polymer or polymer dosage that produces optimum dewatering performance in the short-term may not be the best-performing polymer or optimum polymer dosage for the long-term dewatering mechanisms. In other words, the short-term dewatering optimization may not result in optimization for long-term dewatering.

In order to overcome such challenges and to satisfy the dewatering and consolidation performance criteria post-deposition both in the short-term and long-term under field conditions, several key performance indicators (KPIs) were used for evaluating the performances of the tailings-polymer mixtures during the current experiment. Table 8.3 illustrates the short-term key performance indicators and Table 8.4 illustrates the longer-term key performance indicators for optimal dewatering and settling behaviour.

Table 8.3. Short-term key performance indicators for the optimization of polymer dosage.

Dosage	CST (sec)	Settlement (%)	Net water release	Solids content (%)	Undrained shear strength (Pa)
(ppm)	0 hour	48-hour	48-hour	48-hour	7-day
0	1311 6213	1.53	1.84	31.13	0
400	1247 5849	2.00	-3.07	30.30	0
600 (regular)	1046 5433	2.91	-3.96	29.98	0
600 (NAIT)	1028 4520	5.15	-0.13	30.30	47
800 (NAIT)	980 4324	22.33	17.84	31.95	1224
1000	1054 4498	23.53	15.51	33.03	1443

Table 8.4. Long-term key performance indicators for the optimization of polymer dosage.

Dosage	Settlement (%)	Net water release	Solids content (%)	Yield strength (kPa)	Undrained shear strength (Pa)
(ppm)	90-day	90-day	90-day	90-day	90-day
0	20.62	23.84	37.30	0.00	403
400	22.22	21.10	37.14	0.00	537
600 (regular)	28.16	26.13	39.03	13.74	1257
600 (NAIT)	26.80	23.22	38.18	0.00	1392
800 (NAIT)	27.45	23.27	37.92	54.95	5847
1000	25.74	19.29	35.22	27.48	5320

An examination of the short-term key performance indicators (Table 8.3) showed that an 800-ppm dosage resulted in better dewaterability, water release, and settling behaviour in the short-term in comparison to the 600-ppm (both regular and NAIT) or 1000-ppm dosages. For the convenience of comparing the capillary suction time performances of different polymer dosages, the CST test results were normalized with respect to the 800-ppm (NAIT) CST result and then averaged (Figure 8.33). Similarly, solids settlement results (Figure 8.34), net water release results (Figure 8.35), solids content results (Figure 8.36), and undrained shear strengths (Figure 8.37) were normalized with respect to the 800-ppm (NAIT) results.

Figure 8.33. Averaged normalized CST test results with respect to the 800-ppm results.

Figure 8.34. Normalized settlement results (48-hour) with respect to the 800-ppm results.

Figure 8.35. Normalized net water release (48-hour) with respect to the 800-ppm results.

Figure 8.36. Normalized solids content (48-hour) with respect to the 800-ppm results.

Figure 8.37. Normalized undrained shear strengths (7-day) with respect to the 800-ppm.

When the longer-term key performance indicators were evaluated based on the criteria of solids settlement (%), net water release, solids content, preconsolidation pressure, and undrained shear strength measurements, 600-ppm appeared to exceed the 800-ppm in terms of dewatering and settling performance (Figures 8.38, 8.39, and 8.40). Although 800-ppm and 1000-ppm dosages developed more strength/stiffness (Figures 8.41 and 8.42), such development resulted in relatively slower dewatering and settling rates in the longer-term. By contrast, apparently, relatively less strength/stiffness development in the 600-ppm samples led to a better dewaterability and settlement performance.

Figure 8.38. Normalized settlement results (90-day) with respect to the 800-ppm results.

Figure 8.39. Normalized net water release (90-day) with respect to the 800-ppm results.

Figure 8.40. Normalized solids content results (90-day) with respect to the 800-ppm results.

Figure 8.41. Normalized undrained shear strengths (90-day) with respect to the 800-ppm.

Figure 8.42. Normalized preconsolidation pressure (90-day) with respect to the 800-ppm.

An examination of the normalized 48-hour test results (normalized with respect to the 800-ppm results) showed that 600-ppm dosage (regular and NAIT) resulted in 5 ~ 10% more capillary suction time, 75 ~ 85% less settlement, 90 ~ 95% less net water release, and 5 ~ 6% less solids content in comparison to the 800-ppm (NAIT) dosage, which appeared to be the optimum dosage for dewatering in the short-term based on the CST, settlement, and net water release results. However, in longer-term (90-day), 600-ppm dosage (regular and NAIT) resulted in 3% more settlement, 6% more net water release, and 3% more solids content in comparison to the 800-ppm (NAIT) dosage. After 7 days, 600-ppm developed an undrained shear strength, which was only 4% of the undrained shear strength developed by 800-ppm after the same period. Even after 90 days, 600-ppm had a considerably lower strength (23% of the 800-ppm strength). Similar to the undrained shear strength, after 90 days, 600-ppm developed a relatively small apparent preconsolidation pressure, which was 25% of the preconsolidation pressure developed in 800-ppm at the same point in time. Hence, despite showing suboptimal dewatering and settlement performance in the short-term, 600-ppm showed a better dewaterability and settlement behaviour in longer-term, but relatively low strength/stiffness development. By contrast, increased strength/stiffness in higher dosages (800-ppm and 1000-ppm) (which was due to the structuration effect) impeded the self-weight consolidation at lower effective stresses.

Although it has been an industry practice to conduct a series of laboratory-scale screening experiments on the FFT samples dosed with different polymers or polymer dosages and to select the best performing polymer or optimum polymer dosage for the full-scale or field-scale implementation based on some specific key performance indicators

(KPIs), it has always been a great challenge to predict whether the selected polymer or the polymer dosage would produce an optimal dewatering performance in the fields as well. The CST, solids settlement, solids content, net water release, undrained shear strength and preconsolidation pressure results from this study suggested that a polymer dosage that is most likely to generate an optimal dewatering performance in longer-term may generate a relatively higher CST time (5 ~ 10%) (immediately after the mixing), less settlement (75 ~ 85%) (48-hour), less net water release (90 ~ 95%) (48-hour), and less solids content (5 ~ 6%) (48-hour) in comparison to the apparent optimum polymer dosage (the dosage that produces lowest CST, highest solids settlement, and highest net water release during the laboratory-scale screening experiments). However, the above-mentioned ranges of numbers may vary depending on the polymer types, tailings characteristics or compositions, and mixing/operating conditions. If a polymer dosage is selected from the results of the short-term laboratory-scale screening experiments but also based on the consideration of optimizing the long-term dewatering performance, such polymer dosage may not generate an optimal dewatering and settling rate immediately after the deposition or in the short-term, but the FFT flocculated with such dosage would be less susceptible to the potentially adverse effects of structuration in longer-term, and therefore, would produce an overall optimal performance.

8.4.8 A simple empirical method for predicting the structuration effect in fFFT

In this study, in addition to a 0-ppm FFT sample (the control sample with no polymer), FFT samples were initially dosed with five different polymer dosages in the range of 400 ~ 1000-ppm, deposited in the 10 cm tall polypropylene columns, and later

showed the structuration effect in varying degrees. Based on the measurements of solids settlement (%) or the volume of the water released from the fFFT (mL) or the net water release (NWR) in the columns after 48 hours of deposition and the measurements of undrained shear strength from the 90-day old fFFT samples, a simple empirical method was proposed for predicting the degree of structuration in a particular tailings-polymer mixture. Typically, the capillary suction time (second), solids settlement (%), water released (mL), net water release, and solids content (%) are used as the key performance indicators for the fFFT in the short-term, whereas the undrained shear strength values reflect the thixotropic strength developed in the fFFT with time. Therefore, if the thixotropic strengths measured from the 90-day old fFFT samples are found to correlate well to the 48-hour solids settlements or to the volume of the water released after 48 hours, such correlation could be used to predict the potential thixotropic strength development in the fFFT samples (which would also approximate the degree of structuration in the fFFT samples) based on the measurements of short-term key performance indicators.

