Summary Report on Radon Emanation

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1 Radon Emanation

The phenomena of radon emanation from materials was brought to the forefront at the Fall 1990 water meeting where Prof. R. Steinberg (Drexel University) reported relatively high ^{222}Rn emanation rates from a stainless steel drum. The rate he reported implied that the SNO detector stainless steel liner would be significant source of radon into the H_2O and thus would cause significant disequilibrium in the ^{238}U chain. The Queen's W.E.T. lab was in the process of developing miniature low background Lucas cells for radon detection and monitoring of the ^{238}U chain in ultrapure water and decided to follow up on Steinberg's measurement on stainless steel. This eventually expanded to look at ^{222}Rn emanation from other materials to be used in the SNO detector. At the Univ. of Guelph a device was built to measure emanation by collecting the daughters from ^{222}Rn and ^{220}Rn decay. The CANMET lab at Elliot Lake has also constructed and tested apparatus for measuring radon emanation from materials in vacuum and under water. A report is in preparation.

The gamma and beta which are troublesome in the ^{238}U and ^{232}Th chain occur near the bottom of each chain. Hence disequilibrium at the radon daughter (such as an injection of extra radon) effectively pushes up the radioactive U and Th level even though not a single atom of U or Th has been added. ^{222}Rn in the U chain has a 3.8 day half life so it can be transported throughout the H_2O before it decays. Even though ^{220}Rn has a short half-life (55 sec) its 11 hour Pb daughter can also be transported throughout the water.

All materials submersed in the SNO detector emanates radon to some extent and it is necessary to assess the total amount being injected into the water. The most critical part of H_2O shielding is between the acrylic vessel and the PSUP

and therefore close attention has to be paid to the materials in this region.

The most technically straightforward procedure is to look for radon emanated when a material is inside a vacuum. We examine in detail below the work which has been done on ^{222}Rn and ^{220}Rn emanation into vacuum. Work on radon emanation of materials submersed in water is still in a development stage.

2 Techniques for ^{222}Rn into Vacuum

2.1 WET Lab (Queen's)

The apparatus used at the Queen's WET lab to measure radon emanation includes:

1. An emanation chamber (30 cm diameter, 60 cm long acrylic cylinder with polypropylene end caps and Viton O-ring sealed).

2. A radon board (stainless steel Swagelok connectors, a main liquid-nitrogen cooled stainless steel trap, a secondary volume-reducing stainless steel trap and a connection to the Queen's Lucas cells,

3. Low background Queen's Lucas scintillation cell (acrylic body, ZnS coated sides)

The material to be tested is put inside the emanation chamber and the chamber is pumped with an oil roughing pump for 24 to 48 hours. Thé ultimate pressure reached depends on the material under study and is typically around 0.4 mbar (0.3 Torr). The chamber is then sealed for 3 to 6 days in order for the ^{222}Rn to build up. The chamber is connected to the radon board and the radon is subsequently extracted and transferred into a low background Lucas cell. The entire setup and procedures are discussed in detail in M.-Q. Liu's M.Sc. thesis (Queen's University, 1991).

A typical spectrum from the Lucas cell is shown in figure 1. Note that the signal from alphas striking the ZnS is well above the noise from the PMT and electronics.

The various efficiencies for the above procedure are as follows:

Transfer from emanation chamber to radon trap	72±5%
Trapping in small trap, transfer to a 1.5 inch diam. Lucas cell	75±5%
Detection of each alpha decay in the Lucas cell	62±3%

Total efficiency = $72 \times 75 \times 62 = 33.5\%$

There are three alpha associated with each ^{222}Rn decay so the above total efficiency implies one count detected for each radon in the emanation chamber.

The background of the setup is as follows:

Emanation chamber: 25 to 60 counts per day

Radon board: 5 counts per day

Lucas cell: 3 to 10 counts per day

The background in the Lucas cell depends on how much ZnS is used and the cumulative amount of ^{222}Rn that has been put into it (the 22 year ^{210}Pb persists).

