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Leaching Measurement on Omega Reflector (WET Lab)

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1 Experimental Procedure

The leaching rate of Omega reflector, PMT glass etc. have been studied by mass leaching and conductivity measurements at Oxford and U.B.C.. To infer the radium leach rate from the above data requires several assumptions: the Ra leaching rate is the same as the mass leaching rate and Ra is uniformly distributed in the material. In reality Ra may be more concentrated on the surface or more easily leached out, so a direct Ra leaching measurement is still necessary and would be complementary to the Oxford and U.B.C. data.

A new 90 liter leaching board has been operated since July combined with the Rn board . The leaching board consists of a PVDF tank, a heater, a water pump and a MnO_2 coated acrylic bead column. All the surfaces of the heater, the pump and the tubing are teflon, so we can get a very low background for the leaching study. The cleaning procedure for the leaching board as recommended by the Carleton group is as follows:

1. Wash with TSP detergent, then rinse with water
2. Wash with 1% HCl, then rinse with water
3. Wash with 1% NaOH, then rinse with water
4. Rinse with distilled water and d.d. water

The procedure to run the leaching board and the Rn board is as follows:

a: After meticulous cleaning of the material (similar to that outlined above for the leach rig), submerse it in the ultrapure water (from Carleton) in the leaching rig.

b: Pump water through the MnO_2 coated acrylic beads for two days, then change the beads and pump water through them again for two days.

c: Dry the second batch of beads by using vacuum pumping with a water vapor trap. Connect the dried filter to the Rn board and pump for about half a day. Seal it for two emanation days. After the two days of emanation, transfer the Rn in the filter into a Lucus cell according to the procedures detailed in M.Q. Liu's thesis. Then count the cell for 1 to 2 days.

d: After step c, turn on the heater, keep the temperature at 95°C for three leaching weeks.

f: Turn off the heater, let the temperature go down to 35°C (about three days). Pump water through the MnO_2 filter (new MnO_2), and repeat step c. By comparing the counts and background counts, we can get the Rn counts off the MnO_2 beads which are supported by direct Ra decay and so we can calculate how much Ra has leached out from the material.

2 Efficiency of Measurement

The total efficiency can be divided into the leaching board efficiency and the Rn board efficiency. The efficiency of the Rn board is already well determined in M.Q. Liu's thesis. It consists of Eff(pumping), Eff(detecting)=63% and Eff(transfer)=75%. The Eff(pumping) for the large emanation chamber (50 liters) is 70%. In our case, the column volume is <15 cm^3, so we take Eff(pumping) as 100%. This can be checked subsequently using a radon source.

The efficiency of the leaching board consists of the efficiency which leached radium is trapped on the MnO_2 coated acrylic beads and the efficiency of emanation (how much Rn supported by Ra in the beads can emanate out in the vacuum). The beads have been proven to have a very high efficiency for trapping Ra (greater than 95% according to tests done at Carleton). In our case, the water is circulated over the beads at a rate is about 1 liter/min so in two days pumping the total volume of water is cycled more than 40 times and so we assume Eff(trapping) is 100%.

We measured a MnO_2 column spiked with radium from the Carleton group. This column is exactly the same as the column used in the leaching board so the efficiency of Rn emanating from beads into vacuum could be determined. The spiked column was made by passing a known volume of standard ^{226}Ra solution through it. The total activity was 10 Bq. The way we measured radon from the spiked column is as follows:

a: pump the spiked column for several hours, then seal it at time T_0.

b: at time T_1 open the column and transfer Rn into a Lucas cell. The emanation time is T_1-T_0.

c: at time T_2, we start to count the cell. T_2 is about 4 hours after T_1 so Rn and its daughter will reach secular eqilibrium in the period. Stop counting at T_3. The counting time is T_3-T_2 and the number of counts is ΔN.

d: in secular equilibrium, three counts corresponds to a Rn decay. We calculate from the formulae below to determine how many Rn atoms were inside the cell at time T_2 and the activity of the source:

$$\Delta N/3 = N \times [(1-\exp(-\lambda \times (T_3-T_2))]$$

where N is the number of Rn at the time T_2 and λ is the Rn decay constant.

The number of Rn inside the cell — N_0 at the time T_1 is given by

$$N_0 = N/\exp(-\lambda \times (T_2-T_1))$$

The number of Rn inside the column — N_c at time T_2 is

$$N_c = N_0/[\text{Eff(transfer)}*\text{Eff(detecting)}*\text{Eff(pumping)}]$$

If P is the disintegration rate of Ra in the MnO_2 then

$$N_c = P/(\lambda \times [1-\exp(-\lambda \times (T_1-T_0))])$$

	t1-t0 sec	t2-t1 sec	t3-t2 sec	ΔN	N_c	P sec^{-1}
1	10800	14400	19757	8089	1.44×10^5	13.5
2	9900	12600	11661	3887	1.16×10^5	11.9
3	6600	33000	5332	1074	7.28×10^4	11.1

The measured activity of the spiked source is 12±1.5 Bq and Eff(emanation) and Eff(pumping) are 100%.

