

Estimates of Gas Leakage through the Deck SNO-STR-92-097

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The physics requirements for the cover gas system have been laid out in the cover gas design criteria document [1]. There it was assumed that there would be a total leakage of about 20 l of LN₂ per day and an allowance for no more than about 10% of that amount for leaks from each of the PMT, PSUP and AV systems. In this document an estimate of the total seal length has been used to estimate the quality of seal that would be required to meet such a criterion.

Estimates

In order to develop some idea of the demands that the above requirements place on the sealing of the various components, both the required clearance for a gasket seal on components and the tolerable size and concentration of pinholes was estimated. These can be considered as alternative ways in which the leakage budget might be reached.

A rough estimate of the required clearance for a gasket can be obtained

Conclusions

The above estimates indicate that the required separation tolerance for joint surfaces is of the order of 5 to 15 μm , and that pinholes should be significantly smaller than 0.1 mm and at a frequency of less than about 10 per foot. Greatest attention will have to be paid to the sealing of the cable bundles and cable feedthroughs.

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^{222}Rn emanation into vacuum

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Abstract

A low-background ZnS scintillator cell based on a design by Lucas has been developed for ^{222}Rn detection. Typical cells have 63% detection efficiency and 3 counts per day background. The cells have been used in measurements of ^{222}Rn emanation rate into vacuum from materials to be used under water in the Sudbury Neutrino Observatory (SNO) solar neutrino detector. The results are presented and the impact on detector design is discussed.

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*How are the
counts turned into
Rn ratios? Singles
or doubles or triples?*

1 Introduction

ZnS-lined scintillator cells (Lucas cells) have been used in radon detection for over 30 years [1, 2]. Most of the development work during this time has been concentrated on increasing detection efficiency. On the other hand, all these cells have relatively high background (several counts per minute). A low background, reasonably high detection efficiency radon detector is required to determine the background caused by radon and its progeny in the Sudbury Neutrino Observatory (SNO), a heavy-water (D_2O) neutrino detector under construction near Sudbury, Ontario, Canada [3]. Figure 1 shows the main parts of the detector. Neutrino interactions in the D_2O produce relativistic electrons or free neutrons. The neutrons are thermalized in the D_2O and are subsequently captured, generating γ -rays which in turn produce relativistic electrons. The electrons from either source will produce Čerenkov photons which pass through the D_2O , through the acrylic vessel which contains the D_2O , through the ultrapure H_2O used as background shielding and to the photomultipliers (PMTs) where they are detected.

The most serious source of background in the SNO detector is the radiation from naturally occurring radionuclides. ^{238}U and ^{232}Th and their daughters (particularly ^{214}Bi and ^{208}Tl) can contribute to the background by high energy β and γ -rays emitted in their decay. Monte Carlo calculations [3] shows that the tolerable concentration of the U chain in secular equilibrium is about 15×10^{-14} gU/ g in the H_2O nearest to the acrylic vessel, and 1×10^{-14} gU/ g in the D_2O .

The emanation of ^{222}Rn and ^{220}Rn and the leaching of their parent radium (^{226}Ra ,

^{224}Ra) from materials into water can cause substantial disequilibrium in the water. The leaching of radium in the SNO detector is being studied by SNO collaborators at Oxford and Queen's [4]. There exists a body of literature on radon emanation from building materials (such as bricks, gypsum board, etc.) which have relatively high radium concentration. Measuring the ^{222}Rn emanation rate from low radioactivity detector materials such as stainless steel, signal cables and PMTs is the objective of the work reported in this paper.

By detecting ^{222}Rn , the rate of 2.45 MeV background gamma rays in the SNO detector from ^{214}Bi decay is determined directly even if there is disequilibrium in the radium or preceding long-lived nuclei. ^{222}Rn has a half-life of 3.8 days, but all daughters before ^{214}Bi have fast half-lives. ^{220}Rn , with a 55 second half-life, is more difficult to detect and requires different techniques [4, 5].

In the first section of this paper, the development of low background scintillation cells is described together with test results. Such cells were used in the measurements of ^{222}Rn emanation into vacuum discussed in the second section. These measurements were also carried out in such a way as to distinguish between ^{222}Rn outgassing and ^{226}Ra -supported radon emanation, which is more important in the SNO detector. In the third section the impact of ^{222}Rn emanation in the SNO detector and some further developments on the scintillation cell is discussed.

2 Development of a low-background scintillation cell

A Lucas cell detector consists of a chamber which is coated on the inside with ZnS

scintillator. An activator such as silver is put into the ZnS to make it scintillate. A photomultiplier is coupled to the window of the cell to detect the light emitted when an alpha particle from the decay of radon or its daughters strikes the ZnS. The cell is filled and sealed through a valve. Typically a SwagelokTM Quick-Connect is used because of its automatic shutoff feature when it is disconnected from the filling apparatus.

In order to have high detection efficiency, a large volume cell is often used [6]. However a larger volume needs more ZnS to coat the surface which results in a higher background. The largest volume with minimum surface area is a spherical design. The main factors considered in a new scintillation cell design are described below:

- 1). Cell body material. The material to be used for the cell body must have a low alpha particle surface-emission rate. Ultraviolet-transmitting (UVT) acrylic is one of the best among low radioactivity materials (< 10 ppt U, Th [4, 7]) and is also transparent. Methylene chloride solvent is used to seal an acrylic window to the cell body and to dissolve the acrylic surface to hold the ZnS coating.