When the measured undrained shear strengths (at Day-90) were plotted with the 48-hour solids settlement in the columns and also with the volume of the water released from the fFFT samples after 48 hours of deposition, a linear relationship was observed between the undrained shear strengths (Pa) and the solids settlement (%) (Figure 8.43) and also between the undrained shear strengths (Pa) and the volume of the water released from the fFFT (mL) (Figure 8.44).

Figure 8.43. The linear relationship between the 48-hour settlement and the Day-90 undrained shear strength.

Figure 8.44. The linear relationship between the 48-hour water release and the Day-90 undrained shear strength.

When the undrained shear strengths measured at Day-90 were plotted with the net water release (NWR) measured from the fFFT samples after 48 hours of deposition, a similar linear relationship was observed between the undrained shear strength and the net water release (NWR) (Figure 8.45). Such correlations indicate that it is possible to predict the degree of structuration in a particular tailings-polymer mixture based on the measurements of short-term key performance indicators.

Figure 8.45. The linear relationship between the 48-hour Net water release (NWR) and the Day-90 undrained shear strength.

Although the difference in structured intact clays and remoulded clays are widely studied, there is little information on the rate of structuration (or restructuration) in literature. Nevertheless, it is crucially important to determine the rate of structuration in soft soil deposits, especially in the oil sands tailings deposits because the dewatering performance of the tailings deposits depends on their consolidation behaviour, and the

time-dependent phenomena of creep and structuration could affect their compressibility over time. By drawing a parallel between the structuration behaviour observed in natural clays and the similar behaviour observed in clay-rich fFFT and using the existing literature on determining the degree of structuration in soft soils (which are based on the changes in compressibility curves over time, i.e., Burland, 1990; Liu and Carter, 1999; Zeng et al., 2016), a few preliminary methods have been proposed for predicting the degree of structuration in a particular tailings-polymer mixture (Qi et al., 2018; Narges et al., 2019). Although the degree of structuration in polymer-amended fines dominated tailings appeared to be in the same order as the structuration measured in natural clay deposits (Qi et al., 2018), an empirical method is much needed for determining the degree of structuration for a given tailings deposit under field conditions. Conducting longer-term column dewatering tests on fluid fine tailings of different compositions and characteristics using different polymers, polymer dosages, polymer mixing methods, and column sizes (diameters and heights) and determining the undrained shear strength that develops in fFFT in longer-term could help in generating more data sets and validating and improving the proposed empirical method and its accuracy. Such knowledge may allow the operators to minimize the negative effects of structuration through appropriate deposition management plan (i.e. layered deposition or use of wick drains to shorten drainage paths) or an appropriate chemical amendment (i.e. an appropriate polymer or polymer dosage).

8.4.9 Conclusion

Most of the recent research on dewatering of tailings has focused on initial dewatering that occurs over a short period after adding polymers and these works have

proved the effectiveness of polymers to assist in increasing the solids content, however, only in the short-term. To gain sufficient strength to allow for the reclamation of tailings impoundments, the consideration of longer-term dewatering through consolidation as well as consolidation-related time-dependent phenomena, i.e., creep, structuration, and thixotropy is crucial. This study examined the performances of different polymer dosages as well as the link between the polymer dosage and the creep, thixotropy, and structuration effect in fFFT on the basis of four criteria or performance indicators: (a) dewaterability; (b) settleability; (c) stiffness; and, (d) strength, both in the short-term and longer-term.

Results from the study showed that the structuration effect was most prominent in the samples mixed with higher dosages of the polymer. The changes in compressibility curves (increase in preconsolidation pressure with time and reduced compressibility) due to the structuration effect in the 1000-ppm samples were found to influence their consolidation behaviour in the longer-term. Although optimal flocculation led to a rapid and large initial dewatering and primary consolidation at the beginning, increased stiffness and strength in the fFFT materials impeded their progress with the creep compression or secondary consolidation. Although the 800-ppm (NAIT) samples demonstrated a good dewaterability potential and a considerable settlement performance, the development of high undrained shear strength and stiffness within the 800-ppm (NAIT) samples prevented them from achieving the optimum performance. 600-ppm samples (both regular and NAIT) produced maximum dewaterability and settleability in longer-term, and they were also found less susceptible to the negative effects of structuration as strength/stiffness development in the 600-ppm samples was relatively insignificant (minimal or no

preconsolidation pressure and relatively small undrained shear strength even after 90 days). Performance-wise, there was very little difference between the 600-ppm (regular or 250 rpm@20 sec mixing method) samples and 600-ppm (NAIT mixing method) samples.

The findings of this study are restricted to the conditions that were investigated in the experiments; hence, the findings or the conclusions are not necessarily transferable to other conditions or settings. So, different polymers or polymer dosages, different tailings compositions and properties, different lift heights, etc. may produce different results (Hripko et al., 2018). In this study, a short height (10 cm) was selected for a single, thin lift of tailings deposition so that the influence of creep could be maximized. A single-drainage boundary condition was considered with no evaporation. However, in field conditions, tailings are typically deposited in multiple lifts or much thicker lifts, and there are evaporation and freeze-thaw effects and different boundary conditions. Hence, more research is required to evaluate the optimum performances of the polymers under a wide range of operating conditions and field trials are required for the validation for the results of the tests. Nevertheless, the test results from this study implied that polymers may have a significant influence on the settlement and dewatering behaviour of FFT not only in the short-term but also in the longer-term. Therefore, while selecting an optimum polymer dosage for a particular tailings-polymer mixture, the magnitude of the resulting structuration effect on the compression behaviour should be considered since such effect could influence the settlement performance and dewatering behaviour in the long-term.

Chapter 9: Synthesis of the results

9.1 Introduction

Evidence of structuration has been reported in natural deposits of sedimentary clays and dredged sediments, and such time-dependent behaviour has been observed to influence their long-term settlement and strength development behaviour. Polymers have been found to improve the dewaterability and settling behaviour of clay-rich oil sands tailings over a short time frame; however, it had been unknown whether structuration has any implications for the long-term dewatering and consolidation performance of polymer amended tailings deposits. This research investigated structuration behaviour in polymer amended tailings from an experimental perspective in light of structuration behaviour observed in clays.

When a combination of laboratory-scale experiments was conducted on the tailings samples dosed with different polymer dosages, a change in volume was observed in the samples over time due to the creep compression. The polymer amended tailings samples also developed a thixotropic strength and there was a time-dependent change in the compression behaviour because of the structuration effect. There was an increase in apparent preconsolidation pressure and reduced compressibility, which impeded the self-weight consolidation of tailings in the long-term by limiting the creep compression.

9.2 Structuration effect and the final settlement and slope stability of a tailings deposit

When oedometer tests were performed on the flocculated FFT samples at specific intervals, the preconsolidation pressures determined from the compressibility curves

provided us with a measure of sample stiffness, and therefore, a method to approximate the degree of structuration in flocculated FFT. When the test was conducted on a 63-day old 600-ppm sample, the sample showed some signs of developing stiffness. 91-day old 600-ppm sample showed relatively greater stiffness, but still, there was no distinct apparent preconsolidation pressure or yield stress development (Figure 9.1).

Figure 9.1. The changes in compressibility behaviour in the 600-ppm samples with time.

By contrast, the samples dosed with higher dosages such as 1000-ppm samples developed an apparent preconsolidation pressure and showed stiffness from the beginning, starting from day-14. As the 1000-ppm samples aged, because of the structuration effect, they showed a considerable increase in yield stress and stiffness, and reduced compressibility (Figure 9.2). Day-63 and Day-91 1000-ppm samples showed resistance to the external compression forces, especially, during the first few cycles of 24-hour loading,

when they were subjected to nominal effective stresses. Such an increase in yield stress and stiffness and the subsequent changes in compressibility behaviour in the 1000-ppm samples were clear indications of the structuration effect in the flocculated FFT samples.

Figure 9.2. The changes in compressibility behaviour in the 1000-ppm samples with time.

When the compressibility curve obtained from a 35-day old 1000-ppm undisturbed sample was compared to the compressibility curve obtained from the corresponding remoulded sample, there was a significant difference in their compressibility behaviour (Figure 9.3), which was also reflected in the difference in their void ratios at the same effective stress level. The remoulded sample showed substantially higher compressibility. Such a comparison between the undisturbed and remoulded compressibility behaviour clearly demonstrated the effects of structuration on the compressibility and strength development behaviour of the fFFT samples.

