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Determination of 237Np in Bioassay Fecal Samples

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Determination of ^{237}Np in Bioassay Fecal Samples

A Project

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Dedicated to

My Family and Friends

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Abstract

^{237}Np, a long-lived alpha-emitting actinide, is present in the environment due to releases from atmospheric nuclear weapons tests, reactor accidents, and nuclear reprocessing plants. The determination of long-lived actinides, such as neptunium, is essential for worker monitoring and environmental control. In bioassay and environmental samples, a radiochemical separation is necessary before measurement in order to eliminate any inferences that may occur in a mixture of actinides. In this paper, a method for determining ^{237}Np in fecal samples using TEVA resin is investigated. Using ^{239}Pu as a tracer, tetravalent plutonium and neptunium were successfully separated from other actinides. Samples were electrodeposited and counted using an alpha-spectrometer. Utilizing a stripping solution containing 0.1M HCl, 0.01M HF, and 0.04M Rongalite, average percent recovery for ^{239}Pu was 89.1% and 80.3% for ^{237}Np. This method eliminates radioactive waste accumulated when using ^{241}Am/^{239}Np as a tracer and will not retain a common interfering actinide, uranium. The proposed method is very promising; however, it should be analyzed in more detail before adoption in any bioassay laboratory.

Introduction

Neptunium was first discovered in 1940 when McMillan and Abelson bombarded a thin uranium foil with low-energy neutrons. This bombardment formed ^{239}Np with a half-life of 2.36 days. In 1942, the long-living alpha-emitting radionuclide ^{237}Np was discovered by Wahl and Seaborg.[1] With a half-life of 2.14×10^6 years, ^{237}Np was produced in large quantities from nuclear reactions fueled with natural uranium.[1-2] ^{237}Np and a number of other radionuclides are present in the environment as a result of releases from atmospheric nuclear weapons tests, reactor accidents, and nuclear reprocessing plants.[3-5] Neptunium and other actinides were also

introduced into the environment from the incident at Chernobyl.[6] The activity of ^{237}Np is slowly increasing in the environment due to the decay of its parent isotope, ^{241}Am. In approximately 200,000 years, ^{237}Np will be the dominating transuranium nuclide in the environment.[7]

Neptunium exists in the environment in several oxidation states; Np(III), Np(IV), Np(V), and Np(VI). The most stable oxidation state of neptunium at room temperature is Np(V) [5]. It is often present as NpO_2^+ and called the neptunyl ion.[5] This ion is highly soluble and makes Np more mobile in the environment than other actinides, such as plutonium and americium.[7]

Actinides and other radionuclides are often monitored in the environment, contamination incidences, nuclear site closures, workers in nuclear fuel cycle facilities, research labs, and government agencies.[9-10] Monitoring radionuclides is essential due to the products produced by research projects, routine operation, and waste processing and storage.[10] The determination of long-lived actinides, such as neptunium, plutonium, thorium, and uranium, is significant for worker monitoring, environmental control, and waste management.[11] Bioassay laboratories will analyze urine and/or fecal samples from workers to determine radionuclide content. Urinalysis is often inadequate in determining the total body content of internally deposited radionuclides. The most accurate measurements of radiological data occur from fecal analysis during the early stages of elimination. However, total fecal dissolution is essential and fecal analysis can be difficult due to the presence of insoluble materials.[9]

Various anion exchange resins are typically used in the analysis of numerous actinides. A different type of resin called TEVA is used to separate tetravalent actinides from all other actinides in a sample over a wide range of acidities. TEVA's active component is a quaternary amine that is a mixture of trioctyl and tridecyl methyammonium nitrate (or chloride). This resin has similar properties as that of typical strong base anion exchange resins, but its functional

groups are in a liquid form adding to greater flexibility to coordinate around target anions. In addition, TEVA resin will not retain many commonly encountered cations.[12]

Actinide separation from a variety of matrices should be highly effective when using TEVA resin. Horwitz et al investigated the effectiveness of TEVA resin in simulated waste solutions and found that ^{237}Np recovery was 96%.[12] Recovery of ^{237}Np in synthetic solution was approximately 90%.[11] Maxwell et al illustrated that ^{237}Np and Pu isotopes spiked in water samples had good yields using TEVA resin.[3] The activity of ^{237}Np in peat and lichen in Finland was investigated by Salminen et al. The average recovery of tracer ^{235}Np was 79% in peat and lichen samples.[7] In Poland forest liter, the mean recovery of ^{237}Np was 62.6%.[6] Diodati and Sartori used alpha-spectrometry and TEVA resin to illustrate that neptunium was clearly separated from all other actinides in effluent and low-level nuclear waste samples.[8] TEVA resin has also been used in the determination of actinides in fish and other animal tissue samples. Maxwell et al showed an average recovery of 99.8% for ^{236}Pu tracer and 90% for ^{237}Np matrix spike.[13] Maxwell et al also showed that TEVA resin can be used in the determination of actinides in emergency urine and water samples.[14]

When analyzing for ^{237}Np, it is often hard to find a suitable alpha-emitter tracer.[11] ^{235}Np has been used as a tracer, but can only be detected by x-ray spectrometry or liquid scintillation counting. ^{239}Np is a commonly used tracer, but has a short half-life and can only be detected by gamma-spectrometry.[7] ^{242}Pu can be used as a tracer for ^{237}Np when analyzing by ICP-MS but its energy is not fully resolved from ^{237}Np when analyzed by alpha-spectrometry.[5,15]

The goal of this work was to develop a method to determine ^{237}Np in fecal samples using ^{239}Pu as a tracer. In this proposed method, TEVA resin was used to separate tetravalent neptunium and plutonium from other actinides. This method will eliminate radioactive waste

accumulated from the use of ^{239}Np /^{243}Am as a tracer, the need for gamma-spectrometry, and the interference of ^{238}U in alpha-spectrometry in the determination of ^{237}Np.

