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I am submitting herewith a dissertation written by Rose Ann Boll entitled "Development of an Improved Environmental Actinide Separation Method." I have examined the final copy of this dissertation for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, with a major in Chemistry.

George K. Schweitzer, Major Professor

We have read this dissertation
and recommend its acceptance:

Accepted for the Council:

Associate Vice Chancellor and
Dean of The Graduate School

DEVELOPMENT OF AN IMPROVED ENVIRONMENTAL ACTINIDE SEPARATION METHOD

A Dissertation

Presented for the

Doctor of Philosophy

Degree

The University of Tennessee, Knoxville

Rose Ann Boll

May 1996

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DEDICATION

The author would like to dedicate this work to her parents, Mary and Herman Boll, whose love, support, and encouragement are always with her. This work is also dedicated to the author's brothers and sister, Ed, Mary, John, Paul, Rita, Ralph, Carol, Fred, Ruth, and Anna, because tumbling rocks move each other along and smooth the rough edges.

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And last but not least, the author wishes to thank her constant studying companion, Pythagoras B. B. Bollamb.

ABSTRACT

The focus of this research project was on the separation of five actinide elements (Th, U, Np, Pu, and Am) from each other in aqueous media. Many of the current separation techniques are cumbersome, time consuming, costly, create large volumes of waste, and have moderate recoveries. In developing a separation method, the unique chemistries of the actinides were taken into consideration. These include oxidation state variation, complexation, trace element behavior, hydrolytic behavior and matrix interactions.

The overall process for the determination of actinides in environmental samples can be divided into five general steps: sample solubilization, elimination of matrix interferences, separation of the actinide ions from each other, preparation of separated fraction for counting, and alpha particle counting of the separated samples. Each of these steps was investigated.

The majority of the investigations in this project relates to the most difficult step, the separation of the actinide ions from each other. Selective coprecipitations using zirconium phenylarsonate, calcium-magnesium-yt trium fluorides, neodymium fluoride, and metal iodates were tried. These were unsatisfactory because good percents recovery required large amount of precipitate which resulted in poorly resolved spectra. The new EIChroM TRU-Resin™ extraction resin was investigated and showed good promise for the separation of the actinide ions. Therefore, various tests were conducted in order to develop a separation procedure using the TRU-Resin™ resin. These investigations included evaluations of different eluting reagents, adjustments in the concentration of reagents, separations in the presence of high concentrations of certain actinide ions, changes in pH, and the effects of different redox reagents. The various trials were evaluated by percents recovery of the actinide ions.

The final developed method utilizes a single-one milliliter TRU-Resin™ resin column. In order to cleanly recover the five actinides from a solubilized soil matrix, the sample is passed through the column twice. The sample is first loaded in 5.0-M HCl with hydrogen peroxide as the

redox reagent. This allows the Am and most matrix ions to pass through the column. The Th is eluted first using 1.3-M HCl, then the Np and Pu are eluted together with 0.030-M oxalic acid in 0.5-M HCl. Finally, the U is eluted with 0.10-M ammonium oxalate. A calcium-oxalate coprecipitation is performed on the Am sample and the dissolved precipitate is then reloaded onto the TRU-Resin™ column in 3.0-M HNO_3 with ascorbic acid as the redox reagent. The overall procedure requires approximately 1.5 working days to perform with percents recovery of the five tested actinides in the 80-100% range.

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CHAPTER I

INTRODUCTION

A. The Problem

In our country and the world today there is a growing awareness and concern about the effects of contamination on our environment and our health. One of the contaminates of great concern is radioactive waste. Lawsuits have been filed in an attempt to seek compensation for the effects of previous radiation exposure to the health of many Americans.[1] Yet the environmental contamination problem is not simply in the past but is a present problem and will be a crisis of the future without proper cleanup.

