FROM THE LAB

Sequential Determination of U, Pu, Am, Th and Np in Fecal and Urine Samples with Total Sample Dissolution

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Since the early days of the atomic age, there has been a need to determine accurately the radiological exposure to man from internally deposited radionuclides. Urinalysis has been used extensively for this purpose even though it has been demonstrated that urinalysis alone is inadequate in determining the total body burden from the majority of internally deposited radionuclides. In virtually all cases in which plutonium or other actinides except uranium are present in the body, the initial clearance is virtually exclusively into the feces. The urinary content becomes more important later on to determine the dose to the lungs, and much higher sensitivities are required for its analytical determination than was required earlier for the fecal analysis. Radiochemical analysis of fecal samples during the initial stages after an uptake continues to be the most sensitive and accurate method to obtain analytical data that will be used in dose calculations. The current publication describes a reliable chemical method for the determination of the actinides as well as gamma-emitting radionuclides in fecal and if needed urine samples.

Introduction

Publications dating back to 1964[1-4] document numerous internal exposures and elimination pathways for a variety of radionuclides including activation and fission products as well as actinides. Initially, the predominant path of elimination for the majority of the radionuclides encountered was through the gastrointestinal (GI) tract and into the feces. Only a few specific radionuclides including isotopes of hydrogen, iodine, mercury, cesium and molybdenum-technetium were found in the urine in easily detectable quantities. All the other radionuclides were eliminated so exclusively by way of the GI tract that the nuclide could not be detected in the urine in a 1000-minute count on a 24-hour sample by gamma-ray spectrometry. An accidental exposure to plutonium involving 78 individuals yielded only 6 urine samples that contained detectable activity; while 68 fecal samples from 22 individuals showed easily detectable levels of plutonium. As pointed out previously,[1] the experimental data shows that the path for clearance of most radionuclides that are inhaled or ingested is more

dependent on the physical characteristics of the particles than on the chemical characteristics of the radionuclides.

The disagreeable nature of fecal sampling, storage, transport and analysis is undoubtedly responsible for the reluctance on the part of many facilities to institute a comprehensive fecal monitoring program. Even though urinalysis is routinely used and accepted as an indicator of internal exposure, this does not change the fact that fecal analysis is necessary if the majority of the contamination is to be detected; especially during the early stages of elimination.

Radiochemical analysis of fecal samples continues to be the most sensitive method to obtain accurate analytical data that will be used in dose calculations. It is important to remember that the accuracy of the calculated dose is directly related to, and can never be better than, the accuracy of the analytical work. The chemical procedure used must be reliable, and must address fundamental chemical considerations such as total sample dissolution and isotopic exchange of tracer with the analyte of interest. The chemistry involved in determining the actinides in fecal material accurately is complex and should not be considered trivial. Although there are chemical procedures that produce acceptable results,[5,6] the majority of the procedures being used today include cost-cutting assumptions that make the procedure unreliable, inaccurate and do not address the fundamental concern of dissolution of the very difficult to dissolve refractory forms of plutonium.

Complete dissolution of the sample is undoubtedly one of the most important parts of any chemical procedure. For a desired chemical reaction to take place the element of interest must be in a specific oxidation state; otherwise the intended reaction will not occur. Furthermore, if an analytical procedure is expected to produce an accurate result, the sample and the analyte of interest must be dissolved completely, and isotopic exchange of the analyte with tracer must be guaranteed. These elementary concepts should be well known and should be the foundation from which every analytical procedure is developed. Unfortunately, the initial dissolution of the sample is usually neglected; especially when dealing with compounds that are difficult to dissolve such as quadrivalent oxides, siliceous materials, and samples like feces that contain high concentrations of calcium and phosphate. The omission of the appropriate type of front-end dissolution chemistry, by itself, will lead to the failure of even the finest chemical procedure. Likewise, isotopic exchange of the tracer with the analyte of interest is usually assumed rather than guaranteed. Even when dissolution techniques properly address the matrix and analyte of interest, the rigor necessary to control the oxidation state and prevent hydrolysis are too often overlooked. The importance of the appropriate type of front-end dissolution technique and the conditions needed to avoid hydrolysis of large ter- and quadrivalent elements cannot be over emphasized.[7]

Potassium fluoride and/or pyrosulfate fusions have been used routinely and reliably in this laboratory for over 30 years and are still the dissolution methods of choice for most sample matrices. However, when this method was applied to the dissolution of the ash from an entire fecal sample, a clear pyrosulfate fusion could not be obtained in a reliable fashion. The incomplete dissolution was due to a combination of factors: the large sample size (entire fecal sample taken for analysis), the high concentrations of phosphate and calcium that were present, and the production of insoluble condensed phosphates from dehydrating conditions produced in pyrosulfate fusions. It became obvious that another dissolution technique had to be used to ensure that the chemical procedure could be used reliably on a variety of fecal samples with different sample sizes. Donivan[8] used lithium metaborate to completely dissolve mill tailings for the determination of ^{230}Th; which demonstrated the fundamental principles of sample dissolution described above and contributed heavily to the choice of lithium metaborate by the current authors.