Materials and Methods

Reagents:

Hydrochloric, hydrofluoric, and nitric acids were prepared using reagent-grade acids (Fisher Scientific, Inc.). All other materials used were ACS reagent grade (Fisher Scientific, Inc.). Type I deionized water was acquired from a NANOpure water system. The resin used in this procedure was TEVA Resin® (Aliquat ™336), available from Eichrom Technologies, Inc., (Darien, Illinois, USA). Radionuclide tracers & spikes ^{239}Np /^{243}Am, ^{239}Pu, ^{237}Np, ^{232}U, ^{244}Cm, ^{236}U were obtained from Isotope Products Laboratories (Valencia, California, USA). Radionuclide tracer & spike dilutions were as follows: ^{239}Np /^{243}Am 0.870 dpm/mL; ^{239}Pu 5.16 dpm/mL; ^{237}Np tracer 52.70 dpm/mL; ^{232}U 8.507 dpm/mL. Actinide spike was diluted and the activity of each radionuclide was as follows: ^{239}Pu 4.91 dpm/mL, ^{237}Np 5.47 dpm/mL, ^{244}Cm 2.93 dpm/mL, ^{236}U 4.18 dpm/mL.

Methodology:

Column preparation:

TEVA resin columns were obtained from Eichrom Technologies, Inc. Each column was a cartridge containing 2mL of small particle size (50-100µm) resin. The cartridges were used with a vacuum extraction system (Eichrom Technologies, Inc.) and flow rates of approximately 1-2 drops/second. The vacuum extraction system consisted of a polycarbonate vacuum box with 12 column positions, a polycarbonate liner to collect rinses, a rack to hold 50mL centrifuge tubes, and a vacuum pump.

Sample Preparation:

Prior to sample analysis, each fecal sample was ashed, digested to remove organic matter, and dissolved in 3M HCl. An aliquot of the fecal sample (0.1 times the net sample weight) is transferred into a 400mL beaker, appropriate tracers added, and gently heated on a hotplate (covered) to dryness. A few milliliters of concentrated HCl is added to the dry sample, covered, and heated to dryness. Each sample is redissolved in 5 mL of 2M $Al(NO_3)_3$ and 5 mL of 6M HNO_3. The 2M $Al(NO_3)_3$ was purified to remove any uranium by passing the solution through a column containing anion exchange resin AG1-X8, 100-200 mesh (Eichrom Technologies, Inc.). The presence of Al^{3+} complexes any oxalates, sulfates, phosphates, or fluorides present that may reduce actinide absorption. Np is reduced from a valance state of Np^{5+} to Np^{4+} thru the addition 2mL of Fe/Ascorbic solution. [12] After fifteen minutes, 1mL of 3.5M $NaNO_2$ is mixed well into the sample to ensure that Pu is in its tetravalent state.[4]

There were three different types of samples used. The first type were load solution samples which consisted of 5 mL of 2M $Al(NO_3)_3$ and 5 mL of 6M HNO_3 with the appropriate tracer/spike. 2mL of Fe/Ascorbic solution and 1mL of 3.5M $NaNO_2$ were added to these samples before loaded on to the column. The second type were composite fecal matrix samples, which consisted of a composite of older fecal samples from prior analyses in the laboratory. The third type were individual fecal samples received by the bioassay laboratory. Both composite fecal matrix and fecal samples were treated as stated in the above paragraph.

Column Separation:

TEVA resin cartridges on a vacuum box were preconditioned with 5mL of 3M HNO_3 at a rate of ~2 drops/second. Once preconditioned, the treated fecal sample was loaded onto the column at a rate of ~1 drop/second. The sample beaker was rinsed with 5mL 3M HNO_3 and

added to the column to ensure the entire sample had been introduced. A column rinse of 30mL 3M HNO_3 was added to remove most matrix components, such as uranium (flow rate ~2 drops/second). The column was then rinsed with 20mL of 9M HCl to remove any potential thorium present.[12] A column rinse of 5 mL 6M HCl was added to remove any residual matrix interferences. Neptunium and plutonium were eluted and collected in a 50mL centrifuge tube with 20mL a solution containing 0.1M HCl, 0.01M HF, and 0.04M $HOCH_2SO_2Na \cdot 2H_2O$ (Rongalite) at a flow rate of ~0.5-1 drop/second. Note that some samples in method development stages used a stripping solution of 10 or 20mL 0.1M HCl and 0.01M HF.

Electrodeposition:

The purified fraction containing neptunium and plutonium was evaporated to dryness in a 50mL beaker. (If the stripping solution did not contain rongalite, skip the ashing steps, and redissolve in 0.1M $NH_4HC_2O_4$ once dry.). Once dry, 3-5mL of concentrated HNO_3 and a few drops of 30 wt% H_2O_2 were added to the beaker and evaporated to dryness. This step was repeated 2-3 more times to ensure that the rongalite was destroyed (ashing more than five times may have an adverse affect on electroplating). If the rongalite is not destroyed, it will collect on the disk during electrodeposition and cause interference. The sample was redissolved in 15mL of 0.1M $NH_4HC_2O_4$ by bringing it to a quick boil. The sample was then transferred to a prepared plating cell and 2.5mL of 5M NH_4Cl was added to the cell. The sample was electrodeposited for 2.5 hours at 13-15V onto a 19mm diameter, flat, polished stainless steel disk using the disk as the cathode and a platinum wire loop as the anode. Electrodeposition was then terminated by adding 2mL of 7M NH_4OH to the cell and waiting one minute before disassembling the cell. The disk was rinsed with water, ethyl alcohol, dried, and labeled before alpha spectrometric counting.

