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Process and Monitor Degasser systems for the SNO Heavy Water System

by

Darren R. Grant, B.Sc.

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
Ottawa, Ontario, Canada

September 8, 1998

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acceptance of the thesis

**Process and Monitor Degasser Systems for the SNO
Heavy Water System**

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

Chair, Department of Physics

Thesis Supervisor

Carleton University

Date Sept 24th / 1998

Abstract

The ability of the SNO detector to measure the flavour independent solar neutrino rate depends heavily on the removal of background sources like ^{222}Rn . Two degassing systems have been designed for this task in the D_2O water system. The first is a high efficiency process degasser designed to reduce the radon load, while the second, monitor degasser, is designed to assay the D_2O for residual radon at extremely low concentrations.

The final designs are based on membrane contactor technology. These were tested and found to have good degassing efficiency ($> 97\%$ for O_2) with low vapour losses ($\approx 1 \text{ ml/min}$). The components have been redesigned to meet the low background criteria for SNO. Two vapour recovery systems have been designed to minimize D_2O losses. The Rn assay system has demonstrated its ability to measure ultralow Rn levels ($< 10^{-14} \text{ g}^{238}\text{U/g D}_2\text{O}$).

The systems have now been built, tested and installed in SNO.

For Samantha Sarah Nova

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Chapter 1

Introduction

Within a 22m diameter, barrel-shaped cavity at the 6800ft level of INCO's Creighton Mine near Sudbury, Ontario Canada is a large Cerenkov Detector known as the Sudbury Neutrino Observatory (SNO). The detector consists of approximately 1000 tonnes of heavy water (D_2O) contained in a spherical acrylic vessel. It is designed to detect neutrinos from the Sun and other astrophysical objects, with the unique ability to measure both the flux of electron neutrinos and the total neutrino flux. The detector does this by observing the Cerenkov light produced by recoiling electrons which result from neutrino interactions with the D_2O . Since neutrinos interact only through the weak force in nature, such interactions with the D_2O are rare. As a result, the limiting factors affecting the sensitivity of these measurements are the background sources present in and around the detector, which produce electrons and photons that mimic the signal. The prominent background sources are the radionuclides (^{232}Th , ^{238}U , ^{40}K) and their daughters, which are found naturally in the rock of the cavity, the materials of the detector, and in the water.

This thesis presents a study of the methods used to remove and monitor the presence of ^{222}Rn in the heavy water of the SNO detector due to the ingress of mine

air and emanation of ^{222}Rn into the water by detector components. It documents the large amount of work which has been put into the design and construction of the D_2O degasser systems, without which the SNO detector would be unable to reach its desired level of sensitivity.

1.1 The SNO Detector

The Sudbury Neutrino Observatory Detector consists of 1000 tonnes of high purity heavy water contained in an acrylic vessel (see figure 1.1). The 12m diameter spherical vessel is made up of 125 sheets of 5cm thick ultra-violet transmitting acrylic panels bonded together. At a distance of 2.5m from the acrylic vessel is a uniform geodesic array which provides the structural support for the 9600 SNO photomultiplier tubes (PMTs) (9456 forward facing towards the center of the detector, and 91 outward facing to record activity outside the vessel area). Both the vessel and PMTs are suspended in approximately 7200 tonnes of ultrapure water. The ultrapure water acts as a shield for the background sources in the cavity walls and is cooled to approximately 10°C to reduce photomultiplier tube (PMT) noise, biological activity and to keep the D_2O near its maximum density of 1.106 g/cm^3 (at 11°C , standard pressure). In order to minimize the effects of radioactivity from the PMTs, a low-radioactivity Schott glass was used, and the PMTs are shielded by the additional 2.5m of ultrapure water. The PMT array is completed with the inclusion of aluminum coated reflectors around each PMT. The total photocathode coverage is approximately 40% and the addition of the reflectors increases the effective solid angle coverage to approximately 65%.

The location for the detector was chosen in an attempt to limit the signals from the external background sources (see figure 1.2). At a depth of 2070m below surface only neutrinos and a few energetic muons are able to penetrate to the detector, thus

Figure 1.1: Schematic of the basic components of the SNO Detector

effectively eliminating the cosmic ray background. The cavity was excavated in a region consisting mostly of norite rock, which has been found to have much lower background levels than other rock types, such as granite and diorite. The finishing touch to the cavity is a urylon lining which was sprayed onto the cavity walls to ensure the cavity is water tight, while also providing a barrier to the Radon which emanates from the rock walls.

1.2 Solar Neutrinos

The ultimate goal of the SNO detector is to provide answers to some of the questions which have been plaguing neutrino physics for many years. Present theory states that the Sun and stars produce energy through nuclear fusion processes in their interiors. The most direct test of this theory occurs through the detection of neutrinos produced in the fusion reactions, since neutrinos interact only via the weak force

Figure 1.2: Schematic of INCO's Creighton Mine and the approximate location of the SNO detector. SNO is located 6800 feet below the surface at the 6800 level of Creighton Mine.

and therefore can escape unimpeded from the interior. The main burning reactions in the Sun (table 1.1) are in what is typically known as the pp chain (the primary energy-generating chain for the Sun). One can see that the reaction of interest for SNO, involving ${}^8\text{B}$, is rare; occurring approximately once in every 5000 terminations of the pp chain. Hence the flux of ${}^8\text{B}$ neutrinos, predicted by the standard solar model for a distance of 1 astronomical unit, is much lower than neutrinos produced in the primary pp reaction (see figure 1.3). However, the higher energies of the rare ${}^8\text{B}$ (and hep) neutrinos make them detectable by SNO (and Super Kamiokande), where the neutrino energy threshold of the experiment is approximately 5.5 MeV (discussed in section 1.3).

A number of solar neutrino experiments have been developed to study the electron neutrino fluxes emitted from the various fusion reactions in the interior of the sun (see table 1.2), and have consistently shown a discrepancy of a factor of two or more when compared to the predicted fluxes from solar energy generation models (table 1.3). This discrepancy is called the Solar Neutrino Problem (SNP) and was the motivation for SNO's design and construction. Two possible solutions to the SNP are suggested: that the model of the method for the Sun's energy production is incomplete in some way and the neutrinos are not produced in the expected numbers, or that the neutrinos change flavor (oscillate) in transit to the Earth. Recent results from helioseismology studies of the Sun have been used to examine standard solar model predictions. Helioseismology has progressed to the point where the highly precise measurements of the frequencies may be used to determine the values of the speed of sound to better than 1% in most of the solar interior, and better than 5% in the solar core. The results have demonstrated that the standard solar model predictions for solar opacity, core temperature and fusion reaction rates (driving neutrino production) are "in good shape"[3], implying some new physics for the neutrino is necessary to explain the SNP. A very promising theory put forth by

Reaction	Number	Termination (%)	ν energy (MeV)
$p + p \Rightarrow {}^2\text{H} + e^+ + \nu_e$	1a	100	≤ 0.420
or			
$p + e^- + p \Rightarrow {}^2\text{H} + \nu_e$	1b (pep)	0.4	1.442
${}^2\text{H} + p \Rightarrow {}^3\text{He} + \gamma$	2	100	
${}^3\text{He} + {}^3\text{He} \Rightarrow \alpha + 2p$	3	85	
or			
${}^3\text{He} + {}^4\text{He} \Rightarrow {}^7\text{Be} + \gamma$	4	15	
${}^7\text{Be} + e^- \Rightarrow {}^7\text{Li} + \nu_e$	5	15	(90%) 0.861 (10%) 0.383
${}^7\text{Li} + p \Rightarrow 2\alpha$	6	15	
or			
${}^7\text{Be} + p \Rightarrow {}^8\text{B} + \gamma$	7	0.02	
${}^8\text{B} \Rightarrow {}^8\text{Be}^* + e^+ + \nu_e$	8	0.02	<15
${}^8\text{Be}^* \Rightarrow 2\alpha$	9	0.02	
or			
${}^3\text{He} + p \Rightarrow {}^4\text{He} + e^+ + \nu_e$	10 (hep)	0.00002	≤ 18.77

Table 1.1: The main burning reactions in the Sun; the energy-generating pp chain. Each conversion of four protons to an α -particle is known as a termination of the chain of energy-generating reaction; i.e. $4p \Rightarrow \alpha + 2e^+ + 2\nu_e + 25\text{MeV}$. Hence, the termination percentage is the fraction of terminations of the pp chain in which each reaction occurs [2]. SNO is able to detect the rare neutrinos produced in reactions 8 and 10.

Figure 1.3: The energy spectrum of neutrinos predicted by the standard solar model. Regions of sensitivity for past and present active experiments are shown across the top. [2]

Experiment		Result
Homestake Cl	[4]	2.54 ± 0.14 (stat) ± 0.14 (sys) SNU
Kamiokande	[5]	2.80 ± 0.19 (stat) ± 0.33 (sys) $\times 10^6$ /cm ² /s
Super Kamiokande	[6]	2.42 ± 0.06 (stat) $+0.10/-0.07$ (sys) $\times 10^6$ /cm ² /s
SAGE	[7]	$72 +12/-10$ (stat) $+5/-7$ (sys) SNU
GALLEX	[8]	69.7 ± 6.7 (stat) $+3.9/-4.5$ (sys) SNU

Table 1.2: A Summary of Results from Solar Neutrino Experiments. The values for Kamiokande and Super Kamiokande are measured neutrino fluxes. By comparison, a SNU is the solar neutrino unit; the product of a characteristic calculated solar neutrino flux (cm⁻²s⁻¹) and a theoretical cross section for neutrino absorption (cm²)[2]. The SNU therefore has units of events per target atom per second (taken as 10⁻³⁶ s⁻¹ for convenience).

Mikheyev, Smirnov and Wolfenstein lends support to this second possible solution, stating that the electron neutrinos produced in the sun can be converted to other types of neutrinos with high efficiency as they pass through the solar matter (matter enhanced oscillations: the MSW effect). This particular solution however requires the mixing of neutrino states and that at least one neutrino type has mass. The mixing of neutrino states, analogous to that observed in the quark sector of the standard model of particle physics, is not currently included in the standard model and would be the first evidence of physics beyond the model.

A further result of this solution is that neutrino masses, even if very small, could have a significant impact on the missing dark matter problem in the universe. The basic problem is that in integrating over the total visible matter, one can account for only approximately 10% of the total mass as measured by studies of the rotational

Experiment	Threshold	Comparison to Theory (EXP/BP95)
Homestake Cl	0.8 MeV	0.273 ± 0.021
Kamiokande	7.3 MeV	0.423 ± 0.058
Super Kamiokande	6.5 MeV	0.358 ± 0.012
SAGE	0.233 MeV	0.526 ± 0.101
GALLEX	0.233 MeV	0.509 ± 0.059

Table 1.3: A summary of the results of neutrino experiments compared to the predicted value determined by Bahcall and Pinsonneault (BP95 [9]). The various experiments have different energy thresholds, and are therefore sensitive to neutrinos from different reactions in the sun. Therefore, the variance in the ratio of the experimental results to theory reflect more on the ability to predict the solar neutrino flux on earth as a function of neutrino energy than on experimental inconsistencies.

velocities of spiral galaxies. The velocity rotation curves of luminous objects in the spiral galaxies have shown that the velocity distribution remains flat even at large distances from the galactic center. This is contrary to what one expects since as one moves away from the center (where the light is concentrated and therefore expected to contain most of the mass) it is expected that the velocity of the objects would drop off. The observed flat rotation curves suggest the presence of a halo of unseen matter surrounding the galaxies. Searches have occurred for a number of possible answers to this question, from MACHOs (MAssive Compact Halo Objects; the baryonic contribution to Dark Matter theories) to WIMPs (Weakly Interacting Massive Particles; the non-baryonic component which is subdivided into Hot (massive neutrinos) and Cold (i.e. neutralinos)). The MACHO searches have produced a handful of possible candidates thus far, but the observed number are far too few to account for the required missing mass, and WIMP searches for exotic particles (Cold) have yet to demonstrate any substantial proof to their existence. However, neutrinos (the most abundant particles in the universe) even with a very small mass could represent the dominant source of mass in the universe and hence provide answers to the missing mass problem. Massive neutrinos would also have profound implications on the ultimate fate of the universe: a continued expansion of space in time, or a final catastrophic implosion (the Big Crunch).

1.3 SNO Physics

SNO represents one of three world class facilities in the second generation of neutrino experiments. Super Kamiokande (a 50 kilotonne light water Cerenkov detector designed to study the recoil electron energy spectrum of ^8B neutrinos), and Borexino (a 100 tonne ultrapure liquid scintillator detector designed to study the recoil electron energy spectrum of ^7Be neutrinos) will detect electron-neutrinos produced in

the Sun with real time counting, much lower intrinsic backgrounds and an order of magnitude increase in sensitivity over the first generation experiments. SNO shares these characteristics of the other second generation detectors with the exception that it is sensitive to all three neutrino types rather than only the electron-neutrinos.

SNO uses three primary reactions to measure neutrino flux, direction and energy spectrum:

The charged-current reaction

$$\nu_e + d \Rightarrow p + p + e^- \quad (1.1)$$

has the largest cross section within the 1000 tonnes of D₂O and therefore provides the dominant signal in SNO. In this reaction the Cerenkov light produced by the recoil electron is detected, and therefore backgrounds which must be considered for the reaction are energetic electrons produced due to β decay, and Compton scattered γ -rays. However the reaction is extremely important in that it provides a measurement of the electron-neutrino energy spectrum since monoenergetic electron-neutrinos produce almost monoenergetic electrons of energy $E_\nu - 1.44\text{MeV}$. Should neutrino oscillations occur, the ^8B neutrino spectrum measured by SNO would show a clear distortion when compared to the predicted spectrum (based on standard solar model calculations) and therefore represents a signal for new physics discovery.

The neutrino-electron scattering reaction

$$\nu_x + e^- \Rightarrow \nu_x + e^- \quad (1.2)$$

is sensitive to all 3 neutrino types, although the scattering cross section is higher for ν_e since they interact through both the charged and neutral current processes. The number of ^8B neutrinos detected from this reaction is expected to be an order of magnitude lower than from the charged-current reaction, but the electrons from the elastic scattering process are kinematically constrained to the forward cone. Therefore one can receive directional information on the neutrino fluxes by studying this

reaction. Furthermore, direct comparisons to other experiments can be made by studying the ratio of Charge Current events (from i.e. Super Kamiokande) to Elastic Scattering events of SNO (CC/ES), and vice-versa. Since only the ν_e contribution will have enough events to be statistically significant in the initial operation of SNO (without Neutral Current signal enhancing), a comparison of CC/ES in SNO cannot discern the neutral current component. Comparison of, for instance, the high statistic ES events of Super Kamiokande to SNO's CC events could however clearly give the neutral current signal, although systematic uncertainties in the experiments may dominate the overall error.

