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**MEASUREMENT OF THE AMORPHOUS
COHERENT SCATTER FORM FACTOR
BY USING AN X-RAY POWDER DIFFRACTOMETER**

by

Matthew P. Wismayer
B.Sc. (York University) 1998

A thesis submitted to the
Faculty of Graduate Studies and Research
in partial fulfillment of the requirements
for the degree of
Master of Science

Ottawa-Carleton Institute for Physics
Department of Physics
Carleton University
September 27, 2001

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Submitted by

Matthew P. Wismayer, B.Sc.

in partial fulfillment of the requirements
for the degree of Master of Science

Thesis Supervisor

Chair, Department of Physics

Date: 9 November 2001.

Carleton University

Abstract

A method has been developed to measure the amorphous coherent scatter form factor using an x-ray powder diffractometer. The method consists of four parts: (i). Low angle background was measured using a specially designed sample holder which absorbs the photons that scatter off the sample holder cavity. This allows one to measure the intensity of the incident x-ray beam that passes directly to the detector. (ii). Polarization and beam broadening and/or machine misalignment were characterized by extracting the incoherent signal from the diffraction pattern of powdered graphite. A correction factor $\Lambda(\theta)$ was determined by dividing the incoherent signal by the calculated incoherent cross section. (iii). Diffraction patterns for $2-150^\circ$ were measured for the samples. (iv). The scattering data were corrected for background, polarization and beam broadening and/or machine misalignment. The coherent scatter form factor was extracted by normalizing the high θ data to the free atom form factors. The method was evaluated for water and plastics by taking the form factor ratio from two different machines, at different energies. Over the range $0.102 < x < 5.39 \text{ nm}^{-1}$ where $x = \lambda^{-1} \sin \theta / 2$ the average form factor ratio for water was 0.928.

Table of Contents

Acceptance Sheet	ii
Abstract	iii
Table of Contents	iv
List of Tables	viii
List of Figures	ix
List of Symbols and Constants	xiv

Chapter 1: Introduction

1.1 Conventional X-ray Imaging	1
1.2 Digital Imaging	6
1.3 CT Imaging	7
1.4 Scatter Imaging	9
1.4.1 Previous Investigators	10
1.5 Coherent Scatter Form Factor	13
1.5.1 Form factors for Scatter Imaging	15
1.5.2 Previous Investigators	15
1.6 Thesis Objective and Outline	19
1.6.1 Objective	19
1.6.2 Outline	19

Chapter 2: Theory

2.1 Radiation Interactions	20
2.1.1 Photoelectric Effect	21
2.1.2 Incoherent Scatter	22

2.1.2.1	Single Electron	22
2.1.2.2	Single Atom	23
2.1.2.3	Compound or Mixture	24
2.1.3	Coherent Scatter	25
2.1.3.1	Single Electron	25
2.1.3.2	Single Atom	27
2.1.4	Total Scatter	31
2.2	Inter-Molecular scattering	32
2.2.1	Crystal Diffraction	33
2.2.2	Amorphous Diffraction	34

Chapter 3: Apparatus

3.1	Rigaku Powder Diffractometer	36
3.1.1	Rigaku Specifications	39
3.2	Scintag Powder Diffractometer	40
3.2.1	Scintag Specifications	43
3.3	Sample Holders	44
3.3.1	Type I Sample Holder	44
3.3.2	Type II, III Sample Holders	45
3.3.3	Type IV, V Sample Holders	46
3.3.4	Type VI Insert	47
3.4	Samples	49

Chapter 4: Experiments and Results

4.1	Scattering Model	50
4.1.1	Sample Scatter N_s	50
4.1.2	Monochromator Polarization	52
4.1.3	Background Radiation	53

4.2	Diffraction Experiments	56
4.2.1	X-ray Tube Settings	56
4.2.2	Diffractometer Alignment	57
4.2.3	Background Measurements	60
4.2.3.1	External Radiation N_e	60
4.2.3.2	Sample Chamber Scatter	61
4.2.3.3	Sample Holder Background	63
4.2.3.4	Type VI Background	66
4.2.4	Correction Measurements	68
4.2.5	Water Measurements	72
4.2.6	Tissue Measurements	76
4.2.7	Plastic Measurements	77

Chapter 5: Calculations

5.1	Background Extraction	81
5.2	Corrections	83
5.3	Coherent Form Factor Extraction	88
5.3.1	Coherent Scatter Form Factor: Water	91
5.3.2	Coherent Scatter Form Factor: Beef Muscle	94
5.3.3	Coherent Scatter Form Factor: Plastics	95

Chapter 6: Comparisons and Conclusions

6.1	Rigaku and Scintag Comparison	98
6.2	Comparison to the Literature	100
6.2.1	Coherent Scatter Form Factor Comparison: Water and Beef samples ..	100
6.2.2	Coherent Scatter Form Factor Comparison: Plastic samples	106

6.3	Concluding Remarks and Future Work	112
6.3.1	Concluding Remarks	112
6.3.2	Future Work	113
6.3.2.1	Slit Configuration	114
6.3.2.2	Low Angle Diffractometer	114
6.3.2.3	Synchrotron Diffractometer	114
References		115

List of Tables

Table	Caption	Page
1	Attenuation coefficients for 60 keV photons	3
2	A summary of all sample holders used in the experiment	44
3	Samples scanned by the x-ray diffractometer	49
4	Characteristic K lines for the cobalt spectrum	56
5	Characteristic K lines for the copper spectrum	56
6	Empty chamber to holder ratio for both the Al and St holders	64
7	Fit parameters for the Rigaku data clusters	85
8	Fit parameters for the Scintag data clusters	85
9	Rigaku water and muscle form factors compared to the published literature	101
10	Scintag water and muscle form factors compared to the published literature	101
11	Rigaku plastic form factors compared to the published literature	106
12	Scintag plastic form factors compared to the published literature	107

List of Figures

Figure	Caption	Page
1	Primary radiological imaging	2
2	Projection imaging model	3
3	Use of a parallel grid to reduce x-ray scatter	5
4	First generation CT imaging	8
5	Differential scattering cross sections of liquid water for 60 keV photons	10
6	X-ray diffraction computed tomography, as invented by Harding	13
7	Contour plot of x as a function of photon energy $h\nu$ and scattering angle θ . Variable x ranges from $0.001 - 100 \text{ nm}^{-1}$..	14
8	Carbon interaction cross sections per atom	20
9	Incoherent form factor per atom for hydrogen, carbon and oxygen	24
10	Model for Thomson scatter	26
11	Geometric model for atomic scattering	28
12	Resultant vector $\mathbf{k}_0' - \mathbf{k}_0$	29
13	Coherent scatter form factors for hydrogen, carbon and oxygen	31
14	Cubic unit cell of repetition where $a_x = a_y = a_z$	33
15	Diffraction scan of iodized NaCl. Measured using a Rigaku powder diffractometer as described in Chp.3	34
16	Diffraction scan of pure ethanol. Measured using a Rigaku powder diffractometer as described in Chp.3	35

17	In this photograph the Rigaku diffractometer is shown. The x-ray tube is on the right and the monochromator and detector are on the left	37
18	Technical diagram of Rigaku XPD	37
19	Monochromator energy window (solid lines) superimposed on the cobalt K-fluorescence lines (dashed lines)	38
20	In this photograph, the Scintag diffractometer is shown. The x-ray tube is on the left and the monochromator and detector are on the right	41
21	Technical setup for Scintag XPD	41
22	Monochromator energy window (solid lines) superimposed on the copper K- fluorescence lines (dashed lines)	42
23	Type I holders Rigaku (a) and Scintag (b) slit blocks	45
24	(a) Type II holder, (b) Type III holder	46
25	(a) Type IV holder, (b) Type V holder	47
26	Type VI insert (a) Rigaku, (b) Scintag, (c) Slat spacing for both Scintag and Rigaku holder	48
27	Scattering model to determine the form of N_s	50
28	Scattering geometry for monochromator	53
29	The signal components for a sample and holder	54
30	The signal components for an empty sample holder	55
31	The signal components for type VI insert	55
32	Alignment peak for the Rigaku x-ray tube	58
33	Alignment peak for the Rigaku detector	59
34	Alignment peak for the Scintag x-ray tube	59
35	Alignment peak for the Scintag detector	60

36	Rigaku measurement of N_e	61
37	Rigaku measurement of empty chamber	62
38	Scintag measurement of empty chamber	62
39	Rigaku N_i measurements of the empty chamber and Al type III, IV, V sample holders, measured as in Figure 30 p.55.	63
40	Rigaku N_i measurements of the empty chamber and St type III, IV, V sample holders	64
41	Scintag N_i measurements of the empty chamber and Al type V sample holder	65
42	Rigaku N_i measurement of type VI insert, measured as in Figure 31 p.55	66
43	Rigaku log-log plot of Al V and type VI background	67
44	Scintag N_i measurement of type VI insert, measured as in Figure 31 p.55	67
45	Scintag log-log plot of Al V and type VI background	68
46	Rigaku low angle comparison of NaCl, SiO ₂ and graphite measured as in Figure 29, p. 54	70
47	Rigaku graphite diffraction pattern, N_i	70
48	Scintag N_i comparison of NaCl, SiO ₂ and graphite, measured as in Figure 29, p. 54	71
49	Scintag graphite diffraction pattern, N_i	71
50	Rigaku water volume optimization	73
51	Rigaku distilled water diffraction pattern N_i	74
52	Scintag water volume optimization	75
53	Scintag distilled water diffraction pattern, N_i	75
54	Rigaku beef muscle diffraction pattern, N_i	76

55	Rigaku polystyrene diffraction pattern, N_t	77
56	Rigaku Lexan diffraction pattern, N_t	78
57	Rigaku Nylon diffraction pattern, N_t	78
58	Rigaku polyethylene diffraction pattern, N_t	79
59	Rigaku PMMA diffraction pattern, N_t	79
60	Scintag polystyrene diffraction pattern, N_t	80
61	Scintag PMMA diffraction pattern, N_t	80
62	Rigaku – These are the data set of Chp. 4. Figure 42 p.66 with the peaks removed and 11-point smoothed	82
63	Scintag – These are the data set of Chp.4 Figure 44 p.67 with the peaks removed and 11-point smoothed	82
64	Rigaku N_{sg} data clusters – This is the data of Figure 47 p.70 with the background and peaks removed	84
65	Scintag N_{sg} data clusters – This is the data of Figure 49 p.71 with The background and peaks removed	84
66	The polynomials f_1 , f_2 and f_3 superimposed on the Rigaku graphite clusters	86
67	The polynomials f_1 , f_2 and f_3 superimposed on the Scintag graphite clusters	86
68	Rigaku correction curves for f_1 , f_2 and f_3 polynomials	87
69	Scintag correction curves for f_1 , f_2 and f_3 polynomials	88
70	Flow chart of coherent scatter form factor extraction	90
71	Rigaku coherent scatter form factor for water using the f_1 , f_2 and f_3 polynomials	92
72	Scintag coherent scatter form factor for water using the f_1 , f_2 and f_3 polynomials	92
73	Rigaku coherent scatter form factor for Beef muscle	94

74	Rigaku and Scintag coherent scatter form factor for polystyrene	95
75	Rigaku and Scintag coherent scatter form factor for PMMA	96
76	Rigaku coherent scatter form factor for polyethylene	96
77	Rigaku coherent scatter form factor for nylon	97
78	Rigaku coherent scatter form factor for lexan	97
79	Water form factor ratio of Rigaku to Scintag	99
80	PMMA and polystyrene form factor ratios of Rigaku to Scintag	100
81	Rigaku and Scintag coherent scatter form factors for water compared to Narten and Kosanetzky	104
82	Rigaku and Scintag coherent scatter form factors for water compared to Peplow	104
83	Rigaku coherent scatter form factor for beef muscle compared to Kosanetzky and Peplow	106
84	Rigaku and Scintag coherent scatter form factors for PMMA compared to Kosanetzky and Peplow	108
85	Rigaku and Scintag coherent scatter form factors for polystyrene compared Kosanetzky	109
86	Rigaku coherent scatter form factors for polyethylene compared to Kosanetzky	110
87	Rigaku coherent scatter form factors for Nylon compared to Kosanetzky	111
88	Rigaku coherent scatter form factors for lexan compared to Harding and Peplow	111

List of Symbols and Constants

Symbol	Description
α	$h\nu_0/m_0c^2$
A_0	Cross sectional area of the incident beam
$C(\Phi)$	Image Contrast
c	Speed of light = 2.998×10^8 m/s
δ	Monochromator tilt angle
θ	Photon scattering angle
e	Electron charge unit = 1.602×10^{-19} C
$E(\mathbf{R},t)$	Electromagnetic wave
E_i	Binding energy of the i th shell
$F(x)$	Coherent scatter form factor
$F_{IAM}(x)$	IAM form factor
$F_{KN}(\alpha,\theta)$	Klein-Nishina function
K	Normalization constant K
k	Propagation constant
k	Coulomb force constant $1/4\pi\epsilon_0 = 8.988 \times 10^9$ N•m ² /C
h	Planck's constant = 6.626×10^{-34} J•s
$h\nu_0$	Incident photon energy
$h\nu$	Scattered photon energy

M_i	Mass fraction of the i th species
m_o	Electron rest mass = 9.109×10^{-31} kg
N_t	Total signal measured in counts
N_s	Total scattered signal
N_b	Total background signal
N_e	External signal
N_h	Sample holder signal
N_f	Fringe signal
N_o	Incident scatter signal
n_o	Number density of scattering centres
$P(\theta, \phi, \delta)$	Polarization function
r_o	Classical electron radius, $ke^2/m_o c = 2.818 \times 10^{-15}$ m
$S(x)$	Incoherent scattering form factor
S_i	Total number of photons scattered in the primary beam
$\Lambda(x)$	Correction function
$\lambda_o(x)$	Projection set
λ	Wavelength of radiation
$\lambda^{-1} \sin \theta/2$	x parameter
μ	Linear attenuation coefficient
ν	Frequency of photon
$\rho(r)$	Charge distribution function
ρ_e	Electron density

τ_{photo}	Photoelectric interaction cross section
Φ	Total x-ray fluence
Φ_t	Target x-ray fluence
Φ_b	Background x-ray fluence
ϕ	Detector-scattered beam angle
σ_{coh}	Coherent interaction cross section
σ_{incoh}	Incoherent interaction cross section
$d\sigma_t/d\Omega$	Total differential cross section per unit solid angle
$d\sigma_{\text{incoh}}/d\Omega$	Incoherent differential cross section per unit solid angle
$d\sigma_{\text{coh}}/d\Omega$	Coherent differential cross section per unit solid angle
$\psi(r)$	Atomic wave function
$d\Omega_o$	Detector solid angle element

Chapter 1

Introduction

This chapter provides an overview of the modalities of x-ray imaging: conventional, digital and computerized axial tomography (CAT). Each of these modalities uses primary beam transmission to obtain diagnostic information. Furthermore this chapter will show that the scattered component of the primary beam can be used to provide imaging information. It will be shown that to model a scatter imaging system one must measure the coherent scatter form factor.

1.1 Conventional X-ray Imaging

Conventional x-ray imaging is based on the principle of shadow projection. Figure 1 is a schematic of an x-ray imaging system. A tungsten anode x-ray tube, typically operating at 100 kVp, produces a polyenergetic photon beam with energies between 20-100 keV. The beam is collimated onto the patient by a primary diaphragm. When the x-ray beam enters the patient the photons will be attenuated by air, blood, other liquids, tissue and bone. The photons exiting the patient expose a cassette film. By analyzing the intensity of the exposed film one can determine that bone attenuates x rays more readily than does blood or tissue. This intensity is plotted in Figure 1. The image produced by the x-ray film represents the shadow projection of the dense objects in the body.

Figure 1. Primary radiological imaging [Ref.1 Chp.16. p.559].

A physical model for x-ray imaging is shown in Figure 2. An x-ray beam with photon fluence Φ_0 and photon energy $h\nu_0$ is collimated onto a target object which is embedded in a test phantom. When the beam passes through the phantom or target, photons will be removed from the incident fluence by any one of the following interactions: coherent scatter, incoherent scatter and the photoelectric effect. The probability for any one of these interactions to occur depends on the total linear attenuation coefficient, μ . The value of μ depends on the energy of the photon and the atomic number of the phantom or target. Table 1 lists values of μ for 60 keV photons.