Sample Measurements:

Pu and Np measurements were performed using a EG&G ORTEC Octête-Plus Alpha-spectrometer containing passivated implanted planar silicone detectors. The alpha-spectrometric sample count time was 3,000 minutes. Samples spiked with ^{239}Np /^{243}Am were gamma counted immediately in a liquid scintillation vial using an Ortec Germanium Detector connected to an Ortec DSPEC Plus Digital Gamma-ray Spectrometer. Samples were counted for 60,000 seconds.

Results and Discussion

Actinide Retention:

When the load solution was spiked with ^{239}Np /^{243}Am, ^{239}Np had an average recovery of 96.3% while there was no trace of ^{243}Am. Thus indicating the method does successfully separate americium from neptunium. Another set of load solution samples were spiked with ^{232}U. As seen in Figure 1, the recovery of ^{232}U was less than 1% demonstrating that uranium is not retained on TEVA resin. When the Actinide mixture (^{244}Cm, ^{236}U, ^{239}Pu, ^{237}Np) was used to spike the load solution, only ^{239}Pu and ^{237}Np were recovered (Figure 2). Therefore, TEVA will retain only tetravalent plutonium and neptunium from a mixture containing numerous actinides using the proposed method.

^{237}Np Analysis:

In the load solution, the average ^{239}Pu recovery was 85.1% and the average ^{237}Np recovery was 88.5%. However, in the composite fecal matrix the average recoveries were a bit lower. ^{239}Pu had an average recovery of 60.4% and ^{237}Np had an average recovery of 46.6% (Table 1, 2). The difference in ^{239}Pu and ^{237}Np recovery between the load solution and the composite fecal matrix was due to either a matrix interference or both isotopes were still retained

on the column. Horwitz et al noted that matrix constituents can have an effect on neptunium sorption on TEVA resin, especially high levels of thorium and uranium.[12] A second 10mL fraction of the stripping solution, 0.1M HCl/0.01M HF, was collected from the composite fecal matrix. In Figure 3, the ^{239}Pu and ^{237}Np peaks are clearly visible from the first 10mL 0.1M HCl/0.01M HF fraction collected. Yet in the second 10mL 0.1M HCl/0.01M HF fraction, two smaller peaks appear for both ^{239}Pu and ^{237}Np (Figure 4). The stripping solution was then increased to 20mL 0.1M HCl/0.01M HF and composite fecal matrix were run again. The average recoveries were 59.0% for ^{239}Pu and 58.0% for ^{239}Np. The average percent recovery for ^{239}Pu stayed fairly consistent, while the average percent recovery of ^{239}Np increased slightly by using 20mL of the stripping solution. In order to increase the recovery of both plutonium and neptunium, 0.04M Rongalite, a reducing reagent, was added to the stripping solution. Rongalite was noted as a good reductant to use when stripping plutonium and neptunium from TEVA resin by Maxwell et al.[14] With rongalite added, the average percent recoveries increased to 89.1% for ^{239}Pu and 80.3% for ^{237}Np in composite fecal matrix (Table 1, 2). The recovery of the ^{239}Pu tracer in fecal samples was investigated to determine if a recovery of 90% was achievable. After analyzing 30 different fecal samples, the average percent recovery of ^{239}Pu was 78.8% (Table 1). The recoveries of ^{239}Pu varied from 99.7% to 39.8% in fecal samples. This wide range was more than likely due to the presence of various different fecal matrix interferences.

Conclusion

Tetravalent plutonium and neptunium can be successfully separated through the use of TEVA resin. Although the regions of intrests are close, percent recoveries were near 90% for both ^{239}Pu and ^{237}Np in a composite fecal matrix using rongalite in the stripping solution. This method has numerous advantages over methods currently being used. The separation of

plutonium and neptunium can be achieved using dilute HNO_3, plus uranium no longer appears in the alpha-spectrum as it does with the use of anion exchange resin. Additionally, by using ^{239}Pu as a tracer for ^{237}Np, there is less radioactive waste generation (^{241}Am) and there is no need for gamma counting (^{239}Np). The proposed method for determining ^{237}Np in fecal samples appears very promising. However, this method should be analyzed in more detail before adoption in any bioassay laboratory.

Future Studies

The proposed procedure should be further investigated using numerous fecal samples to determine if it is rugged enough to withstand various matrix interferences. In addition, the use of ^{238}Pu as a tracer for Np determination should be studied since its region of interest is further from ^{237}Np than ^{239}Pu.