A total neutrino flux can be measured using the neutral current reaction (neutrino disintegration of the deuteron)

$$\nu_x + d \Rightarrow \nu_x + p + n \quad (1.3)$$

where the reaction is detected by observing the γ -rays produced in the re-capture of the neutron. Hence, the dominant background which must be minimized are γ 's with an energy greater than 2.223 MeV which photodisintegrate the deuteron and are mistaken for the signal. This particular reaction is independent of neutrino type and therefore provides a *direct measure of the solar ^8B neutrino production independent of oscillation* [1].

The overall expected neutrino signals for SNO (table 1.4) number approximately 20/day (7000+/year) from the three reactions. When looking at the reaction rates for the range of electron energies expected, plotted with the expected background events (figure 1.4), one finds that the threshold energy which the detector can reach is limited primarily by the U and Th internal backgrounds. Although SNO has the ability to make new physics discoveries through measurements of the above mentioned CC/ES ratio and the search for a distortion in the neutrino spectrum due to oscillations, the true discovery potential for SNO lies in the successful measurement

Reaction	High Rate
CC:	12.7/day
NC:	5.5/day
ES:	1.4/day

Table 1.4: Expected signal rates for the SNO detector assuming a 0.5 standard solar model flux as observed at the SNO threshold of 5 MeV. The Neutral Current signal is the predicted rate for the presence of ^3He Neutral Current Detectors (NCDs) present in the acrylic vessel. (If the MSW solution is correct, then the extra neutrino flavours will be observed, and the NC rate will be approximately 11/day, the ES signal approximately 1.6/day, and the CC rate will remain the same.)

of the CC/NC ratio. The CC/NC ratio provides a measurement of neutrino flavor change independent of the solar models, and SNO is the only experiment capable to making this particular measurement (to better than 10σ assuming conservative levels of radioactivity in the detector components and water). For instance, a measurement of the ratio for the case where MSW oscillations occur would give a CC/NC result of 0.43 ± 0.034 (1σ), while the case of neutrino production being 0.51 of the Standard Solar Model gives the result $\text{CC/NC} = 1.0 \pm 0.12$. Due to the small uncertainties present in these measurements and the large separation in the various possible solutions, the SNO detector has the capability of making very confident statements about the physics governing neutrino behavior and energy production processes in the Sun.

Figure 1.4: Plot of SNO neutrino spectrum (Events/year inside D_2O vs. Electron Energy (MeV)) [10]. This represents a simulation of 1 year of data taking (with the exception of the PMT background, which has been scaled up from a simulation of a shorter time period).

1.4 The SNO Water Systems

The SNO water system consists of two separate water systems, one which purifies and recirculates the heavy water and one which maintains the ultrapure light water for the cavity. The purification loops of the systems are designed specifically to meet the critical needs of the experiment. These include the prevention of biological growth, ensuring a maximum attenuation length for the Cerenkov light, and the removal of radionuclides which limit the detector's sensitivity. Both systems must also provide methods for monitoring the residual activity in the water which act as background. The monitoring involves the daunting task of detecting a few atoms of the relatively short-lived radionuclides per ton of water to about a 10% accuracy. The D₂O system has the further complication of having to add and remove the MgCl₂ salt used to enhance the neutral current interactions, while continuing to purify and monitor the water in the presence of the salt.

The light water system (figure 1.5) contains a pretreatment plant for the initial fill, and a series of stages in the recirculation loop to generate and maintain the purity of the light water. Water from the INCO supply lines enters the pretreatment stage where it is passed through a series of microfiltration units, an activated charcoal filter, zeolite softener and into the triple Reverse Osmosis filter which is capable of removing $\geq 99\%$ of the salts, organics and colloids that remain [11]. The water from the Reverse Osmosis output is then "clean" enough to be injected into the purification and recirculation systems. The first stage for purification breaks apart organic compounds to an ionic form via a bank of 185nm wavelength ultraviolet lamps. These ions are then removed by a bed of 12 ion exchange columns designed to give ultrapure water with a conductivity of 18.3 M Ω /cm. Following the IX bed is the large light water Process Degasser which continuously removes ²²²Rn at an efficiency $\geq 95\%$ while simultaneously removing oxygen from the water, assisting in

limiting biological activity. A second bank of ultraviolet lamps with a wavelength of 254nm is present as a biological sterilizer. The water returns to the cavity via a heat exchanger which removes any process heat added during purification. The water is returned to the area between the PMTs and the Acrylic Vessel, where the cleanest water is required. A second loop, the cooling loop, continuously chills the water without any purification other than filtration. Hence, the purification and cooling loops operate in conjunction to maintain the ultrapure light water of the cavity.

The water can be continuously analyzed chemically and assayed for radioactive contaminants from a number of sample points in the purification loop and cavity. For the ^{238}U chain (figure 1.6), the most troublesome background comes from energetic photons of the ^{214}Bi decay. ^{222}Rn is the chosen ^{238}U daughter to monitor since it is long enough lived to easily assay, and the possibility exists for the long-lived ^{226}Ra to be out of secular equilibrium with the lower part of the decay chain. A monitor degasser is used to remove the Rn from the samples and is collected into Lucas cells for counting the extracted atoms on PMTs.

Similarly, in the ^{232}Th chain (figure 1.7), the problematic decay is due to energetic photons from ^{208}Tl . In this case either ^{224}Ra or ^{212}Pb may be monitored since the disequilibrium between the Ra and Pb is likely to be small. The longer lifetime of ^{224}Ra (3.64 days) is more attractive for use in monitoring, especially as one approaches the expected lower limit of ^{232}Th in the SNO detector. The method devised to perform this assay uses small acrylic beads which are coated with MnOx and contained in 1L polypropylene columns. The columns are designed so that they can be installed directly in the water systems and large quantities of water can be circulated through them. The radium in the water collects onto the MnOx coated beads, and once the column is dried it is placed on an Electrostatic Counter (ESC), which measures the decays of the radium daughters. With knowledge of the efficiencies for the ESCs and the columns, the assays of ^{224}Ra (and ^{220}Rn) can be used to

Figure 1.5: Flow Diagram of SNO's Light Water Purification and Recirculation Loops

Figure 1.6: ^{238}U Decay Chain [12]

Figure 1.7: ^{232}Th Decay Chain [12]

infer an equivalent amount of ^{232}Th in the water[11].

The heavy water system (figure 1.8) features more complex plumbing systems than used for the light water system due to the various modes in which the heavy water is run. In general, the heavy water system is designed to purify, recirculate and assay the D_2O while limiting any losses and degradation of the isotopic purity. Similar to the light water system, a bed of ion-exchange columns have been added in the receiving line to remove the high levels of potassium found in the virgin D_2O (present since potassium is used in the separation process of D_2O from H_2O). Columns of MnOx coated beads are to be used to remove and assay Ra, and a large hastelloy Reverse Osmosis unit is present to assist in the removal and assay for Ra and Pb. The R/O also has the important task of performing the desalination of the

Figure 1.8: Flow Diagram of SNO's Heavy Water Assay and Recirculation Loops

MgCl₂ from the D₂O once the MgCl₂ neutral current run is completed. Unique to the heavy water system is the Seeded Ultrafiltration unit (SUF) designed to purify and assay the D₂O of Th, Ra and Pb. The technique involves the coating of the ultrafiltration membranes with a thin layer of DTiO, which has a high affinity for heavy elements such as Th, Ra, and Pb. After passing approximately 100 Tonnes of D₂O through the membranes, the DTiO and captured radionuclides may be collected through chemical processes, reduced to a small sample and counted directly in liquid scintillator (β - α coincidence counters).

The final components to consider in the SNO heavy water purification loop are the D₂O Process and Monitor Degasser systems (PDG and MDG respectively). In the earlier stages of the water system design it was hoped that the levels of Rn present in the D₂O would be low enough to reach the desired equivalent ²³⁸U concentration of 0.01 ppt without the need of a process degasser. Further study brought to light the possibility of Rn entry into the D₂O through small leaks in valves and poor welds, as well as slow diffusion through thin welds in the process piping and the large number of o-rings in the system. Hence a full scale Rn removal system (PDG) and an independent monitoring system (MDG) have been included in the D₂O recirculation loop in order to meet the desired ²³⁸U levels.

The remainder of this report contains a detailed description of the systems designed to remove and monitor the Rn present in the D₂O. The initial designing, construction and commissioning steps of the D₂O PDG and MDG are documented, and a look at the initial backgrounds of the systems are presented. In Chapter 2 a discussion of the Rn problem in SNO and the steps taken to deal with it are described. Chapter 3 contains the theoretical description of the membrane degasser chosen to perform the Rn removal for the SNO D₂O system. Chapter 4 presents the designs of both the process and monitor degassing systems based on preliminary membrane tests, as well as the commissioning steps performed on the constructed systems. Conclusions drawn from the work thus far, and further studies necessary to complete the commissioning of the degasser systems are discussed in Chapter 5.

Chapter 2

The Radon Problem

2.1 Radon As A Background

There are two primary types of backgrounds of concern to SNO. The first is any low energy signal in the detector, which ultimately sets the thresholds above which the detector can be triggered. The primary sources of this background are β and γ rays produced in radioactive decays: from the U and Th chains, and electronic noise in the PMTs. The second background concern is the neutral current background, where any neutron in the detector may mimic a neutrino disintegration of the deuteron. The main contributors here are “high” energy γ ’s (above the deuteron binding energy) from ^{214}Bi (figure 2.1) and ^{208}Tl (figure 2.2) decays, which may photodisintegrate the deuteron to produce neutrons. To a lesser extent, (α, n) reactions on light elements (in the cavity rock) produce some neutrons which could possibly make it into the detector and further contribute to the backgrounds.

Radon is the heaviest of the noble gases, chemically inert and exists in 20 different isotopes of which ^{220}Rn and ^{222}Rn are significant to the SNO detector as sources of backgrounds. As a gas, radon has the ability to emanate from any material which

Figure 2.1: Simplified Decay Scheme for ^{214}Bi [1]

Figure 2.2: Simplified Decay Scheme for ^{212}Bi [1]

contains uranium and thorium. The natural presence of ^{238}U in the rock of the mine leads to elevated levels of ^{222}Rn in the mine and underground lab air. This is monitored in continuing studies (see figure 2.3) and, compared to surface values, is typically 3pCi/l underground compared to 0.1pCi/l on surface. The relatively long half-life of ^{222}Rn (3.8 days) is such that radon which enters the detector due to the ingress of lab air has a very high probability of decaying inside the detector volume. This is due to the fact that at the typical degassing rate of 160 lpm the recirculation system is unable to turn over the entire detector volume within the 3.8 day half-life. Hence, a number of steps have been taken to reduce the radon levels present in the detector due to both the ingress of lab air and emanation of radon from detector components. These steps include the monitoring of material selection for all detector components, the inclusion of a Rn free cover gas system for the underground D_2O storage tanks and the detector, eliminating the ingress of lab air into the SNO water systems (process piping), and the inclusion of both process and monitor degassing systems.

2.2 Rn Mitigation

2.2.1 Material Selection

The process of material selection for the detector components included both a study of U and Th concentrations in all wetted components and the measure of radon emanating from those materials. The U and Th concentrations were determined through detailed studies using direct γ -ray measurement techniques and studies of neutron activation of the materials. The neutron activation analysis (performed by the University of Guelph at the McMaster University Nuclear reactor) gives a measure of Th and U by detecting the γ rays of 27-day ^{233}Pa (312 keV) and 54-hour

Figure 2.3: ^{222}Rn Level Measurements of Underground Lab Air in 1997 and 1998 [14]. The Daily Counts data points are the same as those for units of pCi/l (overlaid), referring to the primary and secondary y-axes respectively. The daily count data is the raw data, without the Lucas cell backgrounds subtracted.

^{239}Np (106 keV, 228 keV and 278 keV) following β -decay after n-capture on ^{232}Th and ^{238}U [15]. This particular analysis, however, only determines the abundance of the long-lived parents in the ^{232}Th and ^{238}U decay chains. Since both chains contain other relatively long-lived daughters (^{228}Th for example) the members of the decay chains may not be in secular equilibrium for modified materials. Since understanding the contribution to the background from ^{208}Tl and ^{214}Bi decays is so critical to SNO, knowledge of the levels in disequilibrium is very important. Therefore direct γ counting of the materials was performed using a low-background ^{76}Ge detector at the University of Guelph. This technique is capable of detecting any disequilibrium in the decay chains. For instance, the ^{238}U chain produces a 1001 keV γ from the decay of ^{234}Pa and a 186 keV γ from combined decays of ^{235}U (185.6 keV) and ^{226}Ra (186.2 keV). Comparisons of these intensities to the γ intensities at the low end of the chain would give a clear indication of any disequilibrium which might exist [15]. Such disequilibrium could occur in the SNO detector due to an ingress of Rn, or the removal of ^{226}Ra or ^{228}Th from the water, which would significantly alter the concentration of important short-lived daughters (i.e. ^{214}Bi and ^{208}Tl) compared to the parents (^{238}U and ^{232}Th). The knowledge of any disequilibrium in the original “wetted” building materials is crucial for SNO since once a material is placed in ultrapure water the radium may leach out of the material, and result in a severe disequilibrium for the radium and its daughters in the water [18].

Radon emanation measurements of the materials were also performed for the major components of the detector. The process involved placing samples of the materials (i.e. stainless steel, signal cables and PMTs) into a leak tight emanation vacuum chamber and performing extractions of the chamber after a certain sealing time. ^{226}Ra can occur in materials in grains and crystals, and the decay of ^{226}Ra can result in the immediate production of ^{222}Rn into the vacuum for Ra close to the surface of the material. For decays which occur deeper in the material or produce

^{222}Rn atoms with insufficient recoil energy to immediately escape the material, the Rn may diffuse out of the material (outgas) over a period of time. The emanation chambers are large enough to allow fairly large sample materials to be studied, providing results which can be used to place expected limits on ^{222}Rn levels in the detector (see table 2.1). It should be noted the contribution from the water recirculation is not typically included in the estimate of the Rn load since the contribution, with the presence of butyl o-rings and a process degasser, is expected to be $\ll 10^2$ Rn/hr if the system remains leak tight and emanation from the process equipment is small. Dozens of materials have been tested, and their contribution to the SNO background estimated. In many cases this has led to direct involvement with industry to produce ultra-low radioactive products under SNO guidance. For example, the acrylic for the Acrylic Vessel, the resin used to manufacture the water system polypropylene piping, and the Schott glass used for the PMTs were each developed with SNO assistance to ensure the “cleanliness” of the final products.