Figure 2. Projection imaging model.

Material	Linear Attenuation coefficient (cm^{-1})
Air	1.86×10^{-2}
Bone	0.480
Muscle	0.180
Blood	0.178

Table 1. Attenuation coefficients for 60 keV photons [Ref.1 App.A].

The fluence exiting the phantom takes the form,

$$\Phi = \Phi_t + \Phi_b, \quad (1)$$

where $\Phi_t = \Phi_{tp} + \Phi_{ts}$ and $\Phi_b = \Phi_{bp} + \Phi_{bs}$ represent the target and background fluences respectively. The components Φ_{tp} , Φ_{ts} and Φ_{bp} , Φ_{bs} represent the primary and scattered fluence of the target and background respectively. The primary target fluence, Φ_{tp} , is attenuated by both target and background.

$$\Phi_{tp} = \Phi_o \exp(-\mu_t D) \exp[-\mu_b (L - D)], \quad (2)$$

where μ_t and μ_b are the attenuation coefficients of the target and background medium respectively. The dimensions D and L are the thicknesses of the target and phantom respectively. The background fluence Φ_{bp} that is transmitted by the phantom is

$$\Phi_{bp} = \Phi_o \exp(-\mu_b L). \quad (3)$$

The contrast of the transmitted fluence is determined by the following.

$$C(\Phi) = 2 \frac{|\Phi_t - \Phi_b|}{\Phi_t + \Phi_b} = 2 \frac{|(\Phi_{tp} - \Phi_{bp}) + (\Phi_{ts} - \Phi_{bs})|}{(\Phi_{tp} + \Phi_{bp}) + (\Phi_{ts} + \Phi_{bs})}. \quad (4)$$

If the scatter components Φ_{ts} and Φ_{bs} are large the denominator of Eq.(4) will increase resulting in reduced image contrast. Various methods have been developed to reduce the scatter exiting the patient.

By decreasing the beam size around the target area one can reduce the volume of tissue available to scatter x-ray photons. Another method to reduce the scattered component is to decrease the peak potential (kVp) of the x-ray tube. To compensate for the reduction in kVp the current (mA) must be increased. This will result in an increase in patient dose. Another method to reduce scatter is patient compression. When a patient is compressed, soft tissue is forced out of the primary beam and hence there is less scattering material present.

The most effective way of reducing scatter is to use an anti-scatter grid. An anti-scatter grid is constructed out of an array of alternating aluminum and lead septa. The septa are positioned parallel to the path of the primary photons. Assuming that the scattered photons have a different trajectory than the target photons, the septa will prevent scatter from reaching the detector. Grids can remove up to 90 % of the scatter which translates into a much improved contrast. A parallel grid geometry is shown in Figure 3.

Figure 3. Use of a parallel grid to reduce x-ray scatter.

Projection imaging produces high contrasts for skeletal structure. When scanning an organ, for example liver, the x-ray fluences from the target and the background will be similar. This will produce a low contrast. To improve the contrast for soft tissue imaging a contrast agent such as barium is ingested by the patient. This agent will increase the attenuation coefficient of the organ thereby reducing the target fluences. The dissimilarity of the target and background fluence will improve contrast levels. Another problem of projection radiography is the loss of depth information caused by projecting a three dimensional body onto a two dimensional image receptor.

1.2 Digital Imaging

In projection imaging there is a direct spatial relationship between the x-ray photon that interacts with the recording medium and the response of that medium [Ref.2 Chp.8 p.192]. For example when x-ray film is exposed, the distribution of x-ray photons transmitted through the body is permanently recorded. This type of image is called analogue. An analogue image will provide accurate visual information, however it is not possible to analyze or manipulate the image data on a quantitative level.

In order to manipulate the image data new detection methods were developed in the field of radiation physics. One of the most commonly used digital x-ray detectors operates on the principle of energy integration. Images are obtained by integrating the total energy deposited in the detector during the whole radiation exposure. Detectors that work according to this principle are the Charged-Coupled Device (CCD) sensors coupled to conventional phosphors and solid-state detectors which are made of semi-conductor materials.

With the advancement of computer technology, the imaging information collected by the digital detector could be stored and displayed on a TV monitor. Imaging information is stored by placing the x-ray data into a matrix which is subdivided into compartments called pixels with a finite size, not always square. In a digitized image the x-ray

interaction is assigned to the appropriate pixel. A coarse pixellation can lead to discontinuous images (poor resolution) while a smaller pixel size will improve the resolution.

The main advantage of digital radiography is the ability to observe and manipulate the image on a display monitor. The computer can analyze small variations in pixel intensities and display the image in terms of a grey scale. The large number of variations would require many shades on the grey scale. Unfortunately the human eye can only detect at best 20 shades of grey. The numerical data can be manipulated so that the small range of intensity to which the human eye is sensitive is matched to the clinically useful information.

1.3 CT Imaging

An imaging modality called computerized axial tomography (CAT) or in short CT avoids the problems of image distortion associated with projection and digital radiography. Figure 4 shows the geometry for a first generation CT scanner. A planar slice of the body is defined when a pencil beam of x rays is passed through the patient. The x-ray tube is translated along the coordinate axis x to generate a plane projection. This process is repeated for many viewing angles θ until the required set of projection data is obtained. The transmitted intensity at position x and angle θ is given by

$$I_{\theta}(x) = I^0_{\theta}(x) \exp \left(- \int_{AB} \mu(x,y) dy \right). \quad (5)$$

where $I^0_{\theta}(x)$ represents the incident intensity and $\mu(x,y)$ is the two dimensional distribution of the linear attenuation coefficient. The expression in the brackets is a line integral of the attenuation coefficient. The integral is determined over the line AB through the patient.

Figure 4. First generation CT imaging [Ref.3 Chp.4 p.100].

The problem of image reconstruction is to recover $\mu(x,y)$ from a set of projections defined as,

$$\lambda_{\phi}(x) = -\ln \left[\frac{I_{\phi}(x)}{I^0_{\phi}(x)} \right]. \quad (6)$$

The reconstruction of $\mu(x,y)$ is done on a rectangular array where each element or pixel is assigned a value μ_i . To display the data on a video screen each element μ_i is scaled relative to water,

$$\text{CT Number} = \frac{\mu_i - \mu_{\text{water}}}{\mu_{\text{water}}} \times 1000. \quad (7)$$

The CT Numbers for different soft tissues are relatively close to each other. The small differences in the CT Number are amplified and mapped out in grey scale as in digital radiography. By increasing the contrast of the display the radiologist can differentiate between soft tissues.

The above reconstruction technique is based on first generation CT. The fourth generation CT scanner uses a rotating x-ray fan beam and a continuous 360° ring of detectors to image the patient. The advantage of this system is shorter data acquisition times which reduces motion artefacts. Another advantage is that an arc of detectors can sample the transmitted and unattenuated x-ray beam to perform real time beam calibration.

1.4 Scatter Imaging

For a 60 keV photon beam over 30 % of the interactions in water consist of coherent or incoherent scatter. Coherent or elastic scatter occurs when an incident electromagnetic wave of frequency ν causes an electron to oscillate and radiate at the same frequency. Incoherent scatter involves the inelastic collision of a photon and an electron. When an x-ray beam interacts with a material such as water multiple elastic scattering events from different electrons will interfere with each other to produce a diffraction pattern. Due to the particle nature of the incoherent interaction no interference occurs between multiple inelastic scattering events.

Researchers have found that by measuring the coherent scatter exiting a patient one can extract diagnostic information for the purposes of scatter imaging. The use of coherent scatter to obtain diagnostic information is a novel alternative to primary imaging.

1.4.1 Previous Investigators

One of the first groups to investigate the fundamentals of scatter imaging was Johns and Yaffe [Ref.4]. By using 60 keV photons they showed that the dominant scattering process at $2.5 - 6.6^\circ$ was coherent scatter. Figure 5 shows the differential scattering cross sections (proportional to the scattering probability) for liquid water. The coherent differential cross section has a maximum at 3.9° . The incoherent signal is negligible in the region $0-3^\circ$. At the angles 20° and 180° the incoherent curve reaches a maximum. The incoherent curve shows no characteristic peaks.

Figure 5. Differential scattering cross sections of liquid water for 60 keV photons. Coherent data obtained from Narten et al [Ref.5]. Incoherent data obtained from Hubbell et al [Ref.6].

Additionally they determined for an 80 kV_p x-ray beam that the magnitude of coherent first scatter is significant. For an abdomen examination the coherent first scatter to primary ratio was 26 % before the anti-scatter grid and 7.5 % after the 12:1 grid. They also found that due to the forward peaked nature of the coherent differential cross section the coherent scatter did not diverge much from the primary beam. This effect is more pronounced for higher energies where the diffraction pattern will shift to lower angles. For example the maximum scattering probability for water occurs at 2.2° for 100 keV photons. In this case air gaps and/or anti-scatter grids become less useful in separating the coherent and primary photons.

A semi-analytic model for back- and forward-scatter imaging was developed by Leclair and Johns [Ref.7]. The simulated targets were biological materials: liver, fat, bone, muscle, blood and brain matter. Assuming a monoenergetic photon beam (10-200 keV) the signal to noise ratio (SNR) and image contrast were evaluated numerically for the back- and forward-scatter models. Results showed for example that for white brain matter versus blood, for thicknesses up to 23 mm, the forward scatter was superior to conventional primary imaging in terms of SNR and image contrast. It was also shown that for liver versus fat, for thicknesses up to 22 mm, the backscatter model was superior to primary imaging.

The scattering model was modified to include the use of a polyenergetic photon beam [Ref.8]. The SNR was calculated for the forward and back scatter models using a 80 or 100 kV clinical x-ray beam, a rectangular beam and a dual peak beam. The targets were biological samples with a range of thicknesses 0.01-40 mm. The SNR was calculated for both polyenergetic and monoenergetic beams. It was found that the SNR for a polyenergetic beam was moderately smaller than for a monoenergetic source. Westmore et al [Ref.9] used a polychromatic source to measure the forward scatter from materials. They found that for the purposes of scatter imaging a material could still be characterized regardless of the polyenergetic source.

Leclair and Johns [Ref.10,11] tested the forward scatter model predictions by designing an apparatus to measure the SNR and contrast for plastic targets. The targets consisted of polymethyl methacrylate (PMMA), polycarbonate, polystyrene, polyethylene and nylon for a range of thicknesses 5-40 mm. Samples of beef, liver and muscle were also examined. A sample of water was also used. Each target was placed at the centre of a 15-cm diameter spherical water phantom. The x-ray source was a tungsten rotating anode x-ray tube operating at 80 kVp with 2.5 mm Al filtration. A pinhole collimator directed the forward scatter onto a high-purity germanium (HPGe) detector. Good agreement between prediction and experiment was obtained for the forward scatter model.

Harding et al [Ref.12] designed a diffraction CT system for the purpose of tissue and material identification. Figure 6 shows a simplified version of the CT apparatus. As in first generation CT, a pencil beam cuts through a slice of the patient. An array of detectors is designed to collect the scatter at several angles θ_i . The scattered radiation is collected for different angles ϕ of the incident beam on the patient. In conventional CT imaging the two dimensional attenuation coefficient map is extracted from the transmitted intensity.

To extract scattering information from diffraction CT one must determine the total number of photons scattered in the primary beam.

$$S_i = N_t \int_X n_o(X) \frac{d\sigma_i}{d\Omega} \Delta\Omega_i dX. \quad (8)$$

where N_t is the number of photons transmitted through the object, n_o is the number density of the scattering centres, $d\sigma_i/d\Omega$ is the total differential cross section at the scattering position X and $\Delta\Omega_i$ is the solid angle of the detector element. By measuring $S_i/(\Delta\Omega_i N_t)$ one can reconstruct the quantity $n_o(d\sigma/d\Omega)$ which is analogous to μ_i in conventional CT imaging.

Figure 6. X-ray diffraction computed tomography, as invented by Harding et al [Ref.12].

To test the accuracy of the CT system, Harding et al [Ref.12] used a powder diffractometer to measure the diffraction patterns of plastics such as polyethylene and biological materials such as bone, muscle and blood. He showed that there was adequate agreement between the diffraction CT system and diffractometer.

1.5 Coherent Scatter Form factor

The coherent differential scattering cross section is proportional to the square of the coherent scatter form factor. The form factor can be analyzed at three levels of complexity: free atom, molecular, and inter-molecular. The free atom form factor describes the interference effects between Z electrons of an atom. The molecular form factor describes the interference between scattering from all electrons of the same molecule. Finally the inter-molecular form factor describes the interference between scattering from all electrons in the material, even on different molecules.

The form factor depends on the energy and scattering angle θ of the photon. This dependence is defined by the x -variable,

$$x = \frac{1}{\lambda} \sin \frac{\theta}{2} \text{ (length}^{-1}\text{)}, \quad (9)$$

where λ is the wavelength of radiation. Equation 9 is proportional to the momentum transfer of the scattering event. The momentum transfer of an elastic scattering event is always conserved. Suppose a characteristic scattering event was measured at the position θ_0 using a photon energy $h\nu_0$. To measure this characteristic event using a higher energy photon, the θ position will shift to a lower value to conserve momentum transfer via Eq.(9). Figure 7 displays a contour plot of Eq.(9) on a log-log scale.

Figure 7. Contour plot of x as a function of photon energy $h\nu_0$ and scattering angle θ . Variable x ranges from 0.001 – 100 nm^{-1} .

1.5.1 Form Factors for Scatter Imaging

To model a scatter imaging system in the diagnostic energy range the coherent form factor must be known. The procedure of extracting the coherent scatter form factor from an imaging system would be nontrivial. The raw scattering data must first be corrected for background radiation which depends on machine geometry such as beam collimation and phantom medium. If the x rays are polychromatic a solid-state detector must be used to collect monoenergetic photons to produce the diffraction pattern. Detector alignment is critical. If the setup uses a monochromator to create a monoenergetic beam one must deal with reduced photon intensities and polarization corrections. Researchers have searched for an experimental setup which would simplify the extraction of the coherent scatter form factor.

1.5.2 Previous Investigators

An accurate way of measuring diffraction patterns is to use a powder diffractometer. Conventionally a powder diffractometer measures the diffraction patterns of crystalline materials such as silicate or graphite. A group lead by Narten [Ref.5] used a powder diffractometer to measure the coherent scatter form factor of water. Monochromatic radiation ($\text{MoK}\alpha = 17.48 \text{ keV}$) was obtained by using a curved NaCl monochromator. The diffraction pattern for triple distilled water was measured for various beam divergences, ranging from 0.5° at the lowest to 4° at the highest. The measured signal ranged from 1×10^5 to 6×10^5 counts for intervals 0.25° and 1° respectively. The observed x range of the machine was $0.39 < x < 12.73 \text{ nm}^{-1}$. The data were collected for eight temperatures between 4°C and 200°C . The sample pressure was kept at 1 atm below 100°C and equal to the vapour pressure above this temperature. Additionally a sample of heavy water (D_2O) was studied at 4°C .

To calculate the coherent scatter form factor, Narten's group corrected their data for background radiation, absorption of the sample, multiple scatter and polarization effects due to the NaCl monochromator. By converting the θ -variable into x -space, Narten then normalized the data to the free atom form factors in the x region $5.57 < x < 12.73 \text{ nm}^{-1}$. In this x -region the x rays scatter off a free atom. There is no inter-molecular interference. To determine the scale of the interaction one must relate the coordinate x to the atomic radius, r , of the atom. The coordinate r is defined as a Fourier conjugate variable which is proportional to the inverse of $2x$. Therefore the x rays will interact with the atom in the region $0.039 < r < 0.089 \text{ nm}$. For example the mean radius of an oxygen atom is 0.066 nm [Ref.13] which lies within the x -region of the x rays. The independent atom model (IAM) is used to calculate the coherent and incoherent form factors for all single atom interactions in the molecule. The single atom form factors were tabulated by Hubbell et al [Ref.6]. After the data were normalized, the coherent form factor was extracted by subtracting off the incoherent radiation. By applying a Fourier analysis to the form factor data Narten extracted the x -ray correlation function which contains information about the position of hydrogen and oxygen atoms.

