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DEMONSTRATION OF AN INSTRUMENT FOR ON-LINE MEASURING OF THE ALPHA RADIATION CONCENTRATION IN HIGH-LEVEL LIQUID WASTE

R. L. Brodzinski, M. J. Barnes, D. T. Hobbs, and S. D. Fink

JULY 12, 2004

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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF ACRONYMS

ADS	Analytical Development Section
ANM	Alpha Neutron Monitor
c/s	counts per second
Cs	cesium
DOE	Department of Energy
DWPF	Defense Waste Processing Facility
HLW	High Level Waste
ICP-ES	Inductively Coupled Plasma – Emissions Spectroscopy
PNNL	Pacific Northwest National Laboratory
s	second
S/N	signal to noise
SPF	Saltstone Production Facility
Sr	strontium
SRS	Savannah River Site
SRNL	Savannah River National Laboratory
SWPF	Salt Waste Processing Facility
TRU	transuranic
WAC	Waste Acceptance Criteria
WPTS	Waste Processing Technology Section

1.0 EXECUTIVE SUMMARY

The Salt Waste Processing Facility and the Actinide Removal Process at the Savannah River Site will remove alpha emitting radioisotopes from High Level Waste to meet regulatory disposal requirements. The removal occurs prior to removal of the radioactive cesium. Both facility designs currently include verification measurement using conventional wet chemistry approaches remote from the operation. Use of an on-line monitor for this purpose offers the opportunity to shorten process cycle time and thereby increase facility throughput.

Personnel from Pacific Northwest National Laboratory (PNNL) designed, built, and functionally tested an on-line monitor, which is capable of measuring the residual transuranic concentrations in processed high-level wastes with a detection limit of 370 Bq/mL (10 nCi/mL) in less than six hours. Personnel from Savannah River National Laboratory (SRNL) assisted in the design and tested the equipment using actual waste.

The monitor measures the neutrons produced by the transuranics, primarily via (α,n) reactions, in the presence of gamma-ray fields up to 1 Sv/h (100 R/h). The optimum design derives in part from Monte Carlo modeling tempered with practical engineering and cost considerations. We demonstrated operation of the monitor in a cell utilizing an actual sample of high-level waste. Results of that demonstration are given, and suggestions for improvements in the next generation system are discussed. Primary findings include the following.

- The waste used for the demonstration contained entrained organic from a prior solvent extraction demonstration as well as precipitated fine particles. Settling of the solids during the experiments complicates interpretation of the results.
- The equipment operated without incident.
- The dynamic response of the equipment exceeded the design requirement suggesting on-line measurements in much less than the requested 6 hours are possible. Until the equipment is tested in a flow through configuration, we can not quantify the response time.
- In the demonstration that most closely approximated proposed operational conditions, the monitor gave slightly positive bias (~25%) in results when compared to conventional wet chemistry results [i.e., 76.7 nCi/g versus 61.1 nCi/g].
- Flushing of the equipment between demonstrations indicated incomplete removal of the residual sample. As much as 5% residual contamination occurred in the most representative demonstration. This value does not represent the behavior in actual deployment within a flow-through system that has a high-velocity flush system.

2.0 INTRODUCTION

Millions of gallons of high-level radioactive liquid waste (HLW) will be processed at the Department of Energy's Savannah River Site (SRS) to remove ^{90}Sr, ^{137}Cs, and selected actinides before sending the resultant salt solution to the relatively low-cost Saltstone disposal facility. The separation process involves batch sorption of strontium and actinides on monosodium titanate, cross-flow filtration to remove the solids, and solvent extraction of the filtrate to remove cesium. To confirm that the process output meets the Waste Acceptance Criteria (WAC) for the Saltstone facility, the program developed a technology to nondestructively monitor the concentrations of the alpha emitting radioisotopes in less than six hours for either a flowing side stream or for a transfer line between process steps [1]. If these concentrations exceed the WAC limits, the batch will need recycled through the process. The WAC call for a maximum alpha concentration of 670 Bq/g (18 nCi/g), and the process requirements specify an instrumental detection level of 370 Bq/mL (10 nCi/mL) within the six-hour measurement time constraint. Because the design specifies measuring the alpha content before removal of the ^{137}Cs from the waste stream, the gamma-radiation dose rate emanating from a process pipe at the measurement point is expected to range between 0.1 and 1 Sv/h (10-100 R/h). This precludes any consideration of measuring the sparse, low-energy characteristic gamma rays from the transuranic (TRU) isotopes.

The transuranic isotopes in SRS tanks produce neutrons either by spontaneous fission or by alpha-neutron (α,n) reactions on light isotopes, principally oxygen and sodium in high-level wastes. Typical tank wastes consist of an alkaline aqueous solution having the chemical composition shown in Table 1. Except for two tanks that have relatively large concentrations of ^{244}Cm, (α,n) reactions are responsible for 97.5-97.9% of the total neutron generation rate. The neutron yield per unit of alpha activity (i.e., the "normalized neutron yield" or neutrons per second per nCi of alpha activity) for typical transuranic isotope ratios in high-level waste is surprisingly insensitive to isotopic composition, and extreme changes in the isotopic composition of the waste stream only result in small changes to the normalized neutron yield. A simple spread sheet allows calculation of the relative neutron yields from various isotopic compositions based on known thick target (α,n) yields [2] and spontaneous fission half-lives [3] coupled with neutron yields per spontaneous fission event [4]. These relative calculated yields can then be used to quite accurately adjust an experimentally determined calibration factor for any specific isotopic composition and thus quantify the alpha radiation concentration.

Table 1. Relevant chemical composition of typical SRS tank wastes.

Element	Molarity
Oxygen	73.1
Sodium	5.6
Lithium	1.15E-4
Boron	8.33E-3
Fluorine	4.69E-3
Magnesium	8.23E-6
Gadolinium	8.59E-6

The neutron emission measurement is, however, non-trivial. At the detection limit of 370 Bq/mL, a typical waste stream only emits ~0.08 n/s/L. Therefore, even if the measurement system background were identically zero and the counting efficiency reached 100%, a 10,000-second count of a one-liter sample would result in ~7.1% uncertainty at the 95% confidence limit. Therefore, the background must be reduced to a value approaching zero and the counting efficiency maximized for a relatively large sample volume to make the measurement within a reasonable time frame.

2.1 DESIGN OF THE MONITOR

A prior report provides a more complete discussion of design considerations and their relative impact [5]. Practical engineering and cost limitations were the primary considerations in designing this prototype neutron monitor. The design team used MCNP4a [6] modeling of all system parameters to determine the optimal dimensions and configurations of gamma-ray shielding, sample neutron moderator/reflector, and external background neutron shielding, given the tradeoff of internally produced cosmic-ray neutron background. A complete description of the as-built system is given elsewhere [7].

Personnel chose a 15.41-cm inside diameter, schedule-40, stainless steel pipe (American Standard 6-inch pipe) for the sample chamber to minimize the time required to make a measurement at the specified accuracy and detection limit. A 3-cm-thick lead gamma-ray shield surrounds the central sample chamber. A 9.5-cm thick moderator/reflector of pure polyethylene surrounds the lead shield and contains 34 3He neutron proportional counters arranged on a single circle near the inner edge of the moderator. The neutron tubes are 1.1-m long, 2.54-cm diameter, and filled to 10-atm pressure. While these tubes are not ideal, they were available at no cost and proved adequate for this demonstration. A 30-cm thick external borated polyethylene shield surrounds the counter canister to reduce the environmental background neutron count rate to a negligible level with respect to the expected sample count rate and cosmogenic production rate. A small gamma-ray sensor is included in the tube module to provide a continuous readout of the gamma-ray dose rate incident on the neutron tubes.