2.2 Low-Background Facility (Guelph)

Guelph has constructed a large emanation chamber-Rn daughter detector based on the use of electrostatic collection of the daughters (A. Howard et al, Nucl. Instrum. Meth. A293, 589 (1990) and Amer. J. Phys. 59, no.6, page 544 (1991). The emanation chamber consists of a 25.4 cm diameter by 81.9 cm long aluminum cylinder (total volume 42 liters of which 20 liters can be used for the sample material) which is flanged in the middle and top. On the top end flange is a HV electrical feedthrough which allows a 400 mm^2 silicon surface barrier detector to be run at 90 volts with a battery and a bias of typically -900 volts to be applied to the whole detector. Material to be measured is placed in the bottom half of the chamber, the chamber is sealed and pumped to 50 mbar (37.5 Torr) for one or two days.

Ultrapure water is flowed through MnO_2 coated acrylic beads. The radium in the water is absorbed by the MnO_2. Two small electrostatic chambers have been constructed for measuring radon emanation from the radium decay in the MnO_2 filters.

A typical alpha energy spectrum is shown in figure 2. The energy resolution is about 40 keV at 5500 keV. The count rate of 6003 and 7687 keV alphas from ^{222}Rn daughters are used for the determination of ^{222}Rn. The cumulative counts as a function of time (100 to 150 hours) has been found to be fitted by two radon components. One component is from radon absorbed onto surfaces when the chamber and material were exposed in air and which desorbs under vacuum. The other component is radon from the decay of radium in the material under study.

The efficiency of their device has been studied with a ^{222}Rn source. The efficiency as a function of pressure and bias voltage was measured and the various contributions are approximately as follows:

Fraction of daughters positively charged	65%
Daughters which follow the electrostatic field lines	80%
Daughters that deposit on the front face of the detector	50%
Alphas that stop in the detector	50%

Hence the total efficiency is about $65 \times 80 \times 50 \times 50 = 13\%$ at the operating conditions used.

The fraction of negatively charged daughters was found to be negligible.

The measurement of ^{220}Rn from radium absorbed onto MnO_2-coated acrylic beads involves one additional factor — the fraction of ^{220}Rn which escapes from the MnO_2. This fraction has been determined to be approximately 65%.

The ^{222}Rn background of the empty chamber is about 10 to 15 counts per day. The background of the silicon surface barrier detector alone in the region of interest is less than a count a week.

3 Results and Comparisons

Table A gives the radon rate into vacuum measured on various materials. For the molecular sieve, there are 640 grams per liter and for silica gel 756 grams per liter.

The fourth column gives the inferred U level by looking at the gamma ray from the radium daughter using the low background detector at Guelph.

The major difference between the Guelph and Queen's measurements is on the molecular sieve. For the 9067 coax cable, J. Bigu (CANMET, Elliot Lake) has recently reported an emanation rate of 0.3±0.1 ^{222}Rn m^{-1}h^{-1}.

Even though Guelph has a lower detection efficiency for ^{222}Rn, they have more sensitivity because of the lower background in their chamber.