- 2). ZnS sample selection. Six different ZnS (silver activated) scintillator samples were tested for their relative light output and background. About 10 mg/cm² of ZnS was sandwiched between two flat pieces of acrylic sheet, taking care to seal the edges and exclude air. After a three day wait to allow radon from residual air to decay away, a PMT was coupled to one side and the background count rate was determined. The relative light output was determined by comparison of the pulse amplitude spectrum from each sample.

There was about a factor of ten variation in the background rate and a factor of

five variation in light output among the six samples tested. The sample from Johnson Associates (Montville, NJ, USA 07045) was selected as the best compromise between light output and background rate.

3). ZnS thickness optimization. The ZnS thickness has to be optimized for light yield and radioactivity background. The ZnS was coated on a flat piece of acrylic by the following deposition method [8]. First the acrylic piece was submersed in a solution of ZnS suspended in ethyl alcohol. The thickness of the ZnS layer was varied by varying the deposition time. After the acrylic piece was taken out from the solution and dried, methylene chloride was used to fix the ZnS onto the acrylic. The ZnS thickness was obtained by the difference in weight before and after the deposition.

Two different geometries were investigated: "transmission" and "reflection". The pulse height spectra obtained using a ^{241}Am alpha source are shown in Figure 2 (a, b) for these two cases. "Reflection" geometry gives an optimum ZnS thickness of about 10 mg/cm^2 , equal to the range of a 5 MeV alpha particle in ZnS. This thickness was chosen for our cells.

what is
'optimum'
?

4). PMT selection. A low noise PMT is preferred for low background measurements. However the light amplitude from the ZnS scintillator is much higher than the PMT noise amplitude, so the choice of PMT is not critical. Also the scintillation light from ZnS(Ag) peaks is in the blue (4500 \AA) region which matches the response of bi-alkaline PMT photocathodes [9].

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5). Cell shape. The shape of the Lucas cell was chosen to maximize the light striking

the PMT and minimize the background. A hemispherical cell with a transparent window was designed. The outside diameter of the cell is two inches to match the diameter of the Philips XP2262B PMT chosen. Coating the cell window with a very thin ZnS layer results in high detection efficiency but some of the pulses are degraded into PMT noise. We chose not to coat the cell window, thus sacrificing detector efficiency, but obtaining pulses clearly separated from the PMT noise.

The hemispherical two-inch diameter scintillation cell designed with the above considerations is shown in Figure 3. Figure 4 shows a typical pulse height spectrum with the cell filled with radon. For comparison, Figure 4 also shows a spectrum obtained from a commercial Lucas cell [10] with a cylindrical shape. For the hemispherical design, the signals are very clearly separated from the PMT noise. Furthermore, the cell background was measured to be 3 counts per day for the new hemispherical cell (surface area = 20 cm^2), as compared to 3000 counts per day for the commercial cell (surface area = 145 cm^2). For the commercial cell, the type and thickness of ZnS, the method of ZnS deposition and the radioactivity of the cell body material (in this case aluminum) together give rise to the higher background.

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o
We can rule out several sources which might produce background scintillations in the cell. Cosmic rays do not produce significant scintillation in the thin ZnS, nor do β and γ -rays [11]. Assuming the air has a ^{222}Rn concentration of 2 pCi per liter [12], then our cell with a volume of 12 cm^3 and a residual pressure of 200 microns would have about 6×10^{-2} counts per day. Acrylic even at a 100 ppt U level would give less than 1 count per day

for our cell design. Hence the background of the cell is mainly from natural radioactivity in the ZnS. The alpha counting rate was measured to be about 15 counts per day per gram of ZnS. If we assume all these counts are from the ^{238}U decay chain alphas and the chain is in secular equilibrium, then the inferred U level is about 2×10^{-9} gU/g ZnS (i.e. 2 ppb).

The cell detection efficiency is defined as the measured alpha decay rate divided by the actual Rn decay rate. An efficiency calibration was done by putting a well determined amount of Rn into the hemispherical scintillation cell and counting. The amount of Rn inside the cell was calibrated by Bigu [16]. The detection efficiency was found to be $3 \times (62 \pm 3 \%)$ compared to 66.6% of the geometric area covered with ZnS. The factor of 3 comes from the three alpha decays associated with each ^{222}Rn decay (^{222}Rn , ^{218}Po and ^{214}Bi) when they come to equilibrium about 3 hours after filling the cell.

Additional background identification was done by recording the time associated with each event. For ^{222}Rn , the alpha from its decay is followed by the ^{218}Po ($t_{1/2} = 31$ minute) alpha. The alpha from the decay of ^{220}Rn is followed by the ^{216}Po ($t_{1/2} = 0.14$ second) alpha. For total rates which are low (as in measuring the scintillation cell backgrounds), two events within 0.5 second of each other have a very high probability of being from ^{220}Rn . It is interesting to note for an accumulated background run of 72 hours on 30 mg of ZnS, we did not observe any ^{220}Rn decays, which indicates there is the equivalent of less than 5 ppb ^{232}Th in the ZnS.

3 ^{222}Rn emanation measurements

$$\bar{y} = 1 - (1-y)^n \quad 7$$

$n = 3$

In materials, ^{226}Ra can occur in the grains, crystals, etc, making up the materials. When ^{226}Ra decays, some of the ^{222}Rn generated close to the surface of the grains can escape into the space between the grains by virtue of their recoil energy. ^{222}Rn trapped deeper inside grains and crystals can escape by diffusing out (outgassing). Only a fraction of the ^{222}Rn created by the decay of radium is given off to the outside; the remainder of the radon undergoes decay in the material. We describe a system and a procedure used to measure the rate at which ^{222}Rn is emanated by a material into vacuum.