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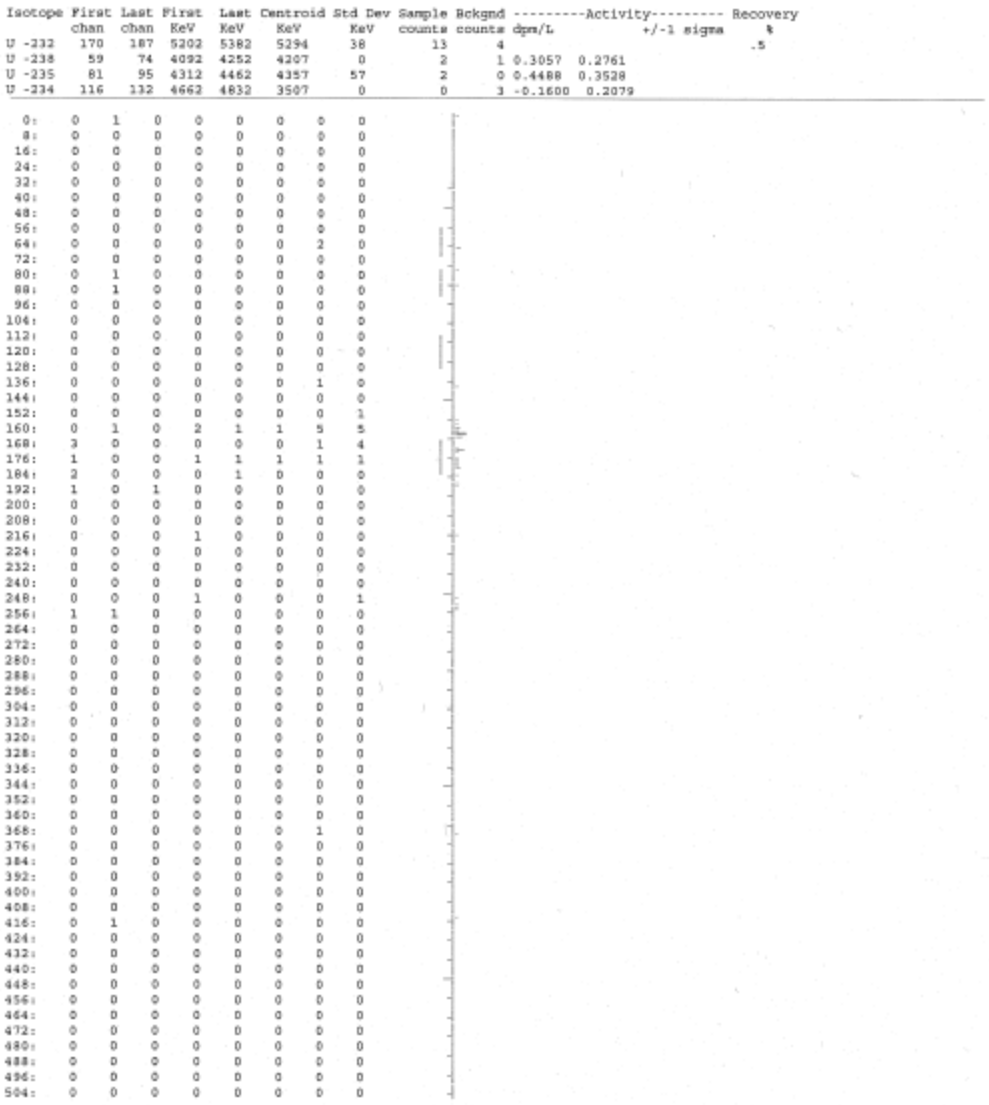
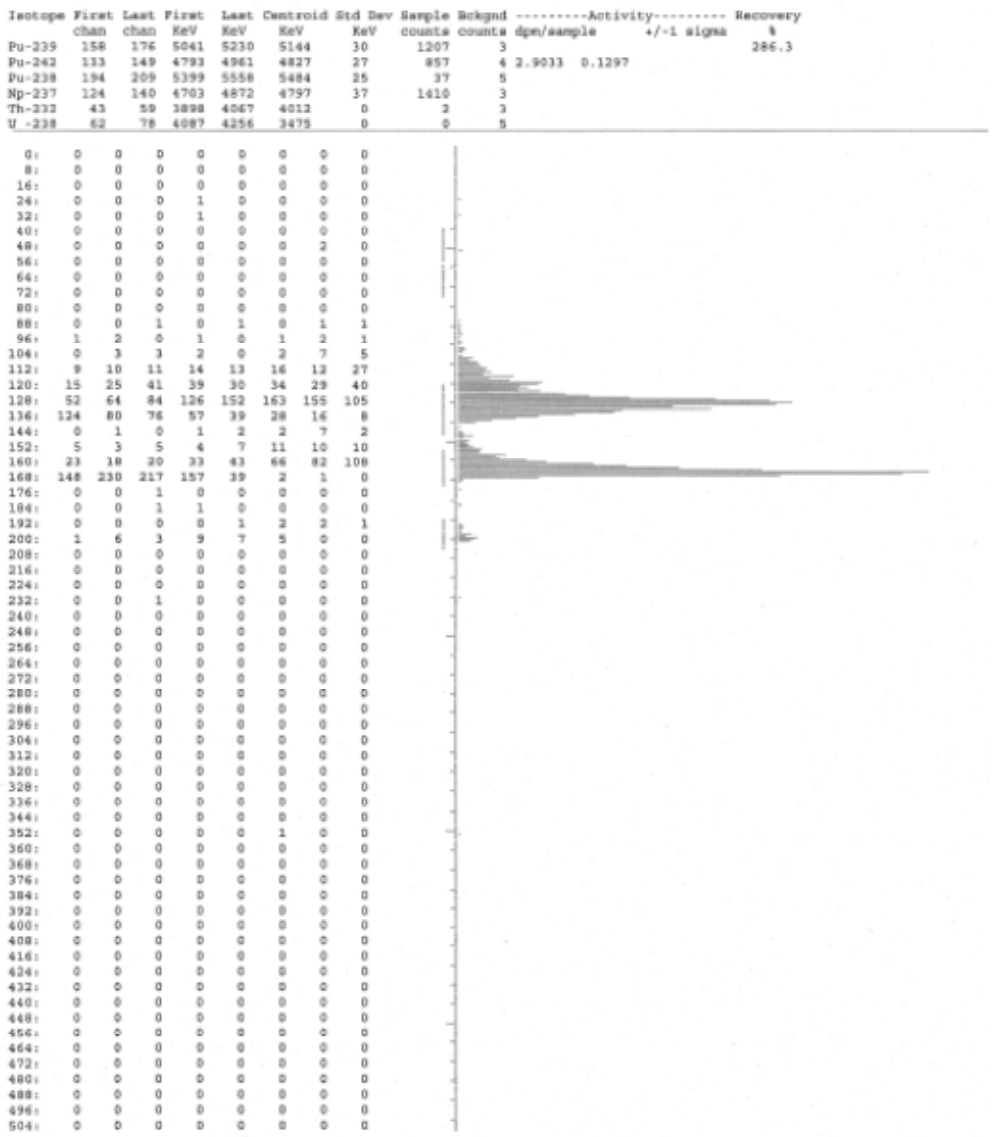