The current publication describes a reliable chemical method for the determination of the actinides in fecal samples using a lithium metaborate fusion as the initial dissolution step. This fusion method was chosen because it eliminates the problems associated with condensed phosphates, totally dissolves the sample, and guarantees isotopic exchange with tracer. Additional chemical separations that can be performed after the initial barium sulfate precipitation, in addition to the

separations presented in the current publication, and the preparation of the reagents can be found in a previous publication[9]. The time needed to analyze six fecal ash samples for the actinides listed above by high-resolution alpha-particle spectrometry is about three days. The chemical yields for all of the actinides determined are routinely better than about 85% (with some as high as 95%) and there are no known radiochemical interferences that adversely affect the accuracy of the results.

Preparation of the Urine Sample

1. Add up to 1 L of urine to a 2-L beaker containing about five carborundum boiling chips and place the beaker on a high temperature hot plate.
2. Add 100 mL of nitric acid, the appropriate isotopic tracers and evaporate the urine sample to about 50 mL.
3. Quantitatively transfer the evaporated urine sample to a plastic sandwich bag containing enough powdered cellulose filtering aid to absorb most of the liquid.
4. Treat the urine sample as described below for a fecal sample.

Preparation and Dry Ashing of the Fecal Sample[10]

The fecal sample should be collected in a plastic bag and placed in an appropriate outer container. If the analysis cannot be started immediately, the sample should be stored in a freezer.

1. Line a 10 x 20 x 5-cm deep aluminum tray with a double layer of vellum (tracing paper). The vellum should cover the entire inside surface of the pan and extend past the pan's vertical sides.
2. Cover the bottom of the lined pan with enough powdered cellulose filtering aid to absorb any excess liquid present in the fecal sample.
3. Place the plastic bag containing the weighed fecal sample into the lined aluminum tray.
4. Slice the bag several times with a scalpel and place the pan on the edge of a covered hot plate. Excess liquid, which will escape during the subsequent steps, should be absorbed in the cellulose powder and should not burn onto the pan.
5. During this initial heating process, ensure that the sample does not foam over the edge of the vellum. As foaming decreases, increase the temperature by moving the pan to the center of a bare hotplate to dry for about four hours.
6. After the fecal sample is dry, ignite the vellum with a small flame of a blast burner.
7. When the sample no longer burns with a flame, place the charred fecal sample in a muffle furnace, and ash the sample for 16 hours at 550 ºC.
8. After cooling, remove the pan from the furnace and transfer the ash to a tarred container.

Chemical Dissolution and Separations

1. Add up to 5 g of fecal ash to a 100-mL platinum dish.
2. Add 5 g of lithium metaborate, 1 g of sodium peroxydisulfate and mix thoroughly with a Teflon stirring rod.
   Note: If lithium metaborate is not available, add 6.2 g of ortho boric acid and 3.7 g of lithium carbonate to produce the metaborate upon heating.
3. Add 0.5 mL of nitric acid. HF should not be used in this step. The residual HF will interfere with the use of Ti(III) prior to the first barium sulfate separation.
4. Add an accurately known quantity of ^{243}Am, ^{236}Pu or ^{242}Pu, ^{229}Th and self purifying[11] ^{232}U tracers at individual activities of about 0.2 Bq.
   Note: Step 4 should be omitted for urine samples as the tracers were added previously. The use of ^{236}Pu will permit the determination of ^{237}Np in the purified plutonium fraction.