The radon emanation system consists of the ^{222}Rn emanation chamber, ^{222}Rn transfer apparatus (“ radon board ”) and hemispherical scintillation cell as shown in Figure 5. The radon emanation chamber is a cylindrical acrylic chamber 30 cm outer diameter and 65 cm long. Its wall thickness is 12 mm, and the ends are sealed with Viton O-rings. The purpose of the radon board is to extract radon from a mixture of gases (O_2 , N_2) with lower freezing points and then transfer it into a scintillation cell. Its design is based on the one used by Key et al [13] in studies of radium distribution in oceans. All the parts of the radon board are made of stainless steel SwagelokTM fittings. Brass wool was put into the traps to increase the ^{222}Rn trapping efficiency.

The radon collection efficiency of the system was calibrated by putting ^{222}Rn from a calibrated source into the emanation chamber, extracting the ^{222}Rn using the “ radon board ” and then putting it into the scintillation cell. The total efficiency is defined as the ratio of the counting rate of the scintillation cell after the extraction to the amount of ^{222}Rn put into the emanation chamber. A $(33 \pm 5 \%) \times 3$ total efficiency was obtained,

which includes $72 \pm 5 \%$ efficiency for pumping the radon out of the emanation chamber into the large trap, a $75 \pm 5 \%$ efficiency for transferring the radon from the large LN_2 trap to the small trap and then into the scintillation cell and $(62 \pm 3 \%) \times 3$ efficiency for detecting a ^{222}Rn decay in the cell. The factor of 3 in the efficiency arises from the three alpha particles emitted in the decay of ^{222}Rn and its daughters.

The background of the system was measured with no material placed inside the acrylic emanation chamber. Contributions to the background come from the acrylic chamber, the radon board and the scintillation cell. The lowest background achieved for the whole system was measured to be about 20 counts per day (where 12 counts per day were from the chamber, 5 from the radon board and 3 from the scintillation cell). It was found that the background rate in the chamber was higher shortly after large amounts of radon were emanated into the chamber by radioactive samples. The higher rate decreased with time at a rate consistent with the hypothesis that it comes from adsorption of ^{222}Rn on the walls of the chamber. The scintillation cell background increases by 1 count a day for every 10^4 ^{222}Rn decays in the cell because of the 22 year ^{210}Pb .

The measurements of radon emanation from materials were performed in the following way. The material for which the ^{222}Rn emanation was to be measured was put inside the emanation chamber and pumped for more than a day. Typically the chamber reached a vacuum of 200 - 500 microns. Then the chamber was sealed in order for the ^{222}Rn to emanate. After a time t_a , the ^{222}Rn in the chamber was extracted and transferred to a scintillation cell. After a 3 hour wait for ^{222}Rn to come to equilibrium, the number

with what?

*Cell pumped out again?
again?*

of counts N_a was obtained for 20 hours of counting. The chamber was sealed and the procedure was repeated for emanation times t_b , t_c , etc. over about 10 days total. By plotting $N_i/(1-e^{-\lambda t_i})$ as a function of the cumulative time, it is possible to distinguish outgassing of absorbed radon from Ra-supported Rn emanation. For ^{226}Ra -supported ^{222}Rn emanation, the function would be a constant value. Contributions from outgassing of absorbed radon produce excess counts for times less than about 4 days. If radon emanation from ^{226}Ra decay was clearly observed, an average value for emanation times much greater than 4 days was determined, together with an uncertainty. In situations with low statistics or without an observable steady emanation rate, only an upper limit for Ra-supported ^{222}Rn emanation could be determined.

The experimental results are summarized in Table 1. In most circumstances, only an upper limit for the ^{222}Rn emanation rate was obtained. As an example of the results, Figure 6 shows the time evolution of the emanated ^{222}Rn for the high-density polyethylene cable. There is some outgassing of absorbed radon initially and after several days all the ^{222}Rn is supported by ^{226}Ra decay in the cable.

A ^{222}Rn emanation rate can be calculated by assuming that it recoils directly out from a ideal smooth surface because of its kinetic energy. This calculated ^{222}Rn emanation rate for known recoil ranges and bulk radioactivity is about 1000 times lower than the observed Ra-supported ^{222}Rn emanation rates. This suggests that ^{222}Rn is diffusing out from the decay of ^{226}Ra contaminants deeper within the material. X

4 Impact on the SNO detector design

The ^{222}Rn emanation rates of the major components of the SNO detector have been measured. If the ^{222}Rn emanation rate into water is similar to that into vacuum, then the total ^{222}Rn emanated from submersed materials in the water can be calculated.

The H_2O is divided into an "inner" volume (1700 tonnes) between the PMT support structure and the acrylic vessel and an "outer" volume (5500 tonnes) between the PMT support structure and the cavity liner. A 99% water-tight seal on the PMT support structure reduces mixing of H_2O in the outer region with the more critical low-radioactivity H_2O inside. There will not be a significant amount of emanated radon in the D_2O because there is very little material other than clean acrylic in contact with it. The ^{222}Rn emanated from the submersed materials in the two volumes of H_2O are presented in Table 2. The last column (supported radon from material emanation) is given by the product of the area or length, the emanation rate and the mean life of ^{222}Rn (3.8 day/ $\ln 2$).

