

Progress Report on the Joint α -Spectroscopy Project of the Wisconsin Department of Natural Resources and the State Laboratory of Hygiene – Radiochemistry Unit.

(March 17, 2000)

Introduction.

To the best of our knowledge no book exists on the subject of α -spectroscopy. Some of the procedures to analyze for the actinides have even undergone significant revisions since this project began. A procedure for the analysis of polonium, which was recommend to us by Perking-Elmer, is now considered to be obsolete by chemists at the Department of Energy. The alternative polonium procedure lacked vital information, and it was necessary to consult with the Department of Energy in order to formulate a workable procedure. The vacuum gauges of the Octête-Plus were misinstalled and miscalibrated, and we have had some problems with the software provided with the instrument. Despite these problems we have progressed to the point where we are almost ready to proceed with the quality assurance tests of the methods, and, subsequently, the analysis of real samples.

Calibration of Vacuum Gauges.

Altogether, the Octête-Plus has nine vacuum gauges: one Granville-Phillips 375 Convectron thermal-conductivity gauge located in the RCAP-2 vacuum manifold and a thermal-conductivity gauge in each of the eight chambers. Initially, the two types of vacuum gauges were not in agreement with each other, although all of the thermocouple gauges were in agreement with one another. For example, Table 1 compares pressure readings taken from the two types of gauges at three different pressures. It is seen that the two gauges did not agree with one another at two lowest pressures and that the Granville-Phillips gauge did not give an accurate measurement of the atmospheric pressure (about 760 Torr).

Table 1. Vacuum Gauge Pressure Readings (January 14, 2000).

	Granville-Phillips Gauge	Chamber 2 gauge
Low pressure (Torr)	0.100	0.43
Medium pressure (Torr)	10	24.9
Atmospheric pressure (Torr)	438	(out of range)

A representative from Granville-Phillips and one from Perkin-Elmer (formerly EG&G ORTEC) were contacted about the vacuum-gauge problem. It became apparent that the Granville-Phillips gauge was misinstalled. The axis of the gauge was supposed to be

oriented in the horizontal plane but had been installed in the vertical plane, and the stem of the gauge was supposed to be oriented in the vertical plane but had been installed in the horizontal plane. After reorienting the Granville-Phillips gauge, it gave a reasonable measurement at atmospheric pressures; however, the Granville-Phillips gauge and the thermocouple gauges were still not in agreement. The Granville-Phillips gauge read about an order of magnitude below the thermocouple gauges at any given pressure. Engineers at Granville-Phillips and Perkin-Elmer informed me that thermal-conductivity gauges, e.g., the Granville-Phillips gauge, are inherently more accurate than thermocouple gauges. This was confirmed by pumping the RCAP-2 vacuum manifold down to the lowest possible pressure, which, from the Octête-Plus Hardware Manual, is around a few mTorr. When this was done, the pressure on the Granville-Phillips gauge was about 13 mTorr. By adjusting some of the fittings on the RCAP-2 manifold, it was easily confirmed that there were small leaks in the vacuum manifold preventing the attainment of a few Torr. However, as is discussed below, we have decided to collect spectra at pressures in the 1 to 10 Torr range (to prevent recoil contamination and contamination by polonium volatilization), so such small leaks are of no consequence, and no more time was spent on optimizing the manifold fittings.

After having properly installed the Granville-Phillips gauge, it was clear that the thermocouple gauges had been miscalibrated. I talked to an engineer at Perkin-Elmer about the calibration procedure for these gauges. Using the Granville-Phillips gauge as a reference, the seven of the eight thermocouple gauges were calibrated in the 1 to 10 Torr range, the range we anticipate using when collecting spectra. It was found that these gauges, tended to wander a little from their initial calibration, but according to an engineer at Perkin-Elmer, this was normal.

Problems arose when calibrating the thermocouple gauge in chamber 4. Initially, the gauge would be calibrated at 10 Torr. Then, the pressure would be lowered without making any adjustments, and when the pressure was returned to 10 Torr (as read on the Granville-Phillips gauge), the thermocouple gauge would read about 8 Torr. None of the other thermocouple gauges displayed this erratic behavior. I informed Dave Crouch of Perkin-Elmer of this problem by e-mail. We have not heard back from Perkin-Elmer about this problem, but have since moved the detector in chamber 4 to chamber 5, which, at the present, is not a problem since there are eight chambers but only four detectors.

Problems with Electrical Contacts.

During the time that the various gauges were being calibrated, two communication problems arose between the computer and the Octête-Plus. These problems were traced back to insufficient electrical contact between a board and its socket and between a power cable and its socket. However, once corrected, these problems have not since recurred.

Later on, a problem developed manifesting itself in somewhat sporadic and excessive background counts on detector 2. This problem was traced to an insufficient electrical contact between detector 2 and its socket. The problem was corrected and has not recurred.

Determination of an Optimal Pressure to Use for Alpha Spectroscopy.

The Perkin-Elmer operating manuals did not seem to have any firm recommendations about what pressures should be used when collecting spectra, although one Perkin-Elmer engineer suggested using a pressure of 10 mTorr. The work of Sill and Olsen¹ suggested that a higher pressure would be effective in reducing recoil contamination of the detectors while scarcely diminishing the alpha particle signal. Since recoil contamination is irreversible, we decided it would be prudent to operate the chambers at the higher pressures.

Sill and Olsen recommended using a pressure corresponding to an “air thickness”, t , of between 12 and 16 $\mu\text{g cm}^{-2}$. The air thickness is defined by

$$t = \rho l, \tag{1}$$

where ρ is the density of the air, and l is the distance between the detector and the sample. For good alpha-spectrum peak resolution, the Perkin-Elmer operating manuals suggest that the distance between a detector and sample be at least equal to the diameter of the detector. Our detectors have a diameter of about 2.76 cm (they have a surface area of 600 mm^2). The value of ρ calculated from Equation (1), using $l = 2.76$ cm and $t = 12$ $\mu\text{g cm}^{-2}$, can be combined with the density of dry air at 20 C and 1 atmosphere (1.205 mg cm^{-3}) and the Ideal Gas Law to give an operating pressure of 2.74 Torr. For $t = 16$ $\mu\text{g cm}^{-2}$, the corresponding operating pressure is 3.65 Torr. Consequently, we have been operating the chambers at a pressure of between 3.0 and 3.3 Torr.

One should keep in mind that there is a tradeoff between detector efficiency and the energy resolution of the alpha peaks. The energy resolution improves as the sample-detector distance is increased, but the detector efficiency decreases as the sample-detector distance is increased. It has come to our attention that some laboratories choose to move their samples closer to the detector, thereby increasing signal but losing resolution. We are in the process of making inquiries to other laboratories about what sample-detector distances are practical.

Energy Calibration of the Passivated Implanted Planer Silicon (PIPS) detectors.

