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Title page

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Radium-226 Analysis Methodology in Savannah River Site High Activity Waste Matrices

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Abstract

Waste cleanup efforts currently underway at the Savannah River Site have created a need to characterize radium-226 levels in the various high activity waste matrices currently in Site inventories. The traditional method our laboratory used for analyzing Ra-226 in higher activity matrices was based on classic cation exchange methodology. Radiochemical separations were often initiated in remotely operated shielded analytical cells followed by additional hands-on separations in radiological hoods. Methodology based on IBC Advanced Technologies SuperLig 640 extractant, mounted in 3M Empore filter media has been developed to streamLine the radium analyses.

Keywords

Radium-226, Ra-226, Radium, Empore, Superlig 640

Introduction

The US Department of Energy's (US DOE) Savannah River Site (SRS) historically produced plutonium and tritium supporting the Nation's nuclear weapons programs. It also performed numerous other radioisotope production programs. SRS also conducted nuclear fuel reprocessing to support the Site's nuclear reactors. While the plutonium production activities have ceased, a portion of the US DOE spent nuclear fuel inventory continues to be reprocessed at SRS to support a non-proliferation effort to blend highly

enriched uranium with depleted uranium to generate low enriched uranium feedstock for commercial nuclear reactor fuel.

High level radioactive waste from both the legacy and the current operations is stored in one of two SRS High Level Waste Tank Farms. The SRS waste tanks are highly caustic (2M OH) and high in sodium (6M Na), resulting in two waste forms (supernate and sludge) with differing characteristic waste distributions. Cs-137 remains soluble in the supernate liquid and dominates the radiological inventory of SRS's high level waste supernate as well as its dried saltcake form. SRS supernate is nominally 6.61E+04 MBq/L Cs-137, SRS Saltcake is nominally 7.33E+03 MBq/L Cs-137[1]. At significantly lower levels other isotopes are present to the limits of their low solubilities in this chemical form. Radioisotopes that are insoluble in the caustic supernate precipitate to form a sticky sludge bottom layer. The primary radioisotopes in that sludge layer are Sr-90 and its daughter Y-90 at nominally 4.11E+05 MBq/L[1]. The sludge layer is also high in actinides, fission products and other activation products.

High-level-radiological-waste cleanup processes being conducted by the Savannah River Site Liquid Waste Programs have created an ongoing need to characterize the radiological inventories of the various SRS Tank Farm waste forms, as well as the products of waste treatment activities that are generated by the SRS Liquid Waste Programs. Although the predominant radioisotopes are similar from waste tank to waste tank, as SRS ran numerous unique radioisotopic production campaigns beyond the core weapons programs, each tank being treated can have unique chemical and radiological distributions, rendering the use of routine analyses for radioisotopes exiting at trace levels problematic. As each tank is a completely new matrix with often-unforeseen interferences, method development is often required building on prior experiences analyzing these sample matrices.

Differences in the relative concentrations of radionuclides and their radiological decay rates produce a large variation in the extent that individual radionuclides contribute to the total radioactivity over time. Since the ability to detect radionuclides is a function of their activity, one criterion for gauging long-term radionuclide contribution from closed

waste tanks is based upon the relative dominance of radionuclides driving activity at the present and into the future. While currently representing only a small fraction of the activity in SRS radioactive waste inventory, Ra-226 and its progeny will eventually represent a more significant portion of the inventories due to its relatively long half-life. In fact, performance assessments conducted in both tank farms concludes that over a 10,000 year period of assessment, Ra-226 is in the top three of radionuclides having the largest contribution to dose to a member of the public (MOP) at the 100-m boundary. The ingrowth of Ra-226 (as radiological parents decay) trends over time in select sectors of the SRS H and F Tank Farms are presented in Figure 1 and Figure 2. This prediction is due to ingrowth of Ra-226 from radiological decay of current Th-230, U-234 and Pu-238 inventories. Current SRS Ra-226 inventories are expected to be quite low as uranium feedstocks brought onto the Site for radioisotopic production and reactor fuel were purified of radium impurities elsewhere.