Kosanetzky et al [Ref.14] studied the diffraction patterns of plastics such as PMMA, and of blood, muscle, fat, liver and bone using a powder diffractometer. The plastic samples were cut to thin slices with a thickness of 3 mm and a diameter of 25 mm . For biological samples a sample holder was constructed with a $12 \text{ mm} \times 20 \text{ mm} \times 3 \text{ mm}$ cavity. The sample holder was covered with a Kapton foil to avoid leakage and drying. The machine used a cobalt target x -ray tube and a graphite monochromator to produce a 6.95 keV photon beam. The scattered intensity was measured from 5° to 100° in steps of $0.05^\circ / 5.0 \text{ s}$. The background radiation was kept small by a compensating divergence slit.

The incoherent and multiple scatter components were corrected by using Monte Carlo simulation. The scattered intensity was also corrected for scatter from the Kapton foil. Additionally a factor $1/\sin\theta$ corrected for variable slit divergence. The differential cross section per unit volume $n_0(d\sigma/d\Omega)$ was extracted by normalizing the scattered intensity to the IAM region. The diffraction patterns for blood, liver and muscle were almost identical to water. The x rays mainly scatter off the tissue's water component.

Evans et al [Ref.15] measured the diffraction patterns of water, olive oil, 19 samples of breast tissue, adipose tissue and whole blood. The samples were contained within 6 mm diameter plastic tubes. The tubes were supported using a wooden clamp and exposed using a highly collimated x-ray beam. The x-ray tube (copper anode) operated at 60 kVp. The energy spread of the x-ray spectrum was reduced by using 0.5 mm Cu filtration. The mean photon energy was 46 keV. The scattered intensity was collected for the range $2^\circ - 12^\circ$. The scattering distributions showed that the peak position of adipose tissue was similar to that of olive oil, $\sim 4^\circ$. The diffraction pattern of breast tissue produced similar peak positions to that of water $\sim 6^\circ$. Evans et al [Ref.15] assumed these results would prove useful in the design of breast phantoms.

Peplow and Verghese [Ref.16] measured the coherent scatter form factor for plastics and biological materials using two energies (8.0 keV, 20 keV) from a synchrotron x-ray beam. A total of 16 samples were measured in this study. They were: PMMA, Lexan, Kapton, deionized water, five pork samples, five beef samples, formaline and human breast tissue in formaline. The raw data were corrected for air scattering, incoherent scattering and multiple scattering by using Monte Carlo based calculations. Their water results were in close agreement with Narten [Ref.5] and Kosanetzky et al [Ref.14].

Tartari et al [Ref.17] measured the diffraction patterns of PMMA, fat and bone. They used a powder diffractometer equipped with a copper target x-ray tube and Ni filter to produce an 8.04 keV photon beam. Each acquisition covered a scattering angle of 3° to 160° . Two other diffractometers were employed as control devices.

The data were corrected for background by subtracting the scatter signal of an empty sample holder from the sample scatter data. The detected spectrum of photons that undergo multiple scatter was subtracted from the scattered intensity. Additionally the data were corrected for sample self-attenuation and incoherent scattering. The coherent scatter form factors were extracted by normalizing the corrected data in the IAM region, $4.0 < x < 6.0 \text{ nm}^{-1}$. It was found that the results for fat and plastics eg. PMMA agreed well with the data of Peplow and Verghese [Ref.16].

Our lab has been measuring the diffraction patterns of plastics and tissues by using an x-ray powder diffractometer. Initial work was done by S. Decossas [Ref.18] and C. Buffet [Ref.19]. They used a diffractometer where the sample holder and detector rotates while the x-ray tube remains stationary. This group studied the normalized intensity patterns of biological materials such as beef muscle, liver, fat and brain matter. Since the sample holder rotates any liquid sample would need to be placed in a sealed container. When analyzing the intensity patterns one must correct for the diffraction pattern of the container cover. Work done by R.R Scharf [Ref.20] measured the biological coherent scatter form factor using a powder diffractometer where the sample holder remained stationary. His water results showed amplitude disagreement with the data of Narten et al [Ref.5] and of Kosanetzky et al [Ref.14]. The results for water and tissues were presented at the Canadian Organization of Medical Physicists (COMP) conference in Sherbrooke, Quebec [Ref.21].

1.6 Thesis Objective and Outline

1.6.1 Objective

The objective of this thesis is to develop a method to measure the diffraction patterns of amorphous materials by using a powder diffractometer. Further more the coherent scatter form factor will be calculated using the diffraction data. The validity of the method will be tested by comparing the coherent scatter form factors from two different powder diffractometers, Rigaku (Cobalt anode) and Scintag (Copper anode).

To perform the data analysis two types of variables are studied: systematic and fundamental. Systematic variables depend on the apparatus used in the experiment. In our experiment the systematic variables are background subtraction excluding external background, polarization due to the graphite monochromator and beam misalignment. Fundamental variables deal with the physics of amorphous and crystal scattering. These variables involve θ - x conversions for the energies 6.941 and 8.064 keV, coherent peak removal for powder diffraction patterns and normalization of amorphous data to the free atom form factors. Results from this work were presented at the Canadian Organization of Medical Physicists (COMP) conference in Kelowna, British Colombia [Ref.22].

1.6.2 Outline

Chapter 2 introduces the theory of x-ray scattering. It starts with basic Thomson scattering which develops the framework for the free atom form factor. In the same chapter the physics of inter-molecular scatter is introduced for amorphous and crystalline materials. Chapter 3 introduces the apparatus, including diffractometer and sample holders. A description of each sample is also given. Chapter 4 covers the procedure for machine calibration and sample measurements. Preliminary results are also shown for background and sample scans. Chapter 5 shows the calculations for correction methods. The final chapter tests the reliability of the experimental method by comparing the results from two different diffractometers. Finally, the form factor data are compared to the published literature.

Chapter 2

Theory

2.1 Radiation Interactions

Consider a photon with energy $h\nu_0 = 60$ keV interacting with an arbitrary medium. The photon will have a probability of undergoing one of the following interactions: photoelectric effect, coherent scatter or incoherent scatter. The probability for any one of these interactions depends on the total interaction cross section which is a function of $h\nu_0$ and atomic number. Let τ_{photo} , σ_{coh} and σ_{incoh} represent the cross sections per atom for photoelectric, coherent and incoherent interactions, respectively. Figure 8 plots the carbon interaction cross sections per atom.

Figure 8. Carbon interaction cross sections per atom [Ref.1 App.A].

2.1.1 Photoelectric Effect

The photoelectric effect occurs when a photon of energy $h\nu_0$ is absorbed by an atom causing ejection of an electron with kinetic energy

$$K = h\nu_0 - E_i > 0, \quad (10)$$

where E_i is the binding energy of the i th shell from which it was ejected. To determine the probability that an electron will be ejected via the absorption of a photon two assumptions are made: (i) the interacting photon's energy must be much larger than the binding energy of the K-shell. This assumption holds true for photons in the medical diagnostic energy range. (ii) The energy of the ejected electron is non-relativistic or $h\nu_0 \ll m_0c^2$. By applying the above assumptions to transition theory [Ref.23 Chp.5 Sec.21], the total photoelectric cross section per electron [Ref.1 Chp.5 p.149] is described by the following.

$$\tau_{\text{photo}} \propto \frac{1}{(h\nu_0)^3} Z^n \left(\frac{\text{area}}{\text{bound electron}} \right). \quad (11)$$

where $n = 3$ for high Z materials and $n = 3.8$ for low Z materials.

2.1.2 Incoherent Scatter

2.1.2.1 Single Electron

Incoherent or Compton scattering involves an incident photon of energy $h\nu_0$ scattering off of a free electron yielding a less energetic photon $h\nu$ and an electron with a kinetic energy $E' = h\nu_0 - h\nu$. The change in photon energy is obtained by applying relativistic energy and momentum conservation to a photon-electron collision [Ref.1 Chp.6 .p174]. The change in photon energy can be related to its scattering angle θ by the equation,

$$h\nu_0 - h\nu = h\nu_0 \frac{\alpha(1 - \cos \theta)}{1 + \alpha(1 - \cos \theta)}, \quad (12)$$

where $\alpha = h\nu_0 / m_0c^2$. The probability that an incoherent photon will be scattered into a solid angle element $d\Omega$ was derived by Klein and Nishina [Ref.23 Chp.5 Sec.22]. This probability is expressed by the incoherent differential cross section,

$$\frac{d\sigma_{\text{incoh}}}{d\Omega} = r_0^2 \left(\frac{1 + \cos^2 \theta}{2} \right) F_{\text{KN}}(\alpha, \theta) \left(\frac{\text{area}}{\text{free electron} \cdot \text{sr}} \right). \quad (13)$$

where $r_0 = ke^2/m_0c^2 = 2.818 \times 10^{-15} \text{ m}$ is the classical electron radius and

$$F_{\text{KN}}(\alpha, \theta) = \frac{1}{[1 + \alpha(1 - \cos \theta)]^2} \left\{ 1 + \frac{\alpha^2(1 - \cos \theta)^2}{(1 + \cos^2 \theta)[1 + \alpha(1 - \cos \theta)]} \right\}. \quad (14)$$

For low energy x rays $F_{\text{KN}}(\alpha, \theta)$ will approach a value of one. In this case the differential cross section will express an elastic scattering event.

2.1.2.2 Single Atom

Equation 13 assumes incoherent scattering from a free electron. Incoherent scattering from an atom must account for the binding energy of the shell. The incoherent form factor (IFF) defined by the function $S(x)$, expresses the effect of binding energy on the scattering interaction. Hubbell et al [Ref.6] calculated the IFF by using the integral,

$$S(x) = \left| \int_V \sum_{i=2}^N \psi_i(r)^* \exp(ix \cdot r) \psi_1(r) dr^3 \right|^2. \quad (15)$$

where ψ_i is a wave function for the i th excited state and ψ_1 is the ground state wave function. The summation is carried out over all allowed excited states of the atom. Hubbell et al [Ref.6] calculated the IFF for $Z = 1-100$ at $x = 0.05 - 10^{10} \text{ nm}^{-1}$. Figure 9 shows the IFF for hydrogen, carbon and oxygen. At low x values the IFF approaches zero. This indicates that no incoherent scatter occurs at low x values. When the IFF approaches Z , the binding effects of the atom are at a minimum. The likelihood of an incoherent event per atom is expressed by,

$$\frac{d\sigma_{\text{incoh}}}{d\Omega} = r_0^2 \left(\frac{1 + \cos^2 \theta}{2} \right) F_{\text{KN}}(\alpha, \theta) S(x) \left(\frac{\text{area}}{\text{atom} \cdot \text{sr}} \right). \quad (16)$$

2.1.2.3 Compound or Mixture

To determine the incoherent form factor for a molecule one must take the sum of the incoherent form factors for each atom in the molecule. In this report all form factors for a molecule will be expressed as free electron per bound electron.

$$S(x) = \frac{1}{Z} \sum_{i=1}^N M_i S_i(x) \left(\frac{\text{free electron}}{\text{bound electron}} \right). \quad (17)$$

where Z is the total number of bound electrons in the molecule, N is the number of atomic species in the molecule, M_i is the number of atoms of the i th species and $S_i(x)$ is the incoherent form factor for the i th species. The incoherent cross section, σ_{incoh} , is obtained by numerically integrating Eq.(16) for all angles.

Figure 9. Incoherent form factor per atom for hydrogen, carbon and oxygen [Ref.6].

2.1.3 Coherent Scatter

2.1.3.1 Single Electron

Single electron or Thomson scattering involves the oscillation of an electron by an electromagnetic (EM) wave of frequency ν_0 which produces an outgoing EM wave with the same frequency ν_0 . Figure 10 shows an unpolarized EM wave incident on an electron at position O. To obtain the intensity of the scattered wave at position P the incident wave is split up into two electric field components, $E_{\parallel}$ and $E_{\perp}$. The components $E_{\parallel}$ and $E_{\perp}$ lie parallel and perpendicular to the plane of scattering respectively [Ref.1 Chp.6 p.169]. For an unpolarized wave, $E_{\parallel} = E_{\perp}$. When the wave scatters off the electron the parallel component will change direction by an angle θ . The perpendicular component does not change direction. The scattered components $E'_{\parallel}$ and $E'_{\perp}$ will be modified by an optical factor depending on the inverse radial distance r from the electron.

To determine the intensity of the scattered wave the sum of the squares of the components $E'_{\parallel}$ and $E'_{\perp}$ is taken.

$$I_{\text{Thom}} = I_0 \frac{r_0^2}{r^2} \left(\frac{1 + \cos^2 \theta}{2} \right). \quad (18)$$

where I_0 is the incident intensity and $r_0 = ke^2/m_0c^2 = 2.818 \times 10^{-15}$ m is the classical electron radius. The factor $(1+\cos^2\theta)/2$ is called the polarization factor where the scattered wave is partially polarized by the interaction. The scattered wave is fully polarized for $\theta = 90^\circ$.

Figure 10. Model for Thomson Scatter.

To determine the probability that a photon will scatter into a solid angle $d\Omega$ the differential cross section must be extracted from Eq.(18). Let $d\sigma_{\text{Thom}} = (I_{\text{Thom}}/I_0)dA$ and $d\Omega = dA/r^2$ where dA is the differential area subtending a cone of scattered photons. By taking the ratio of $d\sigma_{\text{Thom}}$ to $d\Omega$ the Thomson cross section takes the form,

$$\frac{d\sigma_{\text{Thom}}}{d\Omega} = \frac{r_o^2}{2} (1 + \cos^2 \theta) \left(\frac{\text{area}}{\text{free electron} \cdot \text{sr}} \right). \quad (19)$$

The total Thomson cross section is obtained by integrating Eq.(19) over all angles,

$$\sigma_{\text{Thom}} = \frac{8}{3} \pi r_o^2 \left(\frac{\text{area}}{\text{free electron}} \right). \quad (20)$$

2.1.3.2 Single Atom

When an EM wave interacts with an atom scattering will occur from the Z electrons of the atom and to a lesser extent with the nucleus. Coherent scattering from an atom can be described by using a classical wave approach. Figure 11 shows a pair of electrons located at O and $\mathbf{r}_j$ [Ref.24 Chp.1 p.7]. Consider an EM wave incident upon these electrons

$$E_i(\mathbf{R}, t) = E_o \cos(2\pi\nu t - \frac{2\pi}{\lambda} \mathbf{k}_o \cdot \mathbf{R} + \delta). \quad (21)$$

where E_o is the amplitude, $\mathbf{k} = \mathbf{k}_o(2\pi/\lambda)$ is the propagation vector, $\mathbf{R}$ is the spatial coordinate of the incident wave, and δ is the phase constant. Assume for simplicity that the amplitude E_o is linearly polarized which is defined perpendicular to the page. Additionally $\delta = 0$ for an incident wave.

When the wavefront of Eq.(21) interacts with the electrons at O and $\mathbf{r}_j$, each electron will oscillate producing its own EM wave with the same frequency of the incident wave. The waves generated now travel in a new direction defined by the propagation vector $\mathbf{k}' = \mathbf{k}'_o(2\pi/\lambda)$. It can be shown that the wave generated now has a path difference of,

$$\Delta X = (\mathbf{k}'_o - \mathbf{k}_o) \cdot \mathbf{r}_j. \quad (22)$$

Therefore the phase difference takes the form $\delta = (2\pi/\lambda)\Delta X$. Both waves are observed at a point P at a large distance R from the electrons.

The j th scattered wave at position P takes the form

$$E_{sj}(R, t) = \frac{E_o r_o}{R} \cos \left[2\pi \nu t - \frac{2\pi}{\lambda} (R + (\mathbf{k}_o' - \mathbf{k}_o) \cdot \mathbf{r}_j) \right]. \quad (23)$$

where the amplitude E_o has been modified by the classical electron radius r_o and the inverse of the distance R . By rewriting Eq.(23) in terms of exponential notation, the superposition of Z scattered waves at point P is expressed by,

$$E_s(R, t) = \frac{E_o r_o}{R} \exp \left[2\pi i \left[\nu t - \left(\frac{R}{\lambda} \right) \right] \right] \sum_{j=1}^Z \exp(2\pi i / \lambda) (\mathbf{k}_o' - \mathbf{k}_o) \cdot \mathbf{r}_j. \quad (24)$$

Figure 11. Geometric model for atomic scattering.