2.2.2 The Cover Gas System

A second step taken towards the mitigation of Rn in the detector is the inclusion of a cover gas system applied over the detector, cavity and the underground 60 tonne heavy water storage tanks (figure 2.4). Effectively the cover gas provides a static barrier between the high levels of radon in the mine air and the water. The system is designed to produce a nitrogen gas with a radon level a factor of 10^5 lower than that present in the mine air, from the boiling of Liquid Nitrogen stored in a 1000 L dewar. The system cannot be sealed, as even minor fluctuations in the underground pressure (through airflow changes by INCO and external conditions) would place huge stresses on the deck above the cavity and the 60 tonne tanks.

Hence the system must be able to “breathe” to accommodate these changes, and

Component	Area m ²	²²² Rn per m ² per hr	²²² Rn/hr
Acrylic Vessel	452	<0.1	<5 × 10 ¹
Rope	180 m	<0.3 m ⁻¹ hr ⁻¹	<5 × 10 ¹
PMTs	473	<1.6	<8 × 10 ²
Reflectors	673	0.65	4.4 × 10 ²
Plastic PSUP	483	0.8	3.8 × 10 ²
S. Steel	410	<0.3	<1 × 10 ²
Dust (0.4 μg/cm ²)	2491	44 g ⁻¹ hr ⁻¹	4.8 × 10 ²
Total			At most 2.3 × 10 ³ Rn/hr

Table 2.1: Estimate of Emanated Radon Load into the H₂O Between the Acrylic Vessel and PSUP (1700 tonnes) [19].

Figure 2.4: Simplified view of the Cover Gas System [17]

is designed with over/under pressure protection devices and a series of buffer tanks. The over/under pressure protection ensures the structural integrity of the 60 tonne tanks and deck as it will vent these structures if the differential pressure becomes too large. The buffer tanks (1.2 m³ tanks plumbed in series) play the important role of ensuring mine air, which enters the cover gas system during large transients, does not reach the water. This allows the system to breathe without contaminating the N₂ gas. To accomplish this the buffer tanks have a continuous flow of pure nitrogen gas.

In the case of a drastic increase in mine pressure (the most dangerous of possibilities for SNO due to the possible influx of mine air; containing both Rn and H₂O vapour which could degrade the D₂O) the large dead-end volumes will generate a demand for an intake of “air”. The “air” is immediately drawn from the clean buffer

tank supply, as the mine air enters the tanks. Simultaneously, a flow indicator monitoring the cover gas conditions would react to this sudden reverse flow of gas and trigger the cover gas system to increase the N_2 flow rate (from a typical 5-10 lpm for static conditions to as high as 800 lpm for the most severe case [17]). The result is a system which is designed such that even large changes in the air pressure of the mine can be compensated for with a minimized possibility for any contamination of the water.

The ability of the system to meet these design criteria is currently being demonstrated. Samples of the cover gas taken at the exit of the 1000 L dewar have a Rn concentration as low as 0.094 ± 0.013 Rn/l compared to the target of 0.17 Rn/l [20]. Furthermore, extensive sampling of the cover gas over the D_2O in TK03 has shown that the Rn over the water decreases, as was anticipated, to the desired levels. This is shown in figure 2.5. The data points in the lower left show the Rn level in the tank decreasing in time (as residual Rn decays away). The Rn decay curve is shown only to indicate the expected slope if the only process were Rn decay. Clearly there is still some additional source of Rn (probably from emanation), but the level appears to be approaching the goal of 4.5×10^{-14} gU/g D_2O . The remaining points represent a measurement taken from another sample location, and after the tank was exposed to mine air for a brief period of time.

2.2.3 Rn Ingress

A further step was taken to deal with the possible ingress of Rn into the water system process piping, which would then transport Rn into the detector. This non-trivial task began with the realization that Rn could easily diffuse through the urethane o-rings which created the seals of the piping against mine air. Studies of a small subsection of the process piping, the H_2O Monitor Degasser (MDG) closed loop,

Figure 2.5: Current Cover Gas Analysis for TK03. See text for details [21].

showed the presence of an ingress of mine air (Rn) into a leak tight system. It was soon discovered that the mechanism for the ingress was from diffusion. An “o-ring stack” which presented a large surface area of o-rings to ambient air was used to test the diffusive properties for various o-ring materials, and showed clearly the cause of the ingress was diffusion through the urethane o-rings. Other materials studied, such as viton and butyl rubber, demonstrated excellent properties for preventing diffusion of Rn, but have always suffered the drawback of emanating Rn due to the fillers used in these compounds. A great deal of effort resulted in a butyl o-ring filled only with carbon-black, which was measured to have very low Rn emanation levels but still maintained the low diffusive and excellent sealing properties. Hence began the butyl conversion program of the SNO water systems, where some 7000+ o-rings were cleaned, inspected (to meet SNO standards) and installed into the systems. The result of the conversion was measured again with the leak-tight closed loop of the H₂O MDG, where it was observed that the Rn entry rate with the loop filled with water is approximately 450 ± 100 Rn/day. This is consistent with the result for the loop without water present and is greater than a factor of 10-15 lower than the previous measurements with urethane o-rings in the system (6150 ± 500 Rn/day)[23].

After the o-ring conversion was complete, the entire process system was He leak checked. This represented the most stringent testing of the process system's integrity for any region where an ingress of mine air might occur. While the entire system was leak checked, the most demanding criteria were applied to those lines returning to the cavity following the Rn removal stage with the process degasser (PDG). The process involved the draining and drying of the system lines (because H₂O had been used previously in the lines for commissioning tests of the D₂O system), followed by a N₂ pressure test to 20 psig, in order to locate any gross leaks in the piping (pinched o-rings, loose valve nuts; Note: it has been discovered that a loose valve nut, which is leak tight to water, can allow an ingress of 10^4 - 10^5 Rn atoms/day

[23]). The system was released of the N_2 gas and re-pressurized to 10 psig with 4He . 4He leak detectors (operated in “sniffer” mode) were then used to locate any leaks (typically at a sensitivity of $>10^{-5}$ cc/s) present in the system. Studies of these leaks showed the typical cause to be an ill-fitting valve stem or flawed o-ring, both of which were immediately replaceable. The leak testing also demonstrated the integrity of the process piping welds since a “thin weld” could allow the diffusion of Rn into the piping. None of these were found. Following the completion of the leak testing (including repairs and a second round of testing) the process piping was cleared for the acceptance of D_2O underground.

2.2.4 Degasser Systems

The final mitigation step to be discussed is the inclusion of Process and Monitor degassers in the H_2O and D_2O systems. Numerous techniques have been developed in industry to remove radon and oxygen from water, including methods for the removal of radon from drinking water. Current popular methods include aeration techniques, vacuum degassing methods and more recent developments have introduced high efficiency membrane contactors which can function at high flow rates.

Review of the SNO Light Water Degassing System

The principles behind aeration and vacuum degassing are very similar in nature. In general this involves the constant renewal of the interface between gas and liquid phases so that a high efficiency mass transfer from the liquid to gas phase can result. Aeration techniques were considered for use in the SNO systems and would have involved the use of either a packed tower aeration column or a diffused aeration method.

The first method uses a large column filled with packing material over which

the water is sprayed in a very “fine dropped” mist. A thin film forms over the large surface area of the packing material, representing an ideal region for the mass transfer to occur. A sweep gas low in radon and oxygen (perhaps taken from the boiling of liquid nitrogen as is done in the SNO cover gas system) is passed through the column. The radon and oxygen are removed from the water via this gas, and swept out of the column. The second method works on a principle where the water is placed into a series of tanks into which a low radon-oxygen gas is bubbled. The bubbling introduces rapid mixing in the tanks which causes the radon and oxygen in the water to be stripped away with rather high efficiency. Both aeration methods have been successfully used in industry for removing radon and oxygen from water sources. However, the high cost factor associated with producing the clean sweep gas required to degass the large quantities of light water for the detector caused the aeration techniques to be abandoned and a vacuum degassing system was developed (figure 2.6).

The degasser developed for the light water system is very similar in design to the large packed tower column used in aeration methods; i.e. a fine mist of small water droplets is sprayed into the column at the top. The small droplets then form a thin film over the packing material in the column, creating the transition region required for mass transfer to occur. However, rather than using a constant flow of clean sweep gas, the column is connected to a large vacuum system which maintains the chamber near the saturated water vapour pressure of 18 Torr at 20° C [13]. The water vapour acts as a carrier for the radon and oxygen out of the column and to the off gas header or to a point where the radon can be extracted and measured. Care in the choices of the materials used for the degasser’s construction has produced a degassing system which is low in backgrounds, while maintaining high efficiency at high flow rates; measured values of oxygen removal > 99% and radon removal of approximately 98% for typical flow rates of 135 lpm [11].

Figure 2.6: Schematic diagram of the H₂O Process Degasser [11]

D₂O Degasser Design Criteria

The commissioning of the H₂O degassers provided a great deal of knowledge which could be applied to the design of the D₂O degasser systems. In consideration of a general degasser design for the D₂O system, there are four primary criteria which must be addressed:

- Maintain high isotopic purity of the D₂O.
- Low vapour load during the degassing.
- High efficiency for O₂ (and therefore Rn) removal.
- Low background materials to ensure the emanation into the system is minimized.

Since the D₂O is on loan and has a value of approximately \$ 300/l, the first two criteria listed above must take priority. Therefore the chosen degassing systems must not change the D₂O chemically in any way or alter its isotopic purity. Furthermore, a degassing system which possesses a low vapour load is required to ensure the D₂O vapour losses can be minimized and effectively recovered. There are also a number individual criteria applied to the process and monitor sides of the degassing systems. The process degasser of the SNO D₂O system must:

- be capable of handling flows up to 160 lpm.
- possess an extraction efficiency for Rn > 95% and O₂ > 99%.
- operate at an average temperature for the D₂O of 8 to 10°C.
- possess a low enough background so as to not add more Rn than is extracted.

Similarly the monitor degasser must:

- operate at flows up to 20 lpm (the maximum flow rate possible with the diaphragm pump of the system)
- possess extraction efficiencies for $Rn > 80\%$ and $O_2 > 95\%$.
- have a method to collect and count the Rn extracted from the D_2O .
- possess the flexibility to measure from various sampling points in the detector, including before and after the process degasser in order to determine its efficiency.
- have a very low background contribution to the overall D_2O system.

The low background is especially critical for the D_2O monitor degasser (MDG) in order to ensure confident assays of the D_2O at the expected 10^{-14} gU/g level (this level brings the current H_2O MDG to its limit of sensitivity). The systems must also be able to operate in the presence of a corrosive salt, added for the neutral current enhanced run. The solution, after careful study of the numerous options, is a membrane degasser.

Introduction to the Liqui-Cel Membrane Degasser

Membrane contactors are devices that allow a gaseous phase and a liquid phase to come into direct contact with each other, for the purpose of mass transfer, without dispersing one phase into the other [22]. Recent developments in membrane technology, particularly increases in the available capacities and efficiencies, have made it possible for the contactors to be used for large process jobs in industry. The constructing of a vacuum degasser, similar to that used in the light water system, using Hastelloy would have been extremely costly. This is because stainless steel is kept almost completely out of the SNO D_2O system to avoid any possibility of corrosion

in the system during the MgCl_2 run. As well, large vapour losses can be experienced using vacuum degassing methods. Therefore, research into the developing membrane technology began. In particular a hydrophobic contactor designed and constructed by the Hoechst Celanese Corporation (Charlotte, North Carolina, USA), called the Liqui-Cel, showed a great deal of promise as a degassing tool for the D_2O system based on the preliminary information obtained from the company.

The Liqui-Cel Contactor (figure 2.7) is a hydrophobic microporous membrane which is capable of transferring gas to and from a liquid flowing through the membrane over a wide range of flow rates. This characteristic makes the contactor ideal for use in both the high flow case of the PDG (80 lpm) and the lower flow MDG (20 lpm). The fluid flows on one side of the membrane and a vacuum, sweep gas or both is applied to the other side. The microporous-hydrophobic characteristics of the membrane allows the gas and liquid phases to interact without the liquid passing through to the gas side of the membrane. This is an ideal condition for the SNO heavy water system where a minimum tolerance for loss of D_2O is applied. Furthermore, the design of the contactor gives an order of magnitude increase in surface area/unit volume compared to conventional columns which is important for achieving high efficiency degassing. The operation of the contactor is based on the principles of Henry's law, which states that *the amount of gas that will dissolve into water at equilibrium is proportional to its partial pressure in the vapour phase in contact with the water* [22]. Hence if one reduces the partial pressure of the gas in contact with the water, then the amount of gas dissolved in the water will be reduced. This can be accomplished by lowering the total pressure of the gas (applying a vacuum to the gas side of the membrane) or altering the concentration of gases in contact with the water (using a stripping gas which is free of the gas to be removed).

The physical description of the membrane degasser operation which follows, leads

Figure 2.7: (A)-Schematic diagram of a Liqui-Cel Contactor; (B)-Conceptual Diagram of Diffusion across a Hydrophobic Microporous Membrane [25]

one to an optimized design for the system and eventually to the experimental determination of the system efficiencies, backgrounds and the most efficient operating conditions.

Chapter 3

Theory of Membrane Degassing

3.1 Overview

The goal of the SNO membrane degasser is to remove gasses with a high efficiency while processing large volumes of water. The physical processes which occur in a degassing system are a combination of gas desorption (the transfer of gasses from the water to the gas phase) and gas absorption. In this chapter, the rules for these processes will be determined in order to design a system with adequate Rn and O_2 degassing efficiencies.

At equilibrium the amount of gas in the water and vapour phases is a constant, and is governed by Henry's Law, given by:

$$p = Hx \quad (3.1)$$

where p is the gas partial pressure, H is Henry's law constant (a function of the water temperature) and x is the concentration of the dissolved solute at equilibrium [22]. Hence, the concentration of Rn in the water may be reduced by lowering the partial pressure of Rn in the vapour phase in contact with the water. Under operating conditions, the constant flushing of the vapour stream to remove the gasses and

the constant renewal of the solutes in the water phase, ensures that the system is always in disequilibrium. This disequilibrium in turn ensures a constant transfer of solutes (dissolved gasses) from the water to the vapour phase. The efficiency of this process depends on the mass transfer coefficients, flow rates, temperature and contactor geometry. In order to understand the membrane degasser, the dependence of the efficiency on these parameters can be estimated by theory. In practice though, geometrical effects limit the accuracy, and in most cases, empirically determined relations are used to parameterize the contactor performance.

In the following, mass transfer coefficients are introduced and discussed. The efficiency of the contactor is related to the operating parameters and the mass transfer coefficients, which allows an estimate of the performance of the contactor under SNO operating conditions to be made.