The required Ra-226 detection limit can be 8 or more orders of magnitude lower than the activities of shorter-lived radioisotopes commonly present in various SRS sample matrices. Ra-226 is an alpha emitting radionuclide with weak gamma emissions. Therefore, a very efficient separation of Ra-226 from high levels of interfering radionuclides is required for sensitive measurements.

The previously used Ra-226 analysis methodology was based on classic cation exchange technology[4] augmented with decontamination treatments using a number of isotope-specfic extractant clean-up steps. Aliquots of tank waste samples were digested in the Shielded Cells using a sodium peroxide fusion dissolution method. The sample digestions were performed in the Shielded Cells due to the high dose associated with the relatively large sample aliquot sizes required to obtain the low desired Ra-226 analytical minimum detection limits. The digested solutions were then decontaminated from interfering nuclides in the Shielded Cells with a series of extraction resins added in batch-contact mode. Bio-Rad AMP (ammonium molybdophosphate) was added to reduce the Cs-137 concentrations[5,6,7]. Eichrom Strontium resin was added to reduce the Sr-90 concentrations[8]. Eichrom Diphonex resin was added to reduce the Y-90, actinide-isotope, and lanthanide-isotope concentrations[9]. The decontaminated sample solutions were then

removed from the Shielded Cells and transferred to the radiohoods in the analytical laboratories, where they were treated again to the same batch-contact decontamination steps as performed in the Shielded Cells to further improve sensitivity. Following decontamination from interfering radionuclides, radium was extracted from the solution using cation exchange resin. The radium chemical yields were traced with Ra-224 and Ra-226. Yields from this methodology were low and erratic (the mean radium yields were on the order of 10%, but could be as low as 1-3%). Some isotopes, such as Co-60, co-extracted, which caused increased detection limits.

Improvements in these yields and further reduction in interferences were desired so that smaller aliquots of sample could be used to obtain the desired minimum detection limits – thus, eliminating the need for a specific Shielded Cells digestion dedicated to the Ra-226 analysis. Improvements in the yields would also eliminate the need for the specific decontamination steps performed in the Shielded Cells that were previously required for the Ra-226 analysis. The net result of such changes would be a significant reduction in analytical costs and turnaround times.

Several extraction methods were evaluated for testing. Eichrom's Lanthanide resin is reported to have an affinity for radium[10]. However, its selectivity for radium relative to the other elements that would be present in SRS high level waste matrices appears poor. Eichrom also produces an MnO_2-based resin which shows affinity for radium[11,12], but again, does not appear to have the selectivity necessary to effectively extract radium from tank waste samples. 3M produces Empore Radium RAD disks, with the claim that the technology is highly selective for radium, without retaining other alkaline earth metals such as strontium[13]. This characteristic is essential for a radium extractive medium used for SRS tank samples, since strontium-90 is a dominant radioisotope in these samples and is the main source of extremity dose associated with such samples. Available literature suggests that the active extractant in Empore Radium RAD disks is IBC Advanced Technologies molecular recognition extractant Superlig 640[14] . IBC also offers this extractant in a analytical bead form sold as AnaLig® Ra-01. The same reference source indicates that Superlig 640 co-extracts strontium with radium, which conflicts with the 3M literature. Recently published methodolody using IBC Advanced Technologies Sr-90

molecular recognition extractant (based on Superlig 620) to extract Ra-226 implys the the radium molecular recognition product is quite similar to the strontium molecular recognition product[15]. While co-extraction of radium and strontium in evironmental matrices may not be an issue, it is problematic in high level waste forms high in Sr-90.

Experiments were conducted to evaluate the suitability of Empore Radium RAD disks based on the IBC Superlig 640 technology for quantifying Ra-226.