The arrangement of these components is shown in plan view in Figure 1 and in a cutaway view in Figure 2. The particular arrangement of the hardware allows deployment into a biologically shielded cell using an overhead gantry crane to lift and position the apparatus and to utilize two lightweight manipulators in the cell to effect the necessary connections. The sample vessel assembly is deployed, connected, and tested independently of the instrument, which is then lowered onto the sample vessel and later removed. This configuration allows recovery of the instrument from the cell for decontamination and relocation for use in other locations. For ease of decontamination, personnel added plastic wrapping of the exterior surfaces and inserted a Mylar™ sheet into the annular opening to prevent excessive spread of contamination to the instrument.

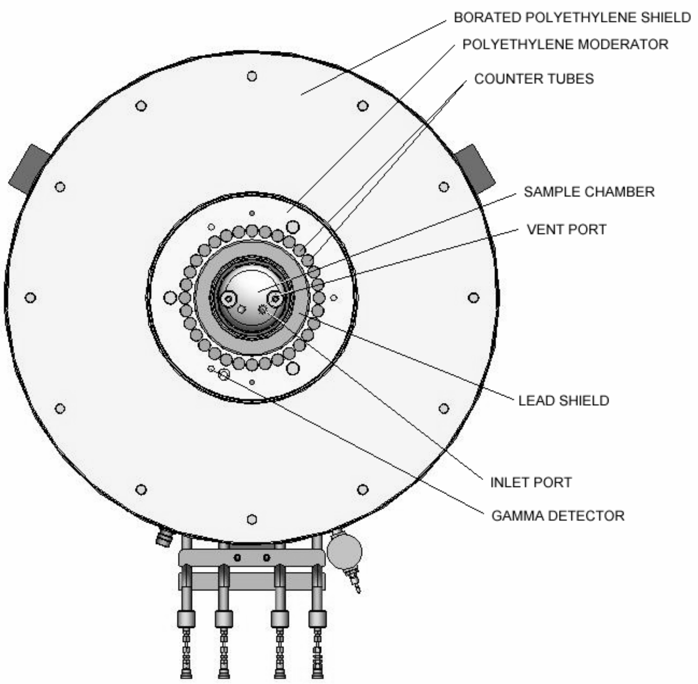


Figure 1. Sectional view of the monitor.

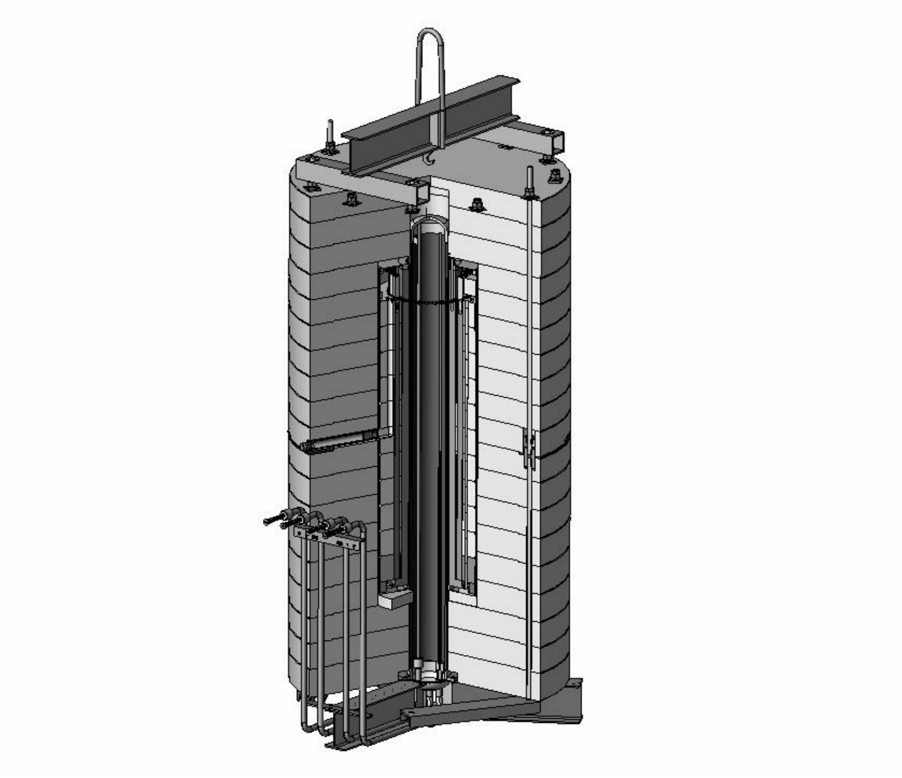


Figure 2. Cutaway view of the monitor.

Each of the ^{3}He tubes is fit with an amplifier/discriminator, and the output signals are fed, through a wired "OR" configuration, to a laptop computer serving as the data acquisition/reduction system. A custom software code processes the signal to subtract background and correct for the system efficiency, producing an output value of actual alpha activity concentration in the sample.

2.2 TANK WASTE SAMPLE ANALYSES

The demonstration used a composite of supernate samples from Tanks 37H available from previous integrated demonstrations of strontium/actinide separations via contact with monosodium titanate and cesium separation by a caustic side solvent extraction process [8]. This material had been stored in a below deck tank in Cell 3 of the Savannah River National Laboratory (SRNL) Shielded Cells Facility since the demonstration.

Personnel obtained approximately 100-mL samples of Tank 37H Composite before and after spiking with the Tank 17.1 and Bounding Waste Composite materials. We diluted duplicate 1.0-mL aliquots of each material into 2M nitric acid. After allowing the acidified samples to stand for a minimum of 2 hours, we poured approximately 5-mL of the acidified samples into shielded sample bottles, removed the samples from the Shielded Cells and submitted the samples to the Analytical Development Section of SRNL for analysis. We repeated this sample preparation protocol on filtered aliquots of the unspiked and spiked Tank 37H Composite. Filtration consisted of pushing 6 – 8 mL of the sample through a 0.45-micron Acrodisc® syringe filter and collecting the filtrate in a clean plastic sample bottle.

Personnel analyzed the samples for gamma activity and alpha activity using established analytical methods including gamma counting, alpha counting following radiochemical separations and inductively coupled plasma mass spectrometry [9].

2.3 FUNCTIONAL TESTING IN ABSENCE OF RADIATION FIELD

SRNL personnel performed several functional tests of the prototype Alpha Neutron Monitor (ANM) in the Mockup Area of the Shielded Cells Facility prior to installing the monitor in Cell 5. The first test determined that the computer controller and neutron counter remained fully functional upon energizing the system after shipment of the equipment from PNNL. (A prior test at PNNL established the functionality of the monitor [10].) At this time personnel recorded a background count with the monitor cavity empty.

The second test consisted of recording counting data with two calibration standards (i.e., blank and spiked) provided by PNNL. The first calibration standard (blank) consisted of a surrogate sample chamber filled with an acidic (pH 4) solution of sodium nitrate, potassium nitrate, and sodium fluoride [11]. The chemical concentrations approximated those in the Tank 37H Composite. The second calibration standard consisted of the same salt solution prepared for the blank calibration standard with the addition of 9.233 nCi/mL of ^{241}Am [10].

The original test plan featured removing the top of the ANM and lowering the chambers containing the calibration standards into the ANM cavity. Due to the labor and time involved with removing and reattaching the top, SRNL personnel modify the plan such that one lowered the monitor onto the pipe containing the calibration standard. To accomplish this method, we constructed a stand for the pipe similar to that used for the tank waste solution stand.

We installed offsets on the bottom of the ANM that lifted the ANM to the height of the steel beams on the sample holder stand. In this configuration the bulk of the ANM weight rests on the floor and not on the three legs of the sample holder. This modification provides a more stable base for the ANM.