5. Heat the platinum dish on the hot plate until the ash is almost dry, taking care not to lose any tracer by splattering.
6. Place the dish on a ring stand and fuse over the full heat of a Fisher blast burner until a clear melt is obtained. If the molten salt turns cloudy, more sodium peroxydisulfate can be added directly to the molten flux to redissolve the insoluble material.
7. Add about 50 mg of KI[12] to the molten flux.
8. Remove the dish from the blast burner and cool the melt to room temperature. The flux should have a convex appearance and should be coalesced into a single mass. The addition of KI increases the surface tension of the flux and allows the flux to be removed easily from the dish.
9. Add the solidified lithium metaborate cake to a boiling solution of 300 mL of water and 100 mL of concentrated hydrochloric acid in a 1-L beaker.
10. Swirl the solution until the cake has dissolved and add 1 mL of 25% hydrazine.
11. If gamma-ray spectrometry is to be performed, this solution can be cooled at this point for counting.
12. Heat the solution to boiling and boil for ten minutes to hydrolyze any condensed phosphates.
13. Dilute the solution to a total volume of 500 mL with water and add 90 g of potassium sulfate. The addition of sodium sulfate at this point is not necessary and may even be counterproductive.
14. After the potassium sulfate has dissolved, add 2 drops of 1% Safranine-O and up to 1 mL of 20% titanium trichloride dropwise. Enough titanium trichloride should be added to reduce any residual ferric ion that is present and produce the colorless leuco form of the indicator. The solution should have a visible violet color of tervalent titanium.
15. Add 10 mL of 0.45% barium chloride dihydrate slowly with rapid stirring.
16. Boil the solution for a full five minutes.
17. Repeat the 10-mL additions of barium chloride and the five-minute boiling periods four more times.
    Note: To obtain high chemical yields of the actinides, each portion of barium sulfate must be allowed to precipitate completely before the next addition is added.
18. Transfer the solution while still hot to six 100-mL glass centrifuge tubes and centrifuge for five minutes.
19. Decant and discard the supernate immediately after centrifuging to avoid precipitating an unnecessary amount of the potassium salt of ter- and quadrivalent titanium.
20. Wash the precipitates into one centrifuge tube with water and add 10 drops of hydrofluoric acid.
21. Centrifuge for five minutes and discard the supernate.
22. Wash the barium sulfate with 30 mL of water, centrifuge for five minutes, and discard the supernate.
23. Add 2 mL of sulfuric acid and heat gently over a low flame of a blast lamp until the barium sulfate has dissolved and the sulfuric acid is fuming. Be careful not to evaporate the solution on the walls to dryness.
    Note: If the sulfuric acid appears black due to the presence of excessive organic material, perform steps 24 and 25, otherwise skip to step 26.
24. Add three drops of nitric acid to the hot sulfuric acid while swirling the solution rapidly to oxidize any residual organic material that is present.
25. After the solution is colorless add two drops of perchloric acid to decompose the nitrosylsulfuric acid that will have formed.
26. Cool the solution to room temperature and add three drops of 20% titanium trichloride.
27. Add 50 mL of reprecipitating solution to the 100-mL centrifuge tube to reprecipitate the barium sulfate.
28. Centrifuge for five minutes and discard the supernate.
29. Wash the barium sulfate with 30 mL of water, centrifuge for five minutes, and discard the supernate.

EDTA dissolution

30. Dislodge the impacted barium sulfate precipitate by flipping the centrifuge tube sharply in a downward motion.

31. Add 20 mL of 0.1-**M** potassium[13] EDTA and heat in a water bath until the barium sulfate has dissolved completely.
    Note: If necessary, place the tube in an ultrasonic bath to disperse the precipitate to ensure complete dissolution.
32. With rapid swirling, add two drops of 30% titanium trichloride followed by two 2.5-g portions of solid potassium hydroxide.
33. Heat in a hot water bath for five minutes to flocculate the titanous hydroxide precipitate.
34. Centrifuge for five minutes, decant and discard the supernate.
35. Wash the precipitate with 10 mL of 0.25-**M** sodium hydroxide directed in a forceful stream from a wash bottle.
36. Centrifuge for five minutes, decant and discard the wash.
    Note: This hydroxide precipitate contains all of the actinides that were present in the original sample.
37. Add 2 mL of perchloric acid to the precipitate and heat gently over the small flame of a blast lamp until the perchloric acid fumes and only about 1 mL remains.
38. Transfer the solution quantitatively to a 50-mL conical polymethylpentene (PMP) centrifuge tube with three successive 5-mL portions of water.
39. Add five drops of 0.5% sodium permanganate and heat in a boiling water bath for five minutes.
40. Add 200 µL of 0.5 mg/mL Nd carrier.
41. Swirl the solution rapidly and add 5 mL of hydrofluoric acid.
42. Immediately transfer the tube to a bath of cold water and let stand for 15 minutes to maximize the yield of americium, thorium and curium.
43. Filter the precipitate as described in "Mounting Fluoride Precipitates…" (Step 118) and collect the filtrate in a 125-mL Erlenmeyer flask.
44. This precipitate contains americium, curium, and thorium and can be counted in an alpha-particle spectrometer to determine if further separations are necessary. If thorium is present, as is usually the case, the filter must be wetashed and a separation of americium from thorium should be performed (Step 100).

Determination of plutonium and uranium

45. To the 125-mL Erlenmeyer flask containing the filtrate and wash from the plutonium-americium separation (Step 43), add 3 mL of sulfuric acid, 0.5 g of boric acid, three drops of 30% hydrogen peroxide and evaporate the solution to fumes of sulfuric acid.
46. Add 4.5 g of anhydrous potassium sulfate and 2.0 g of anhydrous sodium sulfate.
47. Swirl to mix and heat slowly over a blast burner until the salts dissolve.
48. Increase the temperature of the blast burner and continue to heat until the evolution of sulfuric acid fumes have slowed, the residue has dissolved and a clear pyrosulfate fusion is obtained.
49. Cool the pyrosulfate cake to room temperature and add 5 mL of concentrated hydrochloric acid, 35 mL of water and 0.5 mL of 20% ferrous ammonium sulfate.
50. Place the Erlenmeyer on a high temperature hot plate; heat the solution to boiling and boil for five minutes after the cake has dissolved completely.
51. While swirling the solution, add 1 mL of 0.45% barium chloride dropwise.
52. Boil the solution for one minute.
53. Repeat the 1-mL dropwise addition of barium chloride and the one-minute boiling time four more times.
54. Quantitatively transfer the solution and precipitate to a 50-mL conical PMP centrifuge tube.
55. Centrifuge for five minutes.
56. Decant the supernate, which contains the uranium, back into the original Erlenmeyer Flask.
57. Wash the barium sulfate with 5 mL of water, centrifuge for 5 min, and transfer the wash to the Erlenmeyer flask containing the uranium.
    Note: The barium sulfate contains the plutonium and should be treated as described in Step 87 "Determination of Plutonium". The solution in the Erlenmeyer that contains the uranium should be treated as described below without delay.