The scattering model assumed that each electron is a fixed point particle. In reality an atom consists of a diffuse cloud of negative charge characterized by its charge distribution function, $\rho(\mathbf{r})$. The quantity $dZ' = \rho(\mathbf{r})d\mathbf{r}^3$ represents the differential charge distribution of an atom, where $\int \rho(\mathbf{r})d\mathbf{r}^3 = Z$. The scattered amplitude, E_{sj} , no longer originates from the integral charge of the j th electron but from a differential charge dZ' . To obtain the total scattered amplitude we must replace the electrons at $\mathbf{r}_j$ with the charge elements $\rho(\mathbf{r})d\mathbf{r}^3$,

$$E_s(R, t) = \frac{E_0 r_0}{R} \exp 2\pi i \left[vt - \left(\frac{R}{\lambda} \right) \right] \int \rho(\mathbf{r}) \exp [(2\pi i / \lambda) (\mathbf{k}_0' - \mathbf{k}_0) \cdot \mathbf{r}] d\mathbf{r}^3. \quad (25)$$

The integral of Eq.(25) is called the coherent scatter form factor.

$$F(\mathbf{x}) = \int \rho(\mathbf{r}) \exp(4\pi i \mathbf{x} \cdot \mathbf{r}) d\mathbf{r}^3, \quad (26)$$

where $\mathbf{x} = (2\lambda)^{-1} (\mathbf{k}_0' - \mathbf{k}_0)$. The variable $\mathbf{x}$ is obtained by taking the magnitude of the resultant vector $\mathbf{k}_0' - \mathbf{k}_0$ shown in Figure 12.

$$|\mathbf{x}| = \frac{1}{2\lambda} |\mathbf{k}_0' - \mathbf{k}_0| = \frac{1}{\lambda} \sin \frac{\theta}{2}. \quad (27)$$

Figure 12. Resultant vector $\mathbf{k}_0' - \mathbf{k}_0$.

For an unpolarized wave coherent scattering from a single electron is described by Thomson's differential cross section. Coherent scattering for an atom is obtained by multiplying Thomson's differential cross section by the square of the coherent scatter form factor.

$$\frac{d\sigma_{coh}}{d\Omega} = \frac{d\sigma_{Thom}}{d\Omega} F^2(x) \left(\frac{\text{area}}{\text{atom} \cdot \text{sr}} \right). \quad (28)$$

To calculate the function $F(x)$ one requires knowledge about the atom's charge distribution. Classically the charge distribution exists as an array of static point charges. The atomic system is not at all static. Each electron orbits the nucleus where variables such position, momentum, energy and spin are described by the atomic wave functions.

Conventionally atomic wave functions are calculated using the Hartree-Fock method. The Hartree-Fock method is based on the self-consistent-field (SCF) model. This is an independent-particle model in which each electron is assumed to be in the field of the nucleus and is in the average field due to other electrons. Hubbell et al [Ref.6] used the results from the Hartree-Fock method to calculate the charge distribution of the atom.

$$\rho(r) = \sum_{j=1}^Z \psi_j^*(r) \psi_j(r), \quad (29)$$

where $\psi_j(r)$ is the wave function for the j th electron. To solve for the coherent form factor one must substitute the calculated charge distribution into Eq.(26). Hubbell et al [Ref.6] tabulated the coherent form factor for $Z = 1-100$ in the range $x = 0.05-10^{10} \text{ nm}^{-1}$. Figure 13 displays coherent scatter form factors for hydrogen, carbon and oxygen as a function of x . In the direction of the primary photons, $\theta = 0$, an isolated atom scatters like a Thomson electron where the form factor equals the atomic number of the atom. As θ increases the form factor decreases rapidly due to destructive interference.

Figure 13. Coherent scatter form factors for hydrogen, carbon and oxygen [Ref.6].

By substituting the coherent scatter form factor (eg. C_6^{12}) into Eq.(28), the coherent cross section, σ_{coh} , can be determined by numerically integrating over all angles.

2.1.4 Total Scatter

The total probability that a photon will scatter into a solid angle $d\Omega$ is described by the total differential cross section.

$$\frac{d\sigma_t}{d\Omega} = r_0^2 \left(\frac{1 + \cos^2 \theta}{2} \right) [F^2(x) + F_{KN}(\alpha, \theta)S(x)]. \quad (30)$$

2.2 Inter-Molecular Scatter

When light waves interact with an opaque object having a well defined edge, a detailed analysis shows that an array of fringes is formed in the geometric shadow. This experiment first demonstrated the wave phenomenon of diffraction. A.von Laue was the first to postulate and formulate the theory of x-ray diffraction [Ref.25]. He stated that for x-ray diffraction to occur the wavelength of the incident x-ray must have similar dimensions to the scattering target. For example the mean distance between the oxygen atoms of liquid water is 0.29 nm [Ref.5]. For x-ray diffraction to occur between the oxygen atoms the wavelength of the x ray must be similar.

Experiments done by W.L. Bragg [Ref.26 Chp.1 p.10] tested Laue's theory of diffraction. Bragg collimated 0.154 nm x rays onto a crystal target. Three types of crystals were chosen for the experiment: NaCl, KCl and KBr. The diffraction patterns were recorded using a photographic plate. The experiment was repeated for different positions of the photographic plate. The experimental results showed an ordered dot pattern unique to each crystal target. This experiment proved that x rays could produce a diffraction pattern.

At the most sophisticated level, we must now include the effects of interference between scattering from electrons on different atoms or molecules. These interference effects depend on the spatial distribution of the atoms or molecules. There are two types of materials which have distinctly different molecular arrangements. A crystalline material consists of stationary atoms or molecules arranged in an ordered array. Crystalline materials include rock salt (NaCl) and graphite powder. An amorphous material consists of atoms or molecules arranged in a disordered pattern. If the material is a liquid each molecule has no permanent neighbours due to the molecules' translational motion. Liquids at room temperature include distilled water, ethanol and acetone. If the material is a solid the molecules may be arranged in a series of chains. Each molecule will have different arrangements of neighbours. Amorphous solids include plastics such as nylon or polystyrene.

2.2.1 Crystal Diffraction

A crystal is a three-dimensional repetition of some unit of atoms or molecules. Figure 14 shows a series of cubic unit cells. Each cell is defined by the coordinate point (a_x, a_y, a_z) . The cell contains two different kinds of atoms or molecules which are represented by circles and rectangles. For atomic scattering each electron acted as a source of scatter. W.H Bragg [Ref.26] took this concept a step further and used the atoms in each cell as a source of scatter. Additionally he placed these atoms into what are called crystallographic planes defined by the Miller indices (h, k, l) .

Figure 14. Cubic unit cell of repetition where $a_x = a_y = a_z$.

X-ray diffraction occurs when the scatter from neighbouring planes interfere. By using plane geometry Bragg showed that the distance between planes, $d(h, k, l)$, is related to the photon scattering angle θ and the wavelength.

$$\frac{1}{d(h,k,l)} = \frac{2}{n\lambda} \sin \frac{\theta}{2}. \quad (31)$$

When an integral number of wavelengths superimpose a coherent or Bragg peak will take shape. By measuring the peak positions of a crystal diffraction pattern one can extract information on the lattice spacing. Figure 15 shows the diffraction pattern for iodized NaCl. The Bragg peaks are sharp and localized. The signal between the Bragg peaks results from incoherent scatter.

Figure 15. Diffraction scan of iodized NaCl. Measured using a Rigaku powder diffractometer as described in Chp.3.

2.2.2 Amorphous Diffraction

An amorphous material cannot be modeled with respect to Bragg's law since the molecules cannot act as fixed points of scatter. One would assume that with such random order most of the x rays would interfere destructively producing no diffraction pattern. This assumption is proved wrong when one measures the diffraction pattern of pure ethanol. See Figure 16. For the θ range $2 - 40^\circ$ diffraction occurs between the CH_2 molecules. There are no sharp Bragg peaks but the pattern has characteristic maxima at about 12° and 25° . For the angles $75-150^\circ$ the signal levels out at a constant value. This region is called the IAM region where the x rays only interact with the atoms of the

molecule. There is no interference between molecules. To obtain any spatial information on the CH_4 molecule one can extract the coherent scatter form factor. Theoretically one cannot calculate the form factor in the low θ region due to the complex interference effects of molecular scattering. In practice one obtains the amorphous form factor by normalizing the experimental data to the square of the IAM form factor.

Figure 16. Diffraction scan of pure ethanol. Measured using a Rigaku powder diffractometer as described in Chp.3.

The square of the IAM form factor treats each atom in the molecule as an individual source of scattering with no inter-atomic interference. The square of the IAM form factor takes the form

$$F_{\text{IAM}}^2(x) = \frac{1}{Z} \sum_{i=1}^N M_i F_i^2(x) \left(\frac{\text{free electron}}{\text{bound electron}} \right). \quad (32)$$

where Z is the total number of bound electrons in the molecule, N is the number of atomic species in the molecule, M_i is the number of atoms of the i th species and $F_i(x)$ is the form factor for the i th species.

Chapter 3

Apparatus

In this project all x-ray diffraction measurements were done at the National Research Council (NRC) of Canada, Steacie Institute for Molecular Sciences (SIMS), Sussex Drive, Ottawa. The Functional Materials research program has extensive x-ray diffraction facilities, under the direction of Dr. G. Enright.

3.1 Rigaku Powder Diffractometer

The Rigaku Geigerflex x-ray powder diffractometer (XPD) is a machine used to analyze powdered crystals by collecting the scattered intensity as a function of photon scattering angle θ . Figure 17 is a photograph of the Rigaku XPD. The dimensions and components of the apparatus are shown in Figure 18. The Rigaku machine uses a cobalt target x-ray tube which is cooled by a constant circulation of water. The tube's maximum power output is 1.2 kW. The x-ray tube is fitted with a 1° divergence slit (DS). The DS collimates the beam onto the sample. The sample is placed within a cylindrical chamber with a diameter of 8.90 cm and a width (perpendicular to the plane of the drawing) of 4.50 cm. The chamber is sealed by a steel hatch. Two mylar windows (MW) allows x rays to enter and exit the chamber.

When the x rays scatter off the sample the beam will diverge. To collimate the scattered x rays a Soller slit box (SSB) is placed before the detector. A Soller slit box reduces the divergence of the scattered beam by directing the beam through a stack of thin parallel plates. The x rays enter and exit the box through a 1° scatter slit (SS) and a 0.15 mm receiving slit (RS) respectively.

Figure 17. In this photograph the Rigaku diffractometer is shown. The x-ray tube is on the right and the monochromator and detector are on the left.

Figure 18. Technical diagram of Rigaku XPD.

A polyenergetic photon beam scatters off a curved graphite monochromator (GM). To isolate the $K\alpha$ lines from the cobalt spectrum the detector must be positioned at $\phi = 30.97^\circ$. By applying Bragg's relation for crystal diffraction, Eq.(31), the photon energy of 6.941 keV is isolated from the polyenergetic spectrum. To determine the energy bandwidth of the monochromator one must differentiate Eq.(31) with respect to the ϕ variable,

$$\Delta E_{\text{mono}} = \frac{hc}{2(2d)} \frac{\cos \frac{\phi}{2}}{\sin^2 \frac{\phi}{2}} \Delta \phi, \quad (33)$$

where $\Delta \phi$ is the angular divergence of the monochromator ($\sim 0.114^\circ$ or 2 mrad) and $2d$ is twice the spacing of the graphite planes (~ 67.08 nm). The energy bandwidth of the monochromator is $\Delta E_{\text{mono}} = 0.0250$ keV which encompasses $K\alpha_1$ (6.937 keV) and $K\alpha_2$ (6.952 keV). The $K\beta$ (7.673 keV) line falls outside the window of the monochromator. Figure 19 shows the position of the monochromator's energy window with respect to the cobalt K-fluorescence lines.

Figure 19. Monochromator energy window (solid lines) superimposed on the cobalt K-fluorescence lines (dashed lines).

Compton radiation will also pass through the monochromator which depends on the energy of the incident photon and the size of the Compton shift. At the maximum scattering angle used in our experiment ($\theta = 150^\circ$), the Compton shift is $\Delta E_{\text{Comp}} = 0.171$ keV. When an incident photon is within the energy range of 6.916 – 6.966 keV, and undergoes Compton scattering, the energy of the scattered photon will be shifted below the window of the monochromator. The loss of these Compton scatter photons is equalized by more energetic bremsstrahlung photons shifting down in energy into the window of the monochromator. The Compton energy shift is small enough that the $K\beta$ (7.673 keV) line cannot be shifted into the energy window of the monochromator.

The monochromatic photons scatter into a NaI(Tl) scintillation counter. The detector is equipped with a pulse height analyzer which cuts off any harmonics (e.g. bremsstrahlung radiation) which are transmitted through the monochromator. The standard energy resolution of a NaI(Tl) detector is about 20 keV. The tube, sample chamber, and detector are mounted on a goniometer. The goniometer rotates the tube and detector with respect to the sample holder. The entire machine is enclosed in a lead plexiglass radiation shield.

3.1.1 Rigaku Specifications

(I) X-ray tube

Target: Co anode, water cooled, fine focus (0.4 x 8.0 mm)
 Power output: 1.2 kW
 Company: Philips
 Model #: w58122-L

(II) Slits

- (1) X-ray tube: 1° diverging slit (DS).
- (2) Soller slot box (SSB): 1° scatter slit (SS), 0.15 mm receiving slit (RS).
- (3) Detector: 0.45 mm receiving slit (RS).

(III) Sample Chamber

Cylindrical chamber: 8.90 cm diameter, 4.50 cm length.

Mylar window (MW) allows x rays to enter and exit chamber.

(IV) Monochromator

Curved pyrolytic graphite monochromator (GM) with a curvature of 224 mm

Graphite lattice spacing 33.50 nm

(V) Detector

Scintillation counter: NaI (Tl) crystal coupled to a photomultiplier tube.

Company: Rigaku Corporation, Tokyo Japan

3.2 Scintag Powder Diffractometer

The Scintag Advanced Diffraction System is shown in Figure 20. Figure 21 shows the technical setup. The machine uses a copper target x-ray tube which is water cooled. A Soller slit box (SSB)₁ is attached to the x-ray tube. The SSB holds two slits, a 2 mm DS and a 4 mm SS which collimate the beam on the sample. The sample holder sits on an open base. A second slit box (SSB)₂ holds a 0.5 mm SS and a 0.3 mm RS which collimates the beam onto a graphite monochromator (GM).

A scintillation counter is positioned at an angle $\phi = 26.57^\circ$. By applying Bragg's relation for crystal diffraction, Eq.(31), the photon energy 8.064 keV is isolated from the polyenergetic spectrum. The bandwidth of the monochromator is determined by using Eq.(33). The bandwidth of the monochromator is $\Delta E_{\text{mono}} = 0.034$ keV which encompasses $K\alpha_1$ (8.051 keV) and $K\alpha_2$ (8.072 keV). Again the $K\beta$ (8.930 keV) line falls outside the window of the monochromator. Figure 22 shows the monochromator window superimposed on the copper K-fluorescence lines. The goniometer has a radius of 15 cm. The machine is enclosed within a steel hood with a lead glass window.

Figure 20. In this photograph, the Scintag diffractometer is shown. The x-ray tube is on the left and the monochromator and detector are on the right.

Figure 21. Technical setup for Scintag XPD

Figure 22. Monochromator energy window (solid lines) superimposed on the copper K- fluorescence lines (dashed lines).

At the maximum scattering angle used in our experiment ($\theta = 150^\circ$), the Compton shift is $\Delta E_{\text{Comp}} = 0.230$ keV. When an incident photon is within the energy range of 8.030 – 8.098 keV, and undergoes Compton scattering, the energy of the scattered photon will be shifted below the window of the monochromator. The loss of these Compton scatter photons is equalized by more energetic bremsstrahlung photons shifting down in energy into the window of the monochromator. The Compton energy shift is small enough that the $K\beta$ (8.930 keV) line cannot be shifted into the energy window of the monochromator.