We encountered difficulty in lowering the ANM onto the calibration solution chambers due to the square end at the top of the pipes. The square end catches easily on the sides of the ANM cavity which are not perfectly aligned. A rounded top end such as that provided with the sample holder proved sufficient to prevent holdup and performed well in lowering the ANM into place in the Shielded Cells demonstration (see next section).

3.0 DEMONSTRATION IN SRNL SHIELDED CELLS FACILITY

Appendix 1 provides a detailed chronology of events over the course of the demonstration of the ANM in Cell 5 of the SRNL Shielded Cells Facility. The following sections provide a summary of actions, events and observations encountered during the demonstration.

3.1 PREPARATION OF EQUIPMENT AND CELL 5 FOR DEMONSTRATION

After completion of the functional testing, personnel wrapped the exterior of the ANM with plastic wrap and installed a Mylar™ collar ring at the bottom opening to prevent contamination of the external and internal surfaces of the monitor. Personnel removed all equipment and deck plates from Cell 5 and performed extensive cleaning of the cell floor to remove miscellaneous items (e.g., plastic bottles, tools, etc.) that had accumulated over a number of years. After completion of the cleaning, personnel installed two floor grates constructed to provide a level surface on the cell floor for the monitor (i.e., cell floors are sloped to allow liquids to drain for collection).

Personnel lowered the tank sample holder through the cell roof into place on the floor grating. The holder consists of a six inch pipe with two pair of inlet and outlet nozzles for filling and draining solutions in the holder. We oriented the holder such that the fill/drain nozzles faced toward the right side of the cell window (to the northwest) as one faces the cell from the window side.

After installing the tank sample holder we lowered the ANM through the cell roof and over the sample holder until resting on the holder stand and grating. We oriented the monitor such that the electrical power and wiring connector faced to the right as viewed from the cell window. We connected the electrical power and wiring for the monitor via the Knolls Atomic Power Laboratory (KAPL) connector. We supplied power to the control computer and monitor and verified a signal from the neutron monitor.

3.2 DEMONSTRATION TESTING

The first test consisted of acquiring neutron signal with the sample holder empty. We collected this background counting overnight. The overnight counting included acquiring data simultaneously with a multiplicity counter. In principle the multiplicity counter would provide a greater signal to noise (S/N) ratio than the counting instrumentation supplied with the monitor. After the overnight counting, we disconnected the multiplicity counter and recorded an additional hour of data as background.

The demonstration featured a composite supernate sample taken from Tanks 37H previously used in radioactive demonstrations of the strontium/actinide separations and caustic side solvent extraction. The demonstration included two experiments. The first experiment used the Tank 37H Composite. This waste contained alpha activity near the process, or WAC, requirement. The waste suffered precipitation of solids during the long storage period since the earlier demonstration of the solvent extraction process. A visual check of the samples several weeks before testing with the monitor failed to identify the presence of the solids. Thus, personnel made no effort to filter these solids. Furthermore, the sample contained the entrained organic from that demonstration.

For the second experiment, personnel added a source of additional alpha activity to examine performance of the monitor at more extreme concentrations. This material came from a Tank 17.1 sample [11] and from a Bounding Waste Composite [12]. The added material contained a relatively large amount of americium and curium such as present in the worst case wastes for SRS. Adding the new materials caused additional formation of small solid particles. This experiment therefore examined the sensitivity of the monitor to a significantly different distribution of radioisotopes.

Table 2. Chemical composition of Tank 37H Composite.

Species	Concentration	Species	Concentration
Sodium (Na)	5.42 M	Lithium (Li)	0.8 mg/L
Free OH^-	3.4 M	Beryllium (Be)	< 0.2 mg/L
Carbonate (CO_3^{2-})	< 0.2 M	Boron (B)	300 mg/L
Aluminum (Al)	0.5 M	Magnesium (Mg)	0.2 mg/L
Nitrate (NO_3^-)	0.77 M	Gadolinium (Gd)	< 0.2 mg/L
Nitrite (NO_2^-)	0.80 M	Chloride (Cl^-)	< 0.007 M
Sulfate (SO_4^{2-})	<0.007 M	Oxalate ($C_2O_4^{2-}$)	< 0.014 M
Fluoride (F^-)	< 0.013 M	Formate (HCO_2^-)	< 0.04 M
Phosphate (PO_4^{2-})	0.013 M		

Table 3 provides selected radiochemical composition of samples of the Tank 37H Composite taken during the ANM demonstration. Analytical results include those obtained from both unfiltered and filtered samples. Filtration consisted of passing the Composite through an Acrodisc® syringe filter having a 0.45-micron membrane.

Table 3. Radiochemical composition of the Tank 37H Composite before and after Spiking with High Alpha Activity Materials

Analyte	**Unfiltered Tank 37H Composite**	**Filtered Tank 37H Composite**	**Unfiltered Alpha-Spiked Tank 37H Composite**	**Filtered Alpha-Spiked Tank 37H Composite**
Avg ^{137}Cs (nCi/g)	2.46E+05	3.54E+05	2.38E+05	2.32E+05
Std Dev (nCi/g)	2.44E+03	1.59E+05	4.12E+03	3.72E+03
RSD	0.991%	44.99%	1.73%	1.61%
Avg $^{239/240}$Pu (nCi/g)	1.28E+01	3.04E+00	9.24E+00	3.11E+00
Std Dev (nCi/g)	2.57E+00	1.10E+00	5.24E-02	2.23E-01
RSD	20.0%	36.32%	0.568%	7.20%
Avg ^{238}Pu (nCi/g)	3.36E+01	6.88E+00	2.32E+01	8.25E+00
Std Dev (nCi/g)	3.08E+00	2.74E+00	1.01E+00	7.91E-01
RSD	9.17%	39.86%	4.36%	9.60%
Avg Total Pu (nCi/g)	4.64E+01	9.92E+00	3.25E+01	1.14E+01
Std Dev (nCi/g)	5.64E+00	3.85E+00	1.06E+00	1.01E+00
RSD	12.2%	38.78%	3.28%	8.94%
Avg ^{241}Am (nCi/g)	1.47E+01	5.60E-01	2.72E+01	7.98E-01
Std Dev (nCi/g)	7.94E+00	9.56E-04	8.17E-01	6.23E-01
RSD	53.9%	0.17%	3.01%	78.11%
Avg ^{243}Am (nCi/g)	BDL	BDL	2.59E+00	2.25E-01
Std Dev (nCi/g)	-	-	1.24E-01	2.89E-01
RSD	-	-	4.80%	128.44%
Avg Total Am (nCi/g)	1.47E+01	BDL	2.98E+01	1.02E+00
Std Dev (nCi/g)	7.94E+00	-	9.41E-01	9.13E-01
RSD	53.9%	-	3.16%	89.18%
Avg ^{244}Cm (nCi/g)	BDL	BDL	2.77E+02	3.12E+01
Std Dev (nCi/g)	-	-	2.04E+01	3.88E+01
RSD	-	-	7.38%	124.42%
Total Avg Alpha (nCi/g)	6.11E+01	1.05E+01	3.39E+02	4.35E+01
Uncertainty (nCi/g)	1.92E+01	7.69E+00	2.44E+01	4.26E+01
Relative Uncertainty	31.5%	73.42%	7.2%	97.90%
Total Avg Alpha (Bq/mL)	2780	480	15400	1980
Uncertainty (Bq/mL)	870	350	1100	1930

BDL = below detection limit

The Tank 37H Composite exhibits a high gamma activity of approximately 2.4E+05 nCi/g. The gamma activity remained unchanged after spiking the composite with the high alpha activity materials. The total plutonium and total alpha activities of the Tank 37H Composite measured 46.4 ± 5.64 nCi/g and 61.1 ± 19.2 nCi/g, respectively. After filtration, the composite solution exhibited plutonium and total alpha activities at about 17% of the unfiltered values. Thus, nearly 83% of the alpha activity in the Tank 37H Composite is present as un-dissolved solids that can be separated by filtration.