3.2 Mass Transfer Coefficients

The removal of dissolved gasses from the water, or mass transfer, is driven by disequilibrium from saturation of the amount of gas in the vapour and aqueous phases. The mass transfer for the case of say oxygen removal from water involves three sequential steps in the contactor. The first step is the diffusion of the oxygen out of the water to the membrane surface. The second is the oxygen diffusing into the vapour-filled pores in the walls of the hydrophobic hollow fibers. The final step occurs once the oxygen reaches the other side of the pore, where it diffuses into the vacuum or sweep gas [24]. The overall mass transfer coefficient for this type of activity, K , is given by:

$$\frac{1}{K} = \frac{1}{k_L} + \frac{1}{k_M H} + \frac{1}{k_V H}. \quad (3.2)$$

In analogy to the sum of resistors, $1/K$ is known as the total resistance to mass transfer, where K can be thought of as the rate at which mass is transported and

has units cm/s. The values k_L , k_M and k_V are the individual mass transfer coefficients for the liquid, membrane and gas regions [24]. In order to determine the overall mass transfer coefficient (K) it is necessary to estimate the three terms in equation 3.2. Correlations used to determine the coefficients have for the most part been determined from experimental results, although some of the correlations are explained using a combination of chemistry and fluid theory. The overall mass transfer is often dominated by a single term, that which provides the most resistance to transfer. Typically, since the transfer rate is related to diffusion, the region with the highest resistance to diffusion (the liquid region) will therefore dominate K . For a membrane which is made up of hydrophobic microporous fibers, the contribution from the transfer coefficient associated with the membrane (k_M) is found to be small. This is due to the micro-pores of the fibers remaining filled with air, which allows the diffusion through the membrane stage to be very fast and therefore the membrane resistance to have a negligible contribution. Similar arguments can be applied to the gas stream on the other side of the pore. Here too the diffusion is very fast, compared to the diffusion rate in the aqueous stream. Although the k_V and k_M terms could begin to dominate in extreme operating conditions, for normal SNO operation k_L is the dominate term and therefore equation 3.2 reduces to:

$$\frac{1}{K} = \frac{1}{k_L} \quad (3.3)$$

In the case of the SNO D₂O degassers, the membrane contactor chosen makes use of a counter-current cross-flow design, for which the water flows across beds of the sweep-gas filled hollow-fibers. In this particular case, the oxygen mass transfer coefficients (which are not expected to be greatly different from those for radon) have been found to be dependent on not only the liquid flow, but also the geometry of the module. It has been observed that for equal flow rates, contactors with widely spaced fibers have mass transfer coefficients 40% lower than contactors with closely

spaced fibers. The explanation for this is that there are turbulent effects in the contactors downstream of each fiber which in fact enhance the mass transfer by preventing channeling.

Experiments performed to give the best possible conditions for the design of hollow fiber contactors [24] have provided a set of results which can be applied to almost all contactor operations (most importantly those involved at SNO). They have compiled a set of mass transfer coefficients under various operating conditions and geometries. In agreement with the discussion above, they found the performance of the hollow-fiber contactor is almost always controlled by the mass transfer in the liquid phase. A detailed list of the mass transfer correlations is available for the numerous possible flow patterns, which includes the result applicable for the contactors used in SNO [25]. For a perpendicular flow (cross flow) situation, the correlation is given by:

$$\frac{Kd}{D} = 1.4 \left(\frac{dv_o}{D} \right)^{1/3} \quad (3.4)$$

where K is the effective mass transfer coefficient in terms of the feed concentrations, d is the fiber diameter, D is the diffusion coefficient and v_o is the superficial velocity. In general, superficial velocity is defined as local flow rate averaged over a finite area of the porous medium, and is represented by:

$$v_o = v_{ave} / \varepsilon_{vf} \quad (3.5)$$

where ε_{vf} is known as the void fraction or porosity of the membrane (ratio of voids to total volume of the porous medium and voids), and v_{ave} is the average interstitial velocity at any cross section in the bed [30].

In the next section we will derive relationships between the efficiency of the contactor to this mass transfer coefficient and other parameters of the membrane.

3.3 Determination of Liqui-Cel Performance

In deriving the theoretical relations which describe the operation of the Liqui-Cel contactor, the ultimate desire is to achieve a simplified equation which describes the ratio of inlet to outlet dissolved gas concentrations, eventually leading to a relation for degassing efficiency. The general equations are complicated by the detailed geometric effects. However, for dilute solutions, the degassing efficiency is given by:

$$\frac{C_{outlet}}{C_{inlet}} = e^{-KaL/v_o}. [25] \quad (3.6)$$

The justification for this relationship is given below.

If one considers the simple case of water passing around the fibers contained in a cylindrical membrane of a given length ΔL , then the concentration of dissolved gasses of the feed to the exiting solvent can be represented by:

$$\Delta C = fC_{outlet} \quad (3.7)$$

where ΔC is the change in concentrations between the entering feed and exiting solvent, f is the representative rate of change factor in a given period of time, and C_{outlet} is the dissolved gas concentration of the solvent.

One may now begin to think of developing the general relation using a standard nuclear decay analogy, where f represents the rate of decay, or in this particular case the rate at which a solute can be removed from the feed solution using a contactor. In a nuclear decay the general relation is given by:

$$\frac{-dN}{dt} = N\lambda \quad (3.8)$$

which upon intergrating with respect to time becomes:

$$N = N_o e^{-\lambda t} \quad (3.9)$$

where N is the number of atoms from an original number N_0 after a period of time t . λ represents the rate at which the atoms decay (based on the half-life of the atom).

One can see that equation 3.8 can take the same form as equation 3.7 by:

$$\Delta C = -(\lambda \Delta t) C_{outlet} \quad (3.10)$$

Equation 3.7, upon intergration, will become:

$$\ln(C_{outlet}) = -\lambda t + Constant \quad (3.11)$$

and therefore,

$$C_{outlet} = C_{inlet} e^{-\lambda t} \quad (3.12)$$

using the boundary condition that at $t=0$, $C_{outlet} = C_{inlet}$.

In this case, t is actually the residence time of the feed solution in the membrane, given by $\Delta L/v_0$ (Length of the membrane / superficial velocity of the feed in the membrane, having units of time). The quantity λ takes the role of representing the rate at which the dissolved gas can be transferred out of the feed solution while in the contactor. From equations 3.3 and 3.4, the rate of mass transfer is given by K (cm/s). To satisfy the dimensions of equation 3.12, there must be a term with units cm^{-1} included in λ . The obvious choice is to include the geometry of the contractor, i.e. (the effective radius between contacting fibers) $^{-1}$. This is given by looking at the ratio of contactor surface area to total bed volume:

$$a = \frac{\text{Surface Area}}{\text{Bed Volume}} = \frac{n2\pi r L}{\pi R^2 L} \quad (3.13)$$

and reduces to:

$$a = \frac{2nr}{R^2} \quad (3.14)$$

where n is the number of fibers in the membrane, r is the fiber radius and R is the membrane radius. The total number of fibers in a membrane is dependent on the

fiber radii, the total membrane radius and the effective porosity of the membrane (ε):

$$n \cong \frac{\pi R^2 \varepsilon}{\pi r^2}. \quad (3.15)$$

Solving for R^2 in equation 3.15 and replacing R^2 in equation 3.14, one finds:

$$a \cong \frac{2\varepsilon}{r}. \quad (3.16)$$

The typical values for ε range between 0.3 and 0.6, and therefore $a \approx 1/r$. Hence equation 3.12 becomes

$$\frac{C_{outlet}}{C_{inlet}} = e^{-Ka\Delta L/v_o}. \quad (3.17)$$

Equation 3.17 is identical to the general equation 3.6 derived using the historical approach of determining such relations [22]. It represents the simplest form of the ratio of inlet to outlet dissolved gas concentrations. With this general equation in hand, it is now desirable to discuss the implications of what one has seen thus far. The mass transfer coefficients actually describe how fast a membrane contactor can function; i.e. it describes how quickly a mass can move through a medium. From equation 3.17 one can determine the solute concentration in a contactor as a function of the 'residence time' ($\Delta L/v_o$) in the contactor while taking into consideration the absorption/extraction constant (Ka). It turns out the mass transfer coefficient in relation 3.17 determines two important effects for the contactors. The first is contactors will separate with declining efficiency based on the limit of equation 3.17. For instance, if 100 cm of contactor can extract 90% of the solute, then 200 cm will extract 99%, 300 cm 99.9% and so on [25]. The other effect is that the contactors will depend on both fluid mechanics (water and air velocities) as well as chemistry (vapour pressures and any governing reactions) to perform gas absorption/extraction.

3.4 Estimation of Liqui-Cel Performance at SNO

3.4.1 Degasser Efficiency and Parameter Dependence

The degassing efficiency of the system is defined as:

$$\varepsilon_d = \frac{C_{inlet} - C_{outlet}}{C_{inlet}} \quad (3.18)$$

where C_{inlet} is the inlet concentration, and C_{outlet} is the measured outlet concentration [22]. Equation 3.18 can then be rearranged using equation 3.17 giving a relation for the measured efficiencies to those predicted by theory:

$$\varepsilon_d = 1 - \frac{C_{outlet}}{C_{inlet}} = 1 - e^{-KaL/v_o}. \quad (3.19)$$

From equation 3.19, one can see a direct efficiency dependence on the contactor's geometry. The surface area per volume of bed (a) and the length of the contactor (L) are present, as well as the superficial velocity of the feed water. What is not obvious is the temperature dependence of the feed water on the degassing efficiency. In the above equation 3.19, the mass transfer coefficient (K) is dependent on the diffusion coefficient (D), shown in 3.4. It turns out the diffusion coefficient (D) has a direct temperature dependence (see figure 3.1). One can see that for oxygen, the change of the diffusion coefficient with temperature is approximately linear, increasing 3%/°C rise in temperature. One can use this approximation to investigate the degassing efficiency of oxygen from water using a membrane contactor for a number of feed water temperatures over a number of flow rates. One final factor involving the diffusion coefficient which has not yet been discussed is the fact that diffusion coefficients are provided for H_2O , rather than D_2O . Theoretical values for the diffusion coefficient determined with the Wilke-Chang relation (equation 3.20) have shown good comparison to measured values from experiment;

$$D = \frac{7.4 \times 10^{-8} (\phi M_{D_2O})^{1/2} T}{\eta V_{gas}^{0.6}} \quad (3.20)$$

Figure 3.1: The diffusion constant of oxygen in water for various temperatures. Values for 16, 18, 18.2 and 25° C are taken from [27], and the remaining values from [29].

Temperature (°C)	D ₂ O (99.56%)	D ₂ O (100%)
5	1.30	1.31
20	1.24	1.25
40	1.20	1.20
60	1.18	1.18
80	1.16	1.16
95	1.16	1.16

Table 3.1: Ratio of viscosities (D₂O/H₂O), for changes with temperature and isotopic purity of the D₂O.

where ϕ is an association parameter for the solvent water ($\phi = 2.6$), M_{D_2O} is the molecular weight of the solvent (g/mol), T is the absolute temperature (K), η is the viscosity (in centipoises) and V is the molar volume of the solute (cm³/g mol). One can see in equation 3.20 that the diffusion coefficient should vary directly with absolute temperature and inversely with viscosity. Since the viscosity for D₂O is approximately 1.23 times greater than that for H₂O (see table 3.1), then one can argue that the diffusion coefficient for D₂O is 1.23 times lower than for H₂O.

Therefore, in determining the theoretical value for the mass transfer coefficient (K) in order to finally determine the degassing efficiency (ϵ_d), one must take into consideration the temperature dependence of the diffusion coefficient and make the appropriate correction for using D₂O.

3.4.2 Predicted Degassing Efficiencies at SNO

Using equation 3.19 one is able to make reasonable predictions for the overall degassing efficiency for the designed systems. The true value for the mass transfer coefficient (K) is obtained by measuring the performance of the contactor and solving 3.19 for K . However a simple approximation can be made using the schematics provided for the contactors to obtain ε_d . First, considering the terms in equation 3.19, the ratio, R , for C_{outlet}/C_{inlet} given in 3.17 can be evaluated for both the removal of oxygen and radon, giving:

$$\frac{R_{Rn}}{R_{O_2}} = \frac{e^{-K_{Rn}aL/v_o}}{e^{-K_{O_2}aL/v_o}} = e^{-(K_{Rn}-K_{O_2})aL/v_o} \quad (3.21)$$

where K_{Rn} and K_{O_2} are the total mass transfer coefficients for Rn and O_2 respectively, v_o is the superficial velocity of the solvent, and L and a remain the effective length of the membrane and the surface area per volume of bed, respectively. One can then use the correlation represented in 3.4 to replace K_{Rn} and K_{O_2} in equation 3.21. Solving for K in equation 3.4:

$$K = 1.4d^{-2/3}v_o^{1/3}D^{2/3} \quad (3.22)$$

where if $K = K_{Rn}$ or K_{O_2} , $D^{2/3}$ becomes $D_{Rn}^{2/3}$ and $D_{O_2}^{2/3}$ respectively. Therefore, equation 3.21 can be written simply as:

$$\frac{R_{Rn}}{R_{O_2}} = e^{-(D_{Rn}^{2/3}-D_{O_2}^{2/3})1.4d^{-2/3}aLv_o^{-2/3}}. \quad (3.23)$$

Using equation 3.5 the value of v_o can be determined for flows in the PDG and MDG, and the remaining terms in 3.23 are given in the Liqui-Cel specifications (table 3.2), with the exception of the values of D_{Rn} and D_{O_2} . In order to approximate the values of the diffusion coefficients, one can use the basic relation developed empirically by Wilke and Chang (equation 3.20). Given that the molar volumes are 50.5 and 25 cm³/g mol for Rn and O_2 respectively, and M_{D_2O} is 20 g/mol, results in

Fiber Type	PP Hollow Fiber
Housing	Polypropylene
Dimension	4" by 28"
Fiber ID	240 μm
Wall Thickness	30 μm
Effective Pore Size	0.05 μm
Effective Fiber Length	63 cm
Porosity	30%
Effective Area/Volume	3641 m^2/m^3
Potting Material	Epoxy
Maximum Pressure	60 psi
Maximum Temperature	60 $^{\circ}\text{C}$
Maximum Water Flow	30 gpm
Modes of Operation	Vacuum, N_2 sweep, Combo

Table 3.2: Liqui-Cel Characteristics [11].