Determination of uranium

58. Reheat the supernate in the Erlenmeyer containing the uranium to about 50 C and add 200 µL of tellurous acid.
59. Add 1 mL of 25% hydrazine to the solution.
60. Heat the solution for about 10 minutes to reduce most of the iron. When the reduction of the iron is complete, the solution will turn dark from reduction of the tellurium.
61. Add 1 mL of tellurous acid, 2 drops of 1% Safranine-O indicator and heat the solution to boiling.
62. Add about 1 mL of 20% titanium trichloride dropwise until a dense black precipitate of elemental tellurium forms, the colorless leuco form of the indicator is obtained, and about four drops excess has been added.
63. Boil the solution until the tellurium has flocculated completely.
64. Filter the solution while it is still hot through a 25-mm Gelman HT-200 filter paper and collect the filtrate in a clean, tellurium free, 125-mL Erlenmeyer flask.
65. Discard the tellurium precipitate that contains the polonium.
66. Add one drop of 1% Safranine-O to verify the reducing conditions of the solution.
67. If the indicator turns red, add additional drops of titanium trichloride until the colorless leuco form of the indicator is restored.
68. Reheat the solution to boiling and add 1 mL of 0.45% barium chloride dropwise.
69. Boil the solution for one minute.
70. Repeat the 1-mL dropwise addition of barium chloride and the one-minute boiling time four more times.
71. Transfer the solution and precipitate to a 50-mL conical PMP centrifuge tube.
72. Centrifuge for five minutes, decant and discard the supernate.
73. Wash the barium sulfate with 5 mL of water, centrifuge for 5 min, decant and discard the wash.
74. Dislodge the impacted barium sulfate precipitate containing the uranium by flipping the centrifuge tube sharply in a downward motion.
75. Add 3 mL of water, 3 mL of 0.1 **M** potassium EDTA and heat in a water bath until the barium sulfate has dissolved completely.
    Note: If necessary, place the tube in an ultrasonic bath to disperse the precipitate to ensure complete dissolution.
76. With rapid swirling, add 1 drop of 30% titanium trichloride followed by 2 mL of 10 **M** potassium hydroxide.
77. Heat in a hot water bath for five minutes to flocculate the titanous hydroxide precipitate.
78. Centrifuge for five minutes, decant and discard the supernate.
79. Wash the precipitate with 5 mL of 0.25 **M** sodium hydroxide directed in a forceful stream from a wash bottle.
80. Centrifuge for five minutes, decant and discard the wash.
81. Flip the centrifuge tube sharply to dislodge the precipitate and add 15 drops of hydrochloric acid, 4 mL of water and 200 µL of 0.5-mg/mL Nd carrier.
82. Heat the centrifuge tube in a bath of boiling water until the precipitate dissolves completely.
83. Add 3 drops of 20% titanium trichloride and heat in a boiling water bath for 5 minutes.
84. Swirl the solution and add 1 mL of hydrofluoric acid.
85. Immediately transfer the tube to a bath of cold water and let stand for 15 minutes to maximize the yield of uranium.
86. Mount and count as described in "Mounting Fluoride Precipitates…" (Step 118).

Determination of plutonium

87. Dislodge the impacted barium sulfate precipitate containing the plutonium (Step 57) by flipping the centrifuge tube sharply in a downward motion.
88. Add 3 mL of water, 3 mL of 0.1 **M** potassium EDTA and heat in a water bath until the barium sulfate has dissolved completely.
    Note: If necessary, place the tube in an ultrasonic bath to disperse the precipitate to ensure complete dissolution.

89. With rapid swirling, add 1 drop of 30% titanium trichloride followed by 2 mL of 10 **M** potassium hydroxide.
90. Heat in a hot water bath for five minutes to flocculate the titanous hydroxide precipitate.
91. Centrifuge for five minutes, decant and discard the supernate.
92. Wash the precipitate with 5 mL of 0.25 **M** sodium hydroxide directed in a forceful stream from a wash bottle.
93. Centrifuge for five minutes, decant and discard the wash.
94. Flip the centrifuge tube sharply to dislodge the precipitate and add 2 mL of perchloric acid, 15 mL of water and 200 µL of 0.5-mg/mL Nd carrier.
95. Heat the centrifuge tube in a bath of boiling water until the precipitate dissolves completely.
96. Add three drops of 20% ferrous perchlorate and heat in a boiling water bath for five minutes.
97. Swirl the solution and add 5 mL of hydrofluoric acid.
98. Immediately transfer the tube to a bath of cold water and let stand for 15 minutes to maximize the yield of plutonium.
99. Mount and count as described in "Mounting Fluoride Precipitates…" (Step 118).