Table 4 contains the composition of the added material. Addition of the Tank 17.1 and the Bounding Waste Composite materials to the Tank 37H Composite increased the total alpha activity – see Table 3– by a factor of 5.5 from 61.1 ± 19.2 nCi/g to 339 ± 24.4 nCi/g. The total soluble alpha activity increased by a factor of 4.1 from 10.5 ± 7.69 nCi/g to 43.5 ± 42.6 nCi/g. Thus, about 87% of the alpha activity in the spiked Tank 37H Composite material is present in the insoluble solids.

3.3 DEMONSTRATION WITH TANK 37H COMPOSITE

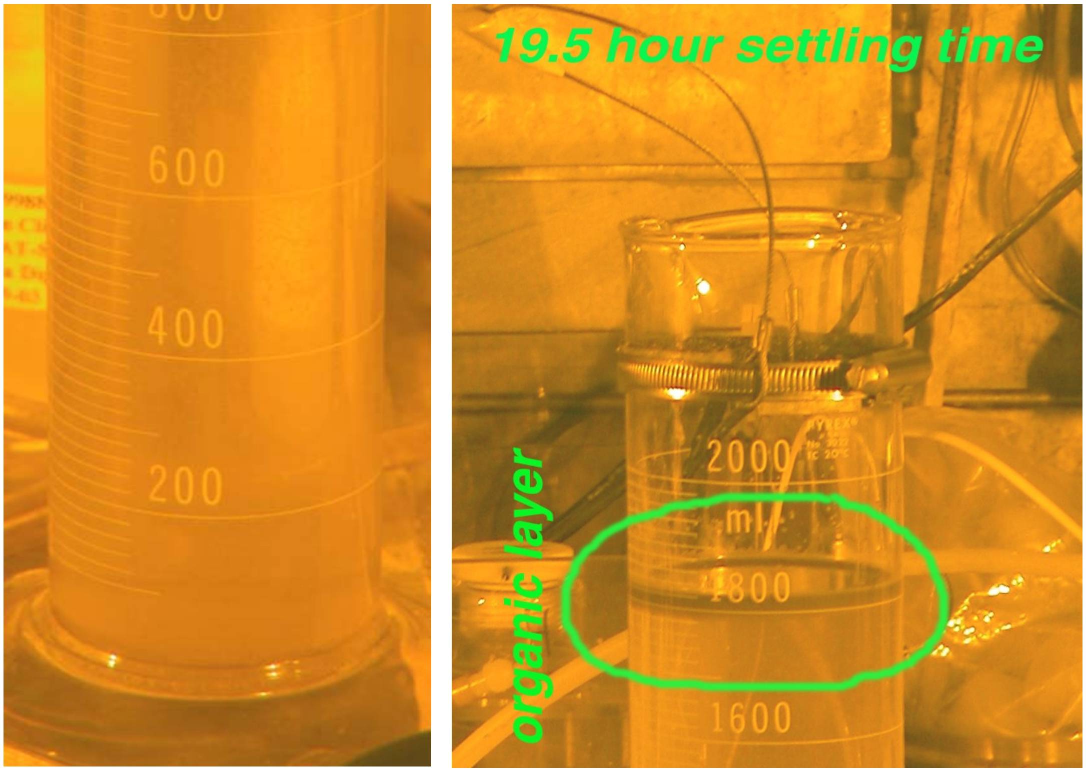
We began pumping the Tank 37H Composite from a below deck tank in Cell 3 into the sample holder in Cell 5 via the Fill/Drain #1 line (F/D #1). Pumping featured a peristaltic pump with Tygon® tubing. After pumping for 15 minutes, the tubing in the peristaltic pump developed a leak. We replaced the tubing and resumed filling the sample holder after about 30 minutes. Shortly thereafter we observed dark solids in the tubing just beyond the peristaltic pump. When pumping we suspended the holding tank contents with an air-driven agitator with a single blade propeller.

Table 4. Radiochemical Composition of Tank 17.1 and Bounding Waste Composite Materials

Component	Tank 17.1 (nCi/g)	Bounding Waste Composite[a] (nCi/g)
^{137}Cs	1.13E+05	5.11E+05
^{154}Eu	4.43E+05	ND
^{155}Eu	6.26E+04	ND
Total Gamma	1.15E+07	ND
^{238}Pu	8.46E+04	3.71E+02
^{239,240}Pu	2.97E+04	3.85E+00
^{241}Am	1.20E+05	ND
^{243}Am/^{239}Np	1.54E+05	ND
^{244}Cm	1.13E+07	ND
Total Alpha	1.15E+07	5.04E+02

[a] Values as reported in Appendix 1 of reference [10] converted to volume basis using a density of 1.25 g/mL

ND = not determined



The photograph on the left shows the presence of the fine solids on the bottom of the cylinder after ~19.5 hours of settling. Notice the solution still contains suspended fines as evidenced by the cloudy appearance. Complete settling required periods as long as ~72 hours. The photo on the right shows the presence of the organic layer at the top of the sample.

Figure 3. Photographs of Tank 37H waste after experiment.

Pumping became difficult with significant pressure build up within a few minutes. At this point we estimated that we had pumped approximately 25 L of the tank supernate out of the Cell 3 tank. The estimated volume capacity of the sample holder is 38 L.

We turned off the pump and disconnected the F/D #1 and the primary vent line (Vent #1) lines. We connected the feed line to the Vent #1 line and opened the secondary vent line (Vent #2) to the overflow carboy. We then began pumping the Tank 37H Composite from the Cell 3 tank to the sample holder through the Vent #1 line. We observed liquid flow for about 10 minutes before finding a collapsed section of the tubing, which we believed was restricting flow. We replaced the entire length of tubing from the tank to the sample and resumed pumping. After pumping an additional 4 L of supernate pumping ceased again.

We reconnected the feed line from the peristaltic pump to the F/D #1 line and a vacuum line on the Vent #1 line on the sample holder. We pulled a vacuum on the primary vent and resumed pumping. After 30 minutes we observed that this arrangement proved ineffective in filling the sample holder.

Inspection of the feed line revealed that small pieces of wood appeared to plug the quick connect at the sample holder inlet nozzle. The pieces of wood originate from the wooden ruler used to measure liquid depth in the Cell 3 tank. We reversed the pump flow to clear the wood chips from the quick connect nozzles.

We installed a 26-L carboy into Cell 3 and pumped about 10 L of supernate from the sample tank into the carboy. We elevated the carboy above the deck plate in Cell 3 so that the carboy height approximated that of the top of the sample holder and began pumping supernate from the carboy into sample holder. This arrangement worked well to fill the sample holder After emptying and refilling the carboy one more time, we observed supernate flowing in the return line from the sample holder indicating that the sample holder was full and, therefore, shut off the feed pump. We collected counting data overnight with the sample holder filled with tank supernate.

The following morning we emptied the sample holder returning the tank supernate material to the below deck tank in Cell 3. Measurement of the tank level indicated an inventory of 47 L of the Tank 37H Composite. We added 1.22 grams of the Tank 17.1 solution and 78.60 grams of the Bounding Waste Composite material to the Tank 37H Composite in the below deck tank. Table 4 provides compositions of the Tank 17.1 and Bounding Waste Composite materials. These materials contain large quantities of alpha activity principally from ^{244}Cm, ^{241,243}Am, and ^{238}Pu.