Experimental

Ra-226 Separation and Analysis

Testing of Empore Radium RAD Disks on SRS Supernatant Samples and Spikes

Initial tests were performed using the 3M Empore Radium RAD disks to extract radium from SRS supernate samples and spiked acid blanks. Two-milliliter aliquots of supernate were acidified to 2M using nitric acid, with the total sample volume being ~38 mL after acidification. The supernatant samples were traced with Ra-224, while the spiked acid blanks had both Ra-224 and Ra-226. Cesium-137 concentrations in the samples were reduced using multiple batch-contact extractions with Bio-Rad AMP. During these Bio-Rad AMP strikes, ~0.2 grams of Bio-Rad AMP was added to 38 mL of acidified sample solution and the combined material was agitated for 30 seconds. After each strike, the Bio-Rad AMP solids were removed via 0.45-µm filtration. Following the Cs-137 removal, radium was extracted from the solutions by passing the solutions through Empore Radium RAD disks at a nominal flow rate of one milliliter per second. The disk was then rinsed with 150 mL of 2N nitric acid, and the rinsed disk was placed into a sealed 2.0 mL test tube for gamma analysis. The samples were assayed using a 70% relative efficent robotic HPGe well gamma spectrometer.

The well geometry gamma spectrometers were chosen for the Ra-226 analysis as the counting geometry of such systems approaches 4 pi. That counting geometry is the most forgiving when counting a potentially non-homogeneous counting vessel, such as a filter paper inserted into a secondary container. Also, as the branching ratio of the primary 186

keV Ra-226 gamma emission is quite small (branching ratio 3.64%)[17], better sensitivities for Ra-226 can be achieved if measuring higher branching ratio gamma-ray emissions from Ra-226 progeny such as the Pb-214 352 keV (branching ratio 35.6%)[17] once it has grown in to equilibrium with Ra-226. Also, as the Pb-214 is an ingrowth product from the gaseous Ra-226 progeny Rn-222, the deposition of Pb-214 may be biased towards the top of the test tube, again making the near 4pi geometry of the HPGe well gamma spectrometer even more desirable. The Radium filters are loaded in test tubes filled with water to minimize any head space in the vials and futher reduce any biases this non-homogeneous deposition might create in the analyses. The samples are also counted after a fourteen-day delay to approach radio-equilibrium between Ra-226 and Pb-214. Finally, two standards are run with every batch, one standard is used as a calibration to compensate for any biases that still could be incurred from measuring Ra-226 from Pb-214 including what has already been dicussed as well as others such as summing effects from the Pb-214 gamma cascades in the highly efficient HPGe well gamma spectrometer.

Assessment of Other Potentially Interfering Isotopes

A test was performed to evaluate the affinity for the Empore Radium RAD disks of four common interfering isotopes found in SRS sludge. Twenty-milliliter aliquots of 2N nitric acid were spiked with Co-60 and Am-241, with Eu-154, and with Sr-90. The samples for each testing group were prepared in triplicate. The flow rates were adjusted to ~1 mL/second with a 50 mL 2N conditioning rinse. The test samples were passed through the Empore Radium RAD disks, and then the disks were rinsed with 150 mL of 2N nitric acid in 3 50-mL increments. The disks containing Co-60 and Am-241 and the disks containing Eu-154 were placed into petri dishes and were analyzed using coaxial high purity germanium gamma spectrometers. The disks containing Sr-90 were rolled up and inserted in liquid scintillation counting (LSC) vials containing 20 mL Perkin Elmer Ultima Gold AB liquid scintillation cocktail and analyzed with a Perkin Elmer 3750 AB liquid scintillation counter.

Reducing the Strontium Retention While Maintaining the Radium Retention

Prior test results indicated quantitative retention of Sr-90 on Empore Radium RAD disks. As bremsstrahlung radiation resulting from Sr-90 and its daughter radioisotope Y-90's beta decay would raise the gamma background continuum of the gamma spectra, the co-retention of Sr-90 and Ra-226 would be quite detrimental for Ra-226 measurement in SRS tank closure samples. Therefore, an alternative rinsing scheme would be necessary (one that significantly reduces the strontium retention) for the Empore Radium RAD disk technology to be useful for tank waste radium separations. Therefore a further review of the literature was conducted to determine what options were available for reducing the strontium retention during Empore Radium RAD disk use. A thesis focusing on a different Superlig extractant, Superlig 620, addressed using citrates to separate strontium from barium.[16] A test evaluating citrates to separate Sr from Ra on the Empore Radium RAD disk (suspected to be Superlig 640) was executed. Two 20-mL aliquots of 2N nitric acid were spiked with Ra-226, and two were spiked with Sr-90. The samples were filtered through Empore Radium RAD disks that had been conditioned with 50 mL of 2N nitric acid. The filters were rinsed with 150 mL of 2N nitric acid. Each filter was then rinsed with 50 mL of 0.5M sodium citrate. The Sr-90 spiked sample disks were placed in LSC cocktail and analyzed by LSC after a period of 10 days (to allow for in-growth of the Y-90 daughter). The disks spiked with Ra-226 were placed in test tubes and analyzed by gamma spectrometry.