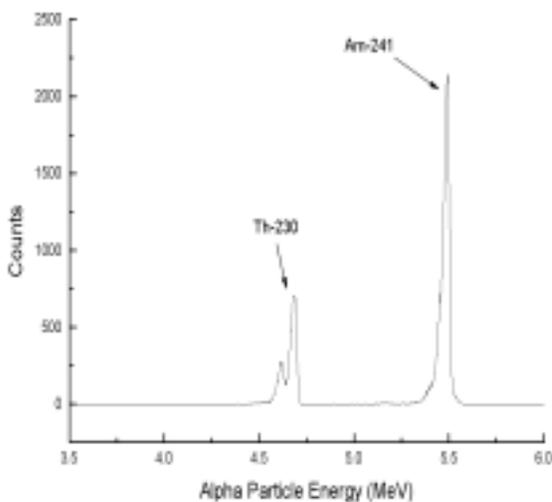
The next step was to calibrate the four PIPS detectors. An energy calibration of the PIPS detectors can be performed by using alpha emitters of known alpha-particle energy. It was determined that the detectors would be calibrated in the energy range of 3.5 to 6.0 MeV because all of the alpha emitters to be studied, and their respective tracers, fall within this energy range (see Table 2).

In the near future, we plan on ordering a calibration standard composed of Np-237, Am-241, and Cm-244 from AEA Technology QSA Inc. Before this can be done, the NRC must license our laboratory for the use of neptunium and curium. This licensing

procedure, which was initiated on December 20, typically takes three months, so approval by the NRC could come quite soon. In the meantime, it was decided to prepare a standard using Am-241, with alpha particle energies of 5.486 and 5.443 MeV, and Th-230 (see Table 2). A solution having an activity of about 100,000 decays per minute for both Am-241 and Th-230 was prepared. The two isotopes were co-precipitated with cerium fluoride using the microprecipitation procedure given in the SOP 4970.25M (see enclosed copy), and outlined in Figure 7. The precipitate was collected on a polypropylene filter and mounted on a stainless steel planchet. The four PIPS detectors were then calibrated using this energy standard. Figure 1 shows a representative spectrum of the Am-241, Th-230 standard collected on February 2.

Table 2. Alpha Particle Energies of Analyte and Tracer Isotopes.

Isotope	Analyte or Tracer	Main peak energy [MeV (branching ratio)]	Secondary peak energy [MeV (branching ratio)]
U-232	Tracer	5.320 (0.686)	5.263 (0.312)
U-234	Analyte	4.856 (0.72)	4.772 (0.28)
U-235	Analyte	4.396 (0.57)	4.336 (0.18)
U-236	Tracer	4.493 (0.74)	4.443 (0.28)
U-238	Analyte	4.195 (0.77)	4.147 (0.23)
Th-228	Analyte	5.424 (0.71)	5.342 (0.28)
Th-229	Tracer	4.842, (0.582)	4.811 (11.4)
Th-230	Analyte	4.684 (0.76)	4.617 (0.24)
Th-232	Analyte	3.994 (0.77)	3.935 (0.23)
Po-209	Tracer	4.882 (1.00)	None
Po-210	Analyte	5.305 (1.00)	None



It is also possible to calculate the absolute efficiency of the detectors with a sample of known activity. However, since our laboratory is not capable of preparing such a standard, the absolute-efficiency calibration will have to wait until we receive the standard from AEA Technology QSA Inc. Because we anticipate employing relative analysis methods using a tracer, making absolute-efficiency calibrations of the detectors is not that important, although we would like to know the detector efficiencies in order to be assured that the detectors are operating properly.

Figure 1. Alpha Spectrum of Energy Standard.

Background Determination, Detector Contamination, and the Proper Handling of Planchets.

Background spectra were taken with the four detectors. The spectra revealed contamination by Am-241 and Th-230, which was most certainly due to the energy standard. The chambers and the sample mounts were cleaned with Radiacwash, and background spectra were collected again. This time only detector 2 showed any significant contamination. Cleaning the chamber and the sample mount once again did not reduce the contamination. A plastic cover was placed over detector 2, and a background spectrum was taken. The spectrum still showed contamination leading to the conclusion that the contamination resided on the surface of the detector 2. (It should be remarked that this is not the same as recoil contamination and should be removable.) Some contamination was removed by swabbing the detector surface with polished water; however, some contamination remains. Removal of the contamination should not be a problem. As will be discussed further below, the cerium fluoride particles from the precipitate seem to have a high affinity for any surface having a low surface tension, such as the detector surface. Using a solvent with a lower surface tension than water, like an organic solvent, should be effective in removing the cerium fluoride particles. I plan on contacting Perkin-Elmer for advice about which solvents can be used. In the meantime, background spectra were collected for detectors 1, 3 and 4 for a period of 2½ days, which showed that contamination of these detectors was minimal if any.

Clearly, great care should be taken in handling the microprecipitate samples. It seems that once a filter is mounted on a planchet, the planchet should only be handled by its edges, and care should be taken not to stir up any air currents that might lead to contamination by the airborne particles.

Recommendations from Dr. Isabel Fisenne at the Department of Energy.

To analyze for Po-210, a representative from Perkin-Elmer recommended that we use procedure Po-01-RC from the EPA's HASL-300 manual, 28th Ed. (see Figure 11). Preparations were made to implement this procedure; however, upon checking the HASL-300 manual, it was found that an alternative procedure was available, Po-02-RC (see Figure 10). The contact person for these procedures is Dr. Isabel Fisenne at the Department of Energy's Environmental Measurements Laboratory. I called Dr. Fisenne on February 4 to inquire about the relative merits of the two procedures. Dr. Fisenne recommended the use of procedure Po-02-RC because of the large sample volumes that must be employed (up to 10 L). It seems that precipitates which appear as the volume of water is reduced can incorporate Po and, as a consequence, interfere with the analysis of Po-210. In addition, procedure Po-02-RC has an improved method for the electrodeposition of polonium.

I also told Dr. Fisenne that we would be analyzing samples for uranium and thorium. She recommended using a neodymium carrier rather than the cerium carrier we had used to prepare the energy standard. She said that cerium is often contaminated with various actinides which could potentially interfere with a uranium or thorium analysis. We

decided to make preparations for the use a neodymium carrier. Two such procedures exist: procedure G-03 Revision 0 from HASL-300, 28th Ed. (see Figure 8) and a procedure developed by Canberra (see Figure). In the meantime, we had decided to check the cerium carrier for actinide contamination.

Check of the Cerium Carrier for Actinide Contamination, and the Development of a Labware Cleaning Procedure.

A blank solution (polished water) was used as the sample for the cerium fluoride microprecipitation procedure. Upon taking an alpha spectrum of the cerium fluoride precipitate, it was found that there was substantial activity from both Am-241 and Th-230. The same filter apparatus that had been used in making the Am-241, Th-230 energy standard was used in this experiment, so it seemed quite clear that the contamination was coming from the filter apparatus. The filter apparatus had been washed once using Radiacwash. Clearly, this cleaning procedure was inadequate. Some time was spent on trying to arrive at an effective cleaning procedure for the filter apparatus. The resulting procedure was to wash the filter apparatus three times with Radiacwash and to use a cotton swab to clean the various nooks and crannies of the apparatus. Upon analyzing a cerium fluoride precipitate prepared with a blank solution after using the new cleaning procedure, it was found that there was little, if any, actinide contamination of the cerium carrier. For example, a spectrum taken using detector 1 on February 15 revealed just seven background counts in a twenty-hour counting period. Thus, our current cerium carrier seems to be fine, although we plan on doing some experiments with a neodymium carrier in the event that future cerium carriers are not contaminant-free.