3 Background

The total background comes from the Lucas cell background, the Rn board background, Ra background from the MnO_2 coated acrylic beads, Ra background in the water and Ra leaching background from the leaching board. The measurement of these backgrounds are listed below:

a: background of Lucas cell (QUAA1) 12±3/day

b: cell+ Rn board 14±4/day

c: filter+MnO_2 +cell+Rn board 12±4/day

d: first time Ra extraction from 65 liters ultrapure water 17±4/day

f: second time Ra extraction from 65 liters of water 11±3/day

g: after heating water to 65°C for 13 days, the overall background 15±4/day

We can see from above that the background basically comes from the Lucas cell itself. If we take the background as 10 counts/day, it means after two days emanation,

$\Delta N/3=N_0*(1-\exp(-\lambda*t))*0.75*0.63$ where the Rn in the column is N_0

$N_0=43$ (counts)

$N_0=0.84*N_{Ra}*\lambda*\Delta t$ (where 0.84 is the Rn decay time correction)

$N_{Ra}=2.1*10^7$

N_{Ra} is the Ra number in the column. This number can give a reasonable limit on the leaching rate from materials.

4 Leaching Measurement of Omega Reflector

We put 6.12 m^2 of single-sided shiny Omega reflector in 80 liters of ultrapure water after it was cleaned according to the following procedure:

a: wash with Decon75 (de-radioactive contamination soap) and rinse with water

b: clean with clean ethyl alcohol

c: rinse with d.d. water

We measured the leaching twice at high temperature (95°) and once at room temperature (20°). The chronological order is as follows:

a: On Aug. 19 after pumping the water through the MnO_2 filter for two days, dry and measure the filter, the background counts was 11/day. Turn on the heater and keep water at 95°. On Aug. 30 turn off the heater. On Sept. 5 the temperature of water was at room temperature, pump water through the filter then count the filter. Three counting results are 40/day, 20/day and 22/day for Omega in water for 18 days. The first number is abnormally higher than the others. We believe the filter may not have been pumped totally dry so there was still some Rn in the column before it is sealed.

b: From Sept. 7 to Sept. 23, Omega sheets was immersed in the water for two weeks at room temperature. After extraction the results are 16/day and 15/day.

c: From Sept. 23 to Oct. 3, the water was kept at 95°C, then the heater was turned off. When the temperature went down to 30°C, pump water through the filter from Oct. 7 to Oct. 9. The results of counting the Rn corresponding to the 20 days of high temperature leaching are 17/day and 19/day.

From above we see the count rate for Omega reflector is just above the background. It is hard to get a number on the leaching rate, but we can obtain an upper limit. Take a maximum number there is 10±3 per day increase above the background. The number of Rn inside the filter after emanation N_0 is calculated by

$$10/3=N_0*(1-\exp(-2.1*10^6*86400))*0.75*0.63$$

$$N_0=0.84*N_{Ra}*\lambda_{Ra}*2\text{days} \text{ (where 0.84 is a decay time correction)}$$

$$A(\text{activity of U chain})=N_{Ra}*\lambda_{Ra}=50.7/\text{day}$$

We assume the U content in Omega is 10 ppb, the leaching rate LR (in $g/m^2/day$) increases a factor of 2 per 10 degree ($2^{7.5}=181$) and that there is secular equilibrium in the U decay chain. After 20 days at 95° C,

$LR*181*20*6.15*10*10^{-9}*6.02*10^{23}*\ln 2/(238*4.5*10^9*365) = 50.7$

Hence the bulk Omega leach rate LR is $<2.13 \times 10^{-4}$ g $A\ell$ m^{-2} day^{-1}.

This upper limit is much larger than the limit of $<5.3 * 10^{-6}$ g/m^2/day as determined by measuring the conductivity of water (UBC). If the Omega material contains 10 ppb U and there is approximately 0.2 m^2 of reflector per PMT, then the above limit we measured implies there is $<4 \times 10^{-9}$ gU per day leached into the water. This can be compared to the 1.7×10^{-5} gU which is in the 1700 tonnes of 10^{-14} gU/g water between the acrylic vessel and the PMT support structure.

5 Work after Omega Reflector

We now have put 5 PMT glass bulbs in the water for studing the leaching from PMT glass. The PMT glass mass leaching has been studied in Oxford. It has a much higher leaching rate (50ng/m^2/day) and a much higher U content (100 ppb) than Omega. If the Ra leaching rate is the same as the mass leaching rate, our Rn count rate will be a few times higher than background. In addition, we have obtained some heavily spiked glass from Oxford. This glass can give a very high Rn count rate, but there may be the possibility of contaminating the leaching board, so we decided to use the spiked glass at the very end as a final test for the efficiency of MnO_2 coated acrylic beads.