3.2.1 Scintag Specifications

(I) X-ray tube

Target: Cu anode, water cooled
Power output: 1.8 kW
Company: ATPS XRD 1000, Holland
Model #: w2200

(II) Slits

- (1) Soller slit box (SSB)₁ attached to X-ray tube: 2 mm diverging slit (DS), 4 mm scatter slit (SS).
- (2) Soller slot box (SSB)₂ attached to detector: 0.5 scatter slit (SS), 0.3 mm receiving slit (RS).

(III) Sample Chamber

Open chamber base

(IV) Monochromator

Curved Pyrolytic Graphite monochromator (GM) with a Curvature of 225 mm.
Graphite lattice spacing 0.335 nm.

(V) Detector

Scintillation counter:
NaI (Ti) crystal coupled to a photomultiplier tube. Company: Applied Research Laboratories, Thermo ARL, Ecublens Switzerland.

3.3 Sample Holders

Holder Type	Description
I	Slit block - Used for aligning the diffractometer.
II	NRC sample holder - Used for powder diffraction scans on the Rigaku machine.
III	Biological sample holder - Used to hold water and tissue samples.
IV	Biological sample holder - Used to hold water and tissue samples with a larger sample cavity than type III.
V	Biological sample holder - Used to hold water and tissue samples with a larger sample cavity than type IV.
VI	Insert - Used to extract background radiation.

Table 2. A summary of all sample holders used in the experiment.

3.3.1 Type I Sample Holder

The type I sample holder is called a slit block which is used to test the alignment of the diffractometer. Two type I holders were supplied by the Rigaku and Scintag companies. Figure 23(a) shows a schematic of the Rigaku slit block. The slit has dimensions 1.1 cm x 0.033 cm (Length x Width). The base of the slit is positioned 0.967 cm from the base of the holder. The Scintag slit block shown in Figure 23(b).

Figure 23. Type I holders Rigaku (a) and Scintag (b) slit blocks.

3.3.2 Type II, III Sample Holders

The type II holder is the standard NRC powder diffraction holder which is designed to fit in the Rigaku powder diffractometer. See Figure 24(a). Its cavity dimensions are 1.49 x 1.39 x 0.076 cm (Length x Width x Depth). The shallow cavity is designed to hold a very fine layer of powder. The surface of the sample lies at the 1.0 cm mark of the holder. For our work a new sample holder, type III, was designed by R.R. Scharf [Ref.20] which could hold water and/or biological samples. The sample holder was fabricated by the Science and Technology Centre (STC) at Carleton University. The sample volume was enlarged from 0.157 cm³ to 1.44 cm³. See Figure 24(b). Two type III holders were constructed out of aluminum (Al III) and stainless steel (St III). Aluminum and steel have large absorption to scatter ratios which would reduce the

background scatter from the edges and base of the sample holder. Also aluminum and stainless steel are easily cleaned and are resistant to rust.

Figure 24. (a) Type II holder. (b) Type III holder.

3.3.3 Type IV, V Sample Holders

By using a fluorescent screen it was determined at low angles that a large portion of the x-ray beam interacts with the edges of the type III holder. The solution was to fabricate a holder with very thin walls. This would increase sample volume and reduce the scattering off the edges of the holder. Holder types IV and V were constructed out of aluminum (Al IV, Al V) and stainless steel (St IV, St V). The geometry of the type IV was similar to type III except the hollowed out cavity was enlarged from 1.44 cm^3 to 3.21 cm^3 . See Figure 25(a). Also the wall thickness was reduced from 0.32 cm to 0.04 cm. The cavity volume was once again increased in the type V holder. This was done by building the walls around the leading lip of the type IV holder. See Figure 25(b). The volume of type V was 4.60 cm^3 . This holder was used for all subsequent Rigaku x-ray scans. The type V holder was also used for the Scintag machine since it was the only holder that could fit the geometry of the chamber.

Figure 25. (a) Type IV holder. (b) Type V holder.

3.3.4 Type VI Insert

The type VI insert was not designed for sample scans but for background analysis. Two inserts were constructed out of stainless steel, one for each of the Rigaku and Scintag machines. The Rigaku type VI insert is shown in Figure 26(a). It has 29 steel strips inserted into the cavity space. Each strip has a thickness of 0.025 cm and a length of 1.90 cm. The spacing between each strip is 0.10 cm. These strips run normal to the incident x-ray beam. These strips allow the x rays to pass into the cavity but prevent any scatter from the walls and the base of the holder from reaching the detector. The Scintag type VI insert is shown in Figure 26(b). It was constructed with 23 steel strips with a thickness of 0.025 cm and a length of 2.39 cm. The spacing between the strips is still 0.10 cm.

Figure 26. Type VI insert (a) Rigaku. (b) Scintag.
(c) Slat spacing for both Scintag and Rigaku insert.

3.4 Samples

Table 3 lists the substances scanned by the diffractometer. The table displays the name, chemical formula and/or composition and the mass of the sample. The salt, water and biological samples were placed in the Al V or St V holder which has a volume of 4.60 cm³. The plastic samples consist of single piece blocks which have the same dimensions as the type II holder. These blocks were constructed without a sample cavity.

Name	Formula/Composition	Mass (grams)
Iodized salt	NaCl	5.44
Silicate powder	SiO ₂	3.78
Graphite powder	C	1.31
Polyethylene ^b	(C ₂ H ₄) _n	5.11
Polystyrene ^b	(C ₈ H ₈) _n	5.83
PMMA ^{a,b}	(C ₅ H ₈ O ₂) _n	6.61
Nylon ^b	(C ₆ H ₁₁ NO) _n	6.38
Lexan ^b	(C ₁₆ H ₁₄ O ₃) _n	6.66
Distilled Water	H ₂ O	3.50
Beef muscle ^b	H:0.1, C: 0.107, N: 0.0275, O: 0.75, Cl: 7.8x10 ⁻⁴	4.90

^a Polymethyl methacrylate

^b Composition from Kosanetzky et al [Ref.15]

Table 3. Samples scanned by the x-ray diffractometers.

Chapter 4

Experiments and Results

4.1 Scattering Model

In order to perform any measurements we must identify the signal components that reach the detector. The total intensity consists of two components: background N_b and sample scatter N_s .

$$N_t = N_s + N_b. \quad (34)$$

4.1.1 Sample Scatter N_s

To determine an expression for the sample scatter N_s , consider the scattering model shown in Figure 27.

Figure 27. Scattering model to determine the form of N_s .

The incident fluence is defined by $\Phi_0 = N_0/A_0$ where N_0 is the number of photons and A_0 is the area of the beam. The beam penetrates the sample surface defined by the dashed line. As the beam travels through the sample it is attenuated by the factor $\exp(-\mu 2L')$ where L' is the variable path length of the photon and μ is the attenuation coefficient of the sample. The beam scatters off a sample element $\rho_e dV'$ where ρ_e is number of electrons per unit volume. Let $dV' = dA' dL'$ where dA' is the area of the beam that interacts with the sample and dL' is the differential length of the sample element. The probability of the scattering event is obtained from the total differential cross section per solid angle, $d\sigma_t/d\Omega$. The fractional number of photons scattered into the detector's solid angle element, $d\Omega_0$, is given by the following relation,

$$\frac{dN_s}{N_0} = \rho_e \frac{dA'}{dA_0} dL' \left\{ \frac{d\sigma_t}{d\Omega} d\Omega_0 \right\} \exp(-2\mu L'). \quad (35)$$

To determine the scatter signal N_s , one must integrate Eq.(35) over the path length which is much larger than the mean free path of the incident photon. By substituting in for $d\sigma_t/d\Omega$ [Chp.2 Eq.(30)] the expression for the total scatter N_s takes the form,

$$N_s = \frac{N_0 \rho_e d\Omega_0 r_0^2}{2\mu} \left[\frac{1 + \cos^2 \theta}{2} \right] [F^2(x) + F_{KN} S(x)]. \quad (36)$$

or

$$N_s = K \left[\frac{1 + \cos^2 \theta}{2} \right] [F^2(x) + F_{KN} S(x)]. \quad (37)$$

and

$$K = \frac{N_0 \rho_e d\Omega_0 r_0^2}{2\mu}. \quad (38)$$

The symbols of Eq.(38) are constants of the scattering model. Constants such as the number of photons N_0 or $d\Omega_0$ are too difficult to measure separately. In practice one measures the constant K by normalizing the scattering data to the IAM form factors, see [Chp.5 Sec.5.3.]

4.1.2 Monochromator Polarization

Equation 36 is only valid for monoenergetic photons. In practice the scattered or incident signal must pass through a monochromator to eliminate the bremsstrahlung spectrum and to suppress unwanted characteristic lines. The monochromator will polarize the beam causing a decrease in fluence. To account for this decrease consider the scattering geometry shown in Figure 28. Let P_1 and P_2 represent two scattering planes defined by the normal vectors $\mathbf{M}_1$ and $\mathbf{M}_2$ [Ref.27]. An unpolarized incident wave defined by the orthogonal set E_π and E_σ scatters off the plane P_1 at an angle θ . The scattered components E_π' and E_σ' become partially polarized (Thomson polarization). The scattered wave encounters another plane P_2 . The second plane is inclined at an angle δ with respect to the x' and z' axes. The components E_π'' and E_σ'' scatter into a detector oriented at an angle ϕ . By decomposing the components E_π'' and E_σ'' into an orthogonal set of EM vectors, the scattering signal N_s can be written as

$$N_s = N_0 P(\theta, \phi, \delta), \quad (39)$$

where,

$$P(\theta, \phi, \delta) = \frac{(\cos^2 \theta \cos^2 \delta + \sin^2 \delta) \cos^2 \phi + \cos^2 \theta \sin^2 \delta + \cos^2 \delta}{1 + \cos^2 \theta}. \quad (40)$$

Equation 40 is known as the polarization factor for a monochromator placed after the scattered beam.

Our experiment uses the powder method, tilt angle $\delta = 0$. The polarization factor can be simplified,

$$P(\theta, \phi) = \frac{1 + \cos^2 \theta \cos^2 \phi}{1 + \cos^2 \theta}. \quad (41)$$

For Eq.(41) to be valid the monochromator must be properly aligned.

Figure 28. Scattering geometry for monochromator.

4.1.3 Background Radiation

The background radiation from the diffractometer N_b consists of three components: external radiation N_e , holder edge scattering N_h or holder edge/base scattering N'_h , and fringe radiation N_f . When the sample is present, the total background signal takes the form.

$$N_b = N_e + N_h + N_f. \quad (42)$$

The N_e component results from radiation from outer space or isotopes in the lead shielding of the diffractometer etc. The component N_h refers to the beam that scatters off the edges of the sample holder. The component N_f consists of the upper half of the primary beam which does not scatter off the sample and goes directly to the detector. This effect only occurs at low angles. Figure 29 shows how the beam scatters when a sample is present.

To measure the background N_b , one must separate the sample scatter N_s from N_f . The obvious approach is to scan an empty sample holder. The total signal from an empty sample holder consists of three components, N_f , N_e and N_h where N_h is the radiation which scatters off the edges and cavity walls of the holder. The total background signal is not the same when a sample is present since $N_h \neq N_h'$. See Figure 30. To solve this problem a slatted device (type VI insert) was designed to absorb any x rays entering the cavity region of the device. Figure 31 shows how the beam scatters for the type VI insert. The signal from the insert takes the form $N_i = N_h$ since $N_s \approx 0$, and $N_b = N_e + N_h + N_i$.

Figure 29. The signal components for a sample and holder.

Figure 30. The signal components for an empty sample holder.

Figure 31. The signal components for type VI insert.

4.2 Diffraction Experiments

4.2.1 X-ray Tube Settings

The Rigaku machine uses a cobalt target x-ray tube. The power supply was set to a potential and a current of 30 kVp and 20 mA respectively (unless otherwise stated these are the potential and current settings for all Rigaku measurements). These settings will produce a polyenergetic spectrum consisting of bremsstrahlung and characteristic radiation. The characteristic lines of cobalt are shown in Table 4.

Characteristic line	Wavelength (nm)	Energy (keV)
$K\alpha_1$	0.1792	6.937
$K\alpha_2$	0.1788	6.952
$K\beta$	0.1620	7.673

Table 4. Characteristic K lines for the cobalt spectrum [Ref.28 Chp.5 p.7].

The Scintag machine uses a copper target x-ray tube. The power supply was set to a potential and a current of 45 kVp and 40 mA respectively (unless otherwise stated these are the potential and current settings for all Scintag measurements). The characteristic lines of copper are shown in Table 5.

Characteristic line	Wavelength (nm)	Energy (keV)
$K\alpha_1$	0.1544	8.051
$K\alpha_2$	0.1540	8.072
$K\beta_1$	0.1392	8.930

Table 5. Characteristic K lines for the copper spectrum [Ref.28 Chp.5 p.7].

4.2.2 Diffractometer Alignment

The type I holder or the slit block is a device used to verify the alignment of the x-ray tube and detector. When the x-ray focal spot and detector are aligned the signal intensity should be symmetric and a maximum at 0° . These two conditions indicate proper alignment.

To verify that the Rigaku machine was aligned the following steps were taken: A slit block was placed in the sample chamber. The slit position of 1.0 cm from the base allows a portion of the focal spot to pass through the slit block. The holder's extra height of 0.15 cm blocks the top half of the x-ray beam from reaching the detector. The tube's potential and current were set to 10 kV and 10 mA respectively. These settings are necessary to protect the NaI detector from the primary beam of the x-ray tube. The detector and the x-ray tube were set to 0° . By selecting the manual oscillation program from the Rigaku program, the x-ray tube was made to oscillate about the 0° mark with an amplitude of 1° while the detector remained stationary. The data were collected every 5 seconds in 0.05° increments (designated as $0.05^\circ/5$ s). The same procedure was repeated for a stationary x-ray tube and an oscillating detector. To verify alignment for the Scintag machine the following steps were taken: The slit block was placed in the chamber. The potential and current were set to 11 kV and 1.0 mA respectively. The x-ray tube was set to scan from -1° to 1° for $0.05^\circ/5$ s. The procedure was repeated for the detector.

All measurements done on the Rigaku and Scintag machines were recorded in counts. The measured signal depends on Poisson statistics where the uncertainty was equal to the square root of the signal. The error of the angular variable was defined as $\frac{1}{2}$ the resolution setting of the machine. For example if the machine was set to scan in 0.05° increments then the error of each point was 0.025° . In most cases the angular uncertainty was considered too small to quote since most amorphous scans have features at least 10° wide.

Figures 32 and 33 show the alignment curves for the Rigaku tube and detector. Both curves have a maximum count signal of $10^6 \pm 10^3$ at 0° . The signal drops to zero at $\pm 0.5^\circ$. Another peak was detected with a count signal of 3.0×10^5 at 1.0° . The main peak is symmetric which shows that the x-ray focal spot and detector were properly aligned at the 1.0 cm mark of the slit block. The secondary peak represents an upper portion of the x-ray focal spot escaping through the slit. Figures 34 and 35 show the alignment curves for the Scintag machine. The maximum signal at 0° was 1475 ± 38 counts for the tube and 1615 ± 40 for the detector. The curves are not as broad as the Rigaku data. The slit width of the Scintag block was 0.01 mm which was smaller than the Rigaku slit. This limits the Scintag's peak intensity and width. There are no secondary peaks for the Scintag curve.

Figure 32. Alignment peak for the Rigaku x-ray tube.

Figure 33. Alignment peak for the Rigaku detector.

Figure 34. Alignment peak for the Scintag x-ray tube.

Figure 35. Alignment peak for the Scintag detector.

4.2.3 Background Measurements

For all background and sample measurements the x-ray tube and detector were set to both rotate at an angle $\theta/2$ with respect to the sample chamber. This scanning technique is called $\theta/2 - \theta/2$ mode.