Table A Summary of ^{222}Rn Emanation Rates

Materials	Queen's ^{222}Rn emanation rate	Guelph ^{222}Rn emanation rate	^{238}U (^{226}Ra) content gU/g
Mole. sieve 13X	1200±120 l^{-1}h^{-1}	5272±100 kg^{-1}h^{-1} 3374±64 l^{-1}h^{-1}	0.59±0.035 × 10^{-6}
Activated charcoal	250±50 l^{-1}h^{-1}		
Silica Gel	440±50 l^{-1}h^{-1}	763±12 kg^{-1}h^{-1} 577±9 l^{-1}h^{-1}	0.197±0.014 × 10^{-6}
Coax cable RG-59	60±30 m^{-1}h^{-1}		
Belden Twinaxial cable	<2 m^{-1}h^{-1}		
Coax cable 8240	6 ± 2 m^{-1}h^{-1}		
Coax cable 9067	<0.6 m^{-1}h^{-1}	60±30 km^{-1}h^{-1}	<10 × 10^{-9}
Kevlar rope (3/8 in. diam.)	<0.3 m^{-1}h^{-1}		0.07 × 10^{-9}
PMT (8 in. diam)	<20 PMT^{-1}h^{-1}		
Low-rad glass 8246	<1.6 m^{-2}h^{-1}		50 × 10^{-9}
Omega reflector	<1.5 m^{-2}h^{-1}	0.65±0.06 m^{-2}h^{-1}	
Black ABS plastic	<1.1 m^{-2}h^{-1}	0.7±0.3 m^{-2}h^{-1}	20±5 × 10^{-9}
White High Den. Polyethy.	<0.9 m^{-2}h^{-1}	0.8±0.1 m^{-2}h^{-1}	
Acrylic	<0.1 m^{-2}h^{-1}		
Aℓ plates	<0.5 m^{-2}h^{-1}		300 × 10^{-9}
MnO_2 coated acrylic beads		1.8±0.5 kg^{-1}h^{-1}	
SS 304L	<15 m^{-2}h^{-1}	<3.2 m^{-2}h^{-1}	<1 × 10^{-9}
SS 304L	<0.3 m^{-2}h^{-1}		
S.S. beam pipe*	370±100 m^{-2}h^{-1}		
Queen's S.S. radon board	30±20 m^{-2}h^{-1}		
Black Viton O-ring	<20 m^{-1}h^{-1}	0.35±.27 m^{-1}h^{-1}	
Gúelph Al electro. chamber		4.9±2.5 m^{-2}h^{-1}	
Foam gasket	<1.2 m^{-2}h^{-1}		

* A second measurement of this beam pipe gave a limit of <15 m^{-2}h^{-1}.

4 ^{220}Rn Emanation

Guelph is the only facility which has measured ^{220}Rn from proposed materials to be used in the SNO detector. The count rate of the 6779 keV alpha from a ^{220}Rn daughter is used. This alpha is well separated from the ^{222}Rn daughter alphas. The background count rate for the empty chamber is 1 to 2 counts per day. They first used their electrostatic device to look at a number of materials with a large range (100 ppb to 62%) ^{232}Th content. Then they looked at materials which have been proposed for use in the SNO detector. Their results are summarized in table B. It was found that the efficiency for detecting ^{220}Rn can be increased if the pressure is lowered and the negative bias on the detector is increased. The efficiency is 4.5% for an operating pressure of 5 mbar (3.8 Torr) and bias of -500 volts.

They did not use a ^{220}Rn source to calibrate and optimize the ^{220}Rn detection efficiency. Rather they used the ^{222}Rn operating conditions for the ^{220}Rn emanation measurements. Subsequently a ^{220}Rn source calibration shows that the ^{220}Rn detection efficiency was 1% compared to 13% for ^{222}Rn at the same operating conditions. Hence the errors in table B are large.

Table B ^{220}Rn Emanation Rates

Material	^{232}Th level gTh/g	^{220}Rn emanation rate
Mole. sieve 13X	$2.1\pm0.11 \times 10^{-6}$	1355 ± 100 h^{-1}kg^{-1}
Belden 9067	$0.077\pm0.025 \times 10^{-6}$	38 ± 24 h^{-1}km^{-1}
Black ABS plastic		<0.36 h^{-1}m^{-2}
MnO_2 acrylic beads		<23 h^{-1}kg^{-1}
White high den. polyeth.		4 ± 2 m^{-2}h^{-1}
Omega reflector		<2.2 m^{-2}h^{-1}
Silica gel	$285\pm21 \times 10^{-9}$	89 ± 20 kg^{-1}h^{-1}
Al electrostatic chamber		8.5 ± 3.0 h^{-1}m^{-2}

5 Radon emanation into water

At the WET lab we have pumped out a beam pipe and measured the radon emanation rate. Then we filled the beam pipe with ultrapure water (the radium level in the supply water has to be less than 10^{-13} gU/g) and let the radon from the beampipe emanate into the water.