Separation of americium and thorium[14]

100. Place the HT-200 filter paper from Step 44 containing the americium and thorium in a 100-mL glass beaker and add 0.5 mL of nitric acid and 10 mL of perchloric acid.
101. Place the beaker on a high temperature hotplate and evaporate to 0.5 mL of perchloric acid without swirling the solution.
102. Add 2 mL of water to the beaker while still hot and transfer the solution to a 60-mL PFA jar.
103. Wash the beaker with another 2 mL of water and combine the wash with the solution in the 60-mL PFA jar.
104. Place the PFA jar on a clay triangle on a high temperature hotplate and evaporate to 50 µL of perchloric acid.
105. Add 4.5 mL of water to the PFA jar while still hot and transfer the solution to a 50-mL PMP centrifuge tube.
106. Add 100 µL of 1% silver nitrate solution, and 200 mg of ammonium peroxydisulfate.
107. Heat in a boiling water bath for 20 minutes.
108. Swirl the solution and add 1 mL of hydrofluoric acid.
109. Immediately transfer the tube to a bath of cold water and let stand for 15 minutes to ensure complete precipitation of the NdF_3.
110. Filter the precipitate containing the thorium and curium as described in "Mounting Fluoride Precipitates…" (Step 118) and collect the filtrate in a 60-mL PFA jar.
111. Add 1 mL of perchloric acid and 3 drops of 30% hydrogen peroxide to the PFA jar.
112. Evaporate the solution to 0.5 mL of perchloric acid.
113. Add 4.5 mL of water to the jar while still hot and transfer the solution to a 50-mL PMP centrifuge tube.
114. Add 200 µL of 0.5 mg/mL Nd carrier, five drops of 0.5% sodium permanganate and heat for five minutes in a boiling water bath.
115. Swirl the solution and add 1 mL of hydrofluoric acid.
116. Immediately transfer the tube to a bath of cold water and let stand for 15 minutes to maximize the yield of americium. Precipitation of americium in the presence of permanganate provides a redundant oxidation step that ensures the total elimination of plutonium.
117. Mount and count as described in "Mounting Fluoride Precipitates…" (Step 118).

Mounting fluoride precipitates for high-resolution alpha-particle spectrometry

118. Wash both sides of a 25-mm 0.2-µm Gelman HT-200 membrane filter paper with 80% ethanol and place the filter right side up in a Gelman

polysulfone filter holder equipped with a stainless steel support screen.

119. Tighten the funnel against the base as tight as possible without wrinkling the filter.
120. Filter the solution slowly by adjusting the vacuum. The rate of filtration should be adjusted so that the individual drops emerge from the outlet of the filtering chimney.
121. Wash the precipitate with 2 mL of water containing five drops of HF.
122. Wash the filter with 2 mL of 80% ethanol. Do not combine the ethanol wash with the main filtrate containing the perchloric acid.
123. Remove the filter containing the purified actinide fraction from the filtering assembly and dry under a 250-watt infrared lamp at a distance of about 10 cm for five minutes.

Results

Figures 1 through 4 show alpha-particle spectra of the separated uranium, plutonium-neptunium, americium and thorium-curium fractions obtained from actual fecal samples spiked with known quantities of various radionuclides. The thorium present was indigenous to the sample and was not added. If curium is present in the sample, it can be quantified in the thorium