3.4 DEMONSTRATION WITH SPIKED TANK 37H COMPOSITE

After mixing the waste in the below deck tank for 1.5 hours, we began filling the sample holder with the spiked Tank 37H Composite. At the start of filling the ANM count rate measured 4.37 c/s. After pumping for 20 minutes the carboy was empty and the count rate measured approximately 72 c/s. We refilled the carboy and then resumed pumping supernate into the sample holder. After emptying the carboy again the count rate measured 128.5 c/s. We refilled the carboy and resumed pumping. After several minutes we observed liquid flowing out of the vessel vent line. We stopped the pump and left unit in the filled position and continued to collect counting data overnight.

The following day we emptied the contents of the sample holder by pumping the supernate back into the below deck tank in Cell 3. We then rinsed the sample holder by pumping approximately 13L of a 0.0077 M NaOH solution through the sample holder from a carboy in Cell 3. We collected the rinse solution in the Cell 3 carboy. We collected counting data post cleaning for about 2 hours and then turned off electrical power to the ANM.

After standing idle for 3 days, we initiated flushing with dilute nitric acid by pumping approximately 16L of 0.39M HNO3 into the sample holder. After filling with the acid, we attempted to pump the acid solution out of the sample holder.

However, we were unable to pump any liquid out of the sample holder presumably due to problems with priming the pump caused by air in the vessel lines. We left the unit in this configuration overnight.

The following day we began collecting counting data. We filled the sample holder with process water to continue flushing the equipment since we wanted to flush the entire surface of the vessel. After filling the holder with process water we collected counting data for additional 2 hours. We then pumped the nitric acid flush solution out of the vessel. This concluded the demonstration of the ANM in the SRNL Shielded Cells.

4.0 RESULTS

Two sealed 15.41-cm diameter, ~2-meter long stainless steel chambers were filled with HLW simulant for determining system background and calibrating the neutron yield from a known alpha activity. One of the chambers contained 713 Bq/mL (19.3 nCi/mL) of ^{241}Am. The efficiency of the counter, based on the ^{241}Am spiked calibration chamber, is ~0.0016 c/s per Bq/mL (~0.06 c/s per nCi/mL) of ^{241}Am alpha activity. This efficiency factor is readily adjusted to accommodate minor differences in the neutron yield from other radionuclide compositions using the calibration spreadsheet described above. This spreadsheet also allows adjustment of the efficiency factor for alternate chemical compositions and for the presence of neutron poisons. While adjustments for chemical and isotopic composition are largely independent of sample and counting geometry, corrections for the presence of neutron poisons in the sample are quite geometry dependent. The corrections for poisons applied here, while less than a 3% effect, were determined by Monte Carlo modeling [6].

More than 99% of the background neutrons observed by the instrument are engendered by cosmic-ray interactions in the internal components of the counter. About 70% of these events occur in the lead gamma-ray shield. As the cosmic-ray flux varies significantly as a function of time and location on the surface of the earth, the total neutron background varies beyond that expected based on Poisson statistics. Measurement of backgrounds need to occur with the blank simulant filled canister in the counting chamber cavity, because background neutrons are produced in the simulant solution, are moderated in the simulant solution, and are absorbed in the simulant solution. Therefore, the combined effects of the salts and solution of the HLW on the background neutron count rate can only be determined by counting a similar solution that does not contain any alpha activity.

The neutron background count rate measured in Richland, WA varied from 3.36 to 3.66 c/s with the blank simulant chamber in place. While observing the neutron pulses with an oscilloscope personnel noted that the vast majority of events appeared to be multiple coincidence events, most of which appeared within 100 µs bursts. This would be the expected behavior if the background neutrons are largely produced by cosmic muon induced spallation reactions. Since, at the required system detection limit of 370 Bq/mL, the expected TRU signal count rate is only ~0.6 c/s, more than 99% of which would be single neutron events, by separating the background events into "singles" and "coincidences" the effective system background can be significantly reduced. A "singles" neutron background rate of 0.492 c/s was measured with a neutron multiplicity counter having the coincidence window width arbitrarily set at 100 µs, which had the effect of increasing the signal-to-noise ratio by a factor of ~7.

After arrival to the Savannah River Site, two background measurements were made with the blank simulant canister in the central cavity at a non-radioactive staging location to verify proper operation of the counting system. These two measurements resulted in background rates of 3.15 and 3.28 c/s with the combined average being 3.24 c/s. Given the lower anticipated cosmic muon flux at the lower geomagnetic latitude of the Savannah River Site, this count rate appeared correct, and we declared the counter to be operating properly.

The counter was then moved into a cell, and a background measurement taken with the sample-counting chamber empty. To minimize the generation of radioactive waste, personnel omitted introduction of the blank waste into the cell. The true system background in the cell is expected to be similar to the 3.24 c/s measured in the staging area, although it might be slightly lower due to some cosmic ray shielding afforded by the thick concrete lids on the top of the cells. The measured empty-chamber background of 4.37 c/s in the cell also appears reasonable, assuming that the presence of a simulant solution would absorb more of the background neutrons than it would generate. In any case, the net count rates from the samples counted in the cell were high enough that the additional uncertainties introduced by not knowing the true backgrounds accurately are negligible.

Personnel introduced an actual sample of high-level waste into the empty chamber and counted overnight. They then removed the HLW sample from the counting chamber and performed another empty chamber background count. Personnel then added a spike of Am/Cm to the original HLW sample, along with a small quantity of "Bounding Waste Solution" that contained additional alpha activity. They introduced this spiked waste solution into the counting chamber and again counted overnight. Personnel sampled and analyzed both the original and the spiked waste solution for TRU concentrations in a wet chemistry laboratory by conventional methods.

Both the original waste solution and the spiked solution contained precipitates, and neither represented a true solution. In fact, the original waste sample contained so much particulate material that pumping problems occurred in transferring the material into the counting chamber. Similarly, when adding the Am/Cm spike to the waste, significant precipitation occurred, and stirring proved necessary to maintain the precipitates in suspension for introduction into the counting chamber.

These precipitates also complicated the laboratory analyses, requiring measurements on both filtered and unfiltered aliquots.

Subsequent measurements showed that settling of the solids required long periods. Even after several days, the solution remained hazy indicating the presence of solids. Hence, we will compare the concentration determined by the monitor to that of the unfiltered sample. For the original sample, this approach seems reasonable, even though the solids account for ~40% of the activity. For the spiked material, the curium is known to rapidly precipitate.[11] Since curium dominates the concentration in the spiked composite (i.e., >80% of the activity) and since curium grossly impacts the calibration factor (as discussed below), this approach will prove less reliable in that instance.

The laboratory results, which represent the best values for the solutions including the suspended particulates, are summarized in Table 5 for both the original waste sample and the spiked waste sample. The loss of plutonium between the original and the spiked sample is attributed to additional precipitation induced when the spikes were added. The ^{239}Pu and ^{240}Pu concentrations are not measured separately and hence are shown combined in the table; for calculations for the monitor we assume an ^{240}Pu isotopic composition of 6% by weight. All values use a measured solution density of 1.227 g/mL.

Table 5. Measured laboratory concentrations of unfiltered TRU isotopes in the solutions and solids of the HLW sample before and after spiking the sample.