Figure 9.3. Different compressibility of undisturbed/remoulded Day-35 1000-ppm samples

When the compressibility curve obtained from the 91-day old 600-ppm undisturbed sample was compared to the corresponding remoulded sample (Figure 9.4), the difference in their compressibility behaviour was not as prominent as it was in the case of 35-day old 1000-ppm samples. Since there was no distinct preconsolidation pressure point on the 600-ppm undisturbed compressibility curve even after 91 days, there was not much of a difference between the 600-ppm undisturbed and remoulded samples in terms of stiffness. Figures 9.3 and 9.4 also showed how the degree/rate of structuration could be different in the FFT samples mixed with different polymer dosages.

Figure 9.4. Different compressibility of undisturbed/remoulded Day-91 600-ppm samples.

Different polymer dosages (in the range of 400 ~ 1000-ppm) resulted in different degrees of structuration effect in the long-term with the higher dosages, i.e., 800-ppm and 1000-ppm showing a relatively prominent structuration effect but also a suboptimal dewaterability. When the effect of structuration was considered, the optimum polymer dosage for the long-term dewatering mechanisms was found to differ from the polymer dosage that produced optimum dewatering performance in the short-term. At the early stage of the experiment, 800-ppm was determined to be the optimum polymer dosage based on the assessment of the short-term key performance indicators (i.e. capillary suction time, 48-hour solids settlement, 48-hour water released, 48-hour net water release). However, in the longer-term, a comparison between the compressibility curves obtained from the 600-ppm, 800-ppm, and 1000-ppm samples (of the same age, i.e., 91-days old) showed that the 600-ppm sample was relatively less stiff and more compressible and had a smaller void ratio at the same effective stress range (0 ~ 100 kPa) (Figure 9.5).

Figure 9.5. The Day-91 undisturbed 600-ppm, 800-ppm, and 1000-ppm compressibility curves showing the variation in the compressibility and strength characteristics of the samples because of different polymer dosages (different polymer dosages resulted in varying degrees of structuration effect in the polymer amended tailings).

Recently, Qi and Simms (2019) proposed an analytical function for estimating the final height of a soft soil/tailings deposit:

$$Final\ height = z + \frac{az[\gamma_s - \gamma_w]^b}{b+1} \dots\dots\dots (9.1)$$

Where $z = \frac{Initial\ height\ (h_0)}{e_0 + 1}$; γ_s = unit weight of solids; and, γ_w = unit weight of water. The parameters a and b can be estimated by fitting the compressibility curve with a power function ($e = a\sigma_v'^b$, where e and σ_v' are void ratio and effective stress respectively).

Now, if we consider a 50-metre tailings deposit and determine the parameters a and b by fitting the Day-91 600-ppm, 800-ppm, and 1000-ppm compressibility curves (shown

in Figure 9.5) with a power function, we can calculate the final height by using the analytical function suggested by Qi and Simms (2019). Different polymer dosages resulted in different final height estimations and final density profiles for the same tailings deposit (Figure 9.6). Both Day-91 800-ppm and Day-91 1000-ppm undisturbed samples resulted in higher void ratios as well as higher final elevations (24.3 m and 23.7 m respectively) owing to their stiffness. By contrast, the Day-91 600-ppm undisturbed sample produced considerably lower void ratios as well as lower final elevation (19.2 m) because of its more compressible nature. Hence, the tailings dosed with lower polymer dosages were found less susceptible to the time-dependent structuration effect, but they displayed the potential for generating better dewatering and consolidation performance in the long-term.

Figure 9.6. Final height estimations and final density profiles of a 50-metre tailings deposit (based on the compressibility curves obtained from the undisturbed fFFT samples).

A fall cone was used to measure the undrained shear strengths developed in the flocculated FFT samples, which also provided a measure of the thixotropic strength gain in the samples. When the measured undrained shear strengths were plotted against the time, the thixotropic strength buildups in the samples were found to vary depending on the polymer dosages. 800-ppm and 1000-ppm samples developed the highest strengths, whereas 600-ppm samples developed a nominal strength (Figure 9.7). Initially, 800-ppm and 1000-ppm samples developed strength at a minimal rate; however, once the rate of initial dewatering or consolidation decreased and the samples started to approach equilibrium in terms of water content, the strength started to increase substantially. A relatively stable condition allowed the fine particles to coalesce and develop stronger interparticle bonding though some additional water was still released from the samples with the development of structuration afterwards. In 1000-ppm samples, the changes in water content were very nominal after Day-42 (Figure 9.8); however, the samples showed significant structuration behaviour. Hence, the change in void ratio or density (due to the dewatering and self-weight consolidation) was thought to be a minor contributing factor in the strength gain of 800-ppm and 1000-ppm samples; rather, it was the thixotropic effect that contributed the most. Since the water content in 600-ppm samples declined uniformly throughout the experiment duration, the strengths developed in 600-ppm samples were also smaller relative to the strengths developed in 800-ppm and 1000-ppm samples.

Figure 9.7. Undrained shear strength development in the flocculated FFT samples.

Figure 9.8. Changes in water content in the flocculated FFT samples.

Field data have shown that the shear strengths of FFT are typically much less than 1 kPa (BGC Engineering Inc., 2010). However, slope stability analyses (performed by other researchers) have suggested that the tailings would require a residual shear strength of at least 20 kPa (Figure 9.9) in order to be sufficiently stable in a gently sloped (6H:1V) tailings deposit so that the deposit could reach a ready-to-reclaim state and also attain a topography similar to the pre-existing natural topography (McKenna et al., 2016). To attain such a level of undrained strength, the tailings would also require reaching a solids content of 70% (equivalent to a gravimetric water content of 43%, which is close to the plastic limit of the tailings) (McKenna et al., 2016). Field trials have shown that polymers allow the dewatering technologies to increase the solids content to at least 45% within 48 hours following deposition (in-line flocculation) and up to 55% (centrifugation) (Matthews et al., 2011; Wells, 2011). However, for adequate densification and shear strength gain of the tailings deposits, the oil sands operators rely on the long-term dewatering mechanisms such as consolidation, evaporation, and freeze-thaw, which work over a longer time frame. The field vane shear strength-void ratio constitutive relationship, which was obtained from the field data indicated that the undrained shear strength of 20 kPa corresponds to a void ratio of 1.5 (Figure 9.10) (Jeeravipoolvarn, 2010). When the final settlement or density profiles were generated for a 50-metre tailings deposit based on the 91-day old 600-ppm, 800-ppm, and 1000-ppm compressibility curves (Figure 9.6), 600-ppm sample produced a final void ratio of less than 1.5 throughout the elevation except for the 4 meters at the top, whereas 800-ppm sample had a final void ratio above 2 kPa. Therefore, for this FFT sample, not only in terms of the final settlement but also in terms of strength development, a polymer dosage of 600-ppm was apparently a better choice than the 800-ppm or 1000-ppm.

Figure 9.9. Limit-equilibrium slope stability analysis for the tailings deposit (from McKenna et al., 2016).

Figure 9.10. Field vane shear strength - void ratio relationship obtained from the field data (from Jeeravipoolvarn, 2010).

Although lower polymer dosages other than the 600-ppm such as 400-ppm showed a dewaterability potential better than the 1000-ppm and comparable to the 600-ppm, the undrained shear strengths developed in the 400-ppm samples were very nominal even after 100 days (~ 0.5 kPa). 400-ppm samples were also found to be too yielding for conducting the consolidation test. Although we do not have the data for the 400-ppm dosage, it is likely that hydraulic conductivity will begin to decrease with the decreasing polymer dosage and affect the performance. Although a very nominal structuration effect in the FFT samples dosed with lower polymer dosages was found to increase the hydraulic conductivity (which in turn helped to accelerate the rate of dewatering and settlement) (Figure 9.11), other factors such as residual shear strength-void ratio relationship should also be considered while determining the optimum polymer dosage for the long-term dewatering of the polymer amended tailings deposits. The variation in polymer dosage has been found to affect the residual shear strength-void ratio constitutive relationship in tailings (Figure 9.12) (Bajwa, 2015).

