Radium Assay of the 60 Ton Tanks

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1. Introduction

Two 60 ton tanks (TK02 and TK03) have been built as SNO heavy water reservoirs. Each tank is 12 feet high and 15 feet in diameter. The tank exterior consists of welded steel plates while the interior is lined (bottom, sides and top) with polypropylene-welded polypropylene-sheets to form a leak-tight liner. The polypropylene (PP) liner minimizes the possibility of radium leaching and radon diffusion from the steel. Each liner was meticulously scrubbed and cleaned with soap and thoroughly rinsed with ultra-pure water. At present there is no nitrogen cover gas inside the tanks – clean lab air goes in or out of the tanks as the water level drops or rises.

We have conducted a series of radium runs on these two 60 ton tanks to determine how they affect the radioactivity level of ultra-pure water stored in them.

2. Experiment

This experiment consists of a series of four runs. IX quality light water was filled into both 60 ton tanks and held there for a time. Then the water in one tank was pumped out and passed through a MnO2 column to extract Radium and the water dumped to drain. For the other tank the water was passed through a MnO2 column and into the second 60 tonne tank. This was then repeated two more times.

The Radium in the column was measured via its decay products by a electro-static radon detector (ESC) on the surface.

The following table shows the history of the four runs:

Run #	Water filling date	Water path
1	May 24-27, 1996	TK02 (10 days) -> Two Columns -> drain
2	May 24-27, 1996	TK03 (17 days) -> Column -> TK02 (water for Run#3)
3	June10-12, 1996	TK02 (20 days)-> Column -> TK03 (water for Run#4)
4	July 2 - 3, 1996	TK03 (12 days)-> Column -> drain

For Run #1, the water passed through two columns in series and then to drain. The idea here was to consider this water as 'fresh' without Radium leached from the tank. The water for Run #2 was put into TK03 at nearly the same time as Run #1 and was held in the tank for 17 days in order to measure any Radium leaching from that tank. This same water was then reused for the next two runs.

With the two columns in series in Run #1 we re-measured the Radium extraction efficiency. For the other three runs, about 50 to 52 tons of water passed through only one (different) column each time. The extraction water flow rate was controlled at around 20 liters per minute and more than 40 hours (6 underground shifts) were needed for each run. Due to the shortage of a supervisor for the graveyard shifts and for other safety reasons, the extraction had to be shut down in the evening for most of runs.

The following table shows the time sequences of the runs:

Run #	Date	Extraction (hr.)	Shutdown (hr.)	Water Passed (T)
1	June 3	7.5	12.0	9.20
	4	12.0	13.25	15.00
	5	10.75	12.0	13.65
	6	11.5	16.5	14.25
	7	Counted		(Total) 52.10
2	June 10	17.5	5.5	21.75
	11	18.5	4.0	22
	12	6.0	18.5	6.4
	13	Counted		(Total) 50.15
3	July 2	5.25	2.0	6.05
	2	6.0	1.75	6.90
	2	6.0	2.0	7.25
	3	27.15	7.75	29.85
	4	Counted		(Total) 50.10
4	July 15	40.85	8.0	50.2
	17	Counted		(Total) 50.2

3. Analysis and Results

3.1 Data Analysis

If the light water contains 1×10^{-14} g/g of ^{238}U and ^{232}Th, the activity would be 11.14 and 3.52 decays per day per ton respectively. As there is a huge difference in the half life of ^{226}Ra and ^{224}Ra it is more useful to note that <u>one ton of water</u> would have <u>11.14 ^{226}Ra decays per day</u> and $3.52 \times 3.6/\ln 2 =$ <u>18.28 ^{224}Ra atoms supported</u> when we analyze the data.

Both ^{226}Ra and ^{224}Ra are absorbed by the MnO_2 beads and thorium to a much lesser extent when water is passed through the column. As the half life of ^{226}Ra is 1600 years, its activity will not change during our run period which is typically 2 days of extraction and 20 days of counting. But ^{224}Ra has an half life of 3.6 days and its activity will decrease rapidly during that period. All time delays for the ^{224}Ra measurement have been corrected for in the analysis.