We had been using the labware cleaning service at the State Hygiene Laboratory for our polypropylene beakers; however, an experiment discussed below indicated that contamination due to the cerium fluoride precipitate was not completely removed by the in-house washing service. Thus, at the present, all plastic labware exposed to the cerium fluoride precipitate is cleaned in the manner described above. (It should be mentioned that plastic labware is used whenever the labware is exposed to hydrofluoric acid, as is the case in the microprecipitation techniques employing either a cerium or neodymium carrier, because the hydrofluoric acid etches glass.)

A communication with a technical applications specialist from Nalge Nunc International (the manufacturer of our polypropylene and polymethylpentene labware) on March 9 suggested that our cleaning procedure could be improved by following our procedure with a concentrated nitric acid rinse. He thought that since the cerium fluoride is insoluble in water, it is attracted to hydrophobic surfaces like those of plastics, or, as discussed above, the surface of the PIPS detector.

Extent of Contamination of U-236, Th-229, and Po-209 Tracers.

The U-236 standard was purchased from Isotope Products Laboratories. Table 2 provides a summary of the quoted activities of the various isotopes in the U-236 standard. (In addition, an alpha spectrum of a U-236 sample, prepared by cerium fluoride

microprecipitation method, was consistent with the data in Table 3.) It is seen that there is significant contamination by U-234, an analyte of interest. Furthermore, the peak energies of U-235, an analyte of interest, and U-236 are quite close (see Table 2), just 47 keV apart, a possible problem if the U-235 activity is small in comparison to that of U-236. The effect that these contaminants have on the determination of the various uranium isotopes is discussed in more detail in the next section.

Table 3. Composition of U-236 Standard.

Nuclide	Atom %	Activity %
U-233	0.002	0.295
U-234	0.116	11.04
U-235	9.20	0.304
U-236	89.38	88.36
U-238	1.306	0.0067

The Th-229 tracer was purchased from the National Institute of Standards and Technology. They reported detecting no alpha-emitting contaminants at the time of preparation (July 1996). However, since the standard was approximately four years old, it

was decided to check it by preparing a small amount of the standard using the cerium fluoride microprecipitation method.

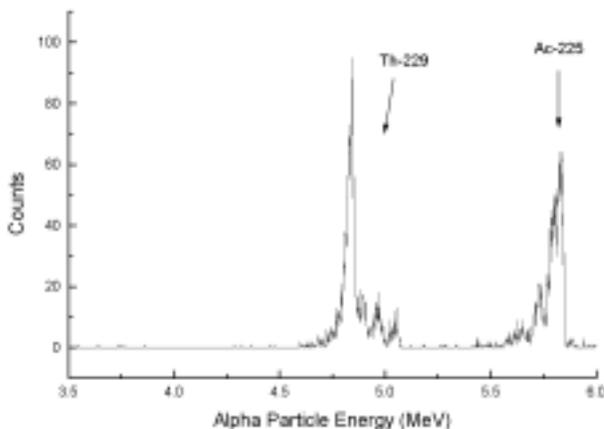


Figure 2. Alpha Spectrum of Th-229 Standard.

The alpha spectrum of the Th-229 standard is shown in Figure 2. The alpha particles due to Th-229 decay show up as a series of peaks in the 4.7 to 5.1 MeV range. The series of peaks in the 5.6 to 5.8 MeV range are due to alpha particles emitted by the decay of Ac-225. Th-229 is a grandparent of Ac-225. Th-229 alpha decays to Ra-225, and the Ra-225 beta decays to Ac-225. From Table 2, it is seen that the alpha-particle

energies due to the decay of the various thorium and uranium isotopes are less than the energies of the Ac-225 peaks. Furthermore, it is shown in the next section that most of the actinium is removed by the cation-exchange columns.

The Po-209 standard was purchase from the National Institute of Standards and Technology. The activity due to Po-209 at the time of preparation (January 1995) was 85.42 Bq/g. Only one contaminant was specified: Po-208, at an activity of 0.106 Bq/g. The main mode of decay of Po-208 is by emitting an alpha particle with an energy of

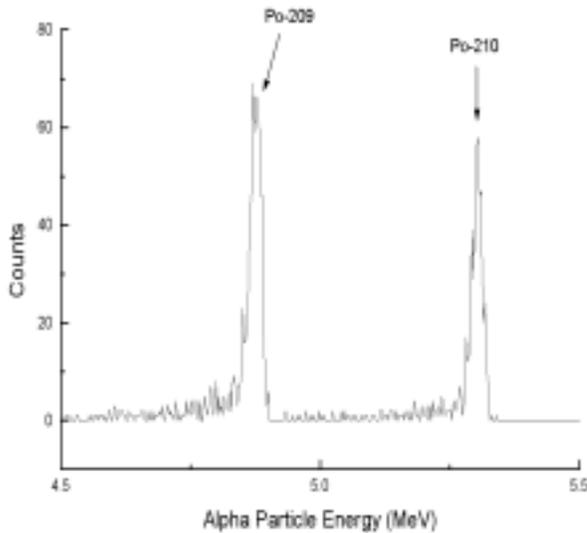


Figure 3. Alpha Spectrum of Po-209 Standard and Po-210 Spike.

5.21 MeV (99+%). This energy is close to that of the Po-210 peak of 5.305 MeV. Since the half life of Po-209 is 105 years and that of Po-208 is 2.898 years, the present activity of Po-209 is about 74 Bq/g and that of Po-208 is about 0.03 Bq/g. Thus, the extent of contamination is only about 0.04%. Figure 3 shows the alpha spectrum of a sample that was prepared using about 1.7 Bq of the Po-209 standard and about 2.0 Bq of a Po-210 spike. The sample was electrodeposited on a nickel planchet as outlined in Figure 10. A peak due to Po-208 is not evident. Since we plan on adding a tracer activity that is approximately at the activity of the sample, we do

not anticipate contamination of the Po-209 standard to be a problem. If the Po-208 and 210 peaks should be comparable in intensity, we should not have a problem discriminating between the two since our instrument resolution is better than 30 keV.

Development of the Procedure for Separating Uranium and Thorium From Each Other and Other Actinides and for Their Subsequent Analysis.

As of February 10, we were confident that our filter-apparatus cleaning procedure and that our cerium fluoride microprecipitation procedure were working well. Having the microprecipitation part of the method working properly before using any of the cation-exchange columns seemed important considering the relative expense of the columns.

Next, we investigated the separation of uranium and thorium from each other and from other actinides using the Eichrom TEVA and UTEVA cation-exchange columns. The tentative procedure for the separation was adapted from Eichrom method ACW01 Revision 1.4 and is summarized in the flowcharts of Figures 12 and 13. (During the course of the work in this part of the project, Eichrom had revised their procedure from Revision 1.4 to Revision 1.5. See SOP 4970.25M for the latest revision.)