$D_{Rn} = 5.07 \times 10^{-8} \text{ T}/\eta(\text{T})$, and $D_{O_2} = 7.74 \times 10^{-8} \text{ T}/\eta(\text{T})$. Hence, equation 3.23 can be written in a general form:

$$\frac{R_{Rn}}{R_{O_2}} = e^{6.24 \times 10^{-6} d^{-2/3} a L v_o^{-2/3} T^{2/3} \eta^{-2/3}}. \quad (3.24)$$

Replacing the characteristic dimensions of the contactor in equation 3.24, it further reduces to:

$$\frac{R_{Rn}}{R_{O_2}} = e^{0.172 v_o^{-2/3} T^{2/3} \eta^{-2/3}}. \quad (3.25)$$

In order to generate a prediction for ε_d using 3.25, one must investigate the direct temperature dependence of the relation. A temperature dependence ($T^{2/3}$) is obvious in equation 3.25, as well as a variation of the viscosity (η) with temperature (shown in figure 3.2). Although the single membrane tests, discussed in chapter 4, were performed with a feed water temperature of approximately 20 °C, the D₂O degasser systems will operate at an average temperature of 10 °C (ranging typically from 8 to 12 °C). In this range the viscosity changes from 1.801 to 1.671 centipoise. To complete the prediction using equation 3.25, one must determine the superficial velocity of water through the Liqui-Cel contactor for the expected D₂O MDG and PDG flow rates. For an MDG flow rate of 20 lpm, the volumetric flow through the module is equivalent to 333.3 cm³/s. This then relates to a superficial velocity of approximately 22.1 cm/s (shell side): $333.3 \text{ cm}^3/\text{s} \times 4/(\pi (8 \text{ cm})^2 0.3)$, where 0.3 is the porosity of the the contactor, and 8 cm is the approximate effective diameter of the membrane. Hence, one finds that the relation R_{Rn}/R_{O_2} varies from approximately 1.88 at 8 °C to 1.96 at 12 °C, and maintains a value of 1.94 at the average operating temperature of 10 °C. One therefore has a reliable relation for determining the Rn degassing efficiencies for heavy water.

In order to make this final prediction for the degassing efficiency of Rn from D₂O, one must first make an estimate of the O₂ degassing efficiency. This is due to the fact that the diffusion coefficients for O₂ have been very well studied in the past for a

Figure 3.2: The viscosity of H_2O for various temperatures [32].

number of temperatures, whereas the value for Rn is not so well known (published as $1.14 \times 10^{-5} \text{ cm}^2/\text{s}$ at 26°C in 1917 [28]). First, one should investigate the accuracy of the Wilke-Chang relation for predicting the diffusion coefficient of water. For instance, at 20°C the Wilke-Chang relation gives a value for the diffusion coefficient of water of $2.15 \times 10^{-5} \text{ cm}^2/\text{s}$ which compares well with the experimentally measured result of $2.1 \times 10^{-5} \text{ cm}^2/\text{s}$ [29]. One can then investigate the result of using the mass transfer correlation (equation 3.22) for the single membrane test conditions, and find that $K = 0.0365 \text{ cm/s}$. Using this result in equation 3.19, one finds that the expected O_2 degassing efficiency for the single membrane test is 97.7%, which is slightly lower than single membrane test results, which gave $>99\%$ at 5 gpm (table 4.1). The higher experimental value is usually attributed to turbulence in the contactor. If one now applies this approach to normal operating temperature conditions of the degasser system for D_2O , one finds that at 10°C the expected diffusion coefficient given by the equation 3.20 is $D = 1.288 \times 10^{-5} \text{ cm}^2/\text{s}$ for O_2 . This results in a total mass transfer coefficient $K = 0.026 \text{ cm/s}$ and an approximate O_2 degassing efficiency of 93.2% for D_2O MDG flow rates. One can now use the relation developed in equation 3.25, from which it was found that at 10°C the value of $R_{\text{Rn}}/R_{\text{O}_2} = 1.94$. Rearranging equation 3.25, one can see that at 10°C , the extraction efficiency for radon ($\varepsilon_{d-\text{Rn}}$) should be about 86.8%. Furthermore, due to the temperature fluctuations of the D_2O passing through the degasser from 8 to 12°C , the O_2 degassing efficiency is expected to fluctuate between 92.4% and 94.0% respectively. This translates to a fluctuation in the Rn degassing efficiency between 85.2% and 88.6%.

Finally, one may perform similar calculations for the D_2O PDG. The anticipated flow rate through the PDG is 80 lpm (through each contactor) which is equivalent to a superficial velocity of approximately 88.5 cm/s. At 10°C the diffusion coefficient remains $1.288 \times 10^{-5} \text{ cm}^2/\text{s}$ and the mass transfer coefficient is $K = 0.0412 \text{ cm/s}$. Therefore, one finds the O_2 degassing efficiency for a single membrane in the D_2O

PDG is 65.6%. The ratio of R_{Rn}/R_{O_2} , for the PDG flow rates at 10 °C, becomes 1.30 and therefore the predicted Rn degassing efficiency is approximately 55.3%. For the PDG design of 2 sets of 3 Liqui-Cel membranes in series, one can then expect the overall degassing efficiency of the PDG to be 95.9% for O₂ and 91.1% for Rn.

Such calculations as these should be interpreted as the lower degassing limits capable by the degassers, since enhancement of the efficiencies due to turbulent effects have been neglected in the approximations. These results and specifications from Hoechst Celanese have resulted in a complete design for the SNO PDG and MDG systems. The discussion of the design of the systems based on the theory developed here follows, and the results of the commissioning of the systems to the current state of the project are presented.

Chapter 4

Degasser Design and Commissioning

A number of possible solutions were studied in the attempt to meet the design criteria for the D₂O degasser systems, including packed aeration columns, vacuum columns and various bubbler systems. During this time, membrane contactor technology was developing at a rapid rate. Early contactor designs placed the water flow on the inside of the microporous hollow fibers and the gas flow on the tube side. This design suffered due to the large resistance to flow on the inside of the fiber causing large pressure drops across the contactor as flow rates were increased. Hence, these contactors were not ideal for large commercial usage unless a number of modules were placed in a parallel flow configuration, which suffered the draw back of higher costs. The next logical step to reduce the pressure drop and therefore increase the possible flow rates was to place the water flow on the tube side of the contactor. The reduced pressure drop would allow one to place contactors in series, which enhanced the efficiency of a single pass. However this configuration suffered from severe channeling of the water and the result was very poor removal performance. In

1993 a baffled device was patented (the Liqui-Cel contactor by the Hoechst Celanese Corp.) which overcame both the above problems [22]. The baffle is placed at the center of the fiber bundle, causing the water to flow radially across the fibers in an even distribution. With this design one has the possibility of high flow rates with a low pressure drop across the contactor and minimal channeling of water, hence maintaining a high efficiency. As such, these devices were attractive to SNO, and a single unit was purchased for evaluation.

4.1 Overview of Measurement Tools

Prior to a discussion of the design and commissioning of the D₂O degasser systems, we should first consider some basic tools needed to make the measurements of the system behavior. These focus on two primary measurements, dissolved oxygen and radon.

4.1.1 O₂ Measurements

Dissolved oxygen measurements are key to monitoring the degassing of water since they can be performed on line. To perform the measurements, a Honeywell Limited Model 7931-Series dissolved oxygen probe has been purchased. When the probe is placed into the sample stream, oxygen diffuses through a membrane and is reduced at the cathode and an equal amount of oxygen is generated at the anode in the following reactions:

Therefore no oxygen or acid is consumed, no water is produced and no net reaction occurs in the probe. The diffusion continues until the oxygen partial pressure

on both sides of the membrane is equal. The probe is designed such that the current necessary to maintain the equilibrium is converted to read out the dissolved oxygen concentration in the solution. It is capable of measuring accurately to ppb dissolved oxygen levels for liquids which have a high resistivity, such as ultrapure H_2O and D_2O . Measurements using the probe demonstrated a pressure dependence of the probes response during rapid pressure changes of the feed water. This is due to the change in the diffusion of the O_2 into the probe head due to the change in water pressure. The probe does correct for pressure and temperature, but cannot react quickly. In order to eliminate this dependence, a flow through adapter has been designed. The probe then discharges to atmospheric pressure and has a constant pressure at the probe head. Furthermore, the flow-through adapter attaches to the probe body with a radial seal. This design modification helps reduce the ingress of Rn and O_2 from the lab air.

4.1.2 Rn Measurements

Although the dissolved oxygen measurements are basically “free” while the degasser system is active, requiring minimal effort in monitoring, what is truly desired is a measure of the Rn levels in the D_2O . In order to do this, one must have a system which is capable of trapping the Rn from the degasser vapour stream and transferring it to a counting device. The Rn must be collected with high efficiency due to the ultra low levels present, and other interferences like water vapour must be removed.

Water Removal

Due to its monetary value, the D_2O must be recovered. For Rn measurements, it is desirable to remove the water vapour early in the extraction process. This is due to the high pressures which water vapour can generate in the Rn trapping regions

Figure 4.1: Schematic of the SNO re-designed Titan trap by FTS

(the Rn board) and in the Rn counting devices (Lucas cells). Furthermore, water which enters the trap regions can form “ice plugs” which impede the transfer of Rn. Therefore the first component of the Rn measurement system is a water vapour trap (Titan trap figure 4.1). The Titan trap is from FTS Systems, Stone Ridge, NY, USA. It is located in the vapour stream which exits the degasser, and allows only Rn and some remaining gasses to enter the Rn trapping region. In general, the Titan trap is a refrigeration coil sealed under vacuum in an large chamber, operating at an average temperature of -55°C . The vapour from the degasser enters at the top of the chamber, flows around the coil which condenses the water in the stream, and the remaining gasses exit to the radon board.

Figure 4.2: Schematic of the SNO Rn Board

The Radon Board

The Rn board (figure 4.2) is the primary device designed to trap and transfer the extracted Rn to a counting device. There have been three Rn boards produced for on site use at SNO; for the H₂O MDG, the cover-gas system and now the D₂O MDG. Although each has slightly different design criteria, the basic designs follow that first described in a paper by Key *et al.* [33].

The gas which exits the Titan trap flows through the primary trap (Trap A) on the Rn board. Trap A is kept at liquid nitrogen temperature, which ensures the Rn is effectively captured, while the bulk of the remaining gasses (typically O₂ and N₂)

pass to the vacuum pump and to the exhaust. At the end of an extraction run, the goal is to transfer the Rn in Trap A to a smaller volume, which is the purpose of the secondary trap (Trap B). Trap A and B are isolated from the rest of the system, but open to one another. During the transfer, Trap A is heated while Trap B is at liquid nitrogen temperature, effectively creating a cryogenic pump for moving the trapped Rn to Trap B. After a given period of time, the Trap B region is isolated. The final step is to transfer this small volume into a counting device (the Lucas cell). This is done by warming Trap B to room temperature and the Rn in the small volume is transferred to an evacuated Lucas cell.

Lucas Cells

Once one has effectively trapped the Rn, the next step is to accurately measure the number of Rn. For SNO, a low background scintillation detector known as a Lucas Cell, was developed (figure 4.3). The SNO Lucas Cell is a small acrylic cylinder, with an empty hemisphere volume in the body, and a clear acrylic window. The hemisphere is evenly coated with a low background scintillation material (activated ZnS) to a thickness which was optimized for the α particle energies of interest. The Rn is transferred from the Rn board to the Lucas cell using self-sealing quick connects, which limit the possible ingress of lab air during transfers. Once the Rn is in the Lucas Cell, the cell is placed on a photomultiplier tube. The α particles produced in the decay of ^{222}Rn (and ^{218}Po and ^{214}Po) which strike the ZnS coated hemisphere produce a small “flash” of light which is viewed by the PMT and recorded by the data acquisition software. For each Rn decay, one expects to see 3 α particles. However, there are losses in efficiency due to the cells geometric coverage, and in low energy pulses which fall below threshold. These combine to give an effective efficiency of 0.63 per α particle. The typical background rates to the Lucas cells are

Figure 4.3: Schematic of the Lucas Cell designed for SNO

on average 10 counts/day from the ZnS and residual activity in the cell. Since the overall collection efficiency for collecting Rn from water into a Lucas cell is about 50%, this system is sensitive to very low radon levels, provided the backgrounds from the rest of the Rn board and degasser can be kept very low as well.

4.2 Single Membrane Tests

The first membrane contactor acquired from Hoesht Celanese Corporation (HCC) for evaluation was predicted to have an O_2 degassing efficiency of $> 99\%$ and a low vapour load of approximately 0.1-0.3 ml/min. These parameters were for typical SNO conditions of a 5 gpm water feed rate and a N_2 sweep gas with vacuum assistance. In the initial experiments, the device (a Liqui-Cel contactor in the commercial polypropylene housing provided by HCC), was installed in parallel with the current

H ₂ O Flow (gpm)	O ₂ levels (ppb)	O ₂ Degassing Efficiency (%)
4.0	31.0	99.5
5.0	35.0	99.4
6.5	57.5	99.1
8.0	88.0	98.6
8.2	91.5	98.5
10.0	138.0	97.7

Table 4.1: The O₂ degassing efficiencies for the Liqui-Cel contactor measured over a range of water flow rates. The incoming feed water had a measured dissolved oxygen level of 6.11 ppm (used as the reference level). A constant nitrogen sweep-gas rate of 5.5 scfh was used in the device.[34]

H₂O MDG and studies of the O₂ removal efficiency were made for a variety of operating conditions. The water vapour load of the device was also measured. The first of the measurements were taken for the case of a varying water flow rate with a constant sweep-gas passing through the membrane (5.5 scfh). The results (table 4.1) showed that very high degassing rates were possible with the device (comparable to the rates presented in the Liqui-Cel specifications). The second experiment measured the O₂ degassing efficiency with a constant water flow rate and different N₂ sweep-gas flow rates (table 4.2).

The results listed in tables 4.1 and 4.2 confirmed a number of the expected characteristics concerning the contactor. For instance, table 4.2 clearly shows that the O₂ degassing efficiency is fairly stable (99.4%) over the range of N₂ flows tested. These efficiencies are taken as lower limits, as the O₂ levels at the output appeared to be monotonically decreasing, independent of any sweep gas flow rate. This is

N ₂ Sweep flow (scfh)	O ₂ levels(ppb)	O ₂ Degassing Efficiency (%)
0 (vacuum only)	177	97.2
5	53.7	99.2
10	41.4	99.3
15	43.2	99.3
20	40.4	99.4
return to:		
5	38.8	99.4
2	38.1	99.4

Table 4.2: The O₂ degassing efficiencies for the Liqui-Cel contactor measured over different sweep gas rates. Feed water flow rate was held at 5 gpm and initial reference feed water dissolved oxygen level of 6.40 ppm. [34].

possible since there is residual O_2 outgassing from the contactor and the system hadn't reached equilibrium during the short runs.

A measurement of the vapour load from the contactor was also made at this time. With a N_2 flow rate of 5 scfh and water flow of 5 gpm, and vapour load of approximately 0.9 ml/min was measured. This was much higher than the average expected 0.2 ml/min vapour load advertised by HCC and became the subject of further R&D. This discrepancy brought to light a number of concerns for the standard HCC housing:

- The contactor possessed poor o-ring sealing surfaces (scaly epoxy), generating favorable conditions for a leak between the aqueous and gas sides.
- The standard housing was not designed to meet SNO specifications for maximum operating water pressures.
- The standard housing made inlet and outlet connections with standard pipe threads, which can be extremely difficult to seal He leak tight.
- Sanitary fittings were used for the large diameter endcap seals. This created a two-fold problem; at higher pressures this sort of seal is prone to leaks, and the large surface of elastomer exposed to the aqueous fluid would act as a significant Rn source (even with the use of "clean" Butyl rubber).
- The materials of construction for the standard housing had not been tested to be Rn clean.
- The sealing surfaces on the membranes were not precisely controlled.