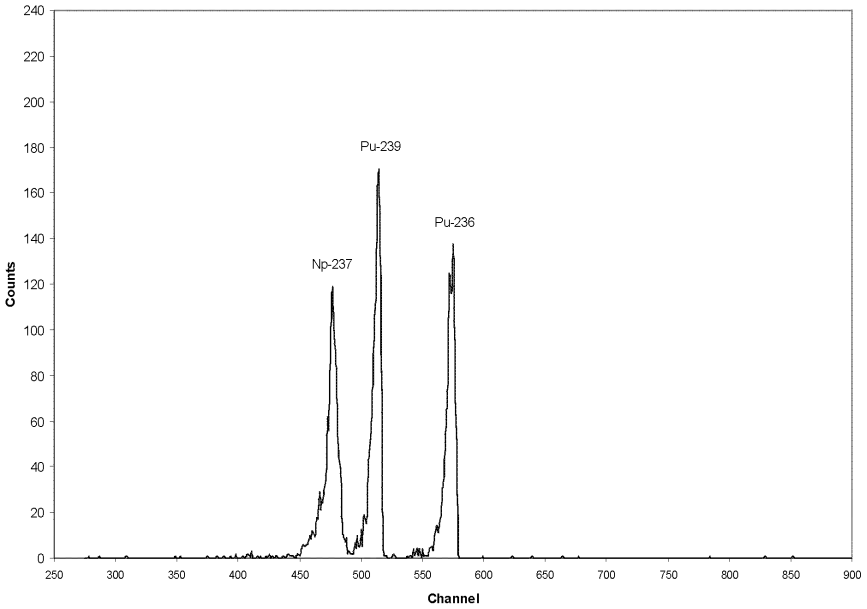

Figure 1 Alpha-particle spectrum of the plutonium-neptunium fraction obtained from an actual fecal sample. Note that the use of ^{236}Pu as an isotopic tracer allows ^{237}Np to be determined quantitatively.

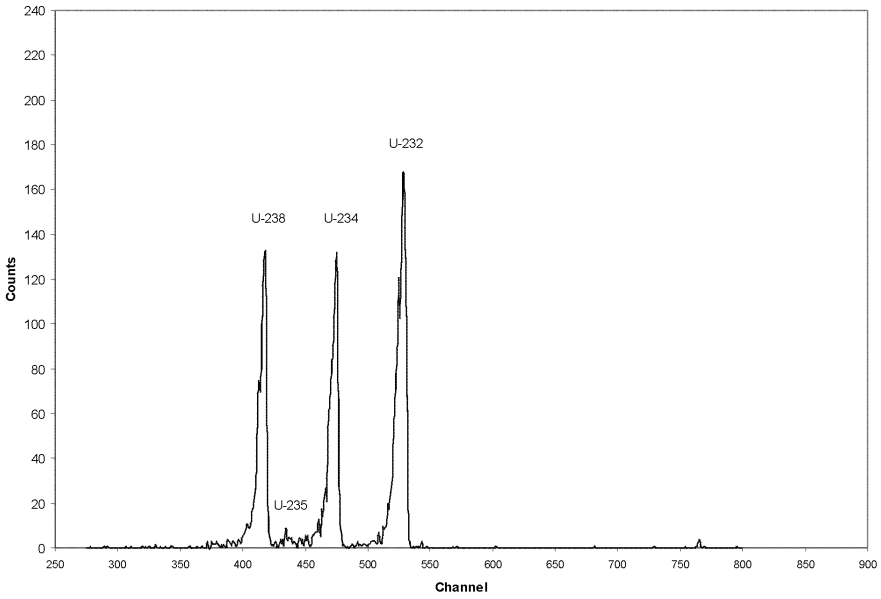

Figure 2 Alpha-particle spectrum of the final uranium fraction obtained from an actual fecal sample.

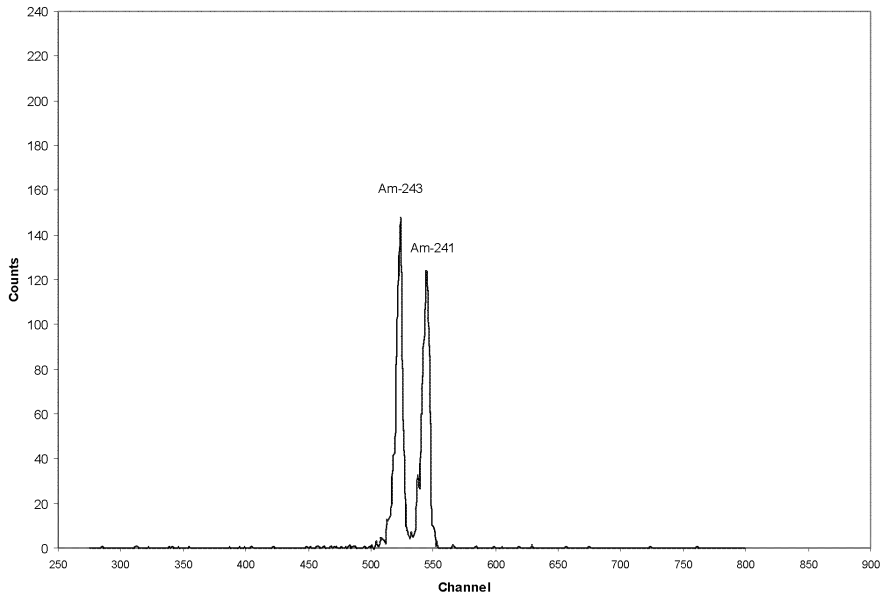

Figure 3 Alpha-particle spectrum of the americium fraction obtained from an actual fecal sample.