Helium gas was then bubbled through the beam pipe and the radon trapped in the radon board. An system efficiency calibration was not done. Assuming a 100% collection efficiency, the preliminary results show that the radon emanation rate into water is probably comparable to that into vacuum.

6 Impact on the SNO Detector

The "white" SNO-87-12 proposal uses 1.5×10^{-14} gU/gH_2O in the Monte Carlo calculations. Recent Monte Carlos have been carried out, incorporating the latest design changes to the SNO detector and including code improvements to more accurately simulate events. Discussions with P. Skensved and B. Robertson indicate that the detector sensitivity to the uranium chain is such that a level of 15×10^{-14} gU/gH_2O probably does not seriously compromise the experiment.

The total amount of radon supported by the 1670 tonnes of water (at 15×10^{-14} gU/gH_2O) between the acrylic vessel and the PMT support structure is 1.5×10^6. This can be compared to the total Rn supported by emanation, which is shown in table C. A foam gasket material will be used to make the support structure water-tight.

It must be remembered that the radon emanation rates used are emanation rates into a vacuum. We have not yet established the corresponding emanation rates into water.

No.	Material	Rn emanation rate into vacuum	Quantity needed	Total ^{222}Rn
1	Schott 8246 PMT glass	<1.6 m^{-2}h^{-1}	9500 each with exposed area 0.0628 m^2	$<1.3 \times 10^5$
2	ABS black plastic in PMT support structure	<1.1 m^{-2}h^{-1}	exposed area 455 m^2	$<6.4 \times 10^4$
3a	ABS black plastic for reflectors	<1.1 m^{-2}h^{-1}	9500 each with exposed area 0.0942 m^2	$<1.3 \times 10^5$
3b	Schott 8246 glass reflectors	<1.6 m^{-2}h^{-1}	same as 3a	$<1.9 \times 10^5$
3c	Omega reflectors	<1.5 m^{-2}h^{-1}	total area 500 m^2	$<1.0 \times 10^5$
4	Acrylic	<1 m^{-2}h^{-1}	400 m^2	$<6 \times 10^4$
5	Kevlar rope	<0.3 m^{-1}h^{-1}	180 m	$<7 \times 10^3$
6	Foam gasket	<1.2 m^{-2}h^{-1}	152 m^2	$<2 \times 10^4$

Total Rn Load

Quantities added	Total Rn supported
1 + 2 + 3a + 3b + 4 + 5 + 6 (glass reflectors)	$<5.8 \times 10^5$
1 + 2 + 3a + 3c +4 + 5 + 6 (Omega reflectors)	$<5.0 \times 10^5$

We require a water-tight PSUP because of the possibility that some radon may get through the plastic liner, that the cable jacket may give significant amounts of radon and that radium leaching may be a problem.

The fraction of ^{220}Rn that comes out of a sample is very low except for fine powder samples (which have a large surface area). The observed rate is consistent with ^{220}Rn recoiling out of the top 300 Angstroms of the material. This is in contrast to ^{222}Rn where it appears to come from deep within the material as its 3.8 day half-life allows it to diffuse out from deeper down. The conclusion is that materials of less than 100 ppb Th do not significantly contribute to the ^{220}Rn in the water.

Radium leaching also contributes to higher radioactivity in the water. At present there are studies underway especially looking at the Omega reflector material (X. Zhu et al, SNO-STR-91-065) and Schott low-radioactivity 8246 PMT glass. At Guelph they will use small electrostatic chambers to look at the radon coming from MnO_2 coated acrylic beads to obtain precise number on the rádium leaching rate.