Most of the radioactive contamination of concern has resulted from the Manhattan project during WWII and the build up of a nuclear arsenal during the cold war.[1] Uranium was bombarded with neutrons in graphite reactors to produce plutonium. This resulted in waste material containing uranium, plutonium, transuranium elements, and decay daughters of these elements.[2]

Part of the initial cleanup process is accurate identification and quantification of what is present in the environmental samples. If this is not identified properly, large amounts of money could be spent to clean up contamination that is not even present or, possibly worse, sites of radioactive contamination could be labeled safe resulting in hazardous exposure to individuals.

The alpha emitting isotopes present in radioactive waste pose a serious threat to the health of our environment. As the elements become part of our air, water, and soil, they are incorporated into the plants and animals that provide food sources for people. This results in ingestion by humans of alpha emitting substances which usually target different organs and can result in extensive destruction of tissue.

The problem that is to be addressed in this work is the separation of five of the actinide elements that are often found in the radioactive waste resulting from the nuclear weapon build up era. These alpha emitting elements are thorium, uranium, neptunium, plutonium, and americium.

Once they are separated into individual elements, they can be identified and quantified using alpha particle detectors.

B. The Need

There is a need to separate the five actinides because no detection method is available that can count all of them together and produce separated alpha emission spectra. The best current equipment for detection of the energies of emitted alpha particles has limited resolution capabilities. PIPS (Passivated Implanted Planar Silicon Detector), made by Canberra Industries, are currently among the best surface barrier detectors on the market. Canberra states that a PIPS detector with a 600 mm^2 active area can provide a resolution of ±25 keV.[3] In practice, especially with a mixture of alpha emitters and the complication of sample scattering and self adsorption, the best that can be obtained is about ±35 keV. Thus with even the best resolution, the energy regions of the emitted alpha particles from the different elements will overlap on the printouts if they are counted together. This results in a need to separate the different elements from each other prior to alpha detection.

Many of the current separation techniques are cumbersome, time consuming, costly, create large volumes of waste, and have moderate recoveries. Therefore, a need exists for a separation method that will be simpler, quicker, more cost effective, less productive of waste, and improve recoveries.

Any separation procedure will need to take into consideration the unique chemistry of the actinides. This includes oxidation state variation, complexation, trace element behavior, and matrix interaction. The actinides show an unusually broad range of oxidation states, ranging from +2 to +7.[4] Some of them favor one oxidation state in aqueous solution (e.g. Th) while others exist in water in four oxidation states when at equilibrium, (e.g. Pu).[4] The five elements need to be detected at extremely low levels (pCi). This means that they exist in very low concentrations (10^{-3} to 10^{-15} M). Their behaviors may take on some unique properties which are characteristic of

trace level solution chemistry (e.g. adsorption).

The separation process will also need to be able to isolate the desired actinide elements in the presence of other substances introduced by the matrix of the sample. For example, a soil matrix may contain high levels of iron, calcium and/or aluminum.

C. Proposed Improvements

The current techniques for separation of low levels of actinide elements include precipitation, solvent extraction, volatilization, anion and cation exchange resins, and recoil separations.[2] In order to separate the elements in a sequential procedure, a mixture of the above techniques is often utilized.

An example of typical market procedures can be demonstrated by the separation methods of the IT Radiochemical Division in use in the fall of 1992.[5] Their procedure for the sequential separation of one sample containing Th, U, Np, Pu, and Am includes the following:

4 columns, (AG1-X8 and AG1-X4 resins)

9 different redox and/or complexing reagents

4 organic eluant and/or organic/acid eluents (about 300 ml)

8 N HNO_3 - 900 ml

conc. HCl and 8-M HCl - 230 ml

dilute HCl - 275 ml

concentrated NH_4OH - 20 ml

2 co-precipitation procedures

9 wet ashing procedures (drydowns)

This procedure generates, per sample, approx. 480 ml of acid waste, 50 ml of base waste, 275 ml of organic waste, and approx. 35 cm^3 of waste resin. After the sample is digested, the procedure requires approximately 8 eight-hour-workdays to complete the separation. Acceptable yields for alpha tracers are between 20% and 120% [International Technologies Analytical Services (ITAS)-

7301]. The normal percents recovery for the above methods usually has the following ranges: Am 90±10%, U 70±10%, Th 45±10%, Np 80±10% and Pu 80±10%.[54]

After review of the current procedures, it was determined that there are several possible improvements that could be developed.