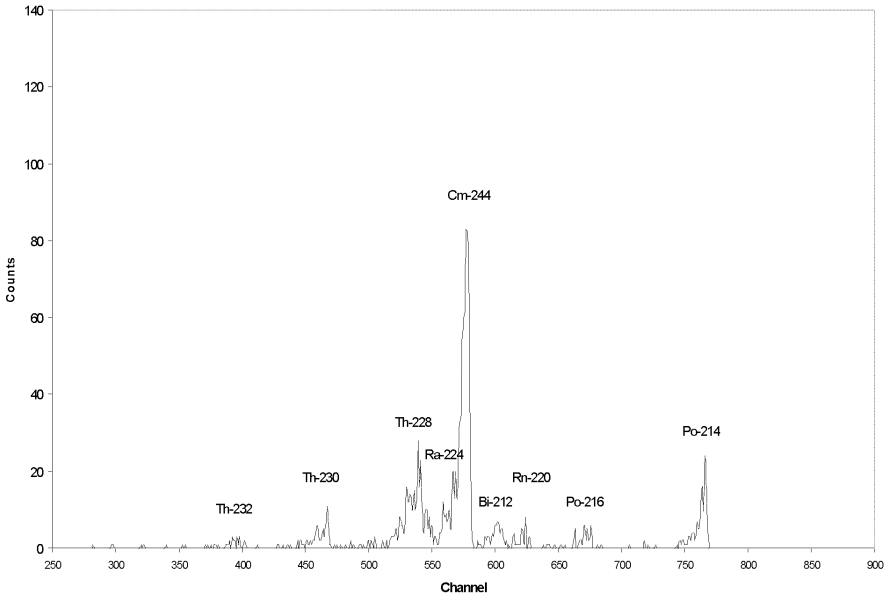

Figure 4 Alpha-particle spectrum of the thorium-curium fraction obtained from an actual fecal sample.

fraction. However, because of the higher solubility due to the actinide contraction, the actual chemical yield of curium will be about 10% lower than the chemical yield calculated from either americium or thorium tracers. If an accurate determination of the curium yield is important, the number of additions of barium chloride for each barium sulfate separation should be increased to a total of nine.[15] It should be noted that the short lived ^{214}Po is only present in the thorium-curium fraction immediately after separation and is undoubtedly due to co-precipitation of 19.8-minute beta-particle-emitting ^{214}Bi.

Figure 5 is a flow diagram outlining the major chemical separations used and is intended to aid the laboratory worker by providing a simplified view of the overall procedure.

Tables 1 and 2 compare two sets of experimental results with their known values. The data in Table 1 was obtained from fecal ash samples that were spiked at RESL with known quantities of uranium, plutonium, americium and neptunium. This data is one example of the many samples that were used as QC standards during the development and testing of this chemical procedure. In some cases the actinides being determined were added before the sample was ashed to ensure that the lithium metaborate fusion would dissolve

Radio-nuclide	Experimental activity	Known activity	Ratio (expt'l/known)
^{241}Am	2.27 ± 0.11 Bq/g	2.24 ± 0.05 Bq/g	1.01 ± 0.05
^{239}Pu	2.32 ± 0.11 Bq/g	2.30 ± 0.05 Bq/g	1.01 ± 0.05
^{238}Pu	2.36 ± 0.11 Bq/g	2.39 ± 0.05 Bq/g	0.99 ± 0.05
^{237}Np	2.02 ± 0.11 Bq/g	1.97 ± 0.04 Bq/g	1.03 ± 0.05
^{238}U	3.22 ± 0.15 Bq/g	3.17 ± 0.06 Bq/g	1.02 ± 0.05
^{234}U	3.10 ± 0.15 Bq/g	3.06 ± 0.06 Bq/g	1.01 ± 0.05

Table 1 Fecal ash sample - internal single blind QC standard prepared at RESL.

Radio-nuclide	Experimental activity	NIST value	Ratio (expt'l/known)
^{241}Am	3.89 ± 0.12 Bq/g	3.89 ± 0.01 Bq/g	1.00 ± 0.03
^{238}Pu	3.91 ± 0.12 Bq/g	3.88 ± 0.01 Bq/g	1.01 ± 0.03
^{238}U	3.96 ± 0.12 Bq/g	3.91 ± 0.02 Bq/g	1.01 ± 0.03
^{234}U	3.81 ± 0.12 Bq/g	3.77 ± 0.02 Bq/g	1.01 ± 0.03
Uncertainties are one standard deviation.			

Table 2 Artificial fecal sample - single blind PE standard prepared by NIST.

the refractory forms of plutonium that will be produced by muffling. The data in Table 2 was obtained from analysis of artificial fecal samples prepared by NIST and analyzed by RESL on a single blind basis for the purpose of laboratory performance evaluation. In all cases, every experimental result agrees with its known value within the statistics of the measurement. All random and the best estimate of any systematic uncertainties encountered anywhere in the entire measurement process have been propagated to the final result. The final uncertainty is expressed as one standard deviation.