Isotope	Original Waste (Bq/mL)	Spiked Waste (Bq/mL)
^{238}Pu	1530 ± 140	1050 ± 50
$^{239/240}$Pu	581± 117	419 ± 238
^{241}Am	670 ± 360	1230 ± 4
^{243}Am	ND	118 ± 5
^{244}Cm	ND	12600 ± 926
Total Alpha	2780 ± 620	15400 ± 1200

The sample chamber was partially filled with the original waste solution when problems occurred due to solids in the waste. Personnel initiated a counting sequence with the sample chamber partly filled, and the filling completed during the running count sequence. The count rate, measured in one-minute intervals, is shown in Figure 4. Note the step-function increase in the count rate from ~7 c/s to ~8.5 c/s when the filling process completed. Also note the steady decline in count rate as a function of time due to particulate material settling to the bottom of the counting chamber. The maximum statistically significant count rate was 8.63 c/s. Assuming the true system background in the hot cell is the same as measured in the cold staging area, 3.24 c/s, the measured net count rate was 5.39 c/s. The calibration factor measured with the ^{241}Am-spiked calibration chamber equaled 0.00166 c/s per Bq/mL. Correcting this calibration factor for the difference in expected neutron yields from the spreadsheet for the pure ^{241}Am spiked calibration chamber and the actual composition of the waste solution results in a calibration factor of 0.00155 c/s per Bq/mL, which when applied to the net count rate of the waste solution results in a measured concentration of 3480 Bq/mL, in reasonable agreement with the laboratory measured value (2780 ± 620 Bq/mL) reported in Table 5, particularly when considering the relatively large uncertainties engendered in both the laboratory analyses and in the cell measurements due to the presence of the precipitates. The monitor value is biased ~25% high relative to the laboratory measurement – i.e., provides a conservatively high result relative to the regulatory objective.

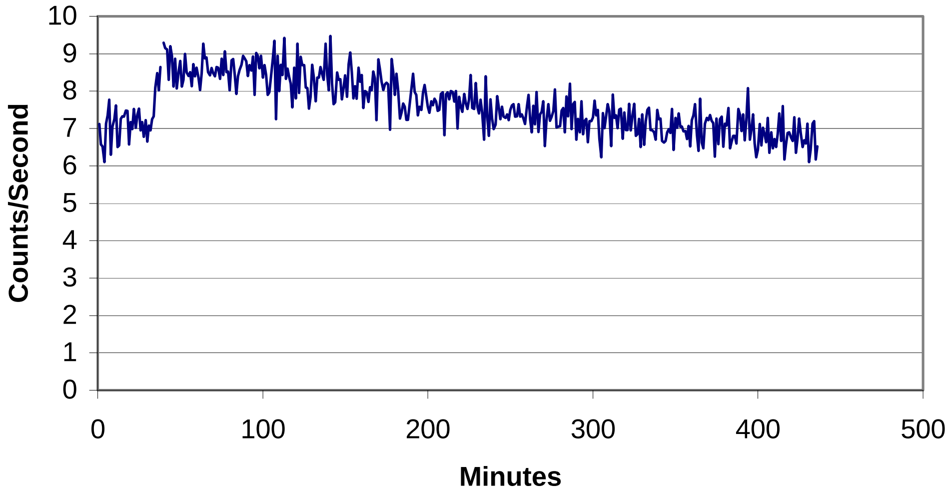


Figure 4. Measured count rate of the spiked HLW solution.

In all cases, the statistical uncertainties in both laboratory and cell measurements are negligible compared to the systematic uncertainties introduced by the presence of the precipitates. Some reasonable measure of these systematic uncertainties is possible in the case of the laboratory analyses, and these are included in the values reported in Table 3. However, no quantitative estimate of the loss in counting efficiency in the cell measurements due to the settling of precipitates is possible. We can only estimate the magnitude of this uncertainty by observing the change in count rate as a function of time.

After the waste solution was removed from the counting chamber, an empty chamber background measurement resulted in 4.58 c/s, about 5% higher than before introduction of the waste solution into the virgin counting chamber. This slight increase could result from residual contamination from the waste solution on the walls of the chamber, or to real change in the cosmic muon introduced neutron background. Since residual contamination certainly existed, we retained the background value as 3.24 c/s.

Personnel then pumped the spiked waste solution into the counting chamber while the counting sequence continued. The filling process completed in two obvious steps with the first step raising the count rate to ~70 c/s and the second raising the count rate to ~130 c/s as shown in Figure 5. The subsequent decaying count rate again demonstrates the precipitation of solids inside the counting chamber. The maximum statistically significant count rate equals 128 c/s, or 125 net c/s due to the spiked waste solution. In this particular case, nearly 99% of the neutrons result from the ^{244}Cm spike, and more than 90% of those are due to spontaneous fission rather than (α,n) reactions. Therefore, the neutron yield per unit of alpha activity for this spiked solution is more than an order of magnitude greater than for the original waste solution. Using the spreadsheet correction for the calibration factor results in a value of 0.0255 c/s per Bq/mL for the spiked solution, or an alpha activity of 4900 Bq/mL. Since this value is only about one third of the laboratory measured value (15,400 ± 1200 Bq/mL) for the solution and solids, it demonstrates that a large fraction of the curium spike precipitated before it was counted. This result is consistent with visual observation during subsequent settling observations on the waste (see Figure 3). Also, the off-line analyses (Table 3) indicate that at least 90%, and possible much more, of the Cm did precipitate.

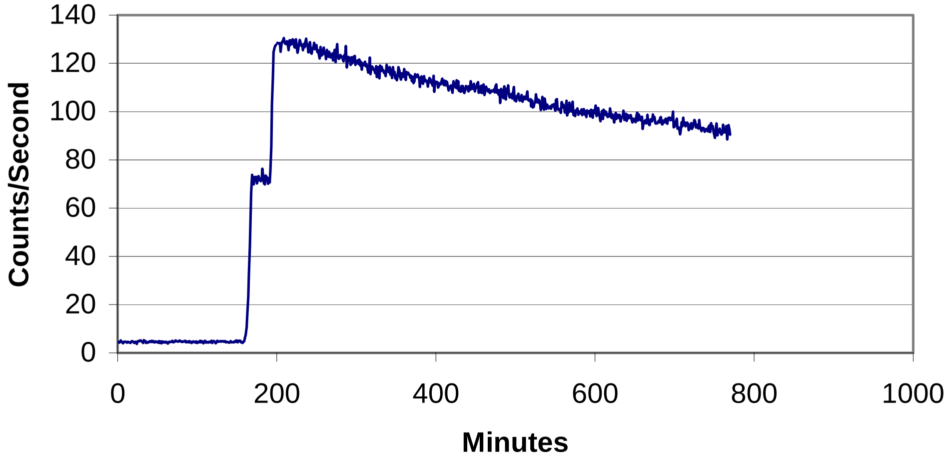


Figure 5. Measured count rate of the spiked waste solution.

Following removal of the spiked waste solution from the counting chamber, the empty chamber background equaled 10.6 c/s, which clearly represents residual contamination inside the counting chamber corresponding to roughly 5% of the activity added to the chamber. Considering the finely divided nature of the precipitates that formed in the spiked solution, that level of residual contamination appears reasonable. Personnel then flushed the sample chamber three times with a dilute sodium hydroxide solution in an attempt to remove the contamination. The empty chamber count rate only dropped to 9.79 c/s, indicating removal of only about 14% of the residual contamination. Personnel considered but did not conduct a dilute acid flush. This concluded the hot cell demonstration, and personnel prepared for removal of the counter assembly from the cell for decontamination and reassignment.

5.0 CONCLUSIONS

Given the sample conditions and constraints under which this demonstration occurred, the monitor provided reasonably accurate measurement. No operational problems occurred with the monitor. Response times were very rapid suggesting required count times much shorter than 6 hours. Accurately determining the response time requires testing in a flow-through configuration. Operation did indicate a potential for residual contamination as high as 5% for the first experiment that most closely matches planned operation. This level of contamination is likely acceptable for operation. Simple flushing with process water or caustic did not prove effective in this configuration but the available flushing velocities proved much lower and less effective than likely typical of field deployment.