Isotope	First chan	Last chan	First KeV	Last KeV	Centroid KeV	Std Dev KeV	Sample counts	Bckgnd counts	Activity dpm/L	+/-1 sigma	Recovery %
U -232	170	187	5202	5382	5294	38	13	4			.5
U -238	59	74	4092	4252	4207	0	2	1	0.3057	0.2761	
U -235	81	95	4312	4462	4357	57	2	0	0.4488	0.3528	
U -234	116	132	4662	4832	3507	0	0	3	-0.1600	0.2079	

0:	0	1	0	0	0	0	0	0
8:	0	0	0	0	0	0	0	0
16:	0	0	0	0	0	0	0	0
24:	0	0	0	0	0	0	0	0
32:	0	0	0	0	0	0	0	0
40:	0	0	0	0	0	0	0	0
48:	0	0	0	0	0	0	0	0
56:	0	0	0	0	0	0	0	0
64:	0	0	0	0	0	0	2	0
72:	0	0	0	0	0	0	0	0
80:	0	1	0	0	0	0	0	0
88:	0	1	0	0	0	0	0	0
96:	0	0	0	0	0	0	0	0
104:	0	0	0	0	0	0	0	0
112:	0	0	0	0	0	0	0	0
120:	0	0	0	0	0	0	0	0
128:	0	0	0	0	0	0	0	0
136:	0	0	0	0	0	0	1	0
144:	0	0	0	0	0	0	0	0
152:	0	0	0	0	0	0	0	1
160:	0	1	0	2	1	1	5	5
168:	3	0	0	0	0	0	1	4
176:	1	0	0	1	1	1	1	1
184:	2	0	0	0	1	0	0	0
192:	1	0	1	0	0	0	0	0
200:	0	0	0	0	0	0	0	0
208:	0	0	0	0	0	0	0	0
216:	0	0	0	1	0	0	0	0
224:	0	0	0	0	0	0	0	0
232:	0	0	0	0	0	0	0	0
240:	0	0	0	0	0	0	0	0
248:	0	0	0	1	0	0	0	1
256:	1	1	0	0	0	0	0	0
264:	0	0	0	0	0	0	0	0
272:	0	0	0	0	0	0	0	0
280:	0	0	0	0	0	0	0	0
288:	0	0	0	0	0	0	0	0
296:	0	0	0	0	0	0	0	0
304:	0	0	0	0	0	0	0	0
312:	0	0	0	0	0	0	0	0
320:	0	0	0	0	0	0	0	0
328:	0	0	0	0	0	0	0	0
336:	0	0	0	0	0	0	0	0
344:	0	0	0	0	0	0	0	0
352:	0	0	0	0	0	0	0	0
360:	0	0	0	0	0	0	0	0
368:	0	0	0	0	0	0	1	0
376:	0	0	0	0	0	0	0	0
384:	0	0	0	0	0	0	0	0
392:	0	0	0	0	0	0	0	0
400:	0	0	0	0	0	0	0	0
408:	0	0	0	0	0	0	0	0
416:	0	1	0	0	0	0	0	0
424:	0	0	0	0	0	0	0	0
432:	0	0	0	0	0	0	0	0
440:	0	0	0	0	0	0	0	0
448:	0	0	0	0	0	0	0	0
456:	0	0	0	0	0	0	0	0
464:	0	0	0	0	0	0	0	0
472:	0	0	0	0	0	0	0	0
480:	0	0	0	0	0	0	0	0
488:	0	0	0	0	0	0	0	0
496:	0	0	0	0	0	0	0	0
504:	0	0	0	0	0	0	0	0

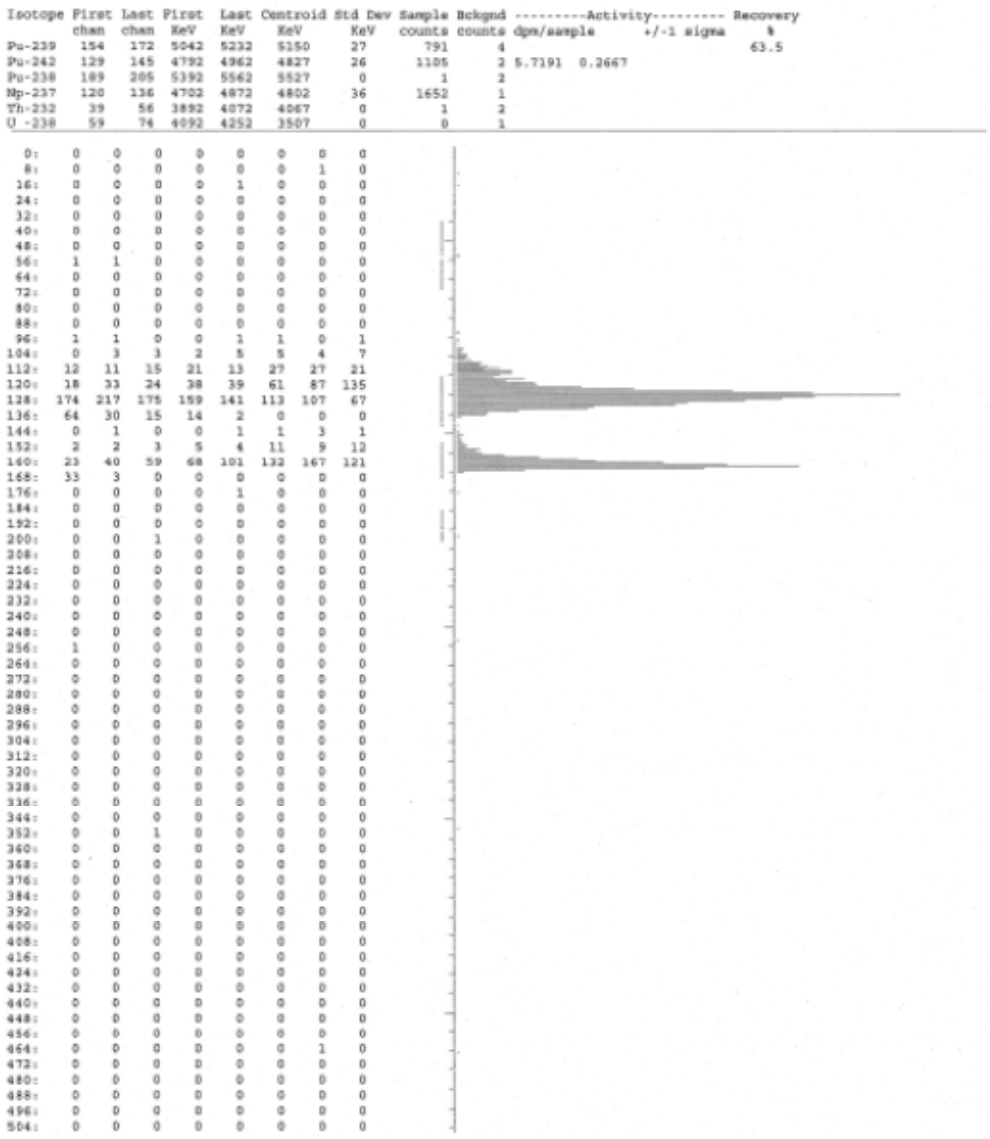
Figure 1: Alpha-spectrum of ^{232}U spike in load solution.