The extraction efficiency is 90% for Radium; the emanation efficiency is 75% for ^{222}Rn and 30% for ^{220}Rn; the detection efficiency is 30% for ^{222}Rn and 20% for ^{220}Rn. So, the overall Radium measurement efficiency is 20% for ^{226}Ra and 5.4% for ^{224}Ra. In this series of runs, the newly designed PP column holders were used and the emanation efficiency for both ^{222}Rn and ^{220}Rn has yet to be re-measured (but we are not expecting a large difference).

3.2 Results

For Run #1 (two columns were used in order to measure the extraction efficiency) no ^{224}Ra was observed in the second column so we deduce a lower limit for the extraction efficiency. From the ^{224}Ra data the extraction efficiency with a MnO_2 column is better than 97%.

The following table shows the results for the four runs from the data analysis.

Run #	226Ra(dpd)	(10^{-14} g/g ^{238}U)	^{224}Ra(atoms*)	(10^{-14} g/g ^{232}Th)	^{228}Th(dpd**)	(10^{-14} g/g ^{232}Th)
1	<33.6	<0.06	1183±162	1.9±0.3	494±149	2.7±0.8
2	115±54	0.21±0.10	969±273	1.5±0.4	<48	<0.3
3	127±49	0.22±0.08	100±268	0.2±0.5	893±341	5.0±1.9
4	28.8±18.3	0.05±0.03	891±155	1.3±0.2	<128	<0.7

* Atoms at the start of counting. The extraction/shutdown sequences and counting delays for each run are different, hence these numbers only give an indication of the absolute number of 224Ra in the column. **See below.

The above table shows that the results are consistent for all four runs. In all cases, the radioactivity of light water from the 60 tonne tanks is at or below the H2O design criteria of 10^{-14} g/g of ^{232}Th and 10^{-13} g/g of ^{238}U.

The D2O design criteria is 3×10^{-15} gTh/g and 4.5×10^{-14} gU/gD2O. The ^{232}Th level based on the measured ^{224}Ra in the 60 tonne tanks are about a factor of five above this criteria.

The results from the ^{228}Th vary largely and its uncertainty is large as well. This is because the existing ^{228}Th background. Also, the extraction efficiency for Thorium is very

low and not accurately known. It is estimated to be about 10% that of Radium and that efficiency is used here.

4. Discussion

4.1 ^{232}Th and ^{238}U Levels

In all four assays, the equivalent ^{232}Th concentrations (inferred from ^{224}Ra) are between 1 to $2*10^{-14}$ g/g. However we observed a much lower level of ^{238}U level (from ^{226}Ra) except in Run #2 which has the first reused water from the previous run. We have already assayed the R/O and IX components of the light water system and have never seen such a high level of U [Report to the Water R and D Review (Lilac Book)].

We can assume that the light water system, especially the R/O system removes Radium very effectively but not for Thorium. Hence, after the R/O system, Radium can only in-grow from Thorium or come from leaching off the walls. As ^{226}Ra has a long half life ($t_{1/2}$=1602 years), we can not detect its activity. On the other hand, ^{224}Ra in-grows very quickly.

The other explanation for the low U level is that the ^{226}Ra background of the detection system (ESC chamber plus circulation pump and loop) is much higher than that of ^{224}Ra (the daughter of 228Th) . This high background increases both statistical and systematic error for the measurement and hence decreases the detection sensitivity.

4.2 Leaching from 60 ton tanks

From the experimental results, it seems that the leaching in the two 60 ton tanks is low from the thick polypropylene liner. The polypropylene sheets were made from the same low radioactivity beads (SV258) as the PP piping used elsewhere in the SNO water system although the sheets were manufactured at a different company.

In contrast we are seeing much higher levels in ultra-pure water that had been in contact with the D2O RO Hastelloy housing (SNO-STR-96-045) and the H2O Process Degasser PP packing/stainless steel chamber (SNO-STR-96-044). A measurement on ultra-pure water shipped in stainless steel drums to the CRL test water plant and run through that setup is also showing high levels of ^{228}Th and 224Ra (work in progress).

4.3 Ramifications for the D2O System

The plan is to fill the first 60 ton tank with purified salted heavy water. Then when it is time to use this brine, the second 60 ton tank will start filling with pure heavy water from the acrylic vessel while the brine from the first tank is emptied into the acrylic vessel. If the above measured contributions of radioactivity come from the 60 ton tank then there would be a serious increase in the 224Ra level in the acrylic vessel.

Differential Spectrum for Run #4