An on-line full-scale separations plant version of this monitor would need to operate uninterrupted for at least five years. Since there are no moving parts to the monitor design – beyond the valves for the associated piping – only electronic failures are anticipated. Such failures would include the ^{3}He tubes or their attached preamplifier/discriminators. The signal output from each of the tube/preamp combinations can be wired independently to a biologically-shielded accessible location where personnel can disconnect any tube/preamp that might become electronically "noisy" or otherwise degrade from the array. The efficiency for a disconnected or otherwise dead tube can be corrected by monitoring a time-tagged neutron source implanted in the monitor. This time-tagged control source would consist of a small ^{252}Cf source with a spontaneous fission rate of about 1 Bq. Each fission will produce an electronic tagging pulse, so that corresponding neutron capture events in the monitor are clearly identified and stored separately as control events. In this manner, real-time operational control of the monitor is guaranteed without increasing the system background and thus diminishing the S/N ratio and sensitivity of the system.

The next generation monitor should incorporate use of a neutron multiplicity counter. The neutron multiplicity counter circuitry effectively increases the S/N ratio by a factor of ~7 by rejecting the majority of coincidence events as "tagged" non-background events. This feature will provide both increased accuracy in the measurements and reduced time to make a determination.

Since >99% of the background neutrons are engendered by cosmic rays, any reduction in cosmic-ray neutron production will further enhance the S/N ratio. The gamma sensitivity of a ^{3}He tube is almost directly proportional to its fill pressure, while the neutron-counting efficiency is not [13]. For example, a 2-atm tube will withstand ~5 times the gamma dose of a 10-atm tube, while the neutron counting efficiency is diminished by less than a factor of two in the 2-atm tube. The simple improvement of using 2-atm tubes will allow the mass of lead gamma-ray shield to be reduced by >44%, with a corresponding reduction in the background count rate. Similarly, both the available signal and the background noise scale linearly with the length of the tube, but since the general figure of merit scales directly with increased signal and inversely with the square root of the background, the longest tubes possible are preferred. Engineering, cost, or installation considerations should be the only reason for selecting shorter tubes. For the same reasons, the design should use 2.54-cm (1-in) diameter tubes rather than 5.08-cm (2-in) diameter tubes. If greater efficiency is required to meet programmatic requirements, the design can incorporate two rows of the smaller diameter tubes, albeit at a greater expense.

Changing to a smaller diameter and thinner pipe for the sample counting chamber, for example American Standard 3-inch schedule-10, and utilizing two rows of ^{3}He tubes will lead to an overall improvement in S/N relative to the current design. First, the counting efficiency provided by two rows of tubes will nearly double that provided by one row, which will nearly compensate for the reduced number of sample neutrons generated in the smaller sample volume. While the number of neutrons generated will decrease by a factor of four, the fraction of those exiting the sample chamber and reaching the counting tubes will greatly increased due to less self-absorption in the sample itself and in the mass of steel composing the chamber walls. Furthermore, the background reduces even more because the mass of steel is less and because the mass of lead gamma-ray shielding is further reduced since the gamma-ray dose emanating from the sample chamber substantially decreases.

Finally, in those circumstances where the gamma-ray dose is not intense, i.e. post-cesium removal in the SRS HLW processing plant, the monitor will significantly benefit from the use of an annular sample chamber and counting tubes both on the inside and outside of the annulus [5].

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7.0 REFERENCES

1. D. D. Walker and S. D. Fink, “Task Technical and Quality Assurance Plan for In-Line Neutron Counter Testing,” WSRC-RP-2002-00539, December 2, 2002.
2. H. Matsunobu, T. Oku, S. Iijima, Y. Naito, F. Masukawa, and R. Nakasima, Data Book for Calculating Neutron Yields from (α,n) Reaction and Spontaneous Fission, JAERI 1324, Japan Atomic Energy Research Institute, Tokaimura, Nakagun, Ibarakiken (1992).
3. R. B. Firestone and V. S. Shirley, eds., Table of Isotopes, 8th edition, John Wiley & Sons, New York, NY 1996.
4. E. K. Hyde, The Nuclear Properties of the Heavy Elements, Prentice Hall Inc., Englewood Cliffs, NJ 1964.
5. R. L. Brodzinski and R. A. Craig, “An Instrument for Monitoring the Transuranic Concentration in Processed High-Level Waste,” PNNL-SA-36413 (2002), Spectrum 2002, Proceedings of the 9th Biennial International Conference on Nuclear and Hazardous Waste Management, Reno, Nevada, August 4-8, 2002.
6. MCNP4a: Monte Carlo N-Particle Transport Code System, distributed by Radiation Shielding Information Center at Oak Ridge National Laboratory.
7. R. L. Brodzinski and R. A. Craig, “An Instrument for Monitoring the Transuranic Concentration in Processed High Level Waste,” PNNL-SA-36413, 2002.
8. M. A. Norato, S. D. Fink, F. F. Fondeur, G. F. Kessinger, R. A. Pierce, and D. D. Walker, “Demonstration of The Caustic-Side Solvent Extraction with Optimized Solvent in the 2-cm Centrifugal Contactor Apparatus using Dissolved Saltcake from Tank 37H,” WSRC-TR-2002-00307, Rev. 1, September 3, 2002.
9. Procedure Manual L16.1, “Analytical Development Section Analytical Operating Procedures (U),” Volume II – Ions, Radiochemistry, Spectroscopy.
10. R. Brodzinski, W. Hensley, N. Holt, M. Knopf, E. Lepel, D. Mullen, T. Samuel and J. Smart, “SRS Prototye In-Line Neutron Monitoring System – Project Summary Letter Report,” PNNL-43237-LR-1001, Rev 0, March 2003.
11. T. B. Peters, D. T. Hobbs, D. P. DiPrete, C. C. DiPrete and S. D. Fink, “Demonstration of Disposal of Americium and Curium Legacy Material Through the High Level Waste System: Results from Baseline Flowsheet Studies,” WSRC-TR-2001-00503, October 23, 2001.
12. S. D. Fink, D. T. Hobbs and T. B. Peters, “Demonstration of MST Efficacy on Removal of Actinides and Strontium in “Bounding Alpha” Waste,” WSRC-TR-2002-00555, December 17, 2002.
13. For a discussion of 3He tube characteristics see the website http://www.gepower.com/dhtml/reuterstokes/en_us/monitors/more_he3.jsp/

Appendix 1

Chronology of Events during Alpha Neutron Monitor Demonstration in SRNL Shielded Cells

Date & Time	Event/Action/Observation
9/16/03	Installed ANM in Cell 5.
9/16/03 1400	Initiated background count including multiplicity counter.
9/17/03 0800	Disconnected multiplicity counter and collected an additional 1 hour of background counts.
9/17/03 0910	Initiated filling sample holder with Tanks 37H Composite from below deck tank in Cell 5. Primary vent line (Vent 1) opened to discharge excess liquid into carboy in Cell 5.
9/17/03 0925	Replaced tubing that had developed leak.
9/17/03 0935	Initiated collecting counting data.
9/17/03 0955	Resumed pumping liquid into sample holder; changed tubing section in pump head approximately every 3 minutes to prevent wearing. Observed small amount of dark solids in tubing past pump head.
9/17/03 1015	Observed considerable head pressure. Stopped pump and released pressure. Resumed pumping. After pumping for a couple of minutes we observed high head pressure. Little liquid flow occurred as dark solids remain in the same position within the tubing.
9/17/03 1035	Estimated that 25 L of supernate pumped out of tank in Cell 5. Count rate at this point is below the background counts with the empty sample holder.
9/17/03 1350	Connected feed pump to primary vent line and resumed pumping at lower speed. Secondary vent line opened to overflow tank in Cell 3.
9/17/03 1415	Observed liquid flow with much lower head pressure.
9/17/03 1615	Stopped pump as liquid not drawn into line from tank due to a collapsed section of tubing. Replaced Tygon® tubing and connected feed line to secondary vent line. Primary vent line serves as the overflow vent.
9/18/03 0830	Initiated pumping liquid into sample holder.
9/18/03 0840	Total liquid pumped out of Cell 5 tank measured at 29L indicating that the sample holder is not filled. Observed spike in counting data corresponding with startup of feed pump.
9/18/03 0940	Pumping no longer effective as liquid level shows no liquid level drop. Checked to see if liquid in primary vent line down from ANM. No liquid observed. Set up vacuum line from Cell 6 to primary vent line.
9/18/03 1235	Began pumping liquid from Cell 3 tank while pulling vacuum on primary vent.
9/18/03 1245	Liquid level in Cell 3 tank dropped approximately ¼” given a pump rate of ca. 60 mL/min. Estimate will take 1.5 – 2 hours to fill sample holder at this rate.
9/18/03 1315	No further drop in liquid level in Cell 3 tank. Pump stopped. Installed carboy in Cell 3.