Discussion

Lithium metaborate does not contain detectable amounts of uranium isotopes, as do the phosphates used in the traditional pyrophosphate fusion. This alleviates blank corrections that can lead to inaccuracy in the final result and improves the sensitivity of the method. Additionally, the problems associated with prolonged heating of phosphates under strong dehydrating conditions can be avoided in the metaborate system. Unlike pyrosulfate fusions, lithium metaborate fusions, under the conditions given, will not dissolve appreciable amounts of platinum from the dish even during prolonged heating at high temperatures. Consequently, uranium can be reduced directly with Ti^{+3} without reducing platinum to the elemental state and degrading the subsequent alpha-particle spectrum.

The solution obtained from dissolving the lithium metaborate glass can be analyzed by gamma-ray spectrometry and is stable for many days. The addition of relatively large quantities of hydrochloric acid was used to keep the solution from precipitating during the time needed for gamma counting but will not adversely affect the chemical yields of the actinides as was the case in previous methods.

It is important to note that the actinides of interest are separated as a group during the initial barium sulfate

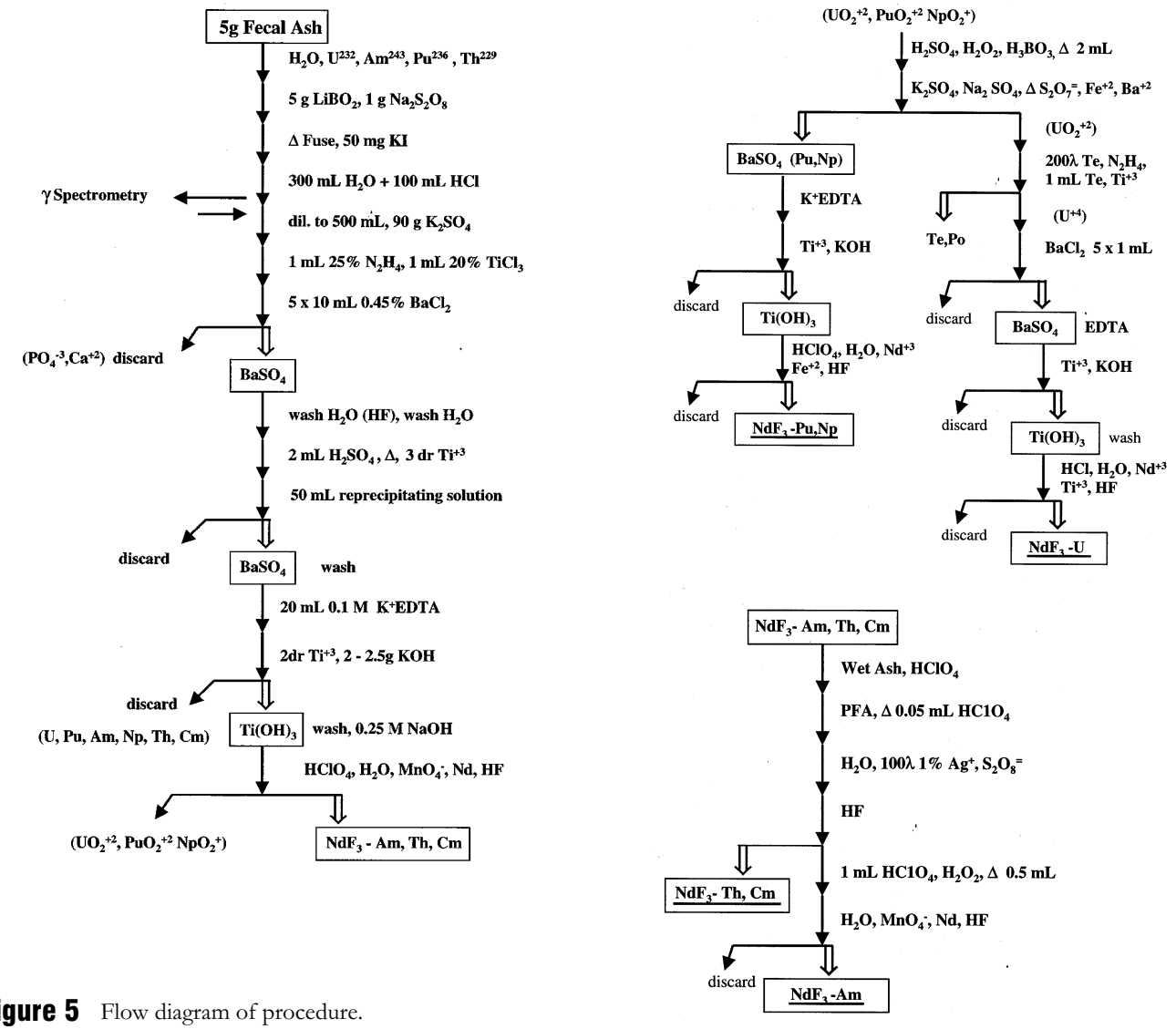

Figure 5 Flow diagram of procedure.

separation from the majority of the sample matrix. This is done to minimize a number of problems associated with the relatively large quantities of phosphates present in fecal samples and avoid precipitation of other insoluble compounds, particularly calcium salts and the potassium salt of titanous titanate.

References

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