Isotope	First chan	Last chan	First KeV	Last KeV	Centroid KeV	Std Dev KeV	Sample counts	Bckgnd counts	Activity dpm/sample	+/-1 sigma	Recovery %
Pu-239	158	176	5041	5230	5144	30	1207	3			286.3
Pu-242	133	149	4793	4961	4827	27	857	4	2.9033	0.1297	
Pu-238	194	209	5399	5558	5484	25	37	5			
Np-237	124	140	4703	4872	4797	37	1410	3			
Th-232	43	59	3898	4067	4013	0	2	3			
U -238	62	78	4087	4256	3475	0	0	5			

0:	0	0	0	0	0	0	0	0
8:	0	0	0	0	0	0	0	0
16:	0	0	0	0	0	0	0	0
24:	0	0	0	1	0	0	0	0
32:	0	0	0	1	0	0	0	0
40:	0	0	0	0	0	0	0	0
48:	0	0	0	0	0	0	2	0
56:	0	0	0	0	0	0	0	0
64:	0	0	0	0	0	0	0	0
72:	0	0	0	0	0	0	0	0
80:	0	0	0	0	0	0	0	0
88:	0	0	1	0	1	0	1	1
96:	1	2	0	1	0	1	2	1
104:	0	3	3	2	0	2	7	5
112:	9	10	11	14	13	16	12	27
120:	15	25	41	39	30	34	29	40
128:	52	64	84	126	152	163	155	105
136:	124	80	76	57	39	28	16	8
144:	0	1	0	1	2	2	7	2
152:	5	3	5	4	7	11	10	10
160:	23	18	20	33	43	66	82	108
168:	148	230	217	157	39	2	1	0
176:	0	0	1	0	0	0	0	0
184:	0	0	1	1	0	0	0	0
192:	0	0	0	0	1	2	2	1
200:	1	6	3	9	7	5	0	0
208:	0	0	0	0	0	0	0	0
216:	0	0	0	0	0	0	0	0
224:	0	0	0	0	0	0	0	0
232:	0	0	1	0	0	0	0	0
240:	0	0	0	0	0	0	0	0
248:	0	0	0	0	0	0	0	0
256:	0	0	0	0	0	0	0	0
264:	0	0	0	0	0	0	0	0
272:	0	0	0	0	0	0	0	0
280:	0	0	0	0	0	0	0	0
288:	0	0	0	0	0	0	0	0
296:	0	0	0	0	0	0	0	0
304:	0	0	0	0	0	0	0	0
312:	0	0	0	0	0	0	0	0
320:	0	0	0	0	0	0	0	0
328:	0	0	0	0	0	0	0	0
336:	0	0	0	0	0	0	0	0
344:	0	0	0	0	0	0	0	0
352:	0	0	0	0	0	1	0	0
360:	0	0	0	0	0	0	0	0
368:	0	0	0	0	0	0	0	0
376:	0	0	0	0	0	0	0	0
384:	0	0	0	0	0	0	0	0
392:	0	0	0	0	0	0	0	0
400:	0	0	0	0	0	0	0	0
408:	0	0	0	0	0	0	0	0
416:	0	0	0	0	0	0	0	0
424:	0	0	0	0	0	0	0	0
432:	0	0	0	0	0	0	0	0
440:	0	0	0	0	0	0	0	0
448:	0	0	0	0	0	0	0	0
456:	0	0	0	0	0	0	0	0
464:	0	0	0	0	0	0	0	0
472:	0	0	0	0	0	0	0	0
480:	0	0	0	0	0	0	0	0
488:	0	0	0	0	0	0	0	0
496:	0	0	0	0	0	0	0	0
504:	0	0	0	0	0	0	0	0