We used Eichrom procedure ACW01 Revision 1.4 to analyze one liter of a sample of water that had had a relatively high gross alpha (21.1 pCi/L by EPA method 900.0) and total uranium (20.7 pCi/L by EPA 908.0). 0.69 Bq of the Th-229 tracer and 1.88 Bq of the U-236 tracer were added to the sample. We collected a good deal of information about the procedure which we have incorporated into the SOP 4970.25M. Upon analysis, it was found that the alpha spectrum of the uranium fraction was contaminated by the Th-229 tracer (see Figure 4). The alpha spectrum of the thorium fraction showed significant

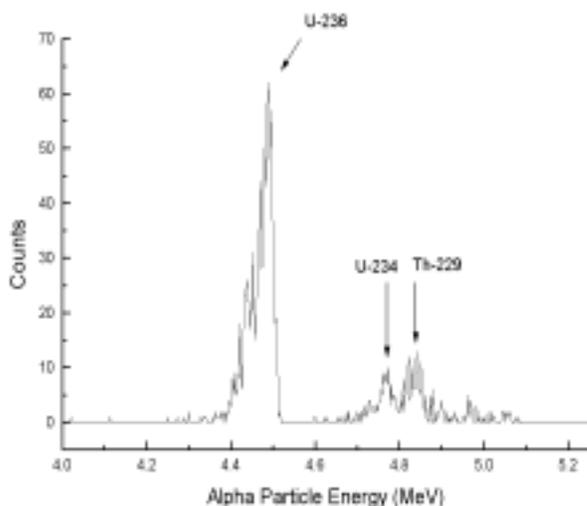


Figure 4. Alpha Spectrum of Uranium Fraction of First Sample.

activity was observed. Although the U-236 tracer contributes to the U-234 peak, it is clear from the relative activities of U-236 and U-234 in the tracer (see Table 3) that much of the U-234 activity is from the sample. Similarly, the sample shows some U-238 activity. Any U-235 which might be present in the sample is obscured by the relatively large U-236 peak occurring at about the same energy (see Table 2).

There are various ways of analyzing this data. One method is called a Region of Interest (ROI) analysis in which the counts in a prescribed energy interval, after background subtraction, are assigned to a particular isotope.

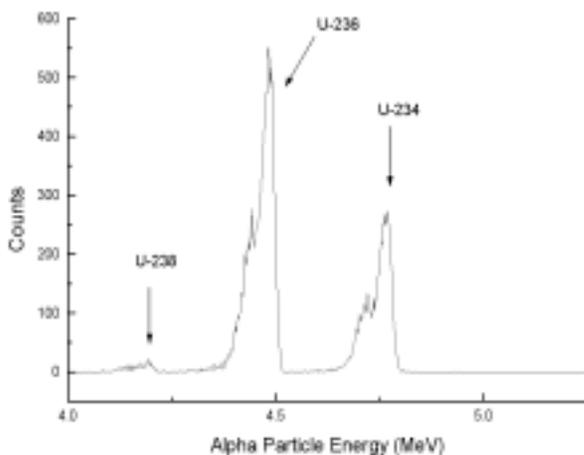


Figure 5. Alpha Spectrum of Uranium Fraction of Second Sample.

contamination by Am-241, which was most certainly acquired during the cerium fluoride microprecipitation step from the polypropylene beaker that was used to make the energy standard. As mentioned above, this compelled us to clean all plastic labware in the same manner as the filter funnel.

The thorium contamination of the uranium fraction may have been corrected by the latest procedure revision (1.5). Figure 5 shows the alpha spectrum of the uranium fraction of a second sample, of similar composition to the first sample. This time no Th-229

activity was observed. However, since there are three well-defined peak regions and four unknown activities, this method of analysis will not work. Another method of analysis is one in which the software fits a Gaussian-like function to the various peaks. One problem with this method is that the software only seems to be capable of accounting for one contaminant in the tracer. Another problem is that the program will have to deconvolute the U-235 peak from the U-236 peak, a task we are not

sure that the software can accomplish with much accuracy. We have contacted Perkin-Elmer about these problems, but, quite frankly, up to this point they have not been very helpful in this regard.

Consequently, we have decided to order a U-232 standard from National Institute of Standards and Technology. The alpha-particle energy of the U-232 peak is well separated from the other peaks (see Table 2). The U-232 standard is not contaminated by other

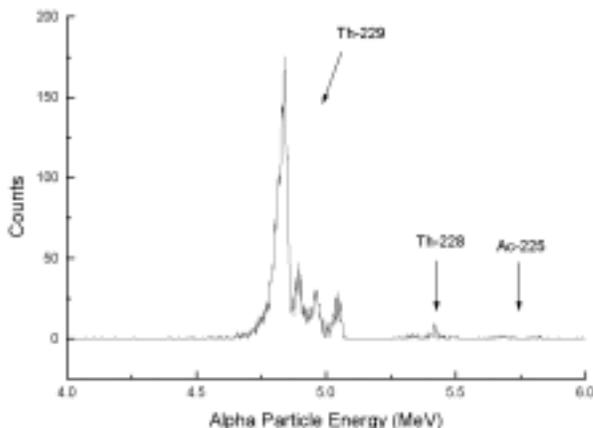


Figure 6. Alpha Spectrum of Thorium Fraction of Second Sample.

uranium isotopes, but has some thorium contamination. However, the thorium contamination can be removed using a TEVA cation-exchange column.

Figure 6 shows the alpha spectrum of the thorium fraction of a second sample. This time little or no Am-241 contamination was detected, and some Th-228 activity of the sample was detected. In addition, there seems to be some activity from Ac-225, the Th-229 standard contaminant, although such activity will not interfere with the analysis of the thorium isotopes.

It should be mentioned that sample preparation takes about 2½ working days. The long sample preparation time is largely due to sample evaporation time and the time it takes for samples to elute on the TEVA and UTEVA cation-exchange columns. I will be attending a one-day conference (Albuquerque, NM on April 27) sponsored by Eichrom, the maker of the TEVA and UTEVA columns, in which various topics will be covered including the handling of large environmental samples and rapid sample preparations using vacuum techniques and their new cartridges.

Development of the Procedure to Analyze for Polonium-210.

As mentioned above Dr. Fisenne of the EPA recommended using procedure Po-02-RC from the EPA's HASL-300 manual (see Figure 10). Initially, some experiments were performed on the electrodeposition step of the procedure. It was found that this part of the procedure worked well for samples prepared from standard Po-209 and Po-210 solutions (see Figure 3). Some experiments were performed to assure ourselves that any thorium or uranium contained in a sample would not be electrodeposited along with the polonium. In one experiment, initiated on February 29, a solution was prepared that contained 0.17 Bq of Th-229, 0.47 Bq of U-236, 0.18 Bq of Po-209, and 0.20 Bq of Po-210. After electrodeposition, a spectrum was collected. No contamination from any thorium or uranium isotopes was detected. The spectrum was practically identical to that of Figure 3.

Procedure Po-02-RC calls for a 10-L volume of water sample which is then reduced to a volume of about 5 ml. Before attempting to evaporate such a large volume, the steps subsequent to the evaporation step and preceding the electrodeposition step were attempted using a small volume of polished water which was acidified with nitric acid and to which was added 10 mg of Pb (which functions as a carrier). In the first of these intermediate steps the sample is digested in a thioacetamide solution, which produces a PbS precipitate, and then redissolved in concentrated hydrochloric acid. The procedure is then repeated using conditions which differ slightly from the first. Some detailed information about the intermediate steps was noted and will be included in the Polonium SOP.