4.2.3.1 External Radiation N_e

The x-ray tube of the Rigaku diffractometer was shut off. The diffractometer was rotated from 2-150°. The machine was set to collect data every 20 seconds per 0.1°. The external background for the Scintag machine was measured by performing the following: The potential and current were set to 10 kVp and 1.0 mA respectively. A lead shield of thickness 0.73 cm was placed in the path of the x rays. The machine was set to the same scan parameters as the Rigaku machine. Figure 36 shows the measurement of $N_t = N_e$ for the Rigaku machine. The plot shows no recognizable distribution. The count average was found to be $\langle N_e \rangle = 0.094$. For the Scintag machine $\langle N_e \rangle = 0.067$.

Figure 36. Rigaku measurement of N_e .

4.2.3.2 Sample Chamber Scatter

The background radiation was measured for the empty sample chambers of the Rigaku and Scintag machines. On the Rigaku machine the diffraction chamber was sealed. The power supply of the x-ray tube was set to the standard settings. The chamber was scanned from 2-150° at 0.1°/5s. The plot shown in Figure 37 shows the background scan of the Rigaku chamber. At $\theta = 5^\circ$ the count signal was 120. The signal levels out at 5 counts at $\theta = 45^\circ$. Within the angular interval 2-20° the majority of the x rays do not scatter off the chamber base but go directly into the detector (fringe radiation N_f). For the angular region 45-150° scattering occurs off the base of the chamber and not off the 1.0 cm sample level. Therefore the x-ray signal was not very strong, about 2-3 counts.

An empty chamber scan for the Scintag machine is shown in Figure 38. The fringe component N_f was detected within the angular region 2-20°. A signal maximum occurs at 25°. This results from increased scatter off the chamber base. At about 38° the pattern shows its first Bragg peak which results from coherent scattering off the steel base plate. A number of Bragg peaks occur between 43.6-150°.

Figure 37. Rigaku measurement of empty chamber.

Figure 38. Scintag measurement of empty chamber.

4.2.3.3 Sample Holder Background

The background signal, $N_b = (N_e + N_f) + N_h$, for the type III, IV and V sample holders were measured using the Rigaku and Scintag diffractometers as shown in Figure 30 p.55. The holders were scanned from 2-150° at 0.1°/5s. Figure 39 superimposes the Rigaku results for the empty chamber and the Al holders on a log-log scale. Figure 40 superimposes the Rigaku results for the empty chamber and the St holders on a log-log scale. The fringe component N_f for both the Al and St holders was lower than the empty chamber scan. When the holders are placed into the machine the front face of the holder blocks a portion of the beam from reaching the detector. This reduces the signal by about 10 counts. From 50-150° the x-rays scatter off the base of the holder. The component N_h was about 2-5 counts. However the background scan for the St III holder shows Bragg peaks at 50° and 56°. The edges of St III holder were wide enough for the beam to scatter off. The edges of St IV and St V were reduced to avoid the scatter peaks. The edges of St IV and St V were reduced to avoid the scatter peaks.

Figure 39. Rigaku N_f measurements for the empty chamber and Al type III, IV, V sample holders, measured as in Figure 30 p.55.

Figure 40. Rigaku N_t measurements of the empty chamber and St type III, IV, V sample holders.

The holder plots of Figures 39 and 40 were so alike that it was too difficult to separate out the holder with the lowest background. To solve this problem, the average ratio of the empty chamber (no holder) to holder signal was calculated for 2-20° and 80-150°. See Table 6. A ratio of 1 means that the empty chamber and holder signals are almost identical. A ratio greater than one means that the empty chamber signal is larger than the holder background. The holder which has the largest ratio at both angular regions was the Al V. The holder Al V had the lowest background signal and was used for all subsequent Rigaku scans.

Holder	Empty/Holder ratio for 2-20°	Empty/Holder ratio for 80-150°
Al III	1.36	1.10
Al IV	1.25	1.16
Al V	1.37	1.27
St III	1.15	1.00
St IV	1.37	1.06
St V	1.19	1.10

Table 6. Empty chamber to holder ratio for both the Al and St holders.

The Scintag signals for the empty sample chamber and the Al V holder were plotted on a log-log scale. See Figure 41. The signal N_f drops to zero at about 50° . From 50 - 125° the holder signal levels out at 5-10 counts. For the range 125 - 150° the signal climbs to 35 counts. This effect may be due the entire beam scattering off the base of the holder. At lower angles the beam is broad which reduces the number of photons striking the base of the holder. The other holder types did not fit in the Scintag machine, and therefore the type V holder was used for all subsequent x-ray scans.

Figure 41. Scintag N_f measurements of the empty chamber and Al type V sample holder.

4.2.3.4 Type VI Background

The background was measured for the type VI insert, where $N_t = N_b = N_h + (N_f + N_e)$ as in Figure 31 p.55. The inserts were placed in the chamber with their slats aligned perpendicular to the x-ray beam. Again the inserts were scanned for 2-150° at 0.1°/5s. Figure 42 shows the Rigaku scan for the type VI insert. Bragg peaks show up at 51.1°, 52.2°, 89.3°, 99.7°, 111.7° and 123.3°. These Bragg peaks may be the result of x rays scattering off the steel strips. Figure 43 compares the background signals of the Al V holder and type VI insert on a log-log plot. For 2-20° the low angle background was reduced by 41% when compared with the Al V holder.

Figure 44 shows the Scintag scan for the type VI insert. A small Bragg peak shows up at 48°. The insert background is compared with the Al3 holder by using a log-log plot. See Figure 45. For 2-20° the low angle background was reduced by 68% when compared with the Al V holder.

Figure 42. Rigaku N_t measurement of type VI insert.
measured as in Figure 31 p.55.

Figure 43. Rigaku log-log plot of Al V and type VI background.

Figure 44. Scintag N_t measurement of type VI insert.
measured as in Figure 31 p.55.

Figure 45. Scintag log-log plot of Al V and type VI background.

4.2.4 Correction Measurements

The projected beam area on the sample was analyzed for the Rigaku and Scintag machines. A piece of fluorescent screen was separated from a Kodak X-Omatic cassette. The screen was then shaped to fit the geometry of the type V sample cavity. The screen was then shimmed to the sample level of 1.0 cm. When the x rays interacted with the screen a projection of the beam was produced. It was determined that the beam became broad and faint for the angular region 2-20°. The area of the beam was not constant throughout the x-ray scan.

An experimental method was devised to extract a scaling function which would correct for the decrease in beam intensity at low angles. The method involves extracting the incoherent signal by removing the coherent peaks from a powdered diffraction pattern. By dividing out the calculated incoherent cross section one can extract a scaling function $\Lambda(\theta)$. This function is machine dependent which not only accounts for beam broadening but any polarization effects of the monochromator. The polarization function, Eq.(41) is replaced with the new correction function or $P(\theta,0) \Rightarrow \Lambda(\theta)$.

To determine the function $\Lambda(\theta)$ we must first measure the incoherent signal of a powder crystal. Three types of powder crystals were scanned: iodized NaCl, SiO₂ and graphite powder. Each sample was placed into the type V sample holder for both the Rigaku and Scintag machines. The sample was made level with the top of the holder. The scattered intensity was collected for 2-150° at 0.1°/5s.

Figure 46 shows the Rigaku diffraction plots of NaCl, SiO₂ and graphite for the low angle region 2-30° as in Figure 29 p.54. The graphite powder curve has a larger scatter component than the diffraction patterns of SiO₂ and NaCl. This may be due to the rough surfaces of SiO₂ or NaCl which act as x-ray absorbers. This systematic absorption is unwanted. If the material absorbs too many photons the ratio of coherent to incoherent scattering will not be accurate. Therefore the graphite pattern was chosen as the correction scan. See Figure 47.

Figure 48 shows the powder scans for the Scintag machine. Again low angle absorption was a maximum for NaCl and SiO₂. Figure 49 shows the Scintag graphite correction scan. The graphite scan for each machine shows a series of Bragg peaks. The first peak occurs at 30.7° and 26.4° for Rigaku and Scintag respectively. The peak shift is due to momentum transfer conservation. To verify this, $x_{\text{Scintag}} = 1.0/0.154 \sin(26.4/2) = 1.48 \text{ nm}^{-1}$ and $x_{\text{Rigaku}} = 1.0/0.179 \sin(30.7/2) = 1.48 \text{ nm}^{-1}$. Therefore $x_{\text{Rigaku}} = x_{\text{Scintag}}$.

Figure 46. Rigaku low angle comparison of NaCl, SiO₂ and graphite, measured as in Figure 29 p.54

Figure 47. Rigaku graphite diffraction pattern. N_p .

Figure 48. Scintag N_t comparison of NaCl, SiO_2 and graphite, measured as in Figure 29, p. 54.

Figure 49. Scintag graphite diffraction pattern, N_t .

4.2.5 Water Measurements

The maximum volume of the Al V sample holder is 4.60 ml. A 3 ml syringe (smallest gradation 0.10 ml) was used to inject liquids into the sample volume. A 0.5 ml syringe (smallest gradation 0.01 ml) was used for any fine volume adjustments. To prepare a liquid sample for a diffraction scan, 4.60 ml of distilled water were injected into the sample holder. The scattering volume which produces an optimum scatter signal was considered to be properly aligned with the incident x rays and detector. The diffractometer was set to measure the intensity as a function of sample volume. The Rigaku diffractometer was set to scan the sample in the range $31.0\text{--}32.0^\circ$ for $1^\circ/60\text{s}$. The Scintag machine was set to scan the sample in the range $27.0\text{--}28.0^\circ$ for $1^\circ/60\text{s}$. These scanning regions were chosen for proper beam coverage of the sample.

By using the syringes the volumes of both samples were varied in 0.1 ml increments accordingly. These measurements must be repeated for every new liquid sample to ensure proper alignment of the scattering volume. After the maximum count rate was determined, the Rigaku and Scintag water samples were scanned for $2\text{--}150^\circ$ at $0.1^\circ/5\text{s}$.

Figure 50 shows Rigaku results for water volume optimization. As the water volume increases the sample was brought closer to the scattering plane of the x-ray beam. This increases the scattering signal. The maximum diffraction signal of water occurs at 3.70 ml. As the volume passes the 3.70 ml mark the surface tension of the water causes the scattering surface to bulge over the 1.0 cm height of the holder. Only a slight bulge will shift the scattering surface out of alignment causing a signal decrease. The experiment could not pass the 3.90 ml mark due to holder volume limits.

Figure 50. Rigaku water volume optimization.

The Rigaku diffraction pattern for 3.70 ml of distilled water is shown in Figure 51. Each point of the plot was smoothed using an 11-point rectangular kernel. Fringe radiation, N_f , occurs between $2-20^\circ$. A broad maximum occurs between $30-35^\circ$ which represent the interference effects between neighbouring water molecules. From $100-150^\circ$ the water scan levels out. This angular range is known as the IAM region.

Figure 51. Rigaku distilled water diffraction pattern N_t .

Similarly for the Scintag machine. Figure 52 shows the result for water volume optimization. The plot is similar to Rigaku except the maximum calibration signal occurred at 3.50 ml. Figure 53 shows the Scintag diffraction plot of 3.50 ml of distilled water. The maximum diffraction signal occurs at about 29° . Note the maximum signal was three times larger than the Rigaku maximum due to the higher power output of the Cu x-ray tube. Again the signal has shifted to conserve momentum transfer.

Figure 52. Scintag water volume optimization.

Figure 53. Scintag distilled water diffraction pattern. N_t .

4.2.6 Tissue Measurements

A sample of beef muscle was scanned on the Rigaku diffractometer. The specimen was prepared by first cutting an oversized sample of beef. The Al V holder was then used as a cookie cutter to extract a portion of muscle that would fill the entire cavity. A glass slide was used to shave off any sections of the specimen that would protrude out of the sample holder. The sample was made level at 1.0 cm. The machine was set to scan from $2-150^\circ$ for $0.1^\circ/5s$. The raw data were smoothed by an 11-point rectangular kernel. Figure 54 shows the diffraction scan of beef muscle.

Figure 54. Rigaku beef muscle diffraction pattern. N_t

4.2.7 Plastic Measurements

The plastic samples Polystyrene, Lexan, Nylon, Polyethylene and PMMA were placed, in turn in the Rigaku diffractometer. The machine was set to scan from $2-150^\circ$ for $0.1^\circ/5s$. The Scintag machine scanned the plastic samples Polystyrene and PMMA for the same scan parameters as the Rigaku. All data sets were smoothed using an 11 point rectangular kernel. Rigaku diffraction scans N_i are shown in Figures 55-59. Scintag measurements are shown in Figures 60-61. For the Rigaku and Scintag scans the IAM region is still between $100-150^\circ$.

Figure 55. Rigaku polystyrene diffraction pattern. N_i .

Figure 56. Rigaku Lexan diffraction pattern, N_t .

Figure 57. Rigaku Nylon diffraction pattern, N_t .

Figure 58. Rigaku polyethylene diffraction pattern. N_t .

Figure 59. Rigaku PMMA diffraction pattern. N_t .

Figure 60. Scintag polystyrene diffraction pattern. N_t .

Figure 61. Scintag PMMA diffraction pattern. N_t .

Chapter 5

Calculations

5.1 Background Extraction

It was determined in the previous chapter that the type VI insert produced the lowest background signal for both the Rigaku and Scintag machines. Localized Bragg peaks were removed from the background scans by using the program mwpeak.for. The program starts to read in the data where no fringe and coherent radiation occur. This is considered the baseline of the background scan. The program then calculates the signal difference.

$$\Delta N = N_i - N_{i+j}, \quad j = 1, 2, 3, \dots, n - i \quad (43)$$

for all subsequent $(i + j)$ th data points. To determine if successive data points N_{i+j} are peak values, Eq.(43) is set to the following condition: $\Delta N \geq \epsilon$, where ϵ is the magnitude of the baseline. If the signal difference is less than ϵ the data point N_{i+j} is not a peak value. The program will then reset the position of the i th data point to the $(i+j)$ th position and retest the ϵ condition for the next data subset. If the signal difference is greater than ϵ the N_{i+j} data point is defined as a peak value. The data point is replaced with the marker $\pi = 3.1415$ which flags the position of the peak. The final section of the program replaced the π markers with an average of 10 points on either side of the marked peaks. The background signals were then smoothed by an 11-point averaging function. Figures 62 and 63 show the Rigaku and Scintag results. Note this program will only work for narrow peaks. If the peaks become too broad the signal difference between the baseline and peak points will fall below the parameter ϵ . The program will then define the new base line, N_i , on a peak position. The end result is partially removed Bragg peaks.

Figure 62. Rigaku – This is the data set of Chp. 4. Figure 42 p.66 with the peaks removed and 11-point smoothed

Figure 63. Scintag – This is the data set of Chp.4 Figure 44 p.67 with the peaks removed and 11-point smoothed.

5.2 Corrections

The following calculations were performed to extract the correction factor $\Lambda(\theta)$. The smoothed background obtained in the previous section was subtracted from the graphite diffraction scan,

$$N_{sg} = N_{tg} - N_b, \quad (44)$$

where N_{sg} and N_{tg} are the scatter and total signals of graphite respectively. After the background was subtracted the coherent peaks were removed from the graphite pattern. Due to graphite's broad Bragg peaks the peak removal program could not accurately separate the coherent signal from the diffraction scan. This problem was solved by identifying each Bragg peak and manually removing the signal.

Figures 64 and 65 show the Rigaku and Scintag diffraction patterns, respectively, with the coherent peaks removed. Each plot starts out with a fringe component N_f . The Rigaku plot shows four data clusters at 48° , 80° , 120° and 150° . The Scintag plot shows clusters at 35° , 75° , 98° and 125° . There is no data cluster for the angle 150° since there was a series of coherent peaks ranging from 125° - 150° . The data clusters shown in both plots represent portions of the incoherent signal of graphite.

$$N_{sg} = K(1 + \cos^2 \theta)[F_{KN}S(x)]\Lambda(\theta), \quad (45)$$

where $F^2(x) \approx 0$ and $S(x)$ is the incoherent form factor for the carbon atom, tabulated by Hubbell et al [Ref.6].

Figure 64. Rigaku N_g data clusters – These are the data of Figure 47 p.70 with the background and peaks removed.