[[35], [31]]

These concerns brought about a new design for the Liqui-Cel housing. The new housing was designed with a radial double o-ring seal (using carbon black filled Butyl

rubber) at a good sealing surface on the membrane. Furthermore, the pipe thread connections for the input/outputs were replaced with direct welds into the endcaps and face-sealing o-rings were used for connection to the process piping. It was felt that using double o-ring seals to separate the aqueous from the gas side would eliminate any possibility of a leak between the two sides (this was the assumed cause of the difference between the HCC predicted vapour load and the measured vapour load).

One such housing was built, and the Liqui-Cel was brought to the Centre for Research in Particle Physics (CRPP) ultra pure water (UPW) plant at Carleton University for testing. Assembly of the membrane into the new housing (the unit which eventually became the MDG degassing unit) was performed in a clean room under controlled conditions. The contactor was then connected to the CRPP UPW system (see figure 4.4). The UPW system has the capability of providing ultra pure water with a resistivity of $18.3 \text{ M}\Omega/\text{cm}$ with a maximum flow rate of 30 lpm.

The vapour loads, measured from the water collected in a liquid nitrogen temperature cold trap for differing feed water (at 20°C) and N_2 flow rates, are listed in table 4.3. One can see that in general there appears to be an increase in the vapour load from the contactor as the water and sweep-gas flow rates are increased. This increase continues and eventually plateaus at approximately 0.9 ml/min . The largest uncertainty in these measurements comes from the efficiency to trap water. In an attempt to confirm the water trapping efficiency a second cold trap was placed down stream of the first trap and showed a total trapping of approximately 1 ml of water at the run end. Looking at runs 3 and 10, for which the water and sweep-gas rates were similar, one can see that there is a discrepancy of 0.15 ml/min , and this is treated as the uncertainty in the measurements. With this error, the vapour load at all but the lowest water and sweep gas flows is consistent with 0.9 ml/min . Hence, one can argue an upper limit of approximately 1 ml/min vapour load from the Liqui-

Run ID	Water Flow (lpm)	N ₂ (lpm)	Time (min)	Vapour Load(ml/min)
1	20	0	15	1.0 $\pm$ 0.13
2	10	0	60	0.75 $\pm$ 0.03
3	30	0	60	0.75 $\pm$ 0.03
4	0	0	135	0.55 $\pm$ 0.01
5	0	0	6	0.50 $\pm$ 0.33
6	0	1.8	45	1.0 $\pm$ 0.04
7	30	1.8	30	0.80 $\pm$ 0.07
8	30	0.6	30	0.90 $\pm$ 0.07
9	30	0.2	85	0.87 $\pm$ 0.02
10	30	0	30	0.90 $\pm$ 0.07

Table 4.3: Water vapour load measurements of the Liqui-Cel for water flows ranging from 0 to 30 lpm and sweep gas flows of 0 to 1.8 lpm (STP)[35]. The uncertainty in the vapour load is estimated to be ± 2 ml of the total volume of water measured (± 1 ml reading error on a graduated cylinder, and ± 1 ml uncertainty on the liquid remaining in the cold trap following emptying for measurement.)

Figure 4.4: Simplified schematic of the CRPP UPW plant with the D₂O MDG Liqui-Cel contactor installed for vapour recovery tests.

Figure 4.5: Liqui-Cel pressure drop measurements plotted with the theoretical curve (provided by HCC [36]) using the CRPP UPW plant, June 1997. Operating mode of feed water temperature at approximately 20 °C and no vapour recovery involved.

Cel contactor in its redesigned housing, contrary to HCC's calculated vapour load of 0.2 ml/min.

The pressure drop across the membrane (from the water feed inlet to the outlet) was also a concern since it determines the possible flow rates which can be achieved through the degassers. The inlet and outlet water pressures were monitored, and the pressure drop as a function of flow was determined (see figure 4.5). The measured pressure drops matched very reasonably the values established by the manufacturer, and extrapolating one finds the pressure drop for a flow rate of 80 lpm (typical flow

Figure 4.6: Simplified schematic of the D₂O PDG and MDG systems.

through a single contactor in the PDG) is <8 psi. This low impedance should not significantly reduce the flows in the D₂O process systems.

From these initial tests a general design of the process and monitor degasser for the SNO D₂O systems was developed (see figure 4.6). The design of the MDG is a single membrane contactor, complete with vapour recovery and radon assay capabilities. The PDG was designed to process the feed D₂O at a rate of 160 lpm (Six Liqui-Cel contactors- 2 sets in parallel of 3 contactors in series, with a flow rate of 80 lpm through each contactor). The sweep-gas for the degassers is taken from the cover-gas supply, at a typical rate of approximately 1 lpm per contactor, and

upon exiting the degassers, enters the vapour recovery system. These designs and further commissioning tests are discussed in the following sections.

4.3 MDG Design

The monitor degasser is a separate entity from the process degasser as it enables one to sample the water from many different points in the detector while the PDG continues to degass the contents of the detector. It is made up of three basic units. The single Liqui-Cel contactor performs the degassing of the water which passes through it using the membrane and new housing design. The sweep gas which exits the Liqui-Cel (including N_2 , O_2 , CO_2 , Rn and D_2O vapour) enters the second unit, a vapour recovery system. This is high efficiency refrigeration coil which acts as a water trap to remove the D_2O from the vapour stream. It was provided by the FTS company, and is called a Titan trap. The gas is then swept (using a vacuum pump) into the third and final component of the MDG; the Rn board. At this point the Rn (and CO_2) are trapped in liquid nitrogen traps from which they are eventually transferred into a Lucas Cell, while the remaining gasses are sent to the exhaust. In addition to the efficiency and vapour load requirements, the MDG has an emphasis on low backgrounds for the device in order to achieve accurate analysis of Rn levels in the water to the expected limits ($1 \times 10^{-14} \text{ g}^{238}\text{U/gD}_2\text{O}$ or approximately 60 Rn atoms/tonne of D_2O).

4.3.1 The MDG Liqui-Cel Studies

The same Liqui-Cel membrane and new housing used in the vapour load tests at the CRPP UPW plant were used as the MDG degassing unit. Upon its return to Sudbury the contactor was dried (by pulling a vacuum on the sealed unit for approximately 3

Blank ID	Rn Production (number of atoms/day)
1	54.7 ± 10.4
2	42.1 ± 6.6
3	21.7 ± 6.0
4	52.7 ± 5.5
Average	41.5 ± 3.8

Table 4.4: The results of the background blanks performed on the MDG Liqui-Cel contactor.

days, through the H₂O MDG FTS unit) and plumbed into a temporary position to the H₂O Rn board (figure 4.7). In this arrangement, blanks for seal times varying from a few days to many weeks could be made on the contactor. A blank is a process which involves the evacuating of the object which is to be studied. After a determined seal time, the object is exposed to the evacuated Rn board's primary trap (Trap A) under liquid nitrogen (LN₂). Anything which is outgassed/emanated by the object into the vacuum over the seal time, is swept through the cooled primary trap using a vacuum pump, as in a standard Rn extraction experiment.

The results of the blanks for the individual Liqui-Cel can be found in table 4.4. The analysis of such blanks requires careful accounting of the Lucas Cell efficiencies and backgrounds, and accounting for the constant decay of the Rn and its daughters.. For instance, for Blank #3 listed in table 4.4, the details of the extraction to the Rn production were:

- Sealing Date: 12:47 August 7, 1997

Figure 4.7: Schematic of the set-up used for performing background blanks on the MDG Liqui-Cel.

- Seal Broken: 9:22 August 18, 1997
- Extraction Start: 9:22 August 18, 1997
- Extraction End: 12:22 August 18, 1997
- Start counting of Lucas cell: 14:40 August 18, 1997
- Lucas cell number: N08
- Counts/Live time: 60/177246
- Background counts/Live time: 115/1005470

. The first step is to calculate the number of signal counts S

$$S = C - \frac{Bt_c}{t_B} \quad (4.1)$$

where C = total counts in the counting time t_c , and B is the number of counts during the background counting period t_B . The time factors in the last term normalize B and C to the same counting period, t_c .

The error on S is ΔS , and is given by

$$\Delta S = \sqrt{\Delta C^2 + \left(\frac{\Delta B t_c}{t_B}\right)^2} \quad (4.2)$$

where $\Delta C = \sqrt{C} \sqrt{3\varepsilon}$ and $\Delta B = \sqrt{B} \sqrt{3\varepsilon}$. The last term comes about due to the fact that the number of counts per radon decay is 3ε , where $\varepsilon = 0.63$ is the single α detection efficiency. This therefore gives the net counts for this run of $S = 39.8 \pm 11.0$ counts.

The number of radon decays this represents is

$$N_D = S/3\varepsilon. \quad (4.3)$$

The next step is to determine the number of Rn atoms present in the Lucas cell at the beginning of counting, N_c :

$$N_c = N_D + N_c e^{-\lambda t_c} = \frac{S}{(1 - e^{-\lambda t_c}) (3\varepsilon)} \quad (4.4)$$

Here λ is the Rn decay constant, $\lambda = 0.181 \text{ days}^{-1}$. The error on N_c is

$$\Delta N_c = \frac{\Delta S}{(1 - e^{-\lambda t_c}) (3\varepsilon)}, \quad (4.5)$$

so that the number of radon atoms in the cell at the beginning of counting was $N_c = 67.6 \pm 18.6$ atoms.

The number of radon atoms present at the start of the extraction can be calculated as

$$N_o = \frac{N_c}{(e^{-\lambda \Delta t}) (\varepsilon_{AB}) \varepsilon_{BL}} \quad (4.6)$$

where Δt is the time separation between the beginning of the extraction, and the start of counting and ε_{AB} is the transfer efficiency from Trap A to B ($\approx 100\%$). ε_{BL} is the transfer efficiency from Trap B to the Lucas cell ($\approx 62\%$ for the D₂O MDG Rn board). Hence, the number of radon at the beginning of the extraction is $N_o = 103.2 \pm 28.5$.

If one assumes the Rn are produced at a constant rate R via emanation and/or leaks, then

$$R = \frac{N_o \lambda}{1 - e^{-\lambda t_s}} \quad (4.7)$$

where t_s is the seal time. This gives the final production rate of $R = 21.7 \pm 6.0$ Rn atoms/day.

Studies of the H₂O MDG revealed that the major source of Rn ingress in the system comes from the stainless steel degassing unit itself (approximately 450 Rn atoms/day [23]). Hence, the Liqui-Cel contactor as a degassing unit has demonstrated an order of magnitude decrease in background compared to the H₂O MDG.

Efficiency Run ID	Transfer Efficiency (%)
1	100.4 ± 1.3
2	98.2 ± 1.1
Average	99.3 ± 1.7

Table 4.5: The results of transfer efficiency measurements performed on the MDG Liqui-Cel contactor to confirm the background blank results.

It is not expected that the results for the single membrane will differ greatly for six present in the PDG, and so a lower limit can be applied for the Rn ingress due to the PDG system (approximately 250 Rn atoms/day).

The question did arise as to whether the results presented in Table 4.4 are reliable due to the complicated internal structure of the Liqui-Cel. The concern was that in the process of a typical blank measurement, not all of the radon emanated would reach the radon board from the Liqui-Cel. This concern was addressed with the measurement of Rn transfer efficiencies in the contactor. The process involved the injection of “hot” Lucas cells into the contactor and performing an extraction to measure the number collected during the typical time scale of a blank run. A “hot” Lucas cell is simply a Lucas cell which is filled with a large, known, number of Rn atoms (from 10^3 to a few 10^5). The radon in the hot cell are then injected at the quick connect shown in figure 4.7. The known amount of radon is recollected through the normal extraction process and returned to the same (now evacuated) Lucas Cell (note: this removes the need to deal with numerous relative efficiencies between Lucas cells). The efficiency to transfer Rn from a Liqui-Cel to the Rn board during a blank measurement has been measured to be close to 100% (table 4.5) and hence there is little retention of Rn within the Liqui-Cel itself. Hot cell transfer efficiency

analysis focuses on the decay time between the start times for counting of a Lucas Cell. For instance, in the second efficiency run shown in table 4.5, the details of the extraction were:

- Lucas cell: N9
- Original hot cell made: 14:55 October 27, 1997
- Original hot cell start counting: 17:22 October 27, 1997
- Original hot cell Counts/Live Time = 24240 counts/ 54563 secs
- Hot cell Injected: 9:11 October 28, 1997
- Evacuated Lucas Cell filled: 13:52 October 28, 1997
- Filled Lucas Cell start counting: 16:13 October 28, 1997
- Filled Lucas cell Counts/Live Time = 38804 counts/ 175774 secs.

If one performs an analysis as done for the blank measurements, one finds the original hot cell contained $1.18 \pm .10 \times 10^5$ Rn atoms at the start of counting. Similarly, one finds the re-filled Lucas Cell contained $6.63 \pm .46 \times 10^4$ Rn atoms at the start of counting. To determine the extraction efficiency, one needs to look at the number of atoms which decay between the two starts of counting (approximately 23 hours), and are therefore lost to the extraction. One quickly finds that approximately 84% of the original number of Rn atoms should be available at the start of counting of the re-filled Lucas Cell. However, if one assumes that the transfer efficiencies to the Rn board from the Liqui-Cel is 100%, and one knows the transfer efficiency from the H₂O MDG Rn board to a Lucas cell is 68%, then one finds that the expected number of atoms in the re-filled Lucas Cell at the start of counting should be $6.75 \pm .60 \times 10^4$ Rn atoms. If we compare this to the number actually measured in the

re-filled Lucas cell, one finds the transfer efficiency for Rn from the Liqui-Cel to the Rn board under normal blank conditions is $98.2 \pm 1.1\%$.

The determination of the backgrounds from the Liqui-Cel represented the end of the “dry” measurements to be performed on the contractor. At this point it was known that the selected device met all of the criteria for a D₂O degasser. The next step in completing the MDG design was to address the problems of the vapour recovery system for the D₂O and the Rn Board.