In the next part of the procedure problems developed when attempting to reduce the volume of a water sample with high gross alpha (21.1 pCi/L by EPA method 900.0) and a sample of Madison tap water that had been acidified with nitric acid and that had 10 mg of Pb carrier added. When the sample volumes were reduced to about 50 mL, a gelatinous precipitate began to appear. Although there was no mention of such a precipitate in procedure Po-02-RC, we proceeded to the next step of the procedure, the thioacetamide digestion, expecting the precipitate to dissolve. During the digestion the gelatinous precipitate did not dissolve but turned black due to the additional precipitation of PbS. An experiment was performed to try to reproduce the gelatinous precipitate under controlled conditions, but the precipitate failed to appear. We guessed that the probable identity of the precipitate was some kind of a silicate. In consulting with an inorganic water chemist at the State Hygiene Laboratory, we learned that the concentration of SiO₂ was probably in excess of 30 mg/L in Madison tap water.

We decided to repeat the experiment to isolate the gelatinous precipitate and to have the precipitate analyzed by the Inorganics Unit of the State Hygiene Laboratory. The experiment was repeated using two 2.5 L Madison tap water samples which were acidified with nitric acid. In addition, Pb carrier was added to one of the samples. A gelatinous precipitate appeared in about equal quantities in both samples, upon evaporation of the sample volumes to about 50 mL. The samples will be ready to be analyzed by March 17.

I also sent an e-mail to Dr. Fisenne at DOE regarding the gelatinous precipitate. She called on March 16, and confirmed that the precipitate was probably a silicate. She recommended an alteration of procedure Po-02-RC in which the gelatinous precipitate is exposed to a hot solution of concentrated nitric and hydrofluoric acids. The acid solution is then evaporated to near dryness. This step is repeated until all of the silicate has dissolved. Presently, we are preparing to implement this part of the procedure.

Recalibration of the PIPS Detectors.

Detectors 1, 2, and 4 have held their initial energy calibration quite well. Detector 3 has drifted out of calibration by about 40 keV. We plan on recalibrating all of the detectors on a regular basis once we receive our standard from AEA Technology QSA Inc. We do not want to use the energy standard that we prepared for the initial calibration, fearing

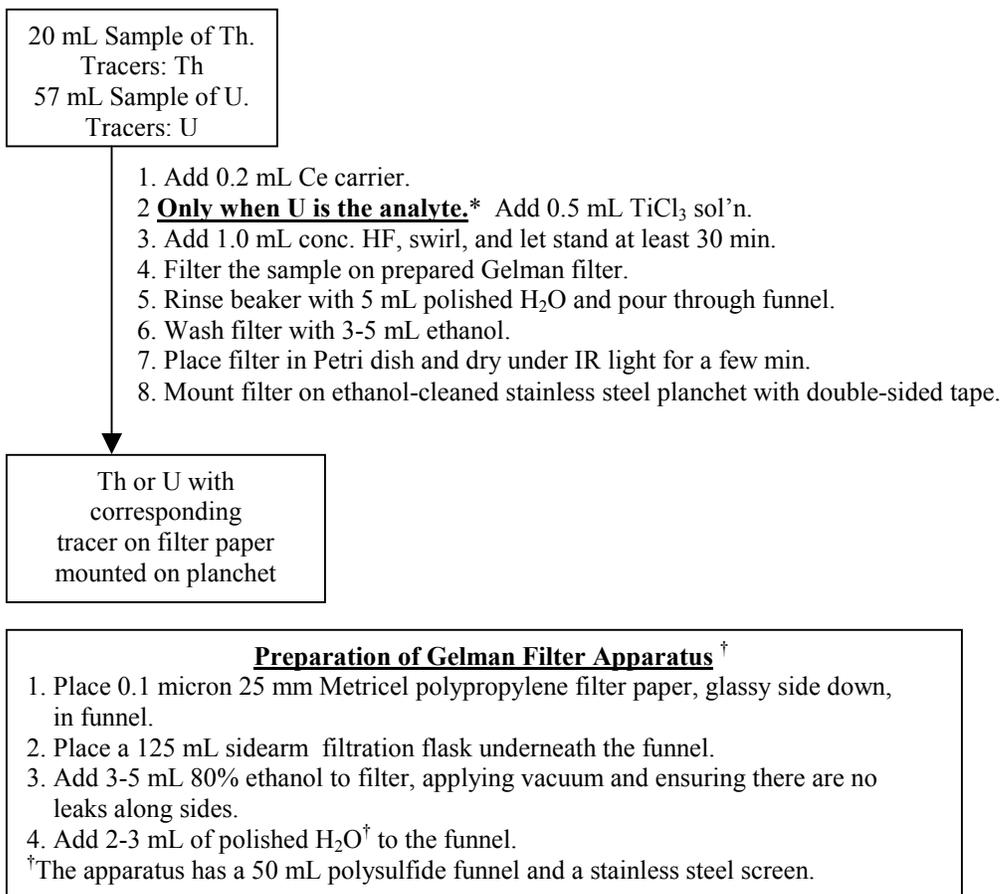
contamination of the chambers and detectors due to the relatively high activity of the standard and the particulate nature of the standard. If need be we can prepare a standard by co-electrodepositing several actinides on a nickel planchet. Contamination from such a standard would be minimal because the electrodeposited actinides would strongly adhere to the nickel.

Conclusions.

We have overcome numerous difficulties presented by the alpha spectrometer and the sample preparation methods. There are still several details which are in the process of being worked out. Despite this we feel we are almost ready to proceed with the quality assurance tests of the methods, and the analysis of real samples.

Co-precipitation of Th, U, Pu, Am, and Cm with CeF₃.

(Adapted from Eichrom method ACW01 Rev. 1.4.)