During the assembly of the detector, some mine dust will be deposited on the surfaces, in spite of extreme care with cleanliness. The final cleanup is expected to reach a level of $0.4 \mu\text{g}$ dust per cm^2 inside the PMT support structure [4] which gives a total of 23 grams of dust over the 5673 m^2 . The dust outside the PMT support structure will be harder to clean up because the surfaces have many hidden crevices. There we are aiming for $4 \mu\text{g}$ of dust per cm^2 which over the 6400 m^2 gives 256 grams of dust.

The total emanated radon in Table 2 can be compared to the design objective for the SNO detector. The 1700 tonnes of H_2O inside the PMT support structure is expected to contain less than $15.0 \times 10^{-14} \text{ gU/g}$ (which supports 1.5×10^6 radon) and the 5300

tonnes of H₂O outside the PMT support structure should contain less than 45.0×10^{-14} gU/g (which supports at least 4.5×10^6 radon). As shown in Table 2, the emanated radon load outside the PMT support structure could be higher than the emanated radon load inside the structure. The H₂O water recirculation system will take water from the outer region, put it through ion exchange resins, high efficiency vacuum degassing and ultraviolet radiation before returning it to the critical inner H₂O volume.

Two other sources of radon are the plastic cavity liner and the cover gas above the H₂O and D₂O surfaces. The design goal for the cavity liner is to have no more than $2 \text{ }^{222}\text{Rn m}^{-2}\text{hr}^{-1}$ penetrating through the liner into the water. Independent measurements indicate that the design goal can be met [16]. If the cover gas is constrained to contain less than 2×10^{-4} pCi/liter of radon, then the exchange of radon into the water will not be a significant problem [14].

5 Further development on scintillation cells

The transfer efficiency can be improved by immersing the scintillation cell into liquid nitrogen while the Rn is being transferred. One effect of doing this is an effective increase of the pressure in the cell by a factor of four due to the lower temperature. The other effect is that the inner surface of the cell becomes a cryogenic pump for radon.

We have developed several cell designs which survive repeated submersion in liquid nitrogen. With this apparatus, nearly all of the Rn collected in the primary ^{222}Rn trap can be transferred into the cell. We are continuing to work on the reliability of the cell design as some have developed cracks in the window to body seal.

6 Conclusion

A low background, high efficiency scintillation cell has been developed for Rn detection for the SNO detector. If the ^{222}Rn emanation rate into water is similar to that into vacuum, then the total Rn emanated into the SNO detector is less than the design objectives.

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Table 1 Experimental Rn emanation rates into vacuum

Materials	^{222}Rn emanation rate hour $^{-1}$	^{238}U content [7] 10 $^{-9}$ g/g (ppb)
Molecular sieve 13X	1200 \pm 120 l $^{-1}$	225 \pm 19
Activated charcoal	250 \pm 50 l $^{-1}$	
Silica Gel	440 \pm 50 l $^{-1}$	197
Coax cable RG-59	60 \pm 30 m $^{-1}$	
Twinaxial PE cable	<2 m $^{-1}$	
Coax cable 8240	6 \pm 2 m $^{-1}$	
Coax cable 9067	<0.6 m $^{-1}$	< 10
Kevlar 3/8" rope	<0.3 m $^{-1}$	0.07
8" PMT	<20 PMT $^{-1}$	
Low-rad. glass	<1.6 m $^{-2}$	50
Aluminum reflector	<1.5 m $^{-2}$	
Black ABS plastic	<1.1 m $^{-2}$	20 \pm 5
White PE	<0.9 m $^{-2}$	
Acrylic	<0.1 m $^{-2}$	
Al plates	<0.5 m $^{-2}$	5
SS 304L [G.Graves]	<15 m $^{-2}$	<1
SS 304L [Sandvik]	<0.3 m $^{-2}$	

Table 2. Rn emanation in the SNO detector

Between the PMT support structure and the acrylic vessel

Material	Quantity	^{222}Rn Emanation rate	Supported ^{222}Rn
Acrylic vessel	452 m ²	<0.1 m ⁻² hr ⁻¹	<6 × 10 ³
Suspension rope	180 m	<0.3 m ⁻¹ hr ⁻¹	<7 × 10 ³
PMT glass	473 m ²	<1.6 m ⁻² hr ⁻¹	<1 × 10 ⁵
Al Reflectors	673 m ²	<1.5 m ⁻² hr ⁻¹	<1 × 10 ⁵
ABS in PMT support	3665 m ²	<1.1 m ⁻² hr ⁻¹	<5 × 10 ⁵
Stainless Steel	410 m ²	<0.3 m ⁻² hr ⁻¹	<2 × 10 ⁴
Mine Dust (0.4μg/cm ²)	23 g	44 g ⁻¹ hr ⁻¹ [15]	1.3 × 10 ⁵
Total			<9 × 10 ⁵

Outside the PMT support structure

Material	Quantity	²²² Rn Emanation rate	Supported ²²² Rn
Stainless Steel	650 m ²	<0.3 m ⁻² hr ⁻¹	<3 × 10 ⁴
Coax cables (a)	190,000 m	<0.6 m ⁻¹ hr ⁻¹	<1 × 10 ⁷
Plastic liner	2000 m ²	2 m ⁻² hr ⁻¹ (b)	5.3 × 10 ⁵
ABS in PMT support	1250 m ²	<1.1 m ⁻² hr ⁻¹	<2 × 10 ⁵
Dust (4μg/cm ²)	256 g	44 g ⁻¹ hr ⁻¹ [15]	1.5 × 10 ⁶
Total			< 1 × 10 ⁷

(a) The coax cables will be bundled and the exposed area is estimated to be 2500 m².