1. The use of a different co-precipitation methods to separate one element or group of elements from the others.
2. The elimination of one or more of the currently used co-precipitation methods by finding a different process to deal with matrix interferences.
3. The use of new extraction resins that could replace some of the more labor intensive columns with shorter and quicker procedures.
4. The development of a sequential procedure where all the desired elements are separated by the loading and eluting of one column.
5. The improvement of the initial sample processing to eliminate matrix interference that complicate the separation process.
6. The reduction of the amount of resin waste by reducing the size of column. This would also reduce the reagent waste volume by decreasing the amount of reagent needed to prepare a column, to elute a column, and to rinse a column.
7. The reduction of cost by reducing reagent usage.
8. The reduction of cost by decreasing the time it takes to do the separation procedure.
9. The increase of income by increasing the number of samples performed in a given time period.
10. The elimination of the disposal expense of mixed waste by replacing the methanol column with an aqueous column procedure.
11. The improvement of tracer recoveries by more efficient loading and eluting procedures.

12. The improvement of tracer recoveries by optimizing the conditions for NdF_3 precipitation prior to counting.

13. The development of a technique that has more matrix tolerance then the current methods.

CHAPTER II

HISTORICAL INFORMATION

A. Actinide Elements

The actinide series consists of the 14 elements that follow actinium on the periodic table, atomic numbers 90-103, and all known isotopes are radioactive. The presence of the 5f shell in the electronic configuration of these elements produces some unique chemical properties. The 5f and 6d shells are of similar energy, therefore the redox chemistry of these elements can be extremely complicated involving many different oxidation states, especially for the elements just after the first two members of the actinide series. As the 5f shell is filled, the increasing nuclear charge brings about a successive contraction, analogous to the lanthanide contraction. The shell will then be better shielded from the fields of neighboring atoms and simultaneously the energy of the 5f level drops, causing the difference between the 5f and the outer orbitals to grow. As a consequence, the participation of the 5f electrons in bonding becomes more difficult, resulting in a very marked stabilization of the trivalent oxidation state for the heavier actinides. [11]

The oxidation states of the earlier actinide elements range from II to VII. The aqueous ions normally formed by the these oxidation states are M^{+2}, M^{+3}, M^{+4}, MO_2^{+}, MO_2^{+2}, and MO_5^{-3}. The M(VII) species have been reported for neptunium, plutonium and americium but are unstable and poorly characterized.[4] These cations are all electron acceptors which coordinate solvent molecules in solvents which exhibit donor properties. The affinity is generally stronger, the more highly charged the acceptor. In the species MO_2^{+} and MO_2^{+2}, the effective charge on the acceptor atom M is considerably higher than the total charge of the entities. These are linear groups in which the oxygens carry a net negative charge.[11] In the case of MO_2^{+2}, the effective charge is thought to be about +3.3 rather than +2.[4]

In aqueous solution, protons may split off from the water molecules of the hydrated actinide

cations resulting in Brönsted acids. In the M^{+3} cations, due to the large radius of the central ion, the repelling force between the protons and the ionic charge is relatively weak. The M^{+4} ions, due to the higher charge of the central ion, form much stronger acids than the M^{+3} ions. However, the acid properties are fairly weak, generally much weaker than for most other hydrated cations of the same charge. An important consideration when dealing with actinide hydrates is that upon the dissociation of a proton, the residual species often condense into polynuclear complexes. The mean number of actinide atoms in these complexes generally increases with pH and with the total concentration of the actinide. The formation of the polynuclear complexes increases as the effective charge of the cation increases, thus the tendency to undergo hydrolysis decreases in the order M+4 > MO_2^{+2} > M^{+3} > MO_2^{+}.[4] The formation of these very high-molecular weight polymers of the solution ions can result in problems. Slow reactions are generally observed to take place in the later stages of the hydrolysis of the cation. Also, once formed, these polymers do not readily disperse in solutions of acidities that would usually prevent their formation. Thus, various complex forms may coexist in appreciable amounts in the same solution.[11]