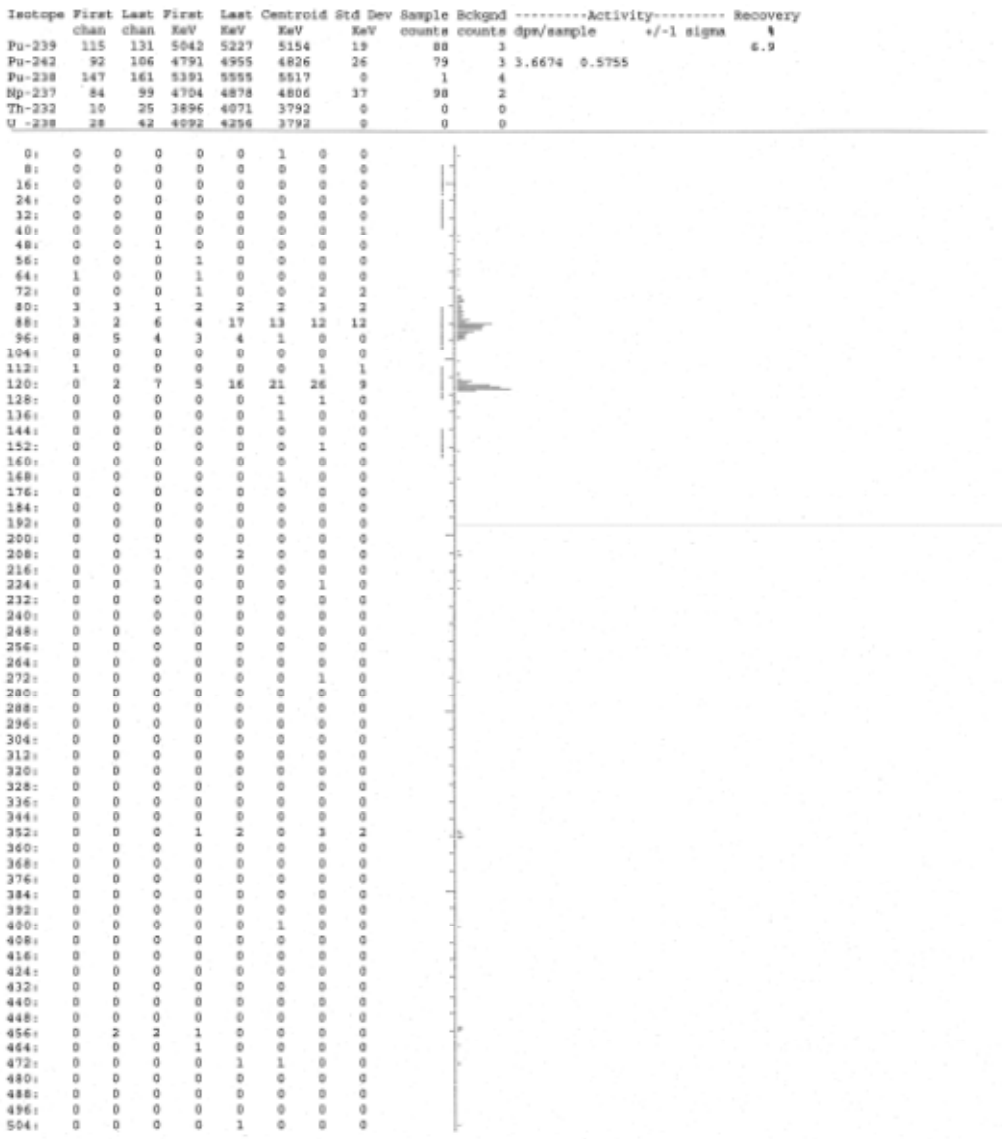
Figure 2: Alpha-spectrum of actinide spike in load solution.



Isotope	First chan	Last chan	First KeV	Last KeV	Centroid KeV	Std Dev KeV	Sample counts	Bckgnd counts	Activity dpm/sample	+/-1 sigma	Recovery %
Pu-239	154	172	5042	5232	5150	27	791	4			63.5
Pu-242	129	145	4792	4962	4827	26	1105	2	5.7191	0.2667	
Pu-238	189	205	5392	5562	5527	0	1	2			
Np-237	120	136	4702	4872	4802	36	1652	1			
Th-232	39	56	3892	4072	4067	0	1	2			
U -238	59	74	4092	4252	3507	0	0	1			

0:	0	0	0	0	0	0	0	0
8:	0	0	0	0	0	0	1	0
16:	0	0	0	0	1	0	0	0
24:	0	0	0	0	0	0	0	0
32:	0	0	0	0	0	0	0	0
40:	0	0	0	0	0	0	0	0
48:	0	0	0	0	0	0	0	0
56:	1	1	0	0	0	0	0	0
64:	0	0	0	0	0	0	0	0
72:	0	0	0	0	0	0	0	0
80:	0	0	0	0	0	0	0	0
88:	0	0	0	0	0	0	0	0
96:	1	1	0	0	1	1	0	1
104:	0	3	1	2	5	5	4	7
112:	12	11	15	21	13	27	27	21
120:	18	33	24	38	39	61	87	135
128:	174	217	175	159	141	113	107	67
136:	64	30	15	14	2	0	0	0
144:	0	1	0	0	1	1	3	1
152:	2	2	3	5	4	11	9	12
160:	23	40	59	68	101	132	167	121
168:	33	3	0	0	0	0	0	0
176:	0	0	0	0	1	0	0	0
184:	0	0	0	0	0	0	0	0
192:	0	0	0	0	0	0	0	0
200:	0	0	1	0	0	0	0	0
208:	0	0	0	0	0	0	0	0
216:	0	0	0	0	0	0	0	0
224:	0	0	0	0	0	0	0	0
232:	0	0	0	0	0	0	0	0
240:	0	0	0	0	0	0	0	0
248:	0	0	0	0	0	0	0	0
256:	1	0	0	0	0	0	0	0
264:	0	0	0	0	0	0	0	0
272:	0	0	0	0	0	0	0	0
280:	0	0	0	0	0	0	0	0
288:	0	0	0	0	0	0	0	0
296:	0	0	0	0	0	0	0	0
304:	0	0	0	0	0	0	0	0
312:	0	0	0	0	0	0	0	0
320:	0	0	0	0	0	0	0	0
328:	0	0	0	0	0	0	0	0
336:	0	0	0	0	0	0	0	0
344:	0	0	0	0	0	0	0	0
352:	0	0	1	0	0	0	0	0
360:	0	0	0	0	0	0	0	0
368:	0	0	0	0	0	0	0	0
376:	0	0	0	0	0	0	0	0
384:	0	0	0	0	0	0	0	0
392:	0	0	0	0	0	0	0	0
400:	0	0	0	0	0	0	0	0
408:	0	0	0	0	0	0	0	0
416:	0	0	0	0	0	0	0	0
424:	0	0	0	0	0	0	0	0
432:	0	0	0	0	0	0	0	0
440:	0	0	0	0	0	0	0	0
448:	0	0	0	0	0	0	0	0
456:	0	0	0	0	0	0	0	0
464:	0	0	0	0	0	0	1	0
472:	0	0	0	0	0	0	0	0
480:	0	0	0	0	0	0	0	0
488:	0	0	0	0	0	0	0	0
496:	0	0	0	0	0	0	0	0
504:	0	0	0	0	0	0	0	0