Figure 9.11. Hydraulic conductivity of the 91-day 600-ppm, 800-ppm, 1000-ppm samples.

Figure 9.12. Undrained shear strength-void ratio relationships of fFFT (Bajwa, 2015).

9.3 Empirical predictors of structuration in flocculated FFT

The study of the structuration effect in polymer amended tailings deposits is especially important since such knowledge bears on the optimization of tailings treatment, deposition, and management plans. But, yet, there exists no consolidation model to describe the structuration effect or predict the full range of consolidation behaviour including structuration in soft soils or deep deposits such as tailings deposits. Although the differences in naturally structured clays and remoulded clays are widely studied, there is little information on the rate of structuration (or restructuration) in literature. Since the performance of the tailings deposits depends on their consolidation, and the structuration effect may change the compressibility of deposited tailings over time, and subsequently, reduce the settlement, it is crucially important that we should be able to predict the degree of structuration in flocculated tailings.

Based on the measurements of short-term key performance indicators for the flocculated FFT samples such as the settlement of the solids in the samples after 48 hours of deposition or the volume of water released from the samples after 48 hours, a simple empirical method was proposed for predicting the thixotropic strength development in a particular tailings-polymer mixture in terms of undrained shear strength, which also approximates the degree of structuration in the flocculated FFT samples. Such knowledge would allow the operators to minimize the negative effects of structuration through an appropriate deposition management method (i.e. layered deposition or use of wick drains to shorten drainage paths) or an appropriate polymer or polymer dosage. Such strategies would also strengthen the industrial efforts in adopting the most effective tailings treatment

and management method, optimizing the long-term dewatering and consolidation performance of the tailings deposits, shortening the time required between the deposition and the reclamation, meeting the current regulatory requirements, minimizing the adverse effects on the environment, and reducing the operational costs.

9.4 Conclusion

Evidently, the flocculated FFT samples that exhibited the highest short-term dewatering potential (i.e. 1000-ppm and 800-ppm) also exhibited the highest degree of structuration in the long-term. Although optimal flocculation and the formation of larger flocs led to a rapid initial dewatering and shorter primary consolidation in 1000-ppm and 800-ppm samples, an increase in strength and stiffness with time impeded the natural creep process or secondary consolidation in those samples and also resulted in a suboptimal dewaterability and settlement performance. By contrast, the samples dosed with relatively lower polymer dosages such as 600-ppm samples developed minimal undrained shear strength and no distinct apparent preconsolidation pressure; however, they showed a considerable and consistent improvement in dewaterability and settleability over time despite showing relatively moderate dewatering and settlement performance at the beginning. Therefore, a lower polymer dosage appeared to be a better choice for producing optimal long-term dewatering and consolidation performance for the FFT deposits.

Chapter 10: Summary and conclusions

10.1 Introduction

For many years, the oil sands industry has been extracting bitumen from the oil sands deposits and playing an important role in the Canadian economy. Owing to an increased demand for energy, the surface mining of oil sands ores has increased manifold, and subsequently, the water-based bitumen extraction process is generating large volumes of tailings. The safe storage and rapid reclamation of these tailings deposits largely depend on the choice of tailings treatment technology, and the subsequent dewatering and consolidation behaviour of the deposited tailings materials. Although some progress has been achieved with the tailings disposal techniques or deposition management strategies and several improved tailings treatment technologies have been developed, the treatment and management of the large volume of tailings in the tailings ponds have remained a major environmental, economic, and social liability. To meet the environmental and safety requirements and minimize the operational costs, it has become a pressing need to improve and advance the current scientific understandings and the prevalent practices of tailings management and overcome the associated challenges, especially, the fundamental problems of slow consolidation and poor dewaterability.

An accurate estimation or prediction of consolidation behaviour is crucial to the proper and sustainable management of tailings during operation, closure, and reclamation. Typically, the large strain consolidation modelling is used to predict the compression behaviour of tailings deposits and determine the final settlement. However, the time-dependent phenomena of creep, thixotropy, and structuration, which the original finite

strain consolidation theory does not consider may change the compressibility and hydraulic conductivity relationships in tailings material over time, which in turn, may produce a final settlement different than the settlement predicted by the model. To incorporate the effects of creep, thixotropy, and structuration into the large strain consolidation theory, reformulate the governing equations, and modify the numerical models, it requires an extensive investigation and assessment of the time-dependent phenomena in FFT.

10.2 Key research objectives

The key objectives of this research project, as described in ‘Chapter 1: Introduction’ were:

- Developing analytical methods to determine the optimum polymer dosage for improving the short-term dewatering and thickening behaviour of fluid fine tailings.
- Investigating the long-term dewatering and consolidation behaviour of polymer amended fluid fine tailings including the time-dependent creep, structuration, and thixotropic effects in flocculated tailings and their implications for the dewatering, consolidation, and strength development performance of tailings deposits.
- Examining the relationship between the changes observed in tailings microstructure during flocculation and over time and the changes observed in macroscopic behaviour of flocculated tailings, i.e., dewaterability, compressibility and strength development.
- Determining the link between the polymer dosage and the time-dependent phenomena in flocculated tailings and suggesting an empirical method for predicting the degree of structuration for a particular tailings-polymer mixture under field conditions.

To address the key objectives, this study investigated the time-dependent effects in polymer amended oil sands tailings such as creep, thixotropy, and structuration, which can

modify the compressibility and strength development behaviour in the long-term, and also examined the influence of an anionic polymer A3338 on the dewaterability, compressibility, and shear strength characteristics of FFT both in the short- and long-term by taking those time-dependent effects into the account from both macroscopic and microstructural points of view. In order to attain a comprehensive understanding of the long-term dewatering and consolidation behaviour of flocculated tailings material, a combination of laboratory experiments was designed and conducted. Different polymer dosages, polymer mixing methods, boundary conditions, and scales of tests were used. Based on an extensive literature review and the findings from the experiments conducted, the time-dependent behaviour of flocculated tailings was described and explained in detail and an empirical method was suggested for predicting the degree of structuration that would result from a particular tailings-polymer mixture.

10.3 Key findings and observations

The key findings of the experimental work and the conclusions drawn from the results are summarised as follows (by chapters):

Chapter 4: The results from the UV-vis spectrophotometry experiments suggested that the UV-vis absorbance-based method could help the development of a process automation system to optimize the polymer dosage for the dewatering of oil sands tailings in real-time, and therefore, could reduce the polymer consumption and minimize the operational costs.

Chapter 5: The torque rheometer experiments demonstrated the potential of the torque rheology-based methods for on-site optimization of polymer dosage for the dewatering of

tailings as the methods were found capable of identifying the best performing dosage based on the measurements of rheological properties of the tailings dosed with different dosages.

Chapter 6: When a combination of lab-scale experiments was conducted on fFFT to investigate the long-term dewatering and consolidation behaviour including the time-dependent creep, thixotropy, and structuration behaviour, a change in volume was observed over time due to the creep. Flocculated tailings developed a thixotropic strength and also a higher preconsolidation pressure and reduced compressibility, which was clearly a structuration effect.

Chapter 7: The results from the similar lab-scale experiments performed on the Leda clay indicated that Leda clay and fFFT showed similar thixotropic and structuration behaviour, which resulted in a similar change in their compressibility and strength characteristics over time and also had implications for their long-term dewatering and consolidation behaviour, though in different degrees. Hence, the structuration effect observed in polymer amended oil sands tailings is not atypical of the structuration effect observed in natural clay deposits.

Chapter 8: Different polymer dosages and mixing protocols were observed to result in different degrees of structuration in fFFT and affect the dewatering and settling behaviour in the long-term significantly. Hence, while evaluating the performances of different polymers or polymer dosages, the magnitude of the resulting structuration effect on the compression behaviour of fFFT should be considered as well. An empirical method was proposed to predict the degree of structuration in a particular tailings-polymer mixture based on the measurements of short-term key performance indicators. Such knowledge would allow the oil sands operators to minimize the negative effects of structuration

through appropriate deposition management plan (i.e. layered deposition or use of wick drains to shorten drainage paths) or an appropriate polymer or polymer dosage.