The citrate wash step showed promise in selectively reducing Sr-90 relative to Ra-226, so a sodium citrate wash was evaluated with two sets of real waste samples spiked with radium tracers. Radium tracer was added to 2 mL of tank sample. The sample was acidified, resulting in a 38 mL solution of ~2N nitric acid. Two blank samples spiked with radium were also analyzed. The samples were subjected to a cesium-removal process, with each being treated with a batch addition of ~0.2 g Bio-Rad AMP, contacted for 30 seconds, and then filtered. The filtrates were then passed through Empore Radium RAD disks that had been conditioned with 50 mL of 2N nitric acid. The disks were rinsed with 150 mL of 2N nitric acid. Then, the disks were rinsed with 100 mL of 0.1M sodium citrate. The disks were then loaded into 2-mL test tubes and assayed using a robotic high purity germanium (HPGe) well gamma spectrometer.

Results were promising, so a more detailed study evaluating a range of concentrations of sodium citrate wash steps was conducted. Six concentrations (0.1M, 0.09M, 0.08M, 0.07M, 0.06M, and 0.05M) of citrate were evaluated. One sample for each concentration was spiked with Sr-90 and one was spiked with Ra-226. Samples of 20 mL 2N nitric acid were generated and spiked with the appropriate isotope. Empore Radium RAD disks were prepared for each sample and were conditioned with 50 mL of 2N nitric acid. The samples were added to their respective filters. The sample filters were rinsed with 150 mL of 2N nitric acid. Each disk was then rinsed with 100 mL of the appropriate concentration of sodium citrate wash. The Sr-90 disks were analyzed using LSC, while the Ra-226 disks were analyzed using gamma spectrometry.

The Sr-90 retention appeared relatively consistent for the range of the citrate concentrations, while the Ra-226 retention appeared to increase as the wash concentration decreased. Therefore, the low end of the citrate waste concentration range was chosen for real waste tests. A 0.05M sodium citrate wash was evaluated with another set of real waste samples spiked with radium tracers. Radium tracer was added to 2 mL of tank sample. The sample was acidified, resulting in a 38 mL solution of 2N nitric acid. Two blank samples spiked with radium were also analyzed. The samples were subjected to a cesium-removal process, each treated with a batch addition of ~0.2 g Bio-Rad AMP, contacted for 30 seconds, and then filtered. The filtrate was then passed through an Empore Radium RAD disk that had been conditioned with 50 mL 2N nitric acid. The disks were rinsed with 150 mL 2N nitric acid. Finally, the disks were rinsed with 100 mL 0.05M sodium citrate. The disks were then loaded into 2-mL test tubes and assayed on a robotic HPGe well gamma spectrometer.

Results and discussion

Testing of Empore Radium RAD Disks on SRS Supernatant Samples and Spikes

Radium recoveries for the two SRS tank waste samples were 87% and 95%, while radium recoveries for the two acid blanks were 84% and 87%. The similarity between the recoveries for the real waste samples and the acid blanks suggests that the additional

constituents in the real waste samples did not reduce the radium recovery. Following the cesium-removal step, the only gamma-emitting isotopes measurable in the supernate were Sn-126 and its daughter radioisotope Sb-126. The Empore Radium RAD disks had no measurable affinity for Sn-126 or Sb-126, as measurements indicated that the Empore Radium RAD disk retentions for the pair were <0.62% and <0.61%. This was encouraging, as it confirmed that the interference from the second most abundant gamma emitters present in SRS supernate matrices are removed using the Empore Rad disk methodology.