(b) Design goal.

Figure Captures:

Fig. 1 Outline of the proposed SNO detector. The detector would be located at a depth of 6800 feet in INCO's Creighton mine near Sudbury, Ontario, Canada.

Fig. 2. Pulse height spectra for different ZnS thicknesses under (a) " transmission " geometry and (b) " reflection " geometry.

Fig. 3 Diagram of a 2 inch diameter hemispherical scintillation cell.

Fig. 4 Pulse height spectrum measured for our cell. The spectrum measured with a commercial cell is also shown.

Fig. 5 Diagram of radon emanation measurement system. The Rn was first trapped in the liquid nitrogen cooled primary trap. Then the primary trap is warmed and the radon was transferred to the smaller trap immersed in liquid nitrogen. Finally the small trap was warmed and the radon was filled into the scintillation cell by free expansion.

Fig. 6 Rn emanation measurement results from Belden high density polyethylene twincoaxial cable [• — before decay correction, Δ — after decay correction, $N_i/(1-e^{-\lambda t_i})$]. The horizontal axis represents the day which the chamber was opened, radon was extracted from the emanation chamber and resealed. It can be seen that the corrected emanation rates are nearly constant after a couple of days, indicating that the Rn is supported by Ra decay.



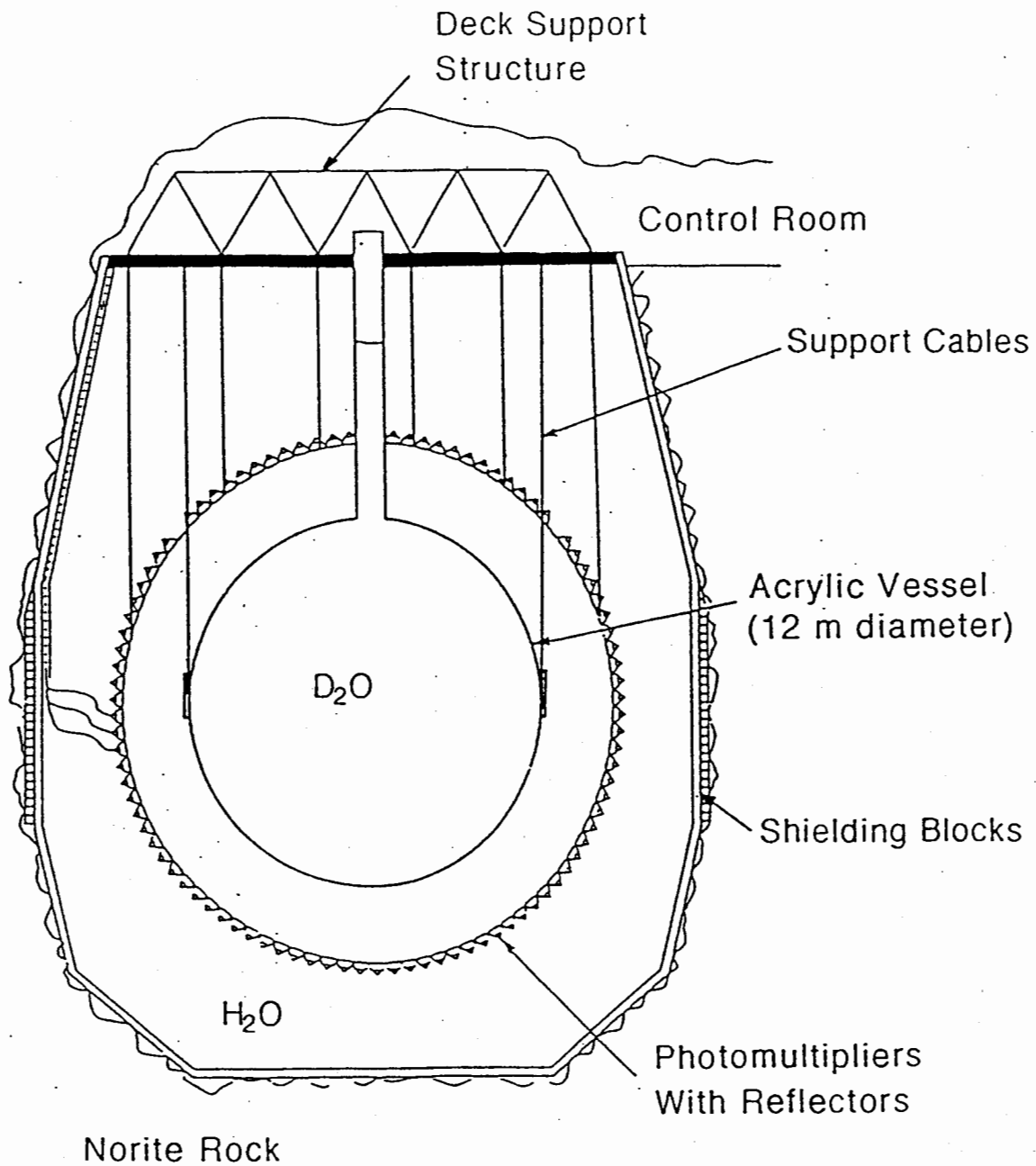


Figure 1 A cross section drawing of the SNO Neutrino Detector

Figure 2 (a)