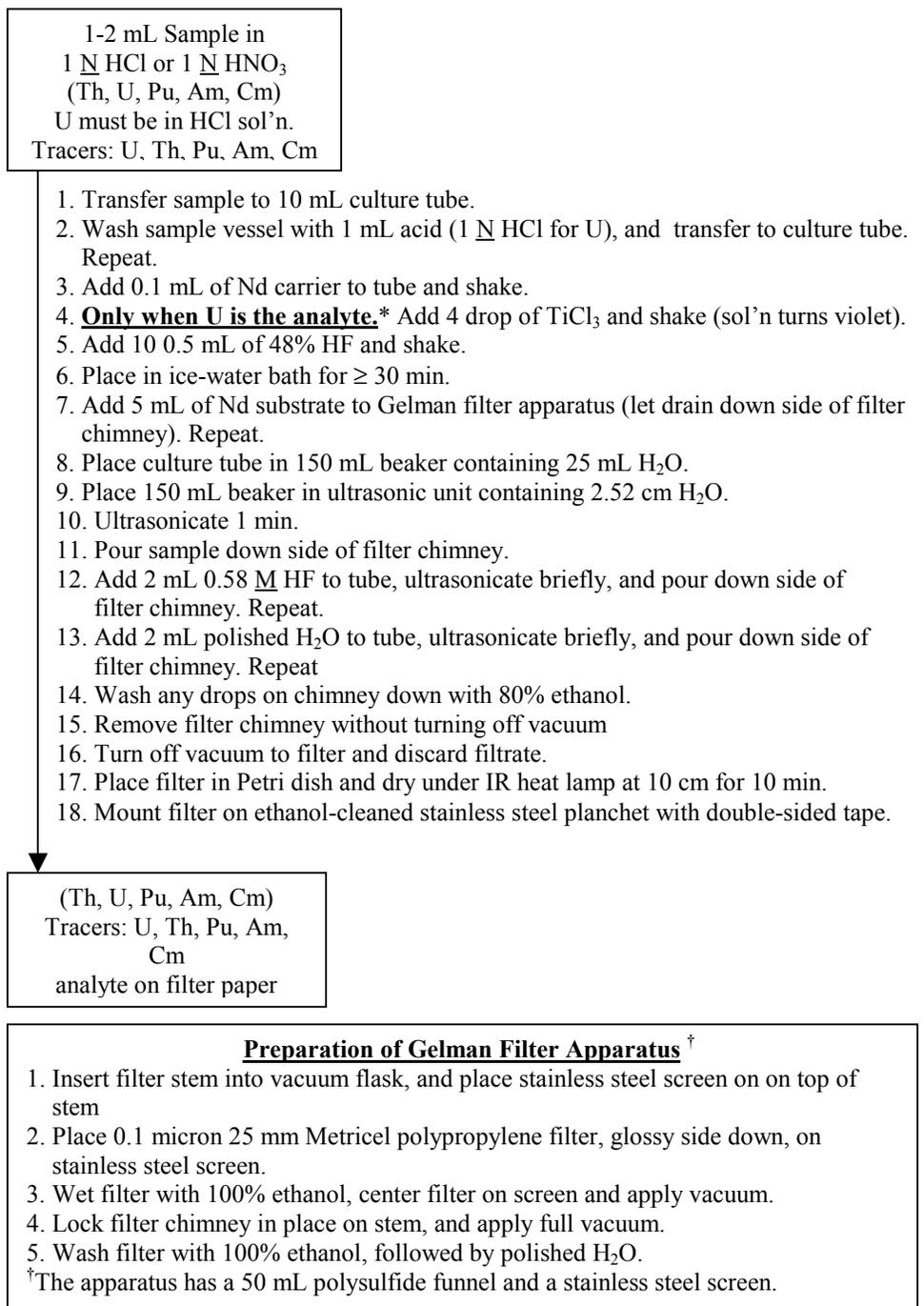


*The U procedure has one more step, the addition of TiCl₃, than the Th, Pu, Am, and Cm procedures.

Figure 7. Co-precipitation of Th, U, Pu, Am, and Cm with CeF₃

Co-precipitation of Th, U, Pu, Am, and Cm with NdF₃.

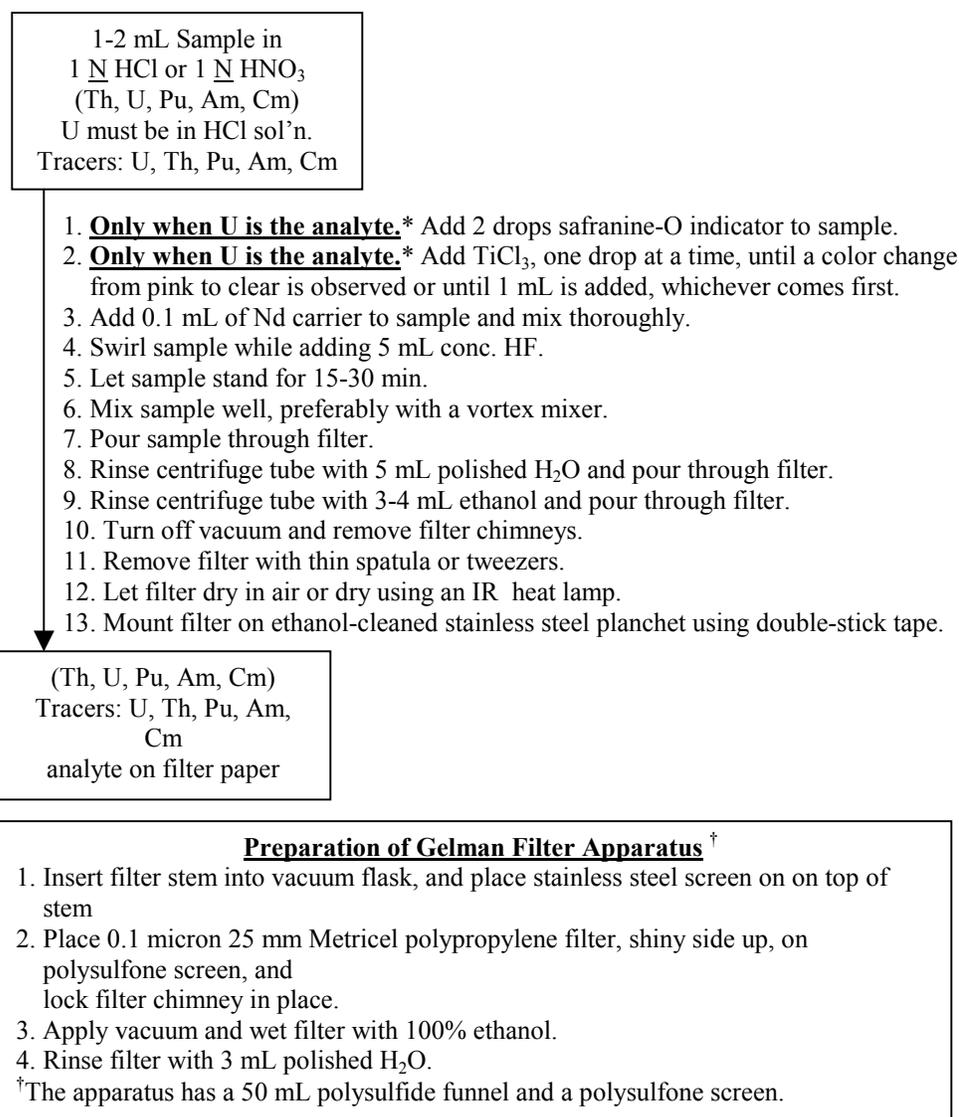
(Adapted from HASL-300, 28th Ed. procedure G-03 Rev. 0.)



*The U procedure has one more step, the addition of TiCl₃, than the Th, Pu, Am and Cm procedures.

Figure 8. Co-precipitation of Th, U, Pu, Am, and Cm with NdF₃.

Co-precipitation of Th, U, Pu, Am, and Cm with NdF₃. (Canberra procedure.)



*The U procedure has two more steps, the addition of safranin-O and TiCl₃, than the Th, Pu, Am, and Cm procedures.

Figure 9. Co-precipitation of Th, U, Pu, Am, and Cm with NdF₃

Po Determination in Water.

(Adapted from procedure Po-02-RC from Vol. 1HASL-300, 28th Ed.)