Assessment of Other Potentially Interfering Isotopes

The test results evaluating the affinity for Empore Radium RAD disks of four common interfering isotopes found in SRS sludge matrices are as follows. Retention for the cobalt, europium, and americium isotopes were all minimal (the Co-60 recoveries were <0.09%, <0.05%, and <0.05%; the Eu-154 recoveries were 0.1%, 0.08%, and 0.004%; and the Am-241 recoveries were <0.08%, <0.02%, and <0.04%). In contrast, the Sr-90 recoveries were quantitative, with measurements of 102%, 105%, and 105%. These high Sr-90 recoveries indicated that an alternative rinsing scheme (one significantly reducing the retention of strontium) would be necessary for the Empore Radium RAD disk technology to be useful for tank waste radium analyses.

Reducing the Strontium Retention While Maintaining the Radium Retention

The test results evaluating the effect of 50 mL 0.5M sodium citrate washes of the Empore Radium RAD disk on Sr-90 and Ra-226 retention follow. On spiked acid aliquots, the 0.5M sodium citrate wash reduced the retention of Sr-90 by a factor of 61 (to slightly less than 2%), while only reducing the retention for Ra-226 by a factor of 2.9 (to ~30%).

On spiked tank waste samples which had been subjected to cesium removal, then passed through the Empore Radium RAD disk, rinsed with the 100mL 0.1M sodium citrate rinse, and then analyzed, the radium recoveries were: a) 39%, 52%, and 52% for the first sample; and b) 46%, 52%, and 63% for the second sample. Under these conditions, on average, the radium recovery was ~50%. On spiked acid blanks that had been subjected

to cesium removal, then passed through the Empore Radium RAD disk, rinsed with the 100mL 0.1M sodium citrate rinse, and then analyzed, the radium recoveries were: a) 62% and 44% for the first set; and b) 43% and 40% for the second set. Under these conditions, on average, the radium recovery was ~47%, which is approximately equal to that seen for the real waste samples.

For Empore Radium RAD disks analyzed following rinses with 0.10M, 0.09M, 0.08M, 0.07M, 0.06M, and 0.05M sodium citrate: a) the Sr-90 recoveries from the spiked acid blanks were 1.6%, 0.9%, 1.8%, 0.3%, 1.9%, and 2.0% respectively; and b) the Ra-226 recoveries from the spiked acid blanks were 43%, 68%, 56%, 95%, 94%, and 72%, respectively. Given that the Sr-90 recoveries were relatively low ($\leq$ 2%) across the 0.05-0.10M sodium citrate concentration range, while the Ra-226 recoveries generally appeared to increase as the citrate concentration dropped, it was thought that use of a 0.05M sodium citrate rinse offered the greatest likely benefit for separating Ra from Sr. A final set of real waste tests was performed under this condition.

Using a 100mL 0.05M sodium citrate rinse with spiked samples being processed through cesium removal and the Empore Radium RAD disk, the Ra-226 recoveries were: a) 95%, 90%, and 86% for the three real waste samples; and b) 83% and 85% for the two spiked blanks. Such recoveries were significantly higher than those obtained using the previous radium method (the cation exchange method) and sufficiently high to justify the pursuit of the new separation method for future SRS Tank Closure Ra-226 analyses.

Current Ra-226 Radiochemical Analyses

The Empore Radium RAD disk methodology has been utilized on twenty-nine samples of SRS Liquid Waste. A flow sheet illustrating the current Ra-226 methodology is provided in Figure 3. Twenty-one samples were from an SRS waste tank that receives SRS supernate that has been chemically treated with a Calixarene treatment[18] to reduce Cs-137 levels by several orders of magnitude from typical supernate levels. These samples are predominated by Cs-137 (nominally 3E4 Bq/mL) followed by Sr-90/Y-90 (nominally 2E3 Bq/mL). These samples were analyzed using the flow sheet of Figure 3, the only resin treatment used was one Bio-Rad AMP treatment to lower the Cs-137 levels prior to

the radium extraction. The thirty samples represent 10 analytical batches analyzed in triplicate over a 2-year period. Each sample is analyzed in duplicate, with one of the duplicates spiked with Ra-226 and analyzed as a matrix spike. The recovery of the matrix spike is applied the results from the non-spiked sample to determine the Ra-226 concentration. Table 1 contains the data from the triplicate matrix spike samples. The performance of the separation is quite consistent across the triplet runs, with an average spike recovery standard deviation of 4.38% among the triplicates. Two blank spikes are also analyzed with each set, one serving as the matrix spike for the second. For quality control purposes, the recovery of the corrected blank spike result must be 75% - 125%.