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3. Low background Queen's Lucas scintillation cell (acrylic body, ZnS coated sides)

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Ultrapure water is flowed through MnO_2 coated acrylic beads. The radium in the water is absorbed by the MnO_2. Two small electrostatic chambers have been constructed for measuring radon emanation from the radium decay in the MnO_2 filters.

A typical alpha energy spectrum is shown in figure 2. The energy resolution is about 40 keV at 5500 keV. The count rate of 6003 and 7687 keV alphas from ^{222}Rn daughters are used for the determination of ^{222}Rn. The cumulative counts as a function of time (100 to 150 hours) has been found to be fitted by two radon components. One component is from radon absorbed onto surfaces when the chamber and material were exposed in air and which desorbs under vacuum. The other component is radon from the decay of radium in the material under study.

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3 Results and Comparisons

Table A gives the radon rate into vacuum measured on various materials. For the molecular sieve, there are 640 grams per liter and for silica gel 756 grams per liter.

The fourth column gives the inferred U level by looking at the gamma ray from the radium daughter using the low background detector at Guelph.

The major difference between the Guelph and Queen's measurements is on the molecular sieve. For the 9067 coax cable, J. Bigu (CANMET, Elliot Lake) has recently reported an emanation rate of 0.3 ± 0.1 ^{222}Rn $m^{-1}h^{-1}$.

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Table B ^{220}Rn Emanation Rates

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No.	Material	Rn emanation rate into vacuum	Quantity needed	Total ^{222}Rn
1	Schott 8246 PMT glass	<1.6 m^{-2}h^{-1}	9500 each with exposed area 0.0628 m^2	$<1.3 \times 10^5$
2	ABS black plastic in PMT support structure	<1.1 m^{-2}h^{-1}	exposed area 455 m^2	$<6.4 \times 10^4$
3a	ABS black plastic for reflectors	<1.1 m^{-2}h^{-1}	9500 each with exposed area 0.0942 m^2	$<1.3 \times 10^5$
3b	Schott 8246 glass reflectors	<1.6 m^{-2}h^{-1}	same as 3a	$<1.9 \times 10^5$
3c	Omega reflectors	<1.5 m^{-2}h^{-1}	total area 500 m^2	$<1.0 \times 10^5$
4	Acrylic	<1 m^{-2}h^{-1}	400 m^2	$<6 \times 10^4$
5	Kevlar rope	<0.3 m^{-1}h^{-1}	180 m	$<7 \times 10^3$
6	Foam gasket	<1.2 m^{-2}h^{-1}	152 m^2	$<2 \times 10^4$

Total Rn Load

Quantities added	Total Rn supported
1 + 2 + 3a + 3b + 4 + 5 + 6 (glass reflectors)	$<5.8 \times 10^5$
1 + 2 + 3a + 3c +4 + 5 + 6 (Omega reflectors)	$<5.0 \times 10^5$

We require a water-tight PSUP because of the possibility that some radon may get through the plastic liner, that the cable jacket may give significant amounts of radon and that radium leaching may be a problem.

The fraction of ^{220}Rn that comes out of a sample is very low except for fine powder samples (which have a large surface area). The observed rate is consistent with ^{220}Rn recoiling out of the top 300 Angstroms of the material. This is in contrast to ^{222}Rn where it appears to come from deep within the material as its 3.8 day half-life allows it to diffuse out from deeper down. The conclusion is that materials of less than 100 ppb Th do not significantly contribute to the ^{220}Rn in the water.

Radium leaching also contributes to higher radioactivity in the water. At present there are studies underway especially looking at the Omega reflector material (X. Zhu et al, SNO-STR-91-065) and Schott low-radioactivity 8246 PMT glass. At Guelph they will use small electrostatic chambers to look at the radon coming from MnO_2 coated acrylic beads to obtain precise number on the radium leaching rate.