10.4 Limitations and recommendations for future study

In the view of the findings and the discussions thereof and by taking the limitations of this study into account, the following recommendations are made for further research on the time-dependent behaviour of flocculated oil sands tailings:

- The findings of this study are restricted to the conditions that were investigated in the experiments; hence, they are not necessarily transferable to other situations or settings (different FFT composition/properties, different polymers/polymer dosages, different boundary conditions, etc.) or to the pilot-scale or field-scale testing. Therefore, it is essential to plan and conduct in-situ testing of existing deposits or field trials in order to determine whether the time-dependent phenomena of creep, thixotropy, and structuration observed during the bench-scale laboratory experiments have the same type or degree of influence on other types of oil sands tailings in the fields. The field measurements of compressibility in tailings deposits could help in estimating the magnitude of structuration effects on compressibility with more accuracy and certainty. Since the ultimate solution to the tailings management problem needs to address both legacy volumes and current tailings productions, both types of tailings should be tested.
- An extensive microstructural investigation of the polymer amended tailings is recommended, which should include the visual observations/qualitative interpretations of the images generated from the samples as well as detailed quantitative analysis of

the tailings fabric (i.e. tracking the changes in number, size, shape, and arrangement of the grains/flocs and pores) so that the microstructural features of polymer amended tailings could be correlated to the macroscopic behaviour (i.e. dewaterability, compressibility, and shear strength gain) in both qualitative and quantitative ways.

- In this study, a simple empirical method was proposed for predicting the degree of structuration in polymer amended tailings, which was based on the correlations established between the short-term key performance indicators for flocculated FFT samples (i.e. 48-hour solids settlement, 48-hour net water release, 48-hour water release etc.) and the undrained shear strengths measured from the aged flocculated FFT samples; but since these linear relationships were not very strong, there is still room for improvement. Such linear relationships could be strengthened by including more data points. Conducting long-term column dewatering tests using FFT samples of different compositions and a wider range of polymer dosages could help in generating a comprehensive set of data on the structuration effect in flocculated FFT, which in turn could help in producing a strong linear relationship between the short-term key performance indicators and the undrained shear strengths developed in aged samples.
- The future research on the application of polymers on oil sands tailings for accelerating the dewaterability of the tailings materials should focus on investigating all the phenomena and processes occurring in the tailings ponds including the time-dependent effects as a whole and evaluate their combined effect on the final goal of reclamation of tailings ponds. A combined analysis of flocculation, sedimentation, consolidation,

freeze-thaw, desiccation, and the time-dependent effects is integral to attaining a comprehensive understanding of overall tailings behaviour.

- The oil sands tailings may experience a wide range of temperatures at different stages of production and pipeline transportation and also when they are deposited and stored in the ponds, where they remain exposed to weather conditions for a long period of time. In addition to time and strain rate, the changes in temperature were found to affect the preconsolidation pressure and compressibility of natural clays and the long-term settlement and strength development behaviour as well. An increase in temperature weakens the interparticle bond strength, and subsequently, causes a partial collapse of the soil structure. However, in this study, the experiments were conducted in the laboratory at room temperature ($\pm 20^{\circ}\text{C}$). Hence, the effects of temperature on the long-term dewatering and consolidation behaviour of clay-rich fluid fine tailings including time-dependent structuration behaviour were not accounted for. Future studies should take the effects of temperature on the long-term behaviour of tailings into account.
- Advanced rheological testing is necessary to simulate the pipeline transportation of flocculated tailings and investigate the influence of pipeline length and diameter, and mixing intensity and duration on the time-dependent behaviour of flocculated tailings.
- To deal with the uncertainty in tailings settlement and strength development predictions, the author recommends formulating/reformulating a numerical model based on the findings from this study, available information on structuration in untreated/flocculated FFT, and the known behaviour of clays regarding structuration.

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Appendices

Appendix A

A.1 Excess pore water pressure buildup and dissipation

Figure A.1.1. Excess pore water pressure dissipation of the DI water (slow deposition).

Figure A.1.2. Excess pore water pressure dissipation of the DI water (rapid deposition).

Figure A.1.3. Excess pore water pressure dissipation of the raw FFT (slow deposition).

Figure A.1.4. Excess pore water pressure dissipation of the raw FFT (rapid deposition).

Figure A.1.5. Excess pore water pressure dissipation of the FFT (400-ppm slow deposition)

Figure A.1.6. Excess pore water pressure dissipation of the FFT (400-ppm fast deposition)

Figure A.1.7. Excess pore water pressure dissipation of the FFT (600-ppm slow deposition)

Figure A.1.8. Excess pore water pressure dissipation of the FFT (600-ppm slow deposition)

Figure A.1.9. Excess pore water pressure dissipation of the FFT (600-ppm fast deposition)

Figure A.1.10. Excess pore water pressure dissipation of the FFT: 800-ppm slow deposition

Figure A.1.11. Excess pore water pressure dissipation of the FFT: 800-ppm slow deposition

Figure A.1.12. Excess pore water pressure dissipation of the FFT: 800-ppm fast deposition

Figure A.1.13. Excess pore water pressure dissipation of FFT (1000-ppm slow deposition).

Figure A.1.14. Excess pore water pressure dissipation of FFT (1000-ppm slow deposition).

Figure A.1.15. Excess pore water pressure dissipation of FFT (1000-ppm fast deposition).

A.2 600-ppm double-drainage column dewatering experiment

When the tailings was dosed with a 600-ppm dosage of polymer and deposited into the double-drainage polypropylene column with a pore water pressure sensor installed into the column, the pore water pressure declined rapidly after the deposition and remained at the same value (within a few mm in pressure head) from Day 5 to Day 30 (Figure A.2.1). After 35 days, the pore water pressure began to show negative values, indicating that the bottom filter paper had dried out and some loss of water had occurred through drying or wicking into the drying filter paper. The flocculated tailings in the replicate columns underwent substantial volume change, declining in gravimetric water content from 200 % to 40% (Figure A.2.2).

Figure A.2.1. Pore water pressure (in hPa) at 1.5 cm (600-ppm double-drainage).

Figure A.2.2. Water content measured at 3 different heights (600-ppm double-drainage).

Figure A.2.3. Compression curves from 600-ppm double-drainage Day-28 and 56 samples.

Appendix B

B.1 Geotechnical characterization test results

Liquid limit and Plastic limit

The Standard British fall cone method (British Standard BS 1377) was used for measuring the liquid limit of the unamended fluid fine tailings (British Standards Institution, 1990). The FFT sample was kept in an open environment for a period so that progressive dewatering could take place under the normal atmospheric condition. The water that was accumulated on the top of the solids as well as the water from the cracks were collected with a syringe and a fine needle. After 21 days, when there was no water film remaining on the surface, but the sample was still completely saturated, a series of fall cone tests were performed. For performing the fall cone test, the sample was placed in a 55 mm diameter and 40 mm deep metal specimen cup. The sample had a level surface and there was no air entrapped within the sample. The cone was positioned in a way so that the tip of the cone just touched the sample surface but did not penetrate it. When the cone was released using the release button, it penetrated the sample. The penetration was allowed to continue for 5 seconds and the dial gauge was used to measure the penetration. After each cone penetration, a layer of FFT was collected from the surface and measured for water content. Then, the next fall cone test was performed on a sample with a water content higher than the previous sample. According to the BS 1377 test procedure, the liquid limit is the water content of a sample which allows the cone to penetrate exactly 20 mm during that period of 5 seconds. Since it was not possible to obtain an exactly 20 mm penetration, the test procedure was performed several times over a range of water contents generating

a range of cone penetration depths (15 ~ 25 mm, as recommended by the British Standard for determining the liquid limit). Then, the variation of penetration with water content was plotted in a log-log scale and the best fitting straight line was drawn through the plotted data points. The water content that corresponded to a penetration of 20 mm was identified at 61%; hence, the liquid limit of unamended FFT was determined as 61% (Houlsby, 1982; Leroueil and Le Bihan, 1996; Feng, 2000, 2001, 2004). Based on the linear logarithmic penetration depth versus logarithmic water content model for the determination of plastic limit from the fall cone test results (Feng, 2001), the plastic limit was estimated to be 28%.

Figure B.1.1. Determination of liquid and plastic limit of unamended FFT using fall cone.