Figure 65. Scintag N_g data clusters - These are the data of Figure 49 p.71 with the background and peaks removed.

To obtain an analytic form of the incoherent signal three different polynomial curves were fit to the data clusters.

$$f_1(\theta) = \frac{A}{\theta^n} + B, \quad (46a)$$

$$f_2(\theta) = \frac{A}{\theta^n} + B + C\theta, \quad (46b)$$

$$f_3(\theta) = \frac{A}{\theta^n} + B + C\theta + D\theta^2. \quad (46c)$$

The program mwfit.for calculated the parameters A, B, C, D and n by using the method of least squares (MLS). For each new value of n the reduced $\chi_r^2 = \chi^2/(X-Y)$ was calculated where X and Y are the number of data points and degrees of freedom respectively. For example the number of degrees of freedom for Eq.(46c) is $Y=4$ (A,B,C,D). Tables 7 and 8 display the Rigaku and Scintag fit parameters respectively, for the polynomials f_1 , f_2 and f_3 . Tables 7 and 8 show that the polynomial, f_3 , has the closest fit to the graphite clusters. Figures 66 and 67 show the polynomials f_1 , f_2 and f_3 fitted to the graphite data clusters.

Polynomial	A ± 62	B ± 0.13	C $\pm 1.54 \times 10^{-3}$	D $\pm 1.0 \times 10^{-5}$	n ± 0.1	χ_r^2
f_1	317.0	5.39	N/A	N/A	1.0	7.96
f_2	2695.0	11.51	-0.009	N/A	2.3	4.78
f_3	4578.0	27.16	-0.462	0.003	2.9	3.36

Table 7. Fit parameters for the Rigaku data clusters.

Polynomial	A ± 252	B ± 0.38	C $\pm 4.59 \times 10^{-3}$	D $\pm 4.0 \times 10^{-5}$	n ± 0.1	χ_r^2
f_1	718.2	83.60	N/A	N/A	1.0	21.8
f_2	15467.7	76.00	0.314	N/A	2.9	5.85
f_3	31725.2	121.10	-1.402	0.012	3.7	3.99

Table 8. Fit parameters for the Scintag data clusters.

Figure 66. The polynomials f_1 , f_2 and f_3 superimposed on the Rigaku graphite clusters.

Figure 67. The polynomials f_1 , f_2 and f_3 superimposed on the Scintag graphite clusters.

Equation 45 and Eq.(46) both represent the incoherent scatter of graphite. Therefore let $N_{sg} = f(\theta)$. By absorbing the scaling factor K into $\Lambda(\theta)$, we have

$$\Lambda(\theta) = \left(\frac{f(\theta)}{(1 + \cos^2(\theta))[F_{KN}S(x)]} \right). \quad (47)$$

The extraction of $\Lambda(\theta)$ was done by the program mwlambda.for. This program performs background subtraction, interpolation of our data to Hubbell's incoherent form factors and finally the calculation of Eq.(47) for the polynomials f_1 , f_2 and f_3 . The inverse of Eq.(47), $\Lambda^{-1}(x)$, is plotted in Figures 68 and 69 for the Rigaku and Scintag machines respectively. Equation 47 was expressed as a function of x which normalized the Rigaku and Scintag data to the same scale. Each figure shows how a different polynomial fit changes the shape of the correction factor.

Figure 68. Rigaku correction curves for f_1 , f_2 and f_3 polynomials.

Figure 69. Scintag correction curves for f_1 , f_2 and f_3 polynomials.

5.3 Coherent Scatter Form Factor Extraction

To extract the coherent scatter form factor the following steps were performed. The background signal N_b was subtracted from the total signal N_t . The data were corrected for polarization and beam misalignment by taking the product,

$$N_s = N_t \Lambda^{-1}(\theta). \quad (48)$$

An expression for N_s can also be obtained by substituting $N_t = K(1+\cos^2\theta)[F^2(x) + F_{KN}(\alpha,\theta)S(x)]\Lambda(\theta)$ into Eq.(48).

$$N_s = K(1+\cos^2\theta)[F^2(x) + F_{KN}(\alpha,\theta)S(x)]. \quad (49)$$

By dividing out the term $(1+\cos^2\theta)$ we are left with the task of calculating the constant K . The constant K was calculated by applying IAM normalization. At high x values the coherent scatter term of Eq.(49) will approach the free atom model.

$$N_s \rightarrow K(1+\cos^2\theta)[F_{IAM}^2(x) + F_{KN}(\alpha,\theta)S(x)], \quad (50)$$

where $F_{IAM}(x)$ is the IAM form factor calculated from Eq.(32) [Chp.2 Sec.2.2.2]. The constant K_i was calculated for each data point N_{s_i} where the indices $i = M_0 \dots M_1$ lie within the high x region. If the signal in the high x -region is approximately constant then the K_i values can be averaged.

$$K = \frac{1}{M_1 - M_0} \sum_{i=M_0}^{M_1} \frac{N_{s_i}}{(1+\cos^2\theta_i)[F_{IAM}^2(x_i) + F_{KN}(\alpha,\theta)S(x_i)]}. \quad (51)$$

By substituting the value K obtained in Eq.(51) into Eq.(49) and then solving for $F(x)$ we then have for the coherent scatter form factor, for any value x .

$$F(x) = \left[\frac{N_s}{K(1+\cos^2\theta)} - F_{KN}S(x) \right]^{\frac{1}{2}} \left(\frac{\text{free } e^-}{\text{bound } e^-} \right)^{\frac{1}{2}}. \quad (52)$$

The form factors were extracted using the program `mwform.for`. Figure 70 summarizes the form factor calculation.

Figure 70. Flow chart of coherent scatter form factor extraction.

5.3.1 Coherent Scatter Form Factor: Water

To extract the coherent scatter form factor of water one must first calculate the IAM for H₂O. By applying the IAM Eq.(32) to the water molecule and then dividing by the number of bound electrons in H₂O (Z=10), the square of the coherent form factor per bound electron can be written

$$F^2_{IAM}(H_2O) = \frac{1}{10} [2F^2_H(x) + F^2_O(x)] \left(\frac{\text{free } e^-}{\text{bound } e^-} \right). \quad (53)$$

Similarly by using Eq.(17) [Chp.2 Sec.2.1.2.2] the incoherent form factor per bound electron is,

$$S(H_2O) = \frac{1}{10} [2S_H(x) + S_O(x)] \left(\frac{\text{free } e^-}{\text{bound } e^-} \right). \quad (54)$$

To calculate the constant K the above equations are substituted into Eq.(51). The summation of Eq.(51) was carried out over the x-range 4.75-5.39 nm⁻¹ for both the Rigaku and Scintag data sets. The quoted x regions represent the tail ends of the diffraction patterns. These x-values were chosen to ensure free atom behaviour. After the form factor has been extracted an 11-point averaging function was applied to smooth the data. The form factor extraction was repeated for the different polynomial fits yielding F₁(x), F₂(x) and F₃(x). Figure 71 shows the Rigaku coherent scatter form factor of water expressed as the square root of a free electron per bound electron. Additionally the coherent free atom form factors are shown for H₂O. Figure 72 shows the Scintag coherent scatter form factors of water.

Figure 71. Rigaku coherent scatter form factor for water using the f_1 , f_2 and f_3 polynomials.

Figure 72. Scintag coherent scatter form factor for water using the f_1 , f_2 and f_3 polynomials.

By examining the Rigaku form factors the shape and height of the curve changes drastically which depends on the type of polynomial used in the correction procedure. As stated in section 5.2 the closest polynomial fit to the graphite clusters is f_3 . This indicates that the most accurate coherent scatter form factor is $F_3(x)$. The peak amplitude of F_3 differs from F_2 and F_1 by about 0.5 and 1.0 respectively. Additionally the peak structure becomes distorted as the height increases. Finally the F_3 plot is higher than the IAM prediction except in the normalization region of 4.79-5.39 nm^{-1} . The plots F_2 and F_1 follow the IAM curve until 5.0 nm^{-1} . The curves rise steeply for $x = 5.0-5.39 \text{ nm}^{-1}$.

The Scintag form factors show little change in shape and amplitude for the different correction factors. According to section 5.2, f_3 is the best fit to the graphite data. Again $F_3(x)$ is the most accurate coherent scatter form factor. In this case F_2 has the highest peak when compared with F_1 and F_3 . For the x -range 3.0-4.0 nm^{-1} the form factors moderately agree with F_3 having the largest amplitude. Again the curves F_1 and F_2 rise steeply for 5.0-6.39 nm^{-1} .

The F_3 form factors are considered the best choice for each machine. When the F_3 form factors are compared with each other there is a drastic disagreement in shape and amplitude. This places doubt on the choice of polynomial, f_3 , used to extract the correction function. The polynomial f_3 fits the data points but may not correctly predict the incoherent baseline. For example in Figure 67 the Scintag incoherent signal shows a quadratic increase in the region 125-150°. This increase may suggest that the signal has coherent structure which is not apart of the incoherent baseline. According to Figures 71 and 72 the $F_2(x)$ form factors agree with respect to shape and amplitude. Apparently the f_2 polynomial predicts the incoherent baseline since its linear term ignores the quadratic increase of the graphite signal. Therefore the correction factor $\Lambda^{-1}_2(x)$ will be used for all subsequent Rigaku and Scintag form factor calculations.

5.3.2 Coherent Scatter Form Factor: Beef Muscle

The Rigaku coherent scatter form factor for beef muscle was obtained by normalizing the scattering data in the x-region $4.75\text{--}5.39\text{ nm}^{-1}$. Figure 73 shows the form factor results for beef muscle and its respective IAM curve. The beef form factors are very similar to that of water having a maximum amplitude of 2.0 at 1.60 nm^{-1} . For $x = 4.0\text{--}5.39\text{ nm}^{-1}$ the plot lies below the IAM region. At $x = 5.0\text{ nm}^{-1}$ the plot starts to steadily increase which is due to the steep behavior of the correction factor.

Figure 73. Rigaku coherent scatter form factor for beef muscle.

5.3.3 Coherent Scatter Form Factor: Plastics

The coherent scatter form factors for plastics were extracted by normalizing the Rigaku and Scintag scattering data in the x region 4.75 - 5.39 nm^{-1} . Figures 74-75 show the coherent form factors of polystyrene and PMMA respectively for Rigaku and Scintag data sets. Figures 76-78 show the Rigaku form factors for polyethylene, nylon and lexan.

Figure 74. Rigaku and Scintag coherent scatter form factor for polystyrene.

Figure 75. Rigaku and Scintag coherent scatter form factor for PMMA.

Figure 76. Rigaku coherent scatter form factor for polyethylene.

Figure 77. Rigaku coherent scatter form factor for nylon.

Figure 78. Rigaku coherent scatter form factor for lexan.

Chapter 6

Comparison and Conclusions

6.1 Rigaku and Scintag Comparison

To test the accuracy of our method the coherent scatter form factors of Rigaku and Scintag are compared with each other. This is done by taking the form factor ratio, $F_R(x) / F_S(x)$ (Rigaku) / $F_S(x)$ (Scintag), of water and PMMA. The ratios are plotted as a function of x where $x = 0.10 - 5.39 \text{ nm}^{-1}$.

Consider the water ratio plotted in Figure 79. In the x -region $0.10\text{-}1.0 \text{ nm}^{-1}$ the plot oscillates erratically. The peak amplitude is about 3.4 at $x = 0.3 \text{ nm}^{-1}$ which indicates that $F_R > F_S$. For example the Scintag and Rigaku functions at 0.3 nm^{-1} are $\Lambda_S^{-1}(x = 0.3) = 0.00018$ and $\Lambda_R^{-1}(x = 0.3) = 0.00072$ respectively. The lower Scintag correction results in a reduced form factor amplitude. A small valley occurs at about $x = 0.58 \text{ nm}^{-1}$ where $F_R < F_S$. At $x = 1.0 \text{ nm}^{-1}$ the oscillations are damped out. The form factor disagreement at low x is related to systematic uncertainty. When the background is subtracted off the graphite patterns, there still remains a large signal or fringe component. When this signal is used to extract $\Lambda_2(x)$ in the x range $0.10\text{-}1.0 \text{ nm}^{-1}$ the function becomes inflated to very large numbers. This results in reduced form factor amplitudes. This uncertainty can only be reduced by finding an alternate method to eliminate or degrade the fringe component N_f .

From $1.0\text{-}5.39 \text{ nm}^{-1}$ the form factors start to agree. The average ratio is about 0.88. This indicates that the Rigaku form factors are slightly lower than the Scintag results. This difference is small enough to suspect the accuracy of the polarization correction. A standard polarization correction is at most 20 %. The average ratio lies within this correction boundary.

Figure 79. Water form factor ratio of Rigaku to Scintag.

The plastic ratios are shown in Figure 80. For the x-region $0.10 - 1.0 \text{ nm}^{-1}$ the PMMA ratio shows form factor oscillations similar to that of water. These oscillations are not large with a maximum value of 0.5 about the 1.0 value. These oscillations may be due to the systematic uncertainties in the background measurements which translates into an inflated correction function $\Lambda(x)$.

From $x = 1.0 - 5.39 \text{ nm}^{-1}$ the average ratio for the PMMA sample is 1.03. This indicates a good curve agreement between the Rigaku and Scintag plots. Again any disagreements may be due to polarization corrections. The ratio calculated for polystyrene exhibits similar behaviour which indicates machine agreement.

Figure 80. PMMA and polystyrene form factor ratios of Rigaku to Scintag

6.2 Comparison to the Literature

6.2.1 Coherent Scatter Form Factor Comparison: Water and Beef Samples

The biological coherent scattering form factors are compared to previous measurements performed by Narten et al [Ref.5], Kosanetzky et al [Ref.14] and Peplow et al [Ref.16]. The form factor data for each author are interpolated to our tabulated results and compared using a percent difference,

$$\% \text{Diff} = \sum_{i=1}^n \frac{|N_i(\text{experiment}) - N_i(\text{literature})|}{nN_i(\text{literature})} \times 100. \quad (55)$$

where N_i (experiment) represents our experimental data and N_i (literature) represents the interpolated published data. The percent difference is calculated for the Rigaku and Scintag x ranges $0.102 - 5.39 \text{ nm}^{-1}$ and $0.102 - 6.27 \text{ nm}^{-1}$ respectively. Tables 9 and 10 list the percent differences calculated for water and beef muscle.

Material	Author	x-range comparison(nm^{-1})	Percent Difference %
Water	Narten ^a	0.102 – 5.39	26.9
	Kosanetzky ^b	0.102 – 5.39	18.6
	Peplow ^c	0.102 – 5.39	20.7
Beef muscle	Kosanetzky ^b	0.102- 5.39	30.3
	Peplow ^c	0.102 - 5.39	23.0

Table.9 Rigaku water and muscle form factors compared to the published literature.
^a [Ref.5], ^b [Ref.14], ^c [Ref.16].

Material	Author	x-range comparison(nm^{-1})	Percent Difference %
Water	Narten ^a	0.102 - 6.27	28.4
	Kosanetzky ^b	0.102 - 6.27	22.5
	Peplow ^c	0.102 - 6.27	21.1

Table 10. Scintag water form factors compared to the published literature.
^a [Ref.5], ^b [Ref.14], ^c [Ref.16].

The percent difference calculated for the Rigaku-Narten comparison of water is 26.9%. To further analyze the form factors three x-regions of the Rigaku curve are studied, low $x = 0.1 - 1.2 \text{ nm}^{-1}$, middle $x = 1.2 - 2.8 \text{ nm}^{-1}$ and high $x = 2.8 - 5.39 \text{ nm}^{-1}$. Figures 81 and 82 plot the results for water from experiment and the literature.

The Rigaku-Narten percent difference for the x-region $0.102 - 1.2 \text{ nm}^{-1}$ is 66.0 %. This large disagreement could be again caused by inaccurate background measurements for both the sample data and the function $\Lambda^{-1}(x)$. When the background signal is subtracted from the graphite pattern a fairly large signal remained. After the coherent peaks are removed the data are divided by the incoherent cross section where $S(x \rightarrow 0) \rightarrow 0$. This behaviour amplifies the scattering data which causes $\Lambda^{-1}(x \rightarrow 0) \rightarrow 0$. In theory a perfect background subtraction would remove the entire signal, $N_h + N_f$, from the graphite pattern. The remaining scatter signal would be miniscule. When the incoherent cross section is divided out the function $\Lambda^{-1}(x)$ will not approach zero as quickly. Narten et al [Ref.5] dealt with this low angle problem by assuming a finite form factor value at $x = 0$. These form factors are calculated using bulk thermodynamic principles.