Figure 3: Alpha-spectrum of the first 10mL fraction of 0.1M HCl/0.01M HF with ^{239}Pu tracer and ^{237}Np spike.



Isotope	First chan	Last chan	First KeV	Last KeV	Centroid KeV	Std Dev KeV	Sample counts	Bckgnd counts	Activity dpm/sample	+/-1 sigma	Recovery %
Pu-239	115	131	5042	5227	5154	19	88	3			6.9
Pu-242	92	106	4791	4955	4826	26	79	3	3.6674	0.5755	
Pu-238	147	161	5391	5555	5517	0	1	4			
Np-237	84	99	4704	4878	4806	17	98	2			
Th-232	10	25	3896	4071	3792	0	0	0			
U -238	28	42	4092	4256	3792	0	0	0			

0:	0	0	0	0	0	1	0	0
8:	0	0	0	0	0	0	0	0
16:	0	0	0	0	0	0	0	0
24:	0	0	0	0	0	0	0	0
32:	0	0	0	0	0	0	0	0
40:	0	0	0	0	0	0	0	1
48:	0	0	1	0	0	0	0	0
56:	0	0	0	1	0	0	0	0
64:	1	0	0	1	0	0	0	0
72:	0	0	0	1	0	0	2	2
80:	3	3	1	2	2	2	3	2
88:	3	2	6	4	17	13	12	12
96:	8	5	4	3	4	1	0	0
104:	0	0	0	0	0	0	0	0
112:	1	0	0	0	0	0	1	1
120:	0	2	7	5	16	21	26	9
128:	0	0	0	0	0	1	1	0
136:	0	0	0	0	0	1	0	0
144:	0	0	0	0	0	0	0	0
152:	0	0	0	0	0	0	1	0
160:	0	0	0	0	0	0	0	0
168:	0	0	0	0	0	1	0	0
176:	0	0	0	0	0	0	0	0
184:	0	0	0	0	0	0	0	0
192:	0	0	0	0	0	0	0	0
200:	0	0	0	0	0	0	0	0
208:	0	0	1	0	2	0	0	0
216:	0	0	0	0	0	0	0	0
224:	0	0	1	0	0	0	1	0
232:	0	0	0	0	0	0	0	0
240:	0	0	0	0	0	0	0	0
248:	0	0	0	0	0	0	0	0
256:	0	0	0	0	0	0	0	0
264:	0	0	0	0	0	0	0	0
272:	0	0	0	0	0	0	1	0
280:	0	0	0	0	0	0	0	0
288:	0	0	0	0	0	0	0	0
296:	0	0	0	0	0	0	0	0
304:	0	0	0	0	0	0	0	0
312:	0	0	0	0	0	0	0	0
320:	0	0	0	0	0	0	0	0
328:	0	0	0	0	0	0	0	0
336:	0	0	0	0	0	0	0	0
344:	0	0	0	0	0	0	0	0
352:	0	0	0	1	2	0	3	2
360:	0	0	0	0	0	0	0	0
368:	0	0	0	0	0	0	0	0
376:	0	0	0	0	0	0	0	0
384:	0	0	0	0	0	0	0	0
392:	0	0	0	0	0	0	0	0
400:	0	0	0	0	0	1	0	0
408:	0	0	0	0	0	0	0	0
416:	0	0	0	0	0	0	0	0
424:	0	0	0	0	0	0	0	0
432:	0	0	0	0	0	0	0	0
440:	0	0	0	0	0	0	0	0
448:	0	0	0	0	0	0	0	0
456:	0	2	2	1	0	0	0	0
464:	0	0	0	1	0	0	0	0
472:	0	0	0	0	1	1	0	0
480:	0	0	0	0	0	0	0	0
488:	0	0	0	0	0	0	0	0
496:	0	0	0	0	0	0	0	0
504:	0	0	0	0	1	0	0	0

Figure 4: Alpha-spectrum of the second 10mL fraction of 0.1M HCl/0.01M HF with ^{239}Pu tracer and ^{237}Np spike.

Matrix	^{239}Pu Recovery %
No Rongalite	
Load Solution (N=8)	85.1 ± 7.1
Fecal Matrix (10mL, N=4)	60.4 ± 2.3
Fecal Matrix (N=12)	59.0 ± 7.9
Rongalite	
Fecal Matrix (N=8)	89.1 ± 3.7
Fecal Samples (N=30)	78.8 ± 17.0

Table 1: Percent recovery of ^{239}Pu ± 1standard deviation in various matrices. N corresponds to the number of samples analyzed.

Matrix	^{237}Np Recovery %
No Rongalite	
Load Solution (N=4)	88.5 ± 2.4
Fecal Matrix (10mL, N=4)	46.6 ± 5.7
Fecal Matrix (N=11)	58.0 ± 18.5
Rongalite	
Fecal Matrix (N=4)	80.3 ± 3.6

Table 2: Percent recovery of ^{237}Np ± 1standard deviation in various matrices. N corresponds to the number of samples analyzed.