Particle size distribution

The particle size distribution of unamended FFT was determined through a combination of wet sieving and sedimentation (hydrometer) analyses. During wet sieving, the particles which passed through the 75-micron sieve (#200 sieve) were collected and dried in an oven. Then, the hydrometer method was used for the quantitative determination of the distribution of particle sizes in that fine-grained portion of FFT (which was finer than the 75-micron sieve and passed through it). The hydrometer method is based on the theory of Stoke's law, which governs the rate of settling of the particle suspended in water. After combining the results of wet sieving and sedimentation (hydrometer) analyses, a complete gradation curve was produced, which showed the percentages of sand, silt and clay in the sample as a percent of the total dry weight (Figure B.1.2).

Figure B.1.2. Particle size distributions of FFT from wet sieve and hydrometer analyses.

Pore water chemistry

The pore water samples, which were collected as bleed water from the unamended FFT, Day-7 fFFT, and Day-42 fFFT samples were sent to an external laboratory (AGAT Laboratories) for the analysis of major cations and anions concentrations, conductivity, pH, total solids and alkalinity. Table B.1.1 shows the results of the pore water chemistry analyses.

Table B.1.1. Pore water chemistry of untreated FFT, Day-7 fFFT, & Day-42 fFFT samples.

Parameter	Unit	Untreated FFT sample	Day-7 fFFT sample	Day-42 fFFT sample
Electrical Conductivity	µS/cm	1590	1610	1660
pH	pH	8.55	8.11	8.23
Total Hardness (As CaCO ₃)	mg/L	79.5	75.2	78
Total Dissolved Solids	mg/L	1050	1080	1110
Total Suspended Solids	mg/L	54	1830	250
Alkalinity (As CaCO ₃)	mg/L	580	487	501
Fluoride	mg/L	3.27	2.86	3.26
Chloride	mg/L	182	171	186

Nitrate as N	mg/L	<0.25	<0.25	<0.25
Sulphate	mg/L	61	142	188
Calcium	mg/L	15.2	14.5	14.1
Magnesium	mg/L	10.1	9.46	10.4
Sodium	mg/L	340	345	360
Potassium	mg/L	14.1	15.7	15.8
Total Iron	mg/L	1.25	2.2	2.95
Total Manganese	mg/L	0.089	0.604	0.089
Iron (Fe ⁺⁺)	mg/L	<0.05	<0.05	<0.05
Iron (Fe ⁺⁺⁺)	mg/L	1.25	2.2	2.95
% Difference/Ion Balance	%	4.14	2.22	4.7

In general, the pore water collected from the fFFT samples showed a slightly higher ionic concentration in comparison to the pore water collected from the unamended FFT sample except for Ca²⁺. The total dissolved solids (TDS) value increased, which was also associated with the increase in the concentrations of the soluble salts including major cations such as sodium (Na⁺) and magnesium (Mg²⁺), and major anions such as sulphate (SO₄²⁻) and chloride (Cl⁻).

Bitumen content (Dean-Stark analysis)

For the current study, an unamended FFT sample was sent to an external laboratory (AGAT Laboratories) to determine the bitumen, solids, and water content of FFT (Dean-Stark analysis). The sample had relatively lower bitumen content (1.41%); however, the solids content (30%) was comparable to the solids content measured from the dry oven test (30% ~ 31%). Results obtained from the Dean-Stark analysis are presented in Table B.1.2.

Table B.1.2. Dean-Stark test data showing the bitumen, solids, and water content of FFT.

Sample	Mass (g)	Hydrocarbons (%)	Water (%)	Solids (%)
Unamended FFT	960.47	1.41	68.45	29.93

Clay content (Methylene blue index test)

Three samples (unamended FFT, Day-7 fFFT, and Day-42 fFFT) were submitted to an external laboratory (AGAT Laboratories) for the methylene blue index (MBI) test. The MBI test is commonly used to measure the clay activity in oil sands ores, froth, and tailings streams (Omotoso, 2011). First, the sample is completely dispersed by using stirring, boiling sonication, and sodium bicarbonate as a dispersing aid. Then, the sample is acidified and titrated with methylene blue (a cationic dye with a strong affinity for the clay surfaces) in 0.5 ~ 1 mL increments. The test comes to an end when a drop of the sample that contains methylene blue is placed on a Whatman filter paper and a permanent blue halo is observed, which indicates that there is an excess of methylene blue (Kaminsky, 2014). Then, the methylene blue index (MBI) is calculated based on the number of

millilitres of methylene blue solution that is added during the test and absorbed a given mass of a sample. The estimated MBI is expressed in meq/100g (milliequivalents of methylene blue per 100 g of sample), which is estimated by multiplying the number of millilitres of methylene blue used to titrate 100 g of a sample by the molarity of methylene blue used (Kaminsky, 2014). Finally, by using the following empirical relationship, which was proposed by Sethi, the percentage of clay in FFT is determined (Yong and Sethi, 1983):

$$\% \text{ Clay} = \frac{MBI \frac{meq}{100g} + 0.04}{0.14}$$

Table B.1.3 shows the results from the MBI test performed on the unamended FFT, Day-7 fFFT, and Day-42 fFFT samples.

Table B.1.3. MBI test data and the percentage of the clay-sized fraction of unamended FFT.

Sample	Mass (g)	Methylene Blue volume (mL)	meq/100g	% clay
Unamended FFT	2.01	6.50	3.93	28.36
fFFT (Day-7)	2.05	9.50	4.63	33.36
fFFT (Day-42)	2.03	13.50	6.65	47.79

Mineralogical composition (X-ray diffraction analysis)

Three samples (unamended FFT, Day-7 fFFT, and Day-42 fFFT) were submitted to an external laboratory (AGAT Laboratories) for the quantitative X-ray diffraction analysis (XRD). Both bulk and clay portions of the samples were analyzed to determine the mineralogical composition. Bulk fraction included >2 micron-sized particles, whereas clay fraction included <2 micron-sized particles. Quantitative phase analyses were done

with the relative intensity ratio (RIR) method. The mineral phases were identified with the International Centre for Diffraction Data (ICDD) PDF-4 Minerals 2018 database in conjunction with the X'Pert HighScore software. The X-ray diffraction data were collected with a Panalytical X-ray diffractometer (XPert-PRO) with a conventional Bragg-Brentano geometry equipped with a copper tube (X-ray source). The equipment was operated at the following settings:

For clay fraction-

Start position [$^{\circ}2\theta$]: 4.0

End position [$^{\circ}2\theta$]: 40.0

Step size [$^{\circ}2\theta$]: 0.03

Scan step time [s]: 0.5

Scan type: Continuous

Generator settings: 40 mA, 45 kV

For bulk fraction-

Start position [$^{\circ}2\theta$]: 4.0

End position [$^{\circ}2\theta$]: 58.0

Step size [$^{\circ}2\theta$]: 0.03

Scan step time [s]: 0.5

Scan type: Continuous

Generator settings: 40 mA, 45 kV

Approximately 3 grams of subsample was taken from each sample for the XRD analysis. Before collecting the subsamples, the samples were made completely

homogenous. Clay fraction was separated from the bulk fraction by using the centrifuging method. Before centrifuging the samples, they were treated in an ultrasonic bath using sodium hexametaphosphate as a deflocculating agent that facilitates the complete disintegration of the matrix from the grains and thus helps to fractionate. The centrifugation method was accomplished in two separate steps. In the first step, the sample was centrifuged at 600 rpm for 5 minutes, which allowed the coarser particle to settle down at the bottom of the tube. However, the clay-sized particles were still there in the fluid in the suspension. Then, they were carefully transferred to another tube. After centrifuging this suspension at 3000 rpm for 20 minutes, the clay-sized particles were collected. This clay fraction was then placed onto a glass slide and dried. Afterwards, the slide was placed in a glycol vapour bath for 24 hours with a goal of identifying the expandable clay minerals, and then, they were separated. A separate portion of clay fraction was placed in diluted hydrochloric acid (HCl) for 24 hours and acidized. Weight fractions were measured for both bulk and clay portions of the samples before analyzing them with the X-ray diffractometer. The three clay fractions were analyzed between 4° to 40° 2θ (Theta) and one bulk fraction was analyzed between 4° to 58° 2θ (Theta). Since the XRD analysis is semi-quantitative (approximately 10% at best), semi-quantitative amounts of clay minerals in the $< 2 \mu\text{m}$ size fraction were determined based on the calculation of basal-peak areas.