Due to the characteristics of the current inventory, the Ra-226 method continues to prove the negative (no quantifiable Ra-226). Across this dataset, with a 2mL sample aliquot, and with counting times of 4 hours on the 14 day counts, the method has an average Ra-226 detection limit (Based on the Curie MDA[19]) of ~ 9.1E-02 Bq/mL.

Four of the samples analyzed were SRS Waste Tank supernate from waste tanks prior to Cs-137 removal. These samples are nominally 5E+06 Bq/mL Cs-137. The samples were analyzed using the protocols outlined in Figure 3, with no Sr-90 removal stage but with two Cs-Removal stages using BioRad AMP. The samples averaged a matrix spike recovery of 99.6% and averaged a blank spike recovery of 101%. Detection limits averaged 4.1E-01 Bq/mL.

The third waste matrix tested was by far the most challenging: four samples of high level waste sludge being prepared for a vitrification treatment to be stabilized for long-term storage as glass. Aliquots of these samples were digested in the Shielded Cells facility using a sodium peroxide fusion technique. Prior to this method development, extensive radiochemistry was required in the Shielded Cells prior to completing the radiochemistry preparations in the radiological hoods. Due to the high yields from this method, small aliquots of dissolved solids were able to be removed from the Shielded Cells and analyzed as outlined in Figure 3, including the Sr-90 reduction and one BioRad AMP Cs-137 extraction. The average Ra-226 matrix spike recovery was 76.1% with a percent standard deviation of 4.71%. The blank spike recovery was 95.8%. The average Ra-226

detection limit was 1.07E+01 Bq/g. The most concentrated radionuclides in these samples were Sr-90/Y-90 (3.9E+07 Bg/g), Cs-137 (4.7E+06 Bq/g), Pu-238 (7.7E+05 Bq/g), Am-241 (1.03E+05 Bq/g), Pu-239/240 (4.0E+04 Bq/g), Eu-154 (3.4E+04 Bq/g), and Co-60 (2.7E+03 Bq/g). As mentioned previously, in the cation exchange based methodology used previously, Co-60 co-extracted with Ra-226, raising analysis backgrounds. With the Empore Radium RAD disk methodology, Co-60 is no longer an interference. With the new methodology, on this problem matrix, chemical yields have risen substantially, method detection limits have improved, and expensive and time-consuming shielded cells operations have been eliminated.

Conclusion

The radium separation scheme that has been developed using the Empore Radium RAD disks has increased the yields of radium by approximately an order of magnitude compared to what was previously being obtained using a cation exchange approach. This increase in yield will allow radiochemistry to be conducted on smaller aliquots of SRS Liquid Waste sample matrices. The use of smaller aliquots of sample allows for the radiochemistry to be conducted entirely in the radiological hoods of the analytical laboratories, as opposed to performing the initial decontamination steps in the Shielded Cells facility. This reduces the costs and time required for Ra-226 analyses on high activity matrices.

340 **Table 1** Ra-226 Spike Recoveries of SRS Tank 50 Triplicate Analyses

Run Date	Average Ra-226 Matrix Spike Recovery of Triplicates	Standard Deviation of Spike Recoveries	Blank Spike Ra-226 Recovery
8/7/2018	74.0%	7.91%	81.1%
2/26/2018	112%	2.55%	101%
7/3/2017	91.8%	4.39%	97.7%
4/25/2017	85.2%	4.71%	122%
1/31/2017	95.4%	3.75%	88.7%
10/20/2016	99.3%	3.50%	107%
7/15/2016	92.8%	3.87%	88.6%
Average	93.0%	4.38%	98.0%