1. Transfer 2.5 L (out of 10 L) of raw water to 4-L beaker.
2. Add 50 mL HNO_3 and 1 mL Pb carrier sol'n.
3. Add 30-80 mBq Po-209 tracer.
4. Evaporate and add aliquots of water until 10 L has been reduced to 25 mL.
5. Transfer sol'n to two 50-mL centrifuge tubes.
6. Reduce volume to 5 mL in steam bath.
7. Add 50 mL of polished H_2O .
8. Adjust pH to 3.5-4.0 with NH_4OH .
9. Add 5 mL thioacetamide sol'n.
10. Digest in steam bath for 1 hr.
11. Cool, centrifuge, and decant supernatant. Discard supernatant.
12. Dissolve precipitate in 2 mL of HCl.
13. Add 50 mL polished H_2O .
- 13 Adjust pH to 3.5-4 with NH_4OH .
14. Add 2 mL of thioacetamide.
15. Digest in steam bath for 1 hr.
16. Cool, centrifuge, and decant supernatant. Discard supernatant.
17. Dissolve precipitate in 1 mL of HCl.
18. Dilute the solution to 25 mL with polished H_2O .
19. Filter sol'n with Whatman No. 41 filter paper into prepared deposition cell (see figure below).
20. Wash filter with hot 0.5 N HCl.
21. Add 1 mL sat'd ascorbic acid sol'n to solution in cell.
22. Place cell in 80°C water bath.
23. Stir with Teflon stir bar for 4 hr. at a speed of maximum agitation.
24. Remove cell from water bath, and pour off sol'n into a beaker.
24. Place disc on a warm hotplate to dry.

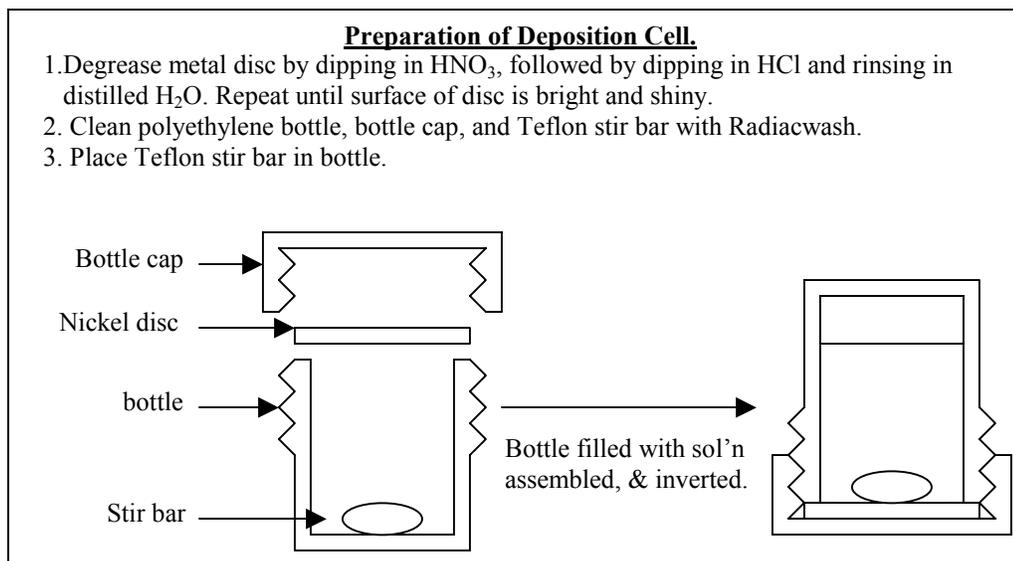


Figure 10. Procedure Po-02-RC: Determination of Po-210 in Water.

Po Determination in Water.

[Adapted from procedures Po-01-RC and Po-02-RC from Vol. 1, HASL-300, 28th Ed. (Nov. 1990)]

1. Place 1 L of sample water in 2-L beaker.
2. Add 50 mL HCl.
3. Evaporate to a volume of 20 mL.
4. Place sol'n in a prepared deposition cell (see figure above).
5. Add 1 mL sat'd ascorbic acid to solution in cell.
6. Place cell in 80°C water bath.
7. Stir with Teflon stir bar for 4 hr. at a speed of maximum agitation.
8. Remove cell from water bath and pour off sol'n into a beaker.
9. Place disc on a warm hotplate to dry.

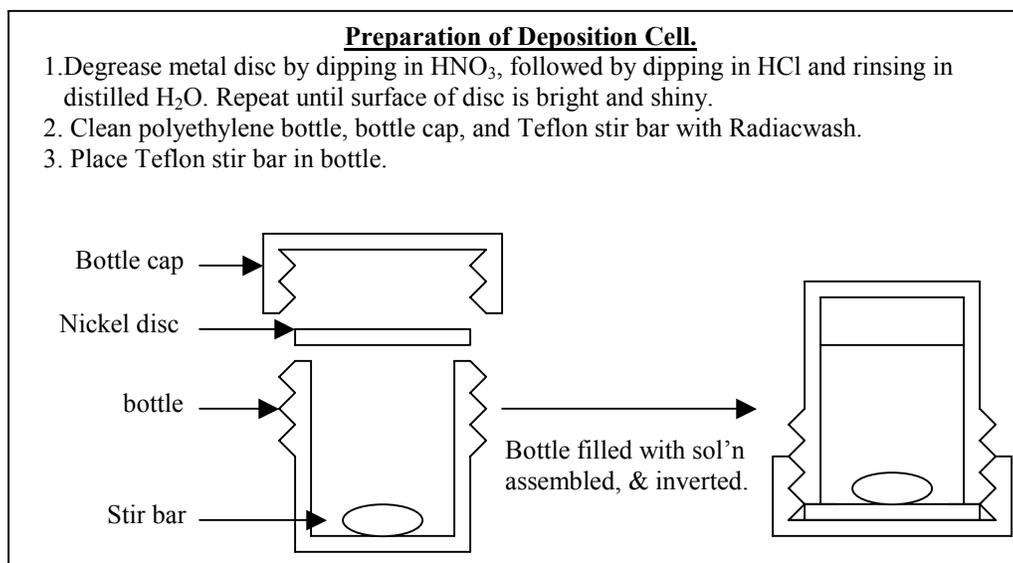


Figure 11. Procedure Po-01-RC: Determination of Po-210 in Water.

Flowchart for U-Th separation: Evaporation option.

(Adapted from Eichrom method ACW01 Rev. 1.4.)