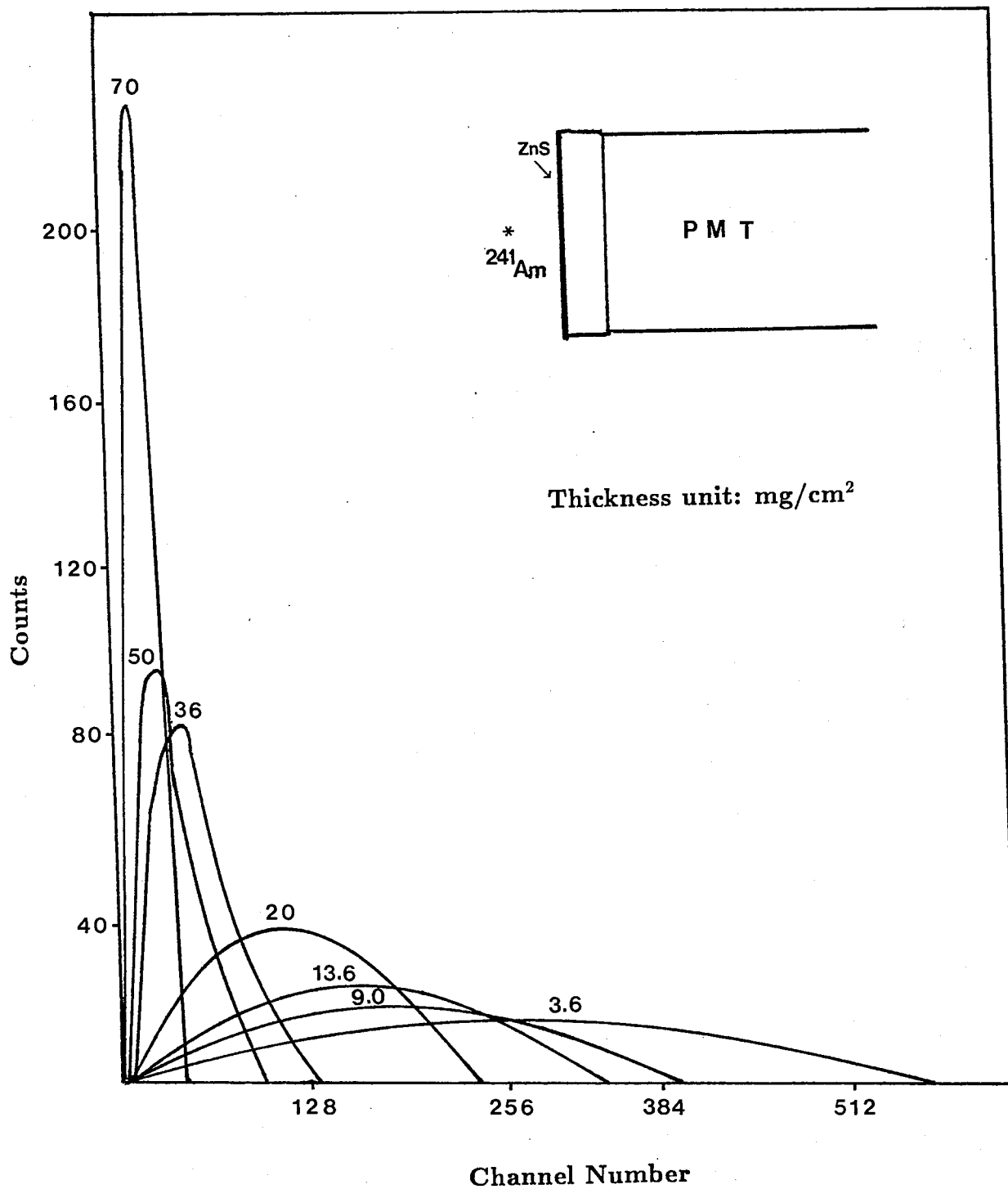


Figure 2 (b)