The non-clay minerals and the associated weight fractions, and the clay minerals ($< 2 \mu\text{m}$) and the associated weight fractions, which were identified and determined by the XRD analysis are summarized in Table B.1.4 and Table B.1.5 respectively.

Table B.1.4. Non-clay minerals in the FFT samples (from the XRD analysis).

Sample	Type of analysis	Weight %	Quartz	P-Feldspar	K-Feldspar	Pyrite	Muscovite	Siderite	Total Non-clay
Unamended FFT sample	Bulk fraction	85.38	40	1	2	2	1	4	51
	Clay fraction	14.62	4	0	0	0	0	1	5
	Bulk and clay	100.00	34	1	2	2	1	4	44
Day-7 polymer-amended sample	Bulk fraction	84.33	40	1	2	2	1	4	50
	Clay fraction	15.67	3	0	0	0	0	0	3
	Bulk and clay	100.00	34	1	1	2	1	3	43
Day-42 polymer-amended sample	Bulk fraction	85.26	35	1	2	2	2	4	46
	Clay fraction	14.74	4	0	0	0	0	1	5
	Bulk and clay	100.00	29	1	2	2	2	4	39

Table B.1.5. Clay minerals in the FFT samples (from the XRD analysis).

Sample	Type of analysis	Weight %	← Kaolinite	Chlorite	Clays Illite	Mixed-layer clays	Smectite →	Total clay
Unamended FFT sample	Bulk fraction	85.38	36	0	14	0	0	50
	Clay fraction	14.62	70	0	23	2	0	95
	Bulk and clay	100.00	41	0	15	TR	0	56
Day-7 polymer-amended sample	Bulk fraction	84.33	36	0	14	0	0	50
	Clay fraction	15.67	71	0	26	0	0	97
	Bulk and clay	100.00	42	0	16	0	0	58
Day-42 polymer-amended sample	Bulk fraction	85.26	43	0	11	0	0	54
	Clay fraction	14.74	69	0	26	0	0	95
	Bulk and clay	100.00	47	0	13	0	0	60

The combined bulk and clay XRD analysis for the unamended FFT sample indicated that the sample consists mainly of kaolinite (41%) [aluminum silicate hydroxide,

$\text{Al}_4\text{Si}_4\text{O}_{10}(\text{OH})_8$] and quartz (34%) [silicon dioxide, SiO_2] with some illite (15%) [potassium aluminum silicate hydroxide, $\text{KAl}_2(\text{OH})_2(\text{AlSi}_3(\text{O}, \text{OH})_{10})$] and a little amount of siderite (4%) [iron carbonate, FeCO_3] (Table B.1.4 and Table B.1.5). Trace amount of mixed-layer clays [illite-smectite, $\text{KAl}_2(\text{OH})_2(\text{AlSi}_3(\text{O}, \text{OH})_{10} - (\text{Ca}, \text{Na})_{0.7}(\text{Al}, \text{Mg}, \text{Fe})_4(\text{Si}, \text{Al})_8\text{O}_{20}(\text{OH})_4 \cdot n\text{H}_2\text{O})$] was identified in the unamended fluid fine tailings sample, but not in the polymer amended samples. The XRD analysis for the clay fraction of the unamended FFT sample indicates that it consists predominantly of kaolinite (70%) with lesser amounts of illite (23%), and a small amount of quartz (4%).

When the XRD analysis was conducted on Day-7 and Day-42 fFFT samples, some minor changes were detected in the relative amounts of the minerals. For example, when compared to the unamended FFT sample, the combined bulk and clay XRD analysis of the Day-7 sample indicated that kaolinite and quartz are still the main components (42% and 34% respectively) with very minor or no change in their amounts (they were 41% and 34% respectively in the unamended FFT), with the almost same amounts of illite (16%) and siderite (3%) as the unamended FFT sample (15% and 4% respectively). In the case of clay fraction, in a similar manner, the amounts of kaolinite and quartz (71% and 3% respectively) did not change significantly from their amounts in the unamended FFT (70% and 4% respectively). However, there was a slight increase in the amount of illite (increased to 26% from 23%) and there were no mixed-layer clays (illite-smectite). Overall, by weight, the clay fraction was 15.67% of the total weight; so, there was a small increase in the clay fraction as it was 14.62% in the unamended FFT.

The combined bulk and clay XRD analysis of the Day-42 fFFT sample indicated that there were some significant changes in the clay mineralogy over time. The percentage of total clay in the combined bulk and clay XRD analysis increased (56% → 58% → 60%), and the percentage of total non-clay decreased (44% → 43% → 39%). As the clay-rich fFFT samples displayed substantial dewatering and volume change behaviour with time, the clay minerals, especially, the kaolinite increased (41% → 42% → 47%), thereby making kaolinite the increasingly dominant clay mineral in fFFT. By contrast, the non-clay minerals such as quartz decreased (34% → 34% → 29%). If we only consider the amount of kaolinite or illite in the clay fraction, the changes were nominal (70% → 71% → 69% and 23% → 26% → 26% respectively); however, the amount of kaolinite in the bulk fraction increased significantly (from 36% to 43%), which contributed to the overall increase of kaolinite in the combined bulk and clay fraction (from 41 to 47%).

Appendix C

C.1 Pore size distribution data (600-ppm single-drainage column experiment)

Figure C.1.1. Pore size distribution data obtained from 600-ppm single-drainage samples.

Figure C.1.2. Change in threshold diameter with time in 600-ppm single-drainage samples.

Figure C.1.3. Log differential pore volume distribution (600-ppm single-drainage samples)

Figure C.1.4. The pore size distribution of Day-1 600-ppm single-drainage sample.

Figure C.1.5. The pore size distribution of Day-7 600-ppm single-drainage sample.

Figure C.1.6. The pore size distribution of Day-14 600-ppm single-drainage sample.

Figure C.1.7. The pore size distribution of Day-28 600-ppm single-drainage sample.

Figure C.1.8. The pore size distribution of Day-56 600-ppm single-drainage sample.

Figure C.1.9. The pore size distribution of Day-72 600-ppm single-drainage sample.

Figure C.1.10. Log differential pore volume distribution of Day-1 600-ppm single-drainage

Figure C.1.11. Log differential pore volume distribution of Day-7 600-ppm single-drainage

Figure C.1.12. Log differential pore volume distribution - Day-14 600-ppm single-drainage

Figure C.1.13. Log differential pore volume distribution - Day-28 600-ppm single-drainage

Figure C.1.14. Log differential pore volume distribution - Day-56 600-ppm single-drainage

Figure C.1.15. Log differential pore volume distribution - Day-72 600-ppm single-drainage

C.2 Pore size distribution data (800-ppm single-drainage column experiment)

Figure C.2.1. Pore size distribution data obtained from 800-ppm single-drainage samples.

Figure C.2.2. Change in threshold diameter with time in 800-ppm single-drainage samples.

Figure C.2.3. Log differential pore volume distribution (800-ppm single-drainage samples)

Figure C.2.4. The pore size distribution of Day-1 800-ppm single-drainage sample.

Figure C.2.5. The pore size distribution of Day-14 800-ppm single-drainage sample.

Figure C.2.6. The pore size distribution of Day-42 800-ppm single-drainage sample.

Figure C.2.7. The pore size distribution of Day-56 800-ppm single-drainage sample.

Figure C.2.8. The pore size distribution of Day-70 800-ppm single-drainage sample.

Figure C.2.9. The pore size distribution of Day-98 800-ppm single-drainage sample.

Figure C.2.10. Log differential pore volume distribution of Day-1 800-ppm single-drainage

Figure C.2.11. Log differential pore volume distribution of Day-14 800-ppm single-drainage.

Figure C.2.12. Log differential pore volume distribution of Day-42 800-ppm single-drainage.

Figure C.2.13. Log differential pore volume distribution of Day-56 800-ppm single-drainage.

Figure C.2.14. Log differential pore volume distribution of Day-70 800-ppm single-drainage.

Figure C.2.15. Log differential pore volume distribution of Day-98 800-ppm single-drainage.