The second region lies between $1.2 - 2.8 \text{ nm}^{-1}$. The Rigaku-Narten percent difference for this x-region is 12.2 %. In this case the peak amplitudes do agree with respect to height and shape. The Rigaku plateau region from $2.2 - 2.4 \text{ nm}^{-1}$ does not match to Narten et al [Ref.5], Kosanetzky et al [Ref.14] or Peplow [Ref.16]. This problem could result from an improper fit of the incoherent signal used to extract the function $\Lambda^{-1}(x)$. To extract the incoherent signal one must first remove the coherent peaks. Since the θ positions of the coherent peaks were not available any signal that resembled a sharp peak was removed from the pattern. This method of extraction could have inadvertently removed parts of the incoherent signal which would introduce systematic uncertainty into the polynomial fit. For the Rigaku graphite pattern this uncertainty is on the order of 5-10 counts.

The final region lies between $2.8 - 5.39 \text{ nm}^{-1}$. The Rigaku-Narten percent difference for this x-region is 7.79 %. This part of the curve shows the best agreement with the authors IAM region. There is a slight height discrepancy in the region $5.0 - 5.39 \text{ nm}^{-1}$ where the experimental plot lies slightly above the IAM prediction.

The percent difference calculated for the Scintag-Narten comparison of water is 28.4%. This value is slightly worse than the Rigaku results. Again three x-regions are analyzed for the Scintag plot. The first region lies between $0.112 - 1.2 \text{ nm}^{-1}$. The Scintag-Narten percent difference over this range is 61.8 %. This difference is due to the inaccurate background subtraction of the graphite diffraction pattern which forces the correction factor $\Lambda^{-1}(x \rightarrow 0) \rightarrow 0$, as explained for the Rigaku extraction process.

The second region lies between $1.2 - 2.8 \text{ nm}^{-1}$. The Scintag-Narten percent difference calculated over this range is 14.7 %. This good agreement can be seen with respect to Scintag's peak amplitude and shape. Scintag's plateau in the region $2.2 - 2.4 \text{ nm}^{-1}$ does not match well with Narten et al [Ref.5], Kosanetzky et al [Ref.14] or Peplow et al [Ref.16]. Again the differences in amplitude are very slight which indicates uncertainty in the polarization correction.

The final region lies between $2.80 - 6.27 \text{ nm}^{-1}$. The Scintag-Narten percent difference over this range is 15.3 %. There is good IAM agreement with the published literature in the x-range $4.0 - 5.39 \text{ nm}^{-1}$. At $x = 5.0 \text{ nm}^{-1}$ the plot climbs steadily above the IAM curve. This variation may be caused by the increasing scatter signal at high x which is more pronounced for the Scintag data set.

Figure 81. Rigaku and Scintag coherent scatter form factors for water compared to Narten [Ref.5] and Kosanetzky et al [Ref.14].

Figure 82. Rigaku and Scintag coherent scatter form factors for water compared to Peplow et al [Ref.16].

The coherent scatter form factor for beef muscle is analyzed. The Rigaku-Kosanetzky percent difference over the x range $0.102 - 5.39 \text{ nm}^{-1}$ is 23.2 %. Figure 83 compares the experimental data with results from Kosanetzky et al [Ref.14] and Peplow et al [Ref.16]. Again the three x -regions of the Rigaku data are compared to the literature.

The Rigaku-Kosanetzky percent difference over the x -region $0.1 - 1.2 \text{ nm}^{-1}$ is 65.1 %. The experimental curve lies below the published results which may be due to the correction function behaviour $\Lambda^{-1}(x \rightarrow 0) \rightarrow 0$.

For the x region $1.2 - 2.8 \text{ nm}^{-1}$ the Rigaku-Kosanetzky percent difference is 10.6 %. For this region the peak amplitude of the beef lies below the published literature. Additionally the shape of the Rigaku curve does not match properly. The uncertainty in polarization corrections cannot account for the sizeable difference in peak amplitudes. Since the maximum amplitude of the beef muscle is reduced this may be a problem of sample leveling. If the beef sample is slightly protruding over the sample level of 1.0 cm. the signal could be reduced by about 100 - 200 counts. This problem could be fixed by performing the optimization procedure used for the water samples.

Finally the Rigaku-Kosanetzky percent difference for the x region $2.80 - 5.39 \text{ nm}^{-1}$ is 4.02 %. This shows good agreement with the IAM region of the published literature. Again the Rigaku plot rises above the IAM prediction at $x = 5.00 - 5.39 \text{ nm}^{-1}$ which may be due to increased high angle scatter.

Figure 83. Rigaku coherent scatter form factor for beef muscle compared to Kosanetzky et al [Ref.14] and Peplow et al [Ref.16].

6.2.2 Coherent Scatter Form Factor Comparison: Plastic Samples

The percent differences for the plastics are shown in Tables 11 and 12. Again we will study the curve characteristics of the low, middle and high x-regions of the plastics.

Material	Author	x-range comparison(nm^{-1})	Percent Difference %
PMMA	Kosanetzky ^a	0.102 - 5.39	18.6
PMMA	Peplow ^b	0.102 - 5.39	21.0
Polystyrene	Kosanetzky ^a	0.102 - 5.39	22.9
Polyethylene	Kosanetzky ^a	0.102 - 5.39	24.0
Nylon	Kosanetzky ^a	0.102 - 5.39	19.0
Lexan	Kosanetzky ^a	0.102 - 5.39	25.9
Lexan	Peplow ^b	0.102 - 5.39	23.0

Table 11. Rigaku plastic form factors compared to the literature.
^a [Ref.14], ^b [Ref.16].

Material	Author	x-range comparison(nm^{-1})	Percent Difference %
PMMA	Kosanetzky ^a	0.117 – 6.27	17.2
PMMA	Peplow ^b	0.117 – 6.27	21.5
Polystyrene	Kosanetzky ^a	0.117 – 6.27	21.2

Table 12. Scintag plastic form factors compared to the literature,
^a [Ref.14], ^b [Ref.16]

The percent difference for the Rigaku-Kosanetzky PMMA form factors is 18.6 %. Figure 84 shows the Rigaku form factors compared to the literature. For the low x-region of 0.102 - 1.2 nm^{-1} the percent difference is 48.2 %. The plastic PMMA shows interference at angles lower than that of water. The first peak occurs at about 0.8 nm^{-1} . This peak is reduced by the correction factor $\Lambda_2^{-1}(x)$. For the x-region 1.2 - 2.8 nm^{-1} the percent difference is 2.5 %. The Rigaku amplitude starts to follow the height and shape of the published curves. The final region $x = 2.80 - 5.39 \text{ nm}^{-1}$ shows a percent difference of 11.6 %. The comparison would be better if not for a slight amplitude decrease in the x-range 3.0 - 4.0 nm^{-1} . The Rigaku comparison in the normalization range, 4.75 - 5.39 nm^{-1} , matches to results of Kosanetzky et al [Ref.14].

The Scintag - Kosanetzky percent difference for PMMA is 17.2%. For the x-region 0.10 - 1.2 nm^{-1} the percent difference is 47.6 %. Again the Scintag correction factor is reducing the size of the first peak. At 1.20-2.8 nm^{-1} the percent difference is 4.80%. At 2.80 - 6.27 nm^{-1} the percent difference is 6.09 %. The rises in the Scintag curve may be due to an increased scatter signal at high x.

Figure 84. Rigaku and Scintag coherent scatter form factors for PMMA compared to Kosanetzky et al [Ref.14] and Peplow et al [Ref.16].

The Rigaku – Kosanetzky percent difference for polystyrene over the x -range $0.102 - 5.39 \text{ nm}^{-1}$ is 22.9 %. Figure 85 shows the form factor results for Rigaku, Scintag and Kosanetzky et al [Ref.14]. For the low x -range of $0.102 - 1.2 \text{ nm}^{-1}$ the percent difference is 36.8 %. At about $x = 0.6 \text{ nm}^{-1}$ a plateau feature on the Kosanetzky et al [Ref.14] curve seems to have been smoothed out on the Rigaku plot. The reduction in amplitude height and/or structure is caused by the correction function's behaviour at low x values. For the x -range $1.2 - 2.8 \text{ nm}^{-1}$ the percent difference is 14.4 %. Even though the amplitudes agree at $x = 1.2 \text{ nm}^{-1}$, the Rigaku curve levels out at a higher amplitude at about $x = 2.4 \text{ nm}^{-1}$. This difference could be the result of an inaccurate polarization correction. The Rigaku - Kosanetzky percent difference for the x -range $2.80 - 5.39 \text{ nm}^{-1}$ is 10.7%.

The Scintag - Kosanetzky percent difference for polystyrene is 21.2 %. The Scintag curve lies slightly below the Rigaku and Kosanetzky et al [Ref.14] curves. For the low x -region of $0.102 - 1.2 \text{ nm}^{-1}$ the percent difference is 40.6 %. Again the plot is suppressed by the Scintag correction factor.

For the x -region $1.2 - 2.8 \text{ nm}^{-1}$ the percent difference is 5.34 %. For this x -range the Scintag plot does not match the height of the Rigaku or published literature. In the x range $2.80 - 6.27 \text{ nm}^{-1}$ the Scintag-Kosanetzky percent difference is 12.6 %. At $x = 5 \text{ nm}^{-1}$ the Scintag plot rises above the IAM prediction which may be a result of increased high angle scatter.

Figure 85. Rigaku and Scintag coherent scatter form factors for polystyrene compared Kosanetzky et al [Ref.14].

The coherent scatter form factors for polyethylene, nylon and lexan are compared in Figures 86 - 88. The Rigaku - Kosanetzky percent difference for polyethylene is 24%. Despite such a sizeable percent difference there is still excellent amplitude agreement for the polyethylene peaks at 1.2 nm^{-1} and 1.4 nm^{-1} . For the x range $2.0 - 5.39 \text{ nm}^{-1}$ the

polyethylene pattern shows multiple peaks which indicates short range structure (large x , short range). To extract the form factor we must normalize the data to the free atom region. These high x peaks lie within the normalization range of $4.75 - 5.39 \text{ nm}^{-1}$ which calls into question the accuracy of the normalization constant K . This problem could be fixed by normalizing the data to higher x values.

The Rigaku - Kosanetzky percent difference for Nylon is 19%. The x range of $0.1 - 1.2 \text{ nm}^{-1}$ shows a reduced amplitude and/or loss of peak structure. This is caused by the behaviour of the function $\Lambda^{-1}(x \rightarrow 0) \rightarrow 0$. Higher x values show good amplitude agreement at about 1.1 nm^{-1} and 1.4 nm^{-1} . The lexan plot shows the worst agreement with a Rigaku-Harding percent difference of 25.9 %. Every section of the curve lies below the results of Kosanetzky et al [Ref.14] or Peplow et al [Ref.16].

Figure 86. Rigaku coherent scattering form factors for polyethylene compared to Kosanetzky et al [Ref.14].

Figure 87. Rigaku coherent scattering form factors for Nylon compared to Kosanetzky et al [Ref.14].

Figure 88. Rigaku coherent scattering form factors for lexan compared to Kosanetzky et al [Ref.14] and Peplow et al [Ref.16].

6.3 Concluding Remarks and Future Work

6.3.1 Concluding Remarks

It was determined that the signal measured from the type VI grid was a reasonable approximation to the background signal $N_b = N_h + N_f + N_e$. The background signal could be improved by reducing the scatter component N_h (scatter off the holder). The component N_h could be reduced by decreasing the thickness of the holder's foil slats. This would reduce the scattering area of the holder which will curtail the production of Bragg peaks.

Another way of reducing the background signal is to eliminate or decrease the fringe component N_f . As mentioned in Chapter 4 the fringe component results from the upper portion of the primary beam entering the detector at low angles. This effect could be reduced by using a smaller slit width. For example the Rigaku machine uses a 1° divergence slit to spread the x-ray beam onto the sample. In the angular region $\theta = 2 - 20^\circ$ the slit divergence is too large to prevent a portion of the beam from entering the detector. By reducing the divergence slit to $\frac{1}{4}$ the size the beam will irradiate a smaller sample area (reduced signal intensity). As a consequence the primary beam will enter the detector only at much smaller angles. Another device which could reduce the signal N_f is called a variable divergence slit. The width of the divergence slit changes in order to keep the beam area on the sample a constant. A variable divergence slit was used on the diffractometer of Kosanetzky et al [Ref.14].

After the background was subtracted, the scattered signal needed to be corrected for beam misalignment and polarization. The correction function $\Lambda_2(x)$ obtained in chapter 5 showed that a majority of the primary photons at low angles were entering the detector and not scattering off the sample. This signal was suppressed by taking the product $N_s \Lambda_2^{-1}(x)$ which would correct for beam broadening and misalignment effects. For $x > 5 \text{ nm}^{-1}$ the correction curve increased in a linear fashion. This increase may be caused by misalignment where there is a lack of photons scattering at high angles.

To increase the accuracy of the correction method the background extraction must be improved. This would give us an accurate incoherent signal at low angles which would improve the correction factor via the polynomial fit. Additionally when the Bragg peaks are removed from the incoherent baseline a knowledge of their peak positions would aid in separating the coherent from the incoherent scatter.

When the Rigaku and Scintag coherent scatter form factors for water were compared to the literature the percentage differences were never better than 18.6 % (Rigaku – Kosanetzky et al [Ref.14] percent difference). The experimental data in the region 0.1 - 1.2 nm⁻¹ showed the greatest deviation of 67% (Rigaku - Narten percent difference). This indicates that the $\Lambda_2^{-1}(x)$ may be over-correcting the scattering data.

The form factors for plastics never showed a better percent difference than 17.2 % (Scintag - Kosanetzky et al [Ref.14] percent difference of nylon). The task of obtaining form factor agreement for plastics was more difficult than that of water or tissue. Most plastic diffraction patterns show coherent peaks at x-values less than 1.0 nm⁻¹. If the function $\Lambda_2^{-1}(\theta)$ was over - correcting the scatter signal a majority of the low angle structure was lost due to amplitude reduction. This effect was pronounced in plastic samples such as PMMA and polystyrene.

Therefore the confidence of our method partly depends on the x-positions of the coherent amplitudes. If coherent peaks or structure exists for values less then 1.0 nm⁻¹ the correction factor will smooth out any uniqueness. Any form factors less than this x-value become suspect.

6.3.2 Future Work

Before any major attempt is made to accurately catalogue the coherent scattering form factor one must improve the mechanics of the machine.

6.3.2.1 Slit Configuration

Work must be done to reduce the background signal or the fringe radiation at low angles. A possible solution to the fringe problem is to experiment with different slit configurations. A smaller slit width means a smaller fringe component. However the smaller slit width reduces the signal intensity. The scan time may need to be increased to obtain recognizable data sets. With the proper slit configuration it may be possible to eliminate any background subtraction correction.

6.3.2.2 Low Angle Diffractometer

To improve the accuracy of the low x range ($0.102 - 1.0 \text{ nm}^{-1}$) one could utilize a low angle diffractometer. Since the machine is designed to work at low angles background due to fringe radiation would be substantially reduced. These data could be used as a control or check for the Rigaku or Scintag correction function $\Lambda^{-1}(x)$ at $x \rightarrow 0$.

6.3.2.3 Synchrotron Diffractometer

For even finer measurements one could use a synchrotron diffractometer. The beam emerging from the synchrotron is linearly polarized (almost). According to Peplow et al [Ref.16] this allows one to use the differential cross section for polarized x rays. They also determined that the coherent differential cross section scales with the square of the form factor. There is no need to correct for Thomson's polarization. Additionally the use of a synchrotron diffractometer would allow one to select different photon energies for the scan. This has the advantage of studying larger x ranges without changing the scattering geometry of the diffractometer.

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