4.3.2 Vapour Recovery Design and Commissioning

In the light water system there is a large FTS water vapour trap (re-designed and constructed from acrylic by SNO) attached to the H₂O MDG. The re-designed FTS for the H₂O system has been measured to emanate very little radon. From the practical experience obtained with the light water FTS, it was hoped that a similar result could be achieved for the D₂O MDG vapour recovery unit. Based on the single membrane test results, a limit of 60 ml/hr water vapour was determined. These results were made with H₂O at 22 °C. For D₂O, with a viscosity approximately 1.3 times higher, and at lower temperatures, we expect this to be somewhat lower. However, the design criteria has been set to cope with 60 ml/hr of D₂O over the course of a normal run (typically 5 hours). It must have a high efficiency for condensing the D₂O in the presence of a 1 lpm (STP) flow of N₂ sweep-gas. Therefore the unit must operate at a temperature which will freeze the D₂O but not the Rn from the contactor. The original unit purchased is known as a Titan trap by FTS. It was designed with a teflon coated stainless steel canister containing a titanium coil. The canister sealed with a butyl rubber gasket (supplied by the company) a glass domed top with inlet and outlet connections for the gas. The refrigerant passes through the inside of the coil, bringing the device to an average operating temperature of

approximately -55°C .

Initial blank measurements of the original Titan trap unit (attached to the H_2O Rn board) were not at all encouraging. The design goal of the D_2O MDG was a system with a total background of < 450 Rn atoms/day production (less than the H_2O MDG which currently functions at the sensitivity limit of $1 \times 10^{-14} \text{ g}^{238}\text{U}/\text{gH}_2\text{O}$). The initial measured emanation rate was $9.74 \pm .25 \times 10^3$ Rn/day, and immediately a source for the Rn in the trap was sought. The most likely source of the Rn was the large butyl rubber gasket used to seal the trap to the glass lid. From past experiences with o-rings, we know that additives to the polymer can be high in radioactivity, or the sealing material may be impervious to Rn. In this case the origin and composition of the rubber material was unknown.

To test the gasket independently, it was placed in a standard 1L polypropylene (PP) column and attached to an Electrostatic counter (ESC) to count the ^{222}Rn (and ^{220}Rn) emanated [11]. After a day of counting, the ^{222}Rn peak region contained well over 2500 counts; an extremely high number for ESC measurements which typically measures a few tens of counts in a given peak after a number of counting days. The gasket was immediately removed from the Titan trap and a replacement acrylic insert was installed. The insert sealed radially to the inside walls of the teflon coated canister and had a face seal to the glass top, with SNO specially-ordered clean viton o-rings.

After installation of the insert, a second series of blanks were made on the trap and glass top combination (see table 4.6). The results showed the typical pattern for emanation of materials. As time progresses the unsupported Rn which was absorbed in the materials is removed (either by decay or through the extractions) and one approaches a plateau for the Rn emanated. One can see that the replacement of the butyl gasket made a significant difference in the Rn background level of the trap, although at about 650 Rn/day, it is not quite low enough to meet the desired

Blank ID	Seal Date	Rn Production Rate
	(Days from Seal)	(Rn atoms/day)
1	0.125	4386 ± 489
2	1.04	2250 ± 374
3	8.82	653 ± 51

Table 4.6: The results of blanks performed on the Titan trap with the butyl rubber gasket replaced with a replacement acrylic insert. The Run ID is for progressive blanks from T_0 (Run 1 was performed 3 hours after sealing time T_0).

sensitivity limit of the MDG.

The next Titan trap component to be studied, and the final item which could be easily replaced, was the glass domed top. The top has a large surface area exposed to the inside of the trap and an unclean glass source could explain the elevated levels measured, since it was expected the teflon coated stainless steel canister should be clean. The glass top was replaced with a “clean” stainless steel plate which was designed and machined specifically for the blanks. The results of a set of measurements in this new design (table 4.7) again showed a Rn production rate of about 750 Rn/day. Since the values were no lower than measured in previous blanks, it was believed that the source must be dust, present during the coating process, trapped under the teflon of the canister.

Therefore the decision was made to replace the entire teflon-coated stainless steel canister and titanium coil. A new trap and coil was designed and machined from clean sources of stainless steel. The glass top was replaced with an acrylic plate, and the need for the sealing insert was eliminated with the inclusion of a face o-ring groove in the new canister. The new trap arrived, was passivated, methanol cleaned

Blank ID	Seal Time	Rn Production Rate
	(days)	(Rn atoms/day)
1	6.76	1033 ± 62
2	5.15	645 ± 23
3	2.54	835 ± 37
4	2.86	762 ± 33

Table 4.7: The results of blanks performed on the Titan trap with the butyl rubber gasket replaced with a replacement acrylic insert and the glass dome replaced with a stainless steel top.

and plumbed into the completed D₂O MDG Rn board. Blanks performed on the new unit have shown a marked improvement in the Rn emanation of the trap region (table 4.8), with Rn production rates below the design goal of 450 Rn/day.

Blank ID	Seal Time	Rn Production Rate
	(days)	(Rn atoms/day)
1	6.20	427 ± 17
2	6.8	356 ± 14

Table 4.8: The results of blanks performed on the “New” Titan trap with the acrylic lid and “clean” viton o-ring.

4.3.3 Radon Board Design and Commissioning

The Rn board (figure 4.2) consists of a primary trap (Trap A), secondary trap (Trap B) and a final region to transfer the trapped Rn to the Lucas cell. In an extraction run, the responsibility of Trap A is to capture the Rn from the gas stream with a high efficiency. The inclusion of brass wool packed into Trap A generates an increased surface area in the trap to collect the Rn, hence obtaining a much higher efficiency. Trap A must also have a high efficiency for transferring the Rn to the secondary trap in order to ensure a maximum number of extracted atoms are available for injection to a Lucas cell. This is much more critical as one approaches the ^{238}U limit in the D_2O and the number of available atoms becomes small. To handle the large amount of sweep gas which must pass through the board to reach the vacuum pump, and still keep the pressure in the trap low, Trap A is formed from 1/2 inch OD stainless steel (SS) tubing. It is connected to the vacuum manifold where a transition to 1 inch OD SS tubing to the vacuum pump creates very efficient pumping.

Trap B acts as a volume reduction trap. Trap A is heated above room temperature after the extraction portion of a run is complete to free the Rn frozen to the interior surfaces. With Trap B immersed in LN_2 , a cryo-pumping of the gasses from Trap A to Trap B occurs. Therefore the collected Rn (and CO_2) is transferred from the large volume region of Trap A (approximately 50 cm^3 in a single U-shaped trap) to the very small volume of Trap B (approximately 30 cm of 1/8 inch looped SS tubing for a volume of 2.4 cm^3).

The final region of the Rn board is dedicated to the transfer of the Rn collected in Trap B into a Lucas Cell. To improve transfer efficiencies, the connections and tubing used in the transfer region are 1/8 inch and all dead volumes are minimized. The transfer is actually performed using Swagelok self-sealing Quick-Connects. The Quick-Connects allow the Lucas Cell connections to the Rn board to be made with

ease and a minimal ingress of mine air into the system during a transfer.

In general, the Rn board must be a very low background device. This includes backgrounds due to emanation of Rn from the materials of the board and ingresses of mine air due to small leaks in the system. The SS tubing which is the primary construction material of the board was passivated and methanol cleaned prior to use in the board in order to remove any residual surface "dirt". In order to make a leak tight system the SS tubing is joined using Swagelok compression fittings, which have been demonstrated in past systems to provide reliable seals for vacuum and pressurized systems over a wide range of temperatures. The reliability of a seal is critical for the Rn board where temperatures are taken from LN₂ temperatures to well above room temperature over a period of a few minutes. Once completed, the integrity of the Rn board was checked using a He leak detector and even the smallest leaks were addressed and repaired, until the board was leak tight to approximately 10^{-8} cm³/s.

After leak checking the newly constructed Rn board, there were two basic measurements which had to be made prior to use; the measurement of blanks on the regions of the board to locate any possible background problem zones which should be replaced, and a measurement of the trapping and transfer efficiencies of the two traps. The blank measurements were performed much the same as the typical blanks described earlier. Initially the Rn board was baked (heated to a very high temperature) and allowed to pump down for a number of hours to remove any water vapour and gasses which might remain in the board from assembly (or prior extractions). At the completion of the pump down, each region of the board was sealed individually. After a given seal time, typically three to five days, the individual regions were each extracted independently into Lucas cells. To measure the transfer regions (quick connects), evacuated Lucas cells were attached to the sealed regions, removed and counted. Trap B was extracted by simply heating slightly, and opening it to the

evacuated transfer region with a Lucas cell attached. Trap A and the intermediate region (defined as the transition region from 1/2 inch to 1/8 inch tubing) blanks were performed by heating the regions and opening them to a LN₂ cooled Trap B. The process of heating was repeated on Trap B and the collected gasses are transferred to a Lucas cell. The results of the blanks (table 4.9) showed that the board was in fact free of any troublesome background regions and the transfer efficiency measurements of the board could proceed.

The critical region of the board to have clean is Trap A since it is the region which is used longest during the extractions. Significant production in Trap A would have large implications for the sensitivity of the Rn board, and therefore the MDG. However, the results in table 4.9 show that the care taken in building Trap A has generated a very low background region. The higher values measured in the intermediate region and Trap B are of little concern since they are used for very short times compared to the total extraction time (typically 1/2 hour), and therefore contribute very little to the overall background of the system. None the less, efforts are underway to locate the sources of these backgrounds.

The measurements of the transfer efficiencies for Trap A and Trap B of the board provide critical values which determine the MDG performance. Transfer efficiencies for the traps again used the "hot" cell injection technique. For measurement of the Rn transfer efficiency from Trap B to the Lucas cell, the method in the past had been to use the volumetric ratio between the two regions. To confirm the validity of this practice a hot cell was attached to the transfer region quick connect and opened to a LN₂ cooled Trap B. The regions were then very slowly evacuated, in the reverse direction to the normal pumping path, through the Rn board and to the vacuum pump. This removed any extraneous gasses and ensured all the Rn in the hot cell was collected in Trap B. Isolating and heating Trap B freed the known amount of collected Rn which was then re-injected into the evacuated Lucas cell,

Rn Board Region	Blank ID	Rn Production Rate (Rn atoms/day)
Trap A	1	1.3 ± 8.5
(5 hours)	2	6.7 ± 5.7
Intermediate	1	51.4 ± 27.9
(10 min)	2	26.9 ± 10.3
	3	15.0 ± 6.9
Trap B	1	27.2 ± 10.5
(20 min)	2	36.7 ± 9.1
Lucas cell regions	No results above lucas cell backgrounds.	

Table 4.9: The results of blanks performed on the various regions of the D₂O Rn board. The times listed for each region indicate the amount of time each region is used in a typical extraction.

Efficiency ID	Transfer Efficiency (%)
Hot cell injection 1	61.6 ± 1.6
Hot cell injection 2	61.9 ± 1.5
Volumetric Ratio of regions	63.8 ± 2.0

Table 4.10: The results of transfer efficiency measurements from Trap B to a Lucas cell using two independent methods.

as in the standard procedure. The result of the transfer was then counted, and a transfer efficiency for radon trapped in Trap B to the Lucas cell is determined (table 4.10). One can see the naive approach to measuring transfer efficiencies (volumetric approach) is consistent with actual Rn measurements, although from this point forward the value used for the efficiency is 61.8 ± 1.0 (sys)%.

The measurement of the transfer efficiency (and also the trapping efficiency) of Trap A to Trap B cannot be done simply by comparing volume ratios. The transfer path is much more complicated and the regions are much larger, so a number of hot cell injections were performed. The Rn board was designed with a sampling port, prior to the entrance to Trap A, for which leak detecting and injections could be performed. The hot cells were injected at this port and extracted to a LN₂ cooled Trap A using the vacuum pump, and a typical transfer of the Rn to a Lucas cell followed. The results (table 4.11) are computed from the initial Rn content of the hot cell compared to the re-filled Lucas cell. The efficiency for transfer from Trap B to the Lucas cell and corrections for the radon decays are included. One can see that the efficiencies from the raw data were very much higher than 100%, and it is very difficult to find a background large enough to suppress the values below 100%. Calculations of the number of ²²²Rn daughters which would remain in the cells to

Transfer ID	Efficiency (%)	Efficiency (%)
	Before Background Subtraction	With Background Subtraction
1	109.6 ± 2.4	105.3 ± 3.5
2	102.9 ± 2.1	98.9 ± 3.3
3	116.9 ± 1.6	
4	111.1 ± 2.1	
5	105.7 ± 1.9	
6	120.1 ± 3.1	
7	117.9 ± 4.5	
8	133.5 ± 6.8	94.0 ± 9.6
Average		100.5 ± 2.3 (stat)

Table 4.11: The results of transfer efficiency measurements from Trap A to Trap B. The second column shows the transfer efficiencies determined from the raw counting. The third column shows the transfer efficiencies with the hot cell back ground subtraction included where measurements were available.

contribute to the background were found to be very small for the time between the evacuating of the Lucas cell and the start time of counting.

In normal use, the Lucas cell backgrounds are of the order of 10 counts per day. However, in this set of measurements, cells with high activity were used in rapid succession. While the radon is captive in the cell, a fraction of it will diffuse into the walls of the cell. During the short extraction period of these measurements (typically 1/2 hour), only a fraction of the radon which diffused into the walls will be removed from the cell. Hence some amount of radon remains within the cell. When the cell is refilled, the partial pressure of Rn is much lower, and so some of

the trapped radon will diffuse back into the active cell volume. This characteristic of the Lucas cell had not been appreciated until it was observed that upon evacuating the Lucas cells, after the hot cell runs, highly elevated count rates occurred for the cells for many days. Studies of these count rates have shown the decay parent is in fact Rn which has remained in the cell. For three of the runs (numbers 1, 2 and 8) in table 4.11, the Lucas cells were evacuated and their backgrounds counted at the completion of the measurements. Based on the observed backgrounds, a background subtraction was attempted for the cells (column 3, table 4.11). The background counts were normalized to the original counting time of the cell, and removed from the total counts (similar to typical cell background subtraction). The background subtraction is not obvious, as the cells have been used in many applications, and the rate at which the Rn is diffusing from the cell is difficult to estimate.

The assumption had always been that the transfer efficiency from Trap A to Trap B was very high (approximately 100%). Again it has been observed this the assumption was consistent with the results. Using the results of the transfer efficiency measurements for both traps, one can quickly determine that of the Rn that enters the Rn board from a degassing run, approximately 62% will be transferred for counting to the Lucas cell. Therefore, the D₂O Rn board has been successfully designed and commissioned as a low background device with sufficient performance to meet the design sensitivity of the MDG.

4.3.4 MDG System Test Results

The three major components of the D₂O MDG have been discussed in detail individually. The design and construction is complete. The commissioning steps under dry conditions (which are primarily concerned with the relevant efficiencies for the devices and overall backgrounds) have been carefully studied. If the connections

Blank ID	Seal Time (days)	Rn Production (Rn atoms/day)
1	2.61	951 ± 53
2	5.20	901 ± 49
Average		925 ± 18

Table 4.12: The results of blank background measurements made on the Titan trap when attached to the D₂O Rn board. The Titan trap configuration had the original SS canister with teflon coating, with the new acrylic gasket and acrylic lid in place.

between the components can be made cleanly, and no significant backgrounds are present in, for instance, the process piping (to which the Liqui-Cel is exposed), then the expectation is that the sum of the backgrounds for the components should be equal to those measured for a total system blank. Here the total system is defined as the Liqui-Cel, Titan trap, Rn board and all plumbing used to connect the components. To check the entire system to ensure that no extra backgrounds were present, total system blanks were performed. The only difference from this system and the final system was that the Titan trap was not in its final state. Instead it still had the original teflon coated canister, the acrylic insert and the new acrylic top. Hence, before measuring the total system blanks, the Titan trap in this configuration had to be measured for backgrounds. The results are shown in table 4.12.