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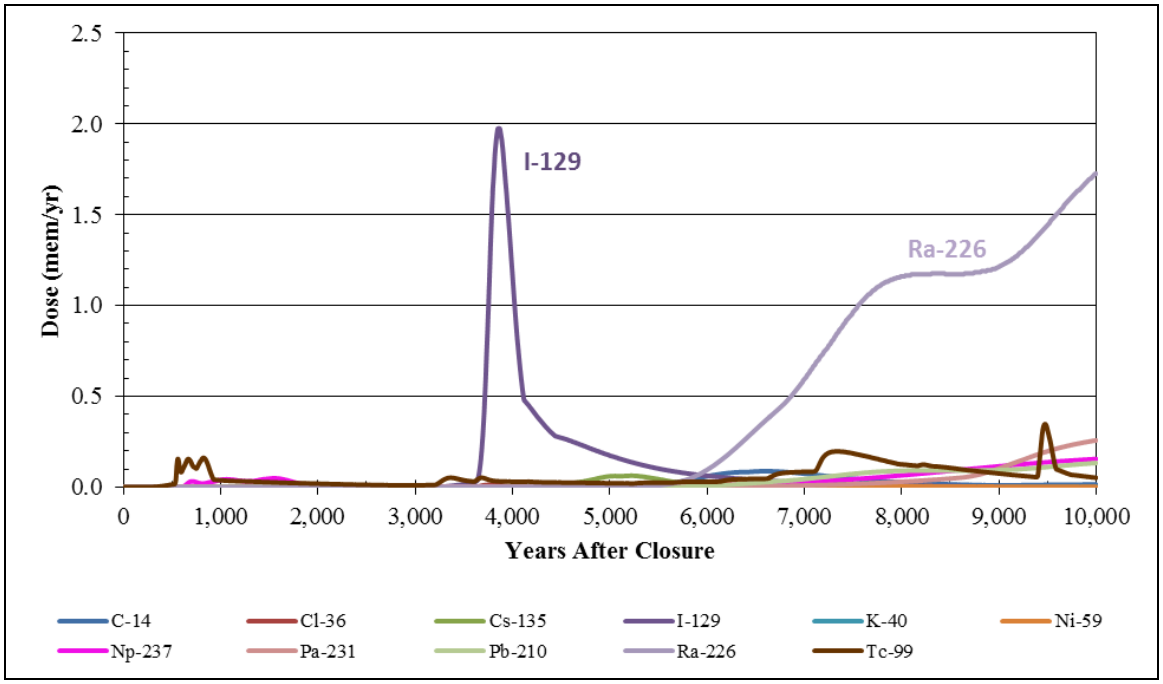
343 **Figure 1** Projected Groundwater Doses Identified in SRS H-TF SA (Sector C)[2]



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346 **Figure 2** Projected Groundwater Doses Identified in SRS F-TF PA (Sector E)[3]