C.3 Pore size distribution data (800-ppm double-drainage column experiment)

Figure C.3.1. Pore size distribution data obtained from 800-ppm double-drainage samples.

Figure C.3.2. Change in threshold diameter with time in 800-ppm double-drainage samples

Figure C.3.3. Log differential pore volume distribution: 800-ppm double-drainage samples

Figure C.3.4. The pore size distribution of Day-1 800-ppm double-drainage sample.

Figure C.3.5. The pore size distribution of Day-7 800-ppm double-drainage sample.

Figure C.3.6. The pore size distribution of Day-14 800-ppm double-drainage sample.

Figure C.3.7. The pore size distribution of Day-28 800-ppm double-drainage sample.

Figure C.3.8. The pore size distribution of Day-42 800-ppm double-drainage sample.

Figure C.3.9. The pore size distribution of Day-56 800-ppm double-drainage sample.

Figure C.3.10. Log differential pore volume distribution of Day-1 800-ppm double-drainage.

Figure C.3.11. Log differential pore volume distribution of Day-7 800-ppm double-drainage.

Figure C.3.12. Log differential pore volume distribution of Day-14 800-ppm double-drainage.

Figure C.3.13. Log differential pore volume distribution of Day-28 800-ppm double-drainage.

Figure C.3.14. Log differential pore volume distribution of Day-42 800-ppm double-drainage.

Figure C.3.15. Log differential pore volume distribution of Day-56 800-ppm double-drainage.

C.4 Scanning electron microscopy images

600-ppm single-drainage sample

Figure C.4.1. Scanning electron micrograph of Day-7 600-ppm single-drainage sample.

Figure C.4.2. Scanning electron micrograph of Day-14 600-ppm single-drainage sample.

Figure C.4.3. Scanning electron micrograph of Day-28 600-ppm single-drainage sample.

Figure C.4.4. Scanning electron micrograph of Day-72 600-ppm single-drainage sample.

600-ppm double-drainage sample

Figure C.4.5. Scanning electron micrograph of Day-1 600-ppm double-drainage sample.

Figure C.4.6. Scanning electron micrograph of Day-7 600-ppm double-drainage sample.

Figure C.4.7. Scanning electron micrograph of Day-21 600-ppm double-drainage sample.

Figure C.4.8. Scanning electron micrograph of Day-35 600-ppm double-drainage sample.

800-ppm single-drainage sample

Figure C.4.9. Scanning electron micrograph of Day-1 800-ppm single-drainage sample.

Figure C.4.10. Scanning electron micrograph of Day-7 800-ppm single-drainage sample.

Figure C.4.11. Scanning electron micrograph of Day-47 800-ppm single-drainage sample.

Figure C.4.12. Scanning electron micrograph of Day-70 800-ppm single-drainage sample.

800-ppm double-drainage sample

Figure C.4.13. Scanning electron micrograph of Day-1 800-ppm double-drainage sample.

Figure C.4.14. Scanning electron micrograph of Day-7 800-ppm double-drainage sample.

Figure C.4.15. Scanning electron micrograph of Day-47 800-ppm double-drainage sample.

Figure C.4.16. Scanning electron micrograph of Day-60 800-ppm double-drainage sample.

Figure C.4.17. Scanning electron micrograph of Day-84 800-ppm double-drainage sample.

Large column (800-ppm single-drainage sample)

Figure C.4.18. Scanning electron micrograph of the large column single-drainage sample (800-ppm), which was obtained from the 0 cm height of the solids.

Figure C.4.19. Scanning electron micrograph of the large column single-drainage sample (800-ppm), which was obtained from the 15 cm height of the solids.

Figure C.4.20. Scanning electron micrograph of the large column single-drainage sample (800-ppm), which was obtained from the 30 cm height of the solids.

600-ppm (NAIT) single-drainage sample

Figure C.4.21. Scanning electron micrograph of Day-3 600-ppm (NAIT) sample.

Figure C.4.22. Scanning electron micrograph of Day-7 600-ppm (NAIT) sample.

Figure C.4.23. Scanning electron micrograph of Day-35 600-ppm (NAIT) sample.

Figure C.4.24. Scanning electron micrograph of Day-91 600-ppm (NAIT) sample.

800-ppm (NAIT) single-drainage sample

Figure C.4.25. Scanning electron micrograph of Day-3 800-ppm (NAIT) sample.

Figure C.4.26. Scanning electron micrograph of Day-7 800-ppm (NAIT) sample.

Figure C.4.27. Scanning electron micrograph of Day-35 800-ppm (NAIT) sample.

Figure C.4.28. Scanning electron micrograph of Day-91 800-ppm (NAIT) sample.

1000-ppm single-drainage sample

Figure C.4.29. Scanning electron micrograph of Day-3 1000-ppm sample.

Figure C.4.30. Scanning electron micrograph of Day-7 1000-ppm sample.

Figure C.4.31. Scanning electron micrograph of Day-35 1000-ppm sample.

Figure C.4.32. Scanning electron micrograph of Day-91 1000-ppm sample.

High-vacuum scanning electron microscopy

800-ppm single-drainage sample

Figure C.4.33. SEM images (high vacuum SEM) from the 7, 21, 35, 56, 70, and 91-day freeze-dried 800-ppm single-drainage samples (500X magnified) (clockwise).

Figure C.4.34. SEM images (high vacuum SEM) from the 7, 21, 56, 70, 91, and 98-day freeze-dried 800-ppm single-drainage samples (1000X magnified) (clockwise).

800-ppm double-drainage sample

Figure C.4.35. SEM images (high vacuum SEM) from the 7, 21, 28, 49, 63, and 91-day freeze-dried 800-ppm double-drainage samples (200X magnified) (clockwise).

Figure C.4.36. SEM images (high vacuum SEM) from the 1, 14, 28, 56, 77, and 98-day freeze-dried 800-ppm double-drainage samples (500X magnified) (clockwise).

Figure C.4.37. SEM images (high vacuum SEM) from the 1, 14, 28, 49, 63, and 91-day freeze-dried 800-ppm double-drainage samples (1000X magnified) (clockwise).

Leda clay sample

Figure C.4.38. Scanning electron micrograph of Day-1 Leda clay sample (1st batch).

Figure C.4.39. Scanning electron micrograph of Day-7 Leda clay sample (1st batch).

Figure C.4.40. Scanning electron micrograph of Day-14 Leda clay sample (1st batch).

Figure C.4.41. Scanning electron micrograph of Day-42 Leda clay sample (1st batch).

Figure C.4.42. Scanning electron micrograph of Day-3 Leda clay sample (2nd batch).

Figure C.4.43. Scanning electron micrograph of Day-7 Leda clay sample (2nd batch).

Figure C.4.44. Scanning electron micrograph of Day-35 Leda clay sample (2nd batch).

Figure C.4.45. Scanning electron micrograph of Day-91 Leda clay sample (2nd batch).

C.5 Optical microscopy images

600-ppm single-drainage sample

Figure C.5.1. Optical microscopy image of 1-hour 600-ppm single-drainage sample.

Figure C.5.2. Optical microscopy image of 2-hour 600-ppm single-drainage sample.

Figure C.5.3. Optical microscopy image of 4-hour 600-ppm single-drainage sample.

Figure C.5.4. Optical microscopy image of 8-hour 600 ppm-single-drainage sample.

Figure C.5.5. Optical microscopy image of 24-hour 600 ppm single drainage sample.

Figure C.5.6. Optical microscopy image of 48-hour 600-ppm single-drainage sample.

800-ppm single-drainage sample

Figure C.5.7. Optical microscopy image of 1-hour 800-ppm single-drainage sample.

Figure C.5.8. Optical microscopy image of 12-hour 800-ppm single-drainage sample.

Figure C.5.9. Optical microscopy image of 24-hour 800-ppm single-drainage sample.

Figure C.5.10. Optical microscopy image of 48-hour 800-ppm single-drainage sample.

200-ppm single-drainage sample

Figure C.5.11. Optical microscopy images of the 200-ppm single-drainage sample (0~24h).

400-ppm single-drainage

Figure C.5.12. Optical microscopy images of the 400-ppm single-drainage sample (0~24h).