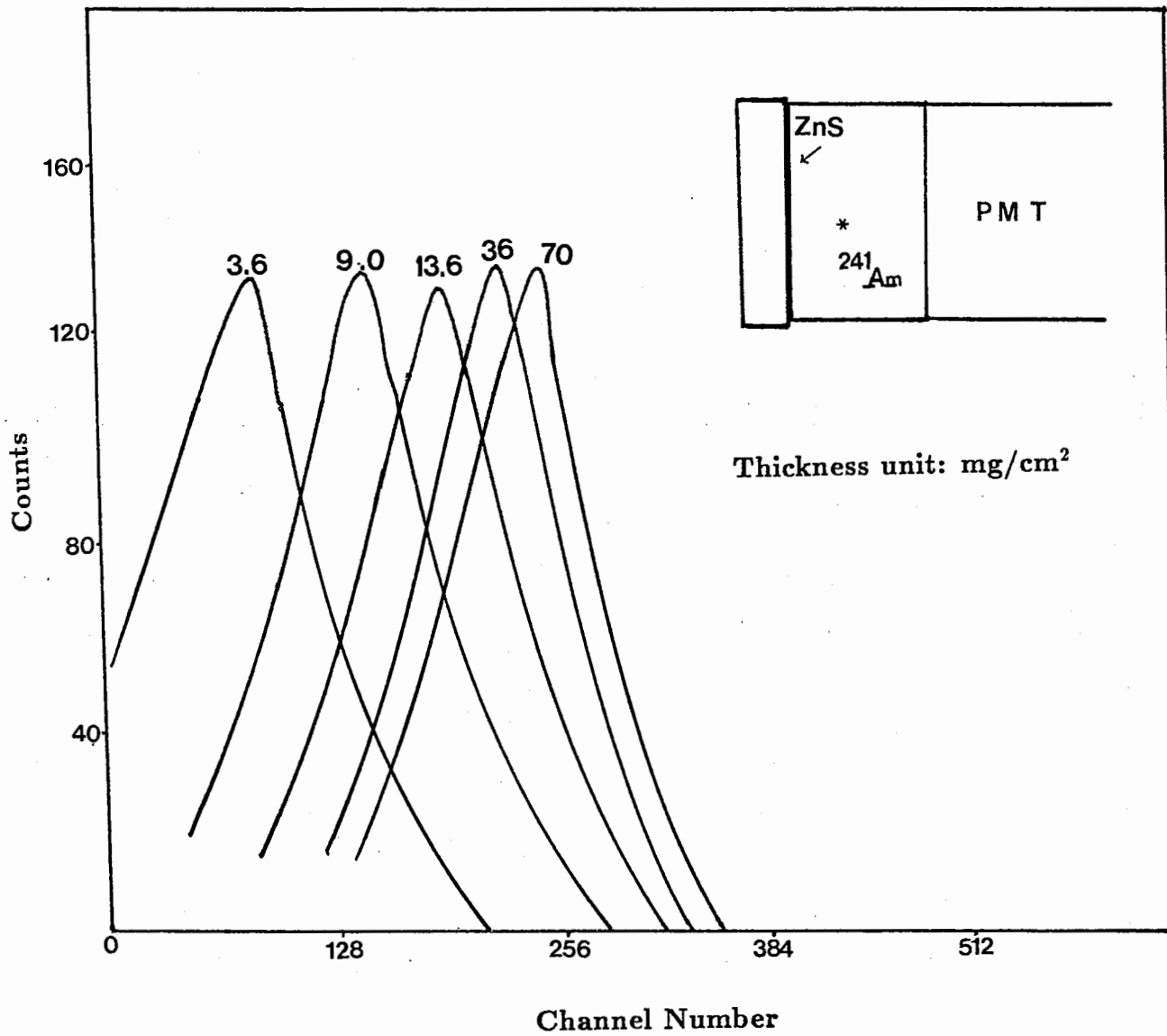


Figure 3.

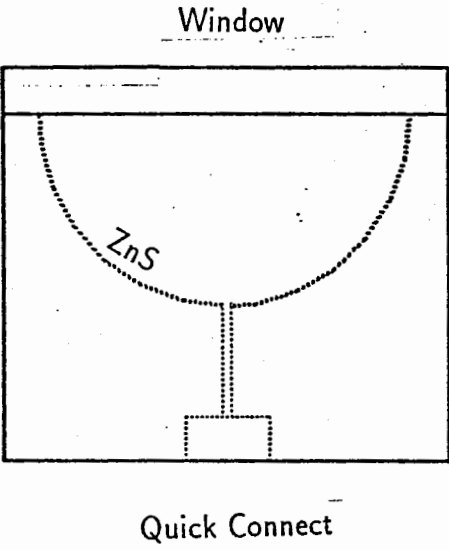
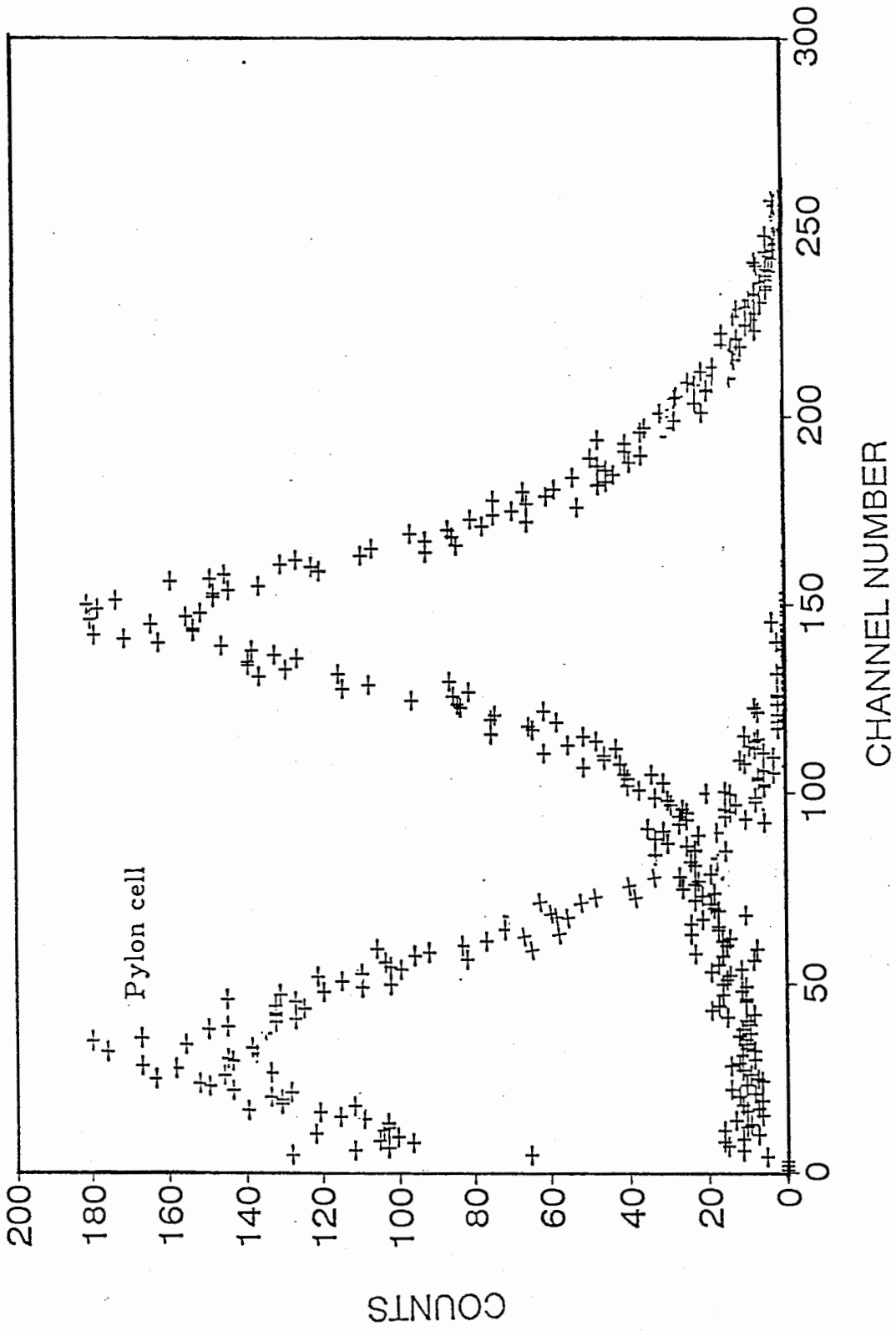