The results of the system blanks compared to those predicted by summing the individual components (Table 4.13) show good agreement. Thus one is able to make a confident prediction for the total background of the system with the new Titan trap. Using the value of the background measured for the Titan trap in its cleanest state (table 4.8) we can estimate the total background for a typical Rn assay. The

Component	Production Rate (Rn atoms/day)
Total system (blank #1)	1056 ± 42
Total system (blank #2)	941 ± 27
Total system (Average)	998 ± 49
Titan trap	925 ± 18
Liqui-Cel	41 ± 4
Rn Board (ave. prod. of Trap A)	4.5 ± 1.9
Sum	971 ± 24

Table 4.13: A comparison of the total system blanks to the sum of the blanks for individual components. The Rn board contribution to background Rn production is taken from Trap A only, since the other regions of the Rn board are always baked and evacuated as standard practice prior to an experiment, and are only exposed for a very brief period.

backgrounds expected from a 5 hour extraction are: 83 atoms from the Titan trap, 10 from the Liqui-Cel and possibly 1 from the Rn board for a total of 94, of which 56 will be transferred to the Lucas cell.

All efficiencies for the MDG system, with the exception of the Rn degassing efficiency of the Liqui-Cel, are known. The expected degassing efficiency for Rn is expected to be no lower than 80%, for the Liqui-Cel at MDG flow rates. Hence we may estimate the MDG performance assuming an activity in the water equivalent to 1×10^{-14} g²³⁸U/gD₂O. At this limit, and with the ²³⁸U daughters in secular equilibrium, one has approximately 60 Rn atoms/tonne in the D₂O. At the typical flow rate of 20 lpm over an extraction period of 5 hours, one can sample 6 tonnes of D₂O, or some 360 Rn atoms. With an 80% Rn removal efficiency, the Liqui-Cel will remove approximately 288 Rn atoms. The efficiency for Rn to move through the Liqui-Cel was seen to be very close to 100%, and transport for Rn through the Titan trap should be similar. Therefore, of the 288 Rn atoms which enter the Rn board, approximately 62% will be transferred to the Lucas cell for counting or 178 Rn atoms. The total Rn in the Lucas cell (a total of 234 atoms of which 56 are background) will produce about 70 counts in the first day of counting. This value is determined by considering the delay from the end of collection to the start of counting, typically 2 hours, (0.98) and the counting efficiency ($0.63 \times 3 = 1.89$), and the probability of decay during the first day of counting ($1 - 0.834 = 0.166$) [23]. Therefore the constructed D₂O MDG clearly has sensitivity for effective Rn studies to the limit of the ²³⁸U concentration with a better than 3:1 signal to background measurement. Furthermore, after the first day of counting the expected 70 counts are well above the Lucas cell backgrounds, with a typical count rate of 10 counts/day. Thus a monitoring system for the Rn concentrations in the D₂O of the SNO experiment at the expected SNO limits has been successfully designed, constructed and is ready to be commissioned with D₂O when it becomes available.

4.4 PDG Design

The design of the SNO D₂O PDG is based on the single membrane tests. The system must be capable of processing 160 lpm of D₂O for extended runs of 24 hour operation. The preferred mode of operation is a vacuum-assisted N₂ sweep mode, for which the vacuum is provided by the vapour recovery system and the sweep-gas is taken from the cover-gas system (approximately 1 lpm per contactor).

4.4.1 Sweep-Gas Control

For continuous operation, it is necessary to automate the regulation of both the water and sweep gas flow rates. The water flow rate through the PDG is controlled at the main recirculation and booster pumps of the system. In the following subsection we describe the instrumentation developed to control the sweep gas flow.

The sweep-gas flow into the PDG is controlled with a mass flow controller using a dial-up potentiometer. Two mass flow controllers (Brook Instruments mass flow controller model 5850TR) were obtained from CRPP, both of which were previously used for a low flow Argon gas mixture. A preliminary study of the mass flow controllers in their initial condition showed a very nice linearity over the entire flow range of the devices. It was determined that the desired PDG sweep-gas flows could be achieved with the mass flow controllers, through the change of two key parts; the orifice plate and the restrictor assembly. Calculations were made to determine the appropriate orifice opening size and restrictor for the flow range of the PDG under operating conditions. The restrictor assembly and orifice plate were obtained, and once the new parts were installed, a test with a calibration device showed the PDG mass flow controller now produced sufficient N₂ flow to supply the PDG with high accuracy over the desired range (0-14 lpm (STP) N₂) (figure 4.8).

It was also imperative that the controllers not contribute significantly to the

Figure 4.8: The calibration curve for the PDG mass flow controller with new orifice and restrictor assembly installed.

backgrounds. Calculations demonstrated that the o-rings present were sufficient to prevent Rn diffusion (< 1 Rn atom entry/day) into the sealed devices. In addition, radon emanation measurements of the controllers sealed under vacuum gave no indication of Rn production above the low Lucas cell backgrounds (10 counts/day). On completion of these tests, the controller has been installed on the PDG system.

4.4.2 The Vapour Recovery System

The PDG vapour recovery system has been designed based on the single membrane vapour load recovery tests. From these tests the expected vapour load is approximately 1 ml/min per contactor. Therefore the PDG vapour recovery system must be sufficient to process a load of approximately 360 ml/hr. It is also required to provide the vacuum to maintain the 30-70 torr outlet pressure of the PDG, in the presence of the N_2 sweep flow of 5-10 lpm(STP).

Two general designs were considered for the vapour recovery system. The first consisted of two FTS type vapour traps (similar to the ones used on the H_2O and D_2O MDGs) used in a parallel configuration. While one of the traps was used to condense the D_2O , the other would be in a defrost cycle, and vice-versa, to give the possibility of continuous operation. The system has the advantage of being plumbed in prior to the vacuum pump, therefore reducing the possibility of downgrading the D_2O with the vacuum pump vapours. The other design considered uses a condenser at the outlet of a dry vacuum pump to condense the D_2O vapour. The advantage of this design is that with the proper choice in condenser (one which contains an auto-drain design) the system becomes capable of continuous operation with minimal manual monitoring. There is still concern over the potential for contaminating the D_2O as the vapour passes through a vacuum pump, but the advantages of such a simple to operate design makes it an ideal choice.

The vapour recovery system is designed with a Pfeiffer dry (oil-free) vacuum pump immediately following the sweep-gas outlet of the PDG. The vacuum pump in actuality draws the sweep-gas through the PDG (providing the necessary 10 psi ΔP across the mass flow controller to reach the desired flow rates) since the cover-gas provides only a stagnant source of clean N_2 . The pump maintains the temperature of the gas which passes through it at approximately 100°C, thereby reducing the amount condensation in the pump prior to entering the condenser. The pump was also chosen based on the internal materials of the device (chosen such that any possible wetted parts do not allow any possibility of corrosion) and the oil-free operation. This which any possibility of contamination due to oil vapours.

Located directly below the pump outlet is the condenser chosen to recover the D_2O from the vapour. The condenser makes use of a toroid cooling coil design which keeps the unit very compact while maintaining a very high efficiency. The condenser flows glycol (from the chiller loop of the lab) through to the inside of the coil at about 4 °C. The temperature of the vapour passing through the condenser is reduced to approximately 10°C and the D_2O in the vapour will collect onto the coils of the condenser. The remaining vapour is sent out to the off-gas header exhaust in the mine. The water (D_2O) in the condenser is gravity drained to a reservoir located physically below the condenser where the D_2O can be tested for any downgrading of the isotopic purity, and chemical impurities prior to returning to the main recirculation loop.

The main PDG unit (the six Liqui-Cels in new housings) has already progressed through the design and construction stages. It is plumbed into the D_2O system water lines and attached to the N_2 cover-gas system with all controllers in place and operational. The PDG vapour recovery system has been installed. The main commissioning steps await the arrival of D_2O .

Chapter 5

Conclusions

5.1 Summary of Results

5.1.1 General Results

We have designed and constructed low background, high efficiency process and monitor membrane degassing systems for the SNO heavy water system. The removal efficiency for O_2 was measured to be $> 99\%$, and is consistent with company specifications, over the water flow rates tested. The Rn removal efficiency, calculated relative to the known O_2 efficiency, has shown it meets the design criteria for the PDG (95%) and is expected to surpass the necessary level of 80% for the MDG.

Despite attempts to lower the vapour loads (redesign of the membrane housings), measurements of the vapour loads from the contactors under numerous operating conditions have demonstrated that the vapour load is higher than originally expected. In order to cope with these increased loads, the vapour recovery systems have been designed to handle a rate of 1 ml/min of D_2O per contactor. Therefore the systems must efficiently condense 60 ml/hr for up to 5 hours for the MDG, and 360 ml/hr during the expected continuous operation of the PDG.

5.1.2 Commissioning Results

The commissioning of the MDG required detailed studies of the Rn backgrounds for the three primary components. Rn background production of the Liqui-Cel contactor was a concern since past measurements of the H₂O MDG degassing unit showed it to be the primary background source in the system. However, the measured Liqui-Cel background (41.4 ± 3.8 Rn atoms produced/day) is a full order of magnitude lower than the H₂O MDG. Confidence in the blanks performed to give the background was demonstrated with an efficiency measurement of $99.3 \pm 1.7\%$ for Rn transfer from the Liqui-Cel to the Rn board.

The vapour recovery system for the D₂O MDG progressed through a series of changes due to the high Rn production rates measured in the original unit. Rn backgrounds for the original system were in excess of 9000 Rn atoms produced/day. Successful redesign of the system has resulted in a vapour recovery system which now has a Rn production rate of approximately 400 atoms/day, while maintaining the high efficiency condensing characteristics of the original system.

The backgrounds for the various sections of the Rn board were measured in detail. It was found the critical regions (Trap A, Trap B and the transfer regions) each satisfied the necessary low background condition for the Rn assay system. The Rn board contribution to the total MDG background during a typical 5 hour assay is < 5 Rn atoms. The transfer efficiencies for Rn through the Rn board have also been studied, and have demonstrated that of the Rn which enters the board during an extraction, 61.8% will enter the Lucas cell for counting.

The studies on the completed D₂O MDG system have demonstrated that the system is capable of performing confident assays of the very low Rn levels in the D₂O. The expected result from the assays is a 3:1 signal to background ratio of Rn in the Lucas cell, for a 1×10^{-14} g²³⁸U/gD₂O. Since the D₂O PDG is in effect a

direct scale-up of the MDG, the result for backgrounds of the MDG Liqui-Cel can be applied to the PDG. Therefore, one can estimate the Rn background contribution to the overall water systems from the PDG. If the 1000 tonnes of D₂O in the Acrylic Vessel contain an average of 60 Rn atoms/tonne, then the D₂O PDG will contribute approximately 250 Rn/day (for continuous operation of the PDG) to the total 60000 Rn present in the Vessel. However, with a 95% Rn removal efficiency and a flow rate of 160 lpm, one can predict the PDG will remove > 13000 Rn/day from the D₂O. Therefore, the background contribution of the PDG can be considered small compared to the remaining Rn in the detector.

5.2 Future Work

There remains a substantial amount of work which has yet to be performed to complete the commissioning of the D₂O degassing systems. Most of these tests require D₂O, which is not yet available for commissioning.

Measurement of the vapour load from the contactors with D₂O has yet to be done. This includes a study of the possible temperature dependence on the vapour loads as the D₂O temperature changes from 20 °C at the start of water fill to 10 °C during normal detector operation. Although it is not expected to exceed the limit set for H₂O (1 ml/min), knowledge of the actual vapour load is extremely important for the PDG vapour recovery system. The current vapour recovery system is designed to condense approximately 98.6% of the D₂O vapour which enters the system in the degasser outlet vapour stream. This is equal to D₂O losses of approximately 4.9 g/hr or 43.8 kg (D₂O)/year (for the case of continuous operation of the PDG). If one assumes a value of \$300/kg, then these losses are equal to approximately \$12000 per year. When the actual D₂O vapour loads have been determined, and when the actual radon content of the D₂O has been measured, the amount of time we need to

run the PDG will be known. If continuous operation is required, it may be necessary to include a secondary recovery system to further reduce losses.

Future study of the MDG vapour recovery system are also required. After the final measurement of the Titan Trap's background (approximately 400 Rn/day), the unit was sent for further electropolishing, followed by ultrasonic cleaning with RadiacWashTM, and a final methanol rinse, in the attempt to removal any residual activity and further reduce the background levels. Final blanks on the cleaned unit need to be performed, and a total system blank and transfer efficiency should be made prior to the start of "wet" commissioning to confirm previous measurements.

Finally, a great deal of effort will be required to measure, with D₂O, the degassing efficiencies of the PDG and MDG for O₂ and, more importantly, Rn. Online O₂ studies will be used to optimize the operating conditions (sweep gas rate for the given flow rates) of the degassers. A new O₂ probe has been acquired for use in D₂O and final installation of the dissolved oxygen probe is required for the degassing systems. This includes deuterating the probe (previously used in H₂O measurements) and commissioning the probe for operation with the D₂O system (wiring and logic programming to the water system monitoring software).

Determining the Rn degassing efficiency will involve flowing D₂O (which is spiked with a known amount of Rn) through a closed loop of piping which includes the degassers and Rn assay system, and performing consecutive Rn extractions during the degassing. The consecutive measurements provide the degassing efficiency curves for the systems, and ultimately will determine the necessary duty cycle for the PDG.

Appendix A

Personal Contributions

In an experiment such as SNO, which is made up of a collaboration of 70 some scientists, it is often difficult to gauge the individual contributions. For the work described in this thesis most of the systems taken through the design and construction stages were either overseen and/or performed by the author.

A major contribution involved the redesign and testing of the MDG vapour recovery system to achieve a low background device. The design and construction of the Rn board was completed as an individual effort, with a guiding hand from the collaboration's Rn experts. For the vapour load measurements of the single Liqui-Cel membrane, I was an active participant of the experiment design and performance teams, including the membrane pressure drop measurements and analysis. The background and transfer efficiency measurements were developed and performed by myself over the course of the research term.

In general, it was my responsibility to ensure an operational process and monitor degassing system was ready for use once the D_2O became available, and I continue to be the primary consultant on the systems as the D_2O commissioning approaches.

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IMAGE EVALUATION TEST TARGET (QA-3)

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