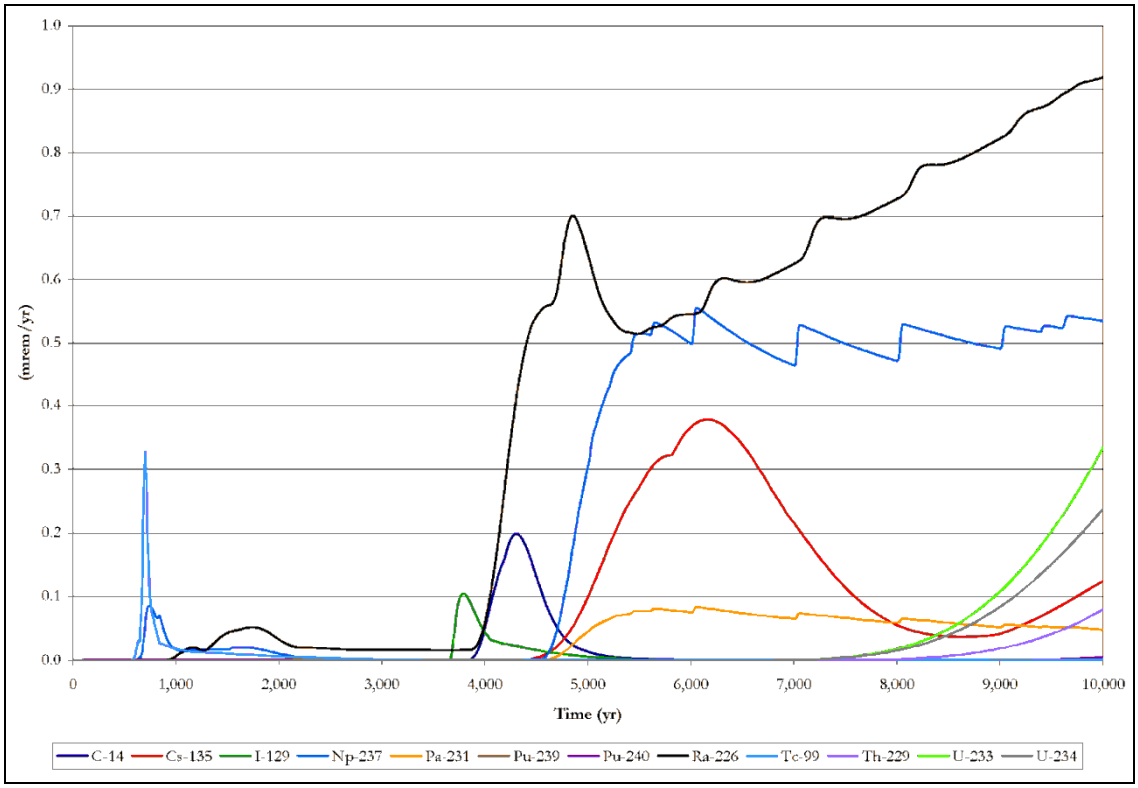

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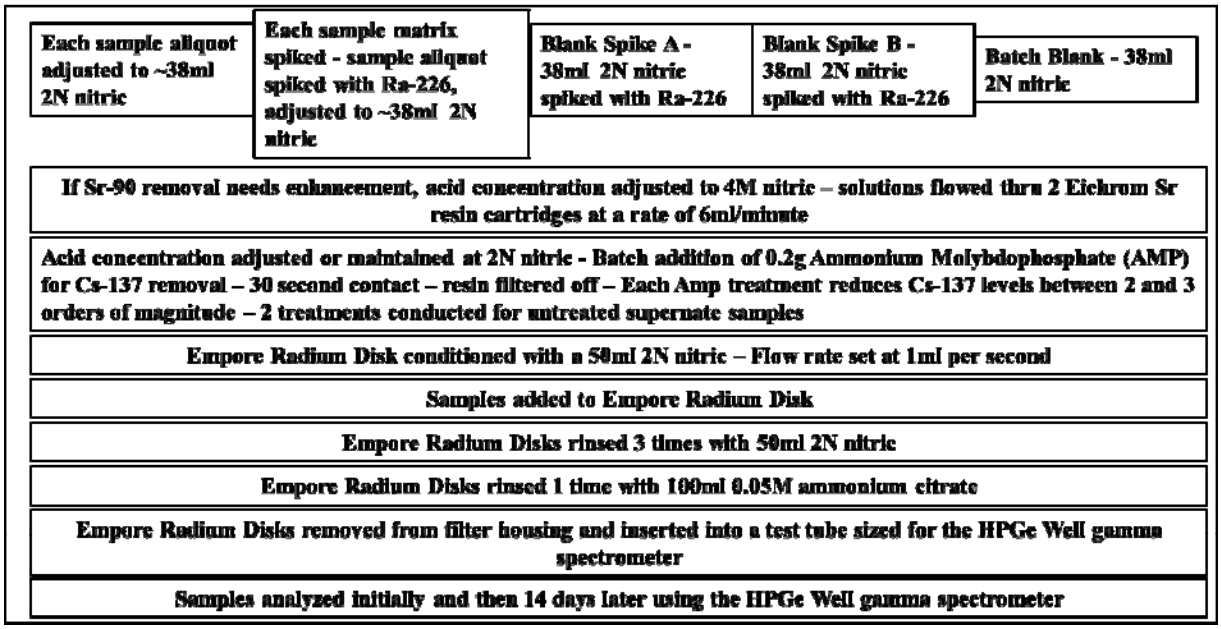
Figure 3. Radium-226 Separation and Analysis Flowsheet



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