Figure 4



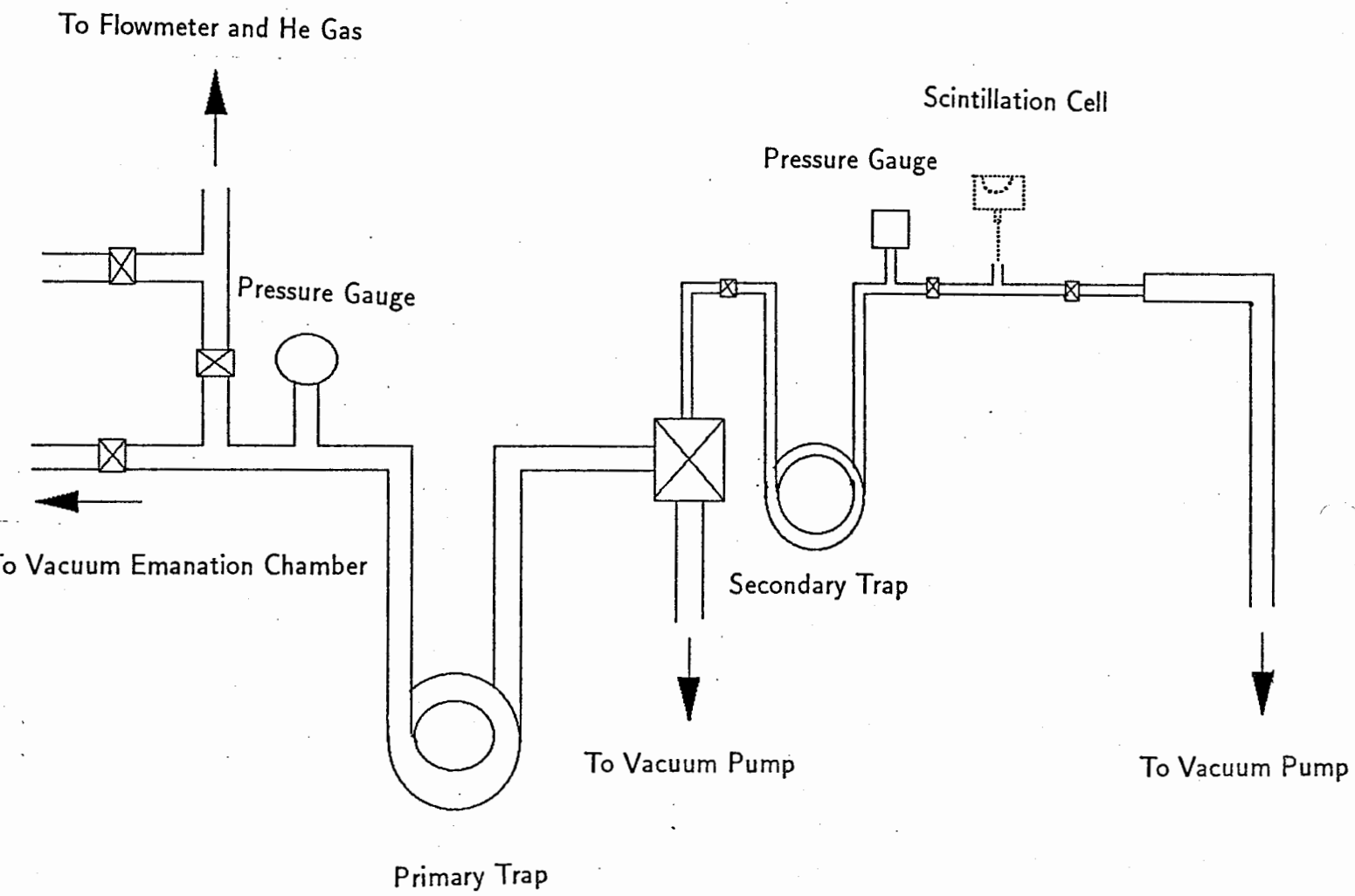


Figure. 5

Figure 6