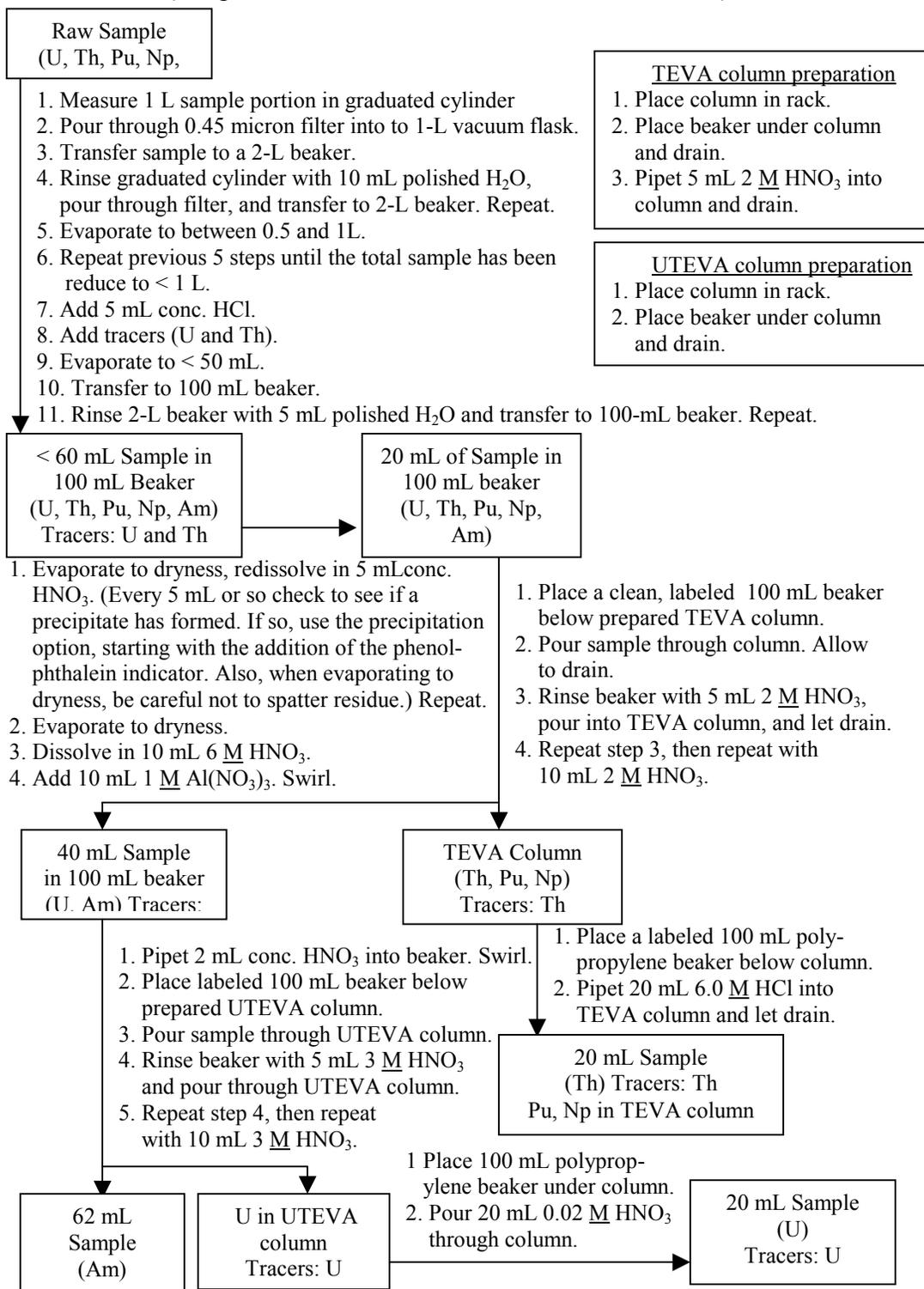


Figure 12. Flowchart for U-Th separation: Evaporation option

Flowchart for U-Th separation: Precipitation option.

(Adapted from Eichrom method ACW01 Rev. 1.4.)

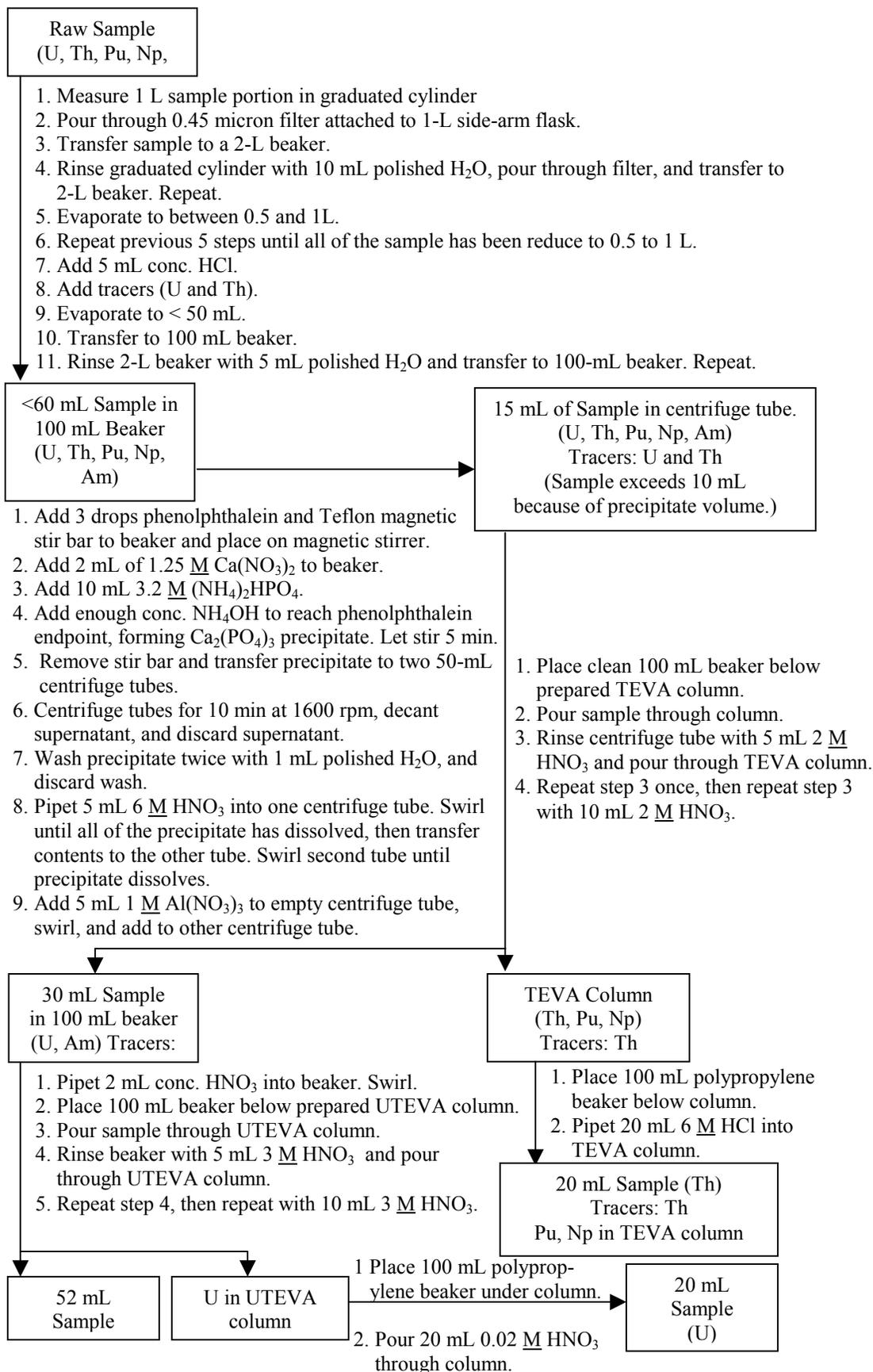


Figure 13. Flowchart for U-Th separation: Precipitation option

References

¹ C. W. Sill and D. G. Olsen *Analytical Chemistry* **42**, 1596-1607 (1970).