9/18/03 1400	Observed small pieces of wood plugging the quick connect on the primary feed line. Reversed liquid flow to clear quick connect.
9/18/03 1545	Observed sticky solids in Cell 3 tank.
9/18/03 1600	Installed new tubing in the cell and pumped approximately 10L of solution from the Cell 3 tank into the carboy. Elevated carboy on back wall of Cell 3 such that the bottom of the carboy is level with top of sample holder in Cell 5.
9/18/03 1645	Initiated pumping of solution from carboy into sample holder through secondary vent line (Vent #2).
9/18/03 1700	Put vacuum trap in place (???). Carboy nearly empty; pumped additional solution from Cell 3 tank into carboy.
9/18/03 1720	Refilled carboy. Cell 3 tank level at 5.25 inches.
9/18/03 1723	Resumed filling sample holder.
9/18/03 1728	Turned feed pump off
9/18/03 1742	Refilled carboy. Cell 3 tank level at 2.75 inches.
9/18/03 1752	Observed liquid exiting through primary vent line indicating that the sample holder is filled. Switched off pump and continued to collect counting data overnight with sample holder filled.
9/19/03 0650	Counting data for last 10 hours shows smooth decline of about 10 nCi/mL. Halted data collection.
9/19/03 0835	Vented both vent lines and empted sample holder to Cell 3 tank. Tank level measured 14.5 inches. Liquid remains in Cell 3 carboy.
9/19/03 0840	Observed that the controller time set to wrong date and time zone. Corrected time to 9/19/03 and Eastern time zone.
9/19/03 0850	Saved all qwe files to folder on zip drive named “backup qwe 9_19_2003”.
9/19/03 0853	Resumed counting of empty vessel. Count rate measured 4.75 c/s.
9/19/03 0854	Dumped solution recovered from vent lines into Cell 3 tank. Cell 3 tank level at 16.25 inches.
9/19/03 1115	Added 1.22 grams of Tank 17.1 solution into the Cell 3 tank.
9/19/03 1145	Added 78.60 grams of the Bounding Waste Composite material to the Cell 3 tank.
9/19/03 1146	Began agitation of Cell 3 tank contents.
9/19/03 1300	Stopped agitation of Cell 3 tank contents.
9/19/03 1315	Cell 3 tank level measured at 16.25 inches.
9/19/03 1318	Began filling Cell 3 carboy with spiked Tank 37H Composite from Cell 3 tank.
9/19/03 1340	Connected feed supply from carboy to primary feed line on sample holder (Feed #1). Counting data collected in file named Run #3 9_19_03.dat followed y 9202003.dat.
9/19/03 1345	Background count rate measured 4.37 c/s.
9/19/03 1350	Initiated pumping of liquid from Cell 3 carboy into sample holder.
9/19/03 1400	Cell 3 carboy empty. Pump stopped. Count rate measured approximately 72 c/s. Refilled Cell 3 carboy.
9/19/03 1426	Resumed filling sample holder from Cell 3 carboy. Count rate measured approximately 70.6 c/s.

9/19/03 1435	Cell 3 carboy empty. Stopped pump. Refilled Cell 3 carboy. Count rate measured approximately 128.5 c/s.
9/19/03 1449	Resumed filling sample holder from Cell 3 carboy.
9/19/03 1453	Sample holder filled. Stopped pump. Cell 3 tank level measured 1.0 inches. Cell 3 carboy contains approximately 5L of solution.
9/19/03 1454	Left system to collect counting data overnight.
9/20/03 0711	Halted data collection. Count rate dropped overnight to approximately 65 c/s. Changed file names to Run2Files1 and 2, respectively.
9/20/03 0723	Initiated pumping waste solution from sample holder back into Cell 3 below deck tank. Opened both vent lines.
9/20/03 0727	No liquid flow. Pulled vacuum on both vent lines. Collected approximately 300 mL of solution.
9/20/03 0740	Observed that the count rate increased upon pumping liquid out of sample holder. Suspect that the pumping suspended settled solids that have high alpha activity.
9/20/03 0755	Still experiencing problems pumping out of the sample holder.
9/20/03 0820	Checked tubing for plugging by pumping in and out of feed tank. No plugging observed.
9/20/03 0830	Initiated pumping out of sample holder through Fill/Drain #1 line. Observed good liquid flow now. Problem was unprimed pump.
9/20/03 0910	Feed vessel empty. Stopped pump. Collected approximately 100 mL of the solution from the Cell 3 tank for radiochemical analyses.
9/20/03 1029	Initiated pumping 0.0077M sodium hydroxide solution from Cell 3 carboy into sample holder through Vent #2. Observed small leak behind quick connect fitting on Vent #1. Stopped pump and connected feed pump to Fill/Drain #1 line using Vent #2 to vent the sample holder.
9/20/03 1043	Cell 3 carboy with 0.0077M NaOH solution is empty. Stopped pump.
9/20/03 1044	Reversed pump flow and emptied sample holder back into Cell 3 carboy.
9/20/03 1057	Sample holder empty. Reversed flow and pumped dilute caustic solution back into the sample holder.
9/20/03 1106	Cell 3 carboy empty. Reversed pump flow and emptied sample holder back into carboy.
9/20/03 1114	Sample holder empty. Reversed flow and pumped dilute caustic solution back into the sample holder.
9/20/03 1122	Cell 3 carboy empty. Reversed pump flow and emptied sample holder back into carboy.
9/20/03 1130	Sample holder empty. Initiated collecting counting data with empty sample holder - file name “Run 3 Post Cleaning”.
9/20/03 1255	Turned on feed pump and observed a small amount of liquid exiting the sample holder. Count rate immediately decreased from 10 to 8.5 count s^{-1}.
9/20/03 1257	Placed the dilute NaOH cleaning solution in Cell 4 for temporary storage.
9/20/03 1332	Stopped data collection and shut down ANM. Unhooked wiring connector to the ANM in Cell 4.
9/23/03 afternoon	Pumped approximately 16L of a 0.38M nitric acid solution into sample holder from a carboy in Cell 3 through the Vent #2 line. Attempted to

	pump the acid solution out of the sample holder but did not observe any liquid flow. Left sample holder with dilute nitric acid solution overnight.
9/24/03 0900	Reconnected wiring to ANM and began collecting counting data – file named “nitric acid flush 9-24-03”.
9/24/03 1005	Initiated pumping process water into sample holder.
9/24/03 1020	Completed filling sample holder with process water.
9/24/03 1145	Count rate measured approximately 17 c/s.
9/24/03 12:15	Pumped dilute nitric acid out of sample holder. Disconnected electrical wiring to ANM.
9/30/03	Removed ANM and sample holder from Cell 5. ANM temporarily stored in 773-A, Room E-079. Sample holder discarded to solid waste.