The complexes formed between the early actinide cations and the halide anions are important. All actinide cation acceptors with oxidation states of III or greater form strong complexes with fluoride ions in aqueous solution. With the fluoride ion, the strength of the complexes follow the general pattern: M^{+4} > MO_2^{+2} > M^{+3} > MO_2^{+}. In contrast, the complexes formed with the heavier halides are all very weak. The strengths of the complexes decrease as the size of the halide ion increases. The simple fluorides, MF_3 and MF_4, are relatively insoluble compounds. Fluorides of the actinyl ions (MO_2^{+}, MO_2^{+2}) are very soluble in water as are the heavier halides of all the oxidation states of the actinides. One must remember that hydrolytic reactions are prone to occur when solutions are not sufficiently acidic thus changing the complex. However, the fluoride ion forms such strong complexes with the actinide cations that the complexes are relatively resistant to hydrolysis.[11]

The nitrate, sulfate, and phosphate anions coordinate through oxygen to the actinide

acceptors yet the complexes they form are not the same. The nitrate anion forms very weak complexes, no stronger in fact than those of the heavy halide ions. Extensive complex formation only occurs at quite high nitrate concentrations. Sulfate, on the other hand, forms a much stronger complex. This is believed to be due to increased electrostatic interaction with the higher charge of the anion. With the sulfate ion, the strength of the complexes follow the general pattern: $M^{+4} > MO_2^{+2} = M^{+3} >> MO_2^{+}$. The various phosphate ions (PO_4^{-3}, HPO_4^{-2}, $H_2PO_4^{-}$) act preferably as bridges, usually forming slightly soluble aggregates with the actinide cations. Generally, different complexes are formed, each one stable within certain limits of phosphate concentration and acidity of the solution. With increasing acidity, the hydrogen ions become more and more efficient competitors for the oxygen donor atoms of the phosphate ions.[11] See Appendix B, Table III[1] for complex formation constants.

B. Chemistry of the Pertinent Actinides

The actinide elements of specific interest in this project are thorium, uranium, neptunium, plutonium, and americium.

1. Thorium, Th

Thorium has the atomic number 90. The electronic configuration of the neutral atom is $[Rn]6d^27s^2$. [13] The only stable oxidation state of thorium in aqueous solution is +4. The +3 oxidation state has been shown to exist in solids only.[15] Thorium compound chemistry resembles that of tetravalent cerium and, to some extent, tetravalent titanium, zirconium, and hafnium.[14] In aqueous solution the hydrated Th^{+4} is probably 8- or 9-coordinated.[15]

Because Th^{+4} is a highly charged ion, its aquated cation has a tendency to split-off protons and function as a fairly strong acid. This hydrolysis is followed by polymerization. The aquated, dimeric ion, $[Th_2(OH)_2^{+6}]$, probably involves two hydroxide bridges and appears to

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[1]Tables III through CXIII can be found in Appendix B.

predominate until about 2.5 protons have split off per thorium cation. As the hydrolysis proceeds, the degree of polymerization starts to increase very rapidly, and before the number of protons split off per thorium cation has reached 3, the complexes seem to contain, on the average, well over 100 thorium ions. Eventually, the continued complexation will yield an amorphous precipitate of thorium hydroxide.[11,15]

The hydrolytic behavior of Th^{+4} appears quite different from that of U^{+4} and Pu^{+4}. The first step in the hydrolysis of these latter two species is the monomeric reaction: $M^{+4} + HOH <---> MOH^{+3} + H^{+}$. This is followed by slow irreversible polymerization of the hydrolysis products. For thorium, on the other hand, the monomeric reaction is unimportant and even in very dilute solution various polymeric species, such as $Th_4(OH)_8^{+8}$, are formed reversibly, with various equilibria set up between them.[19]

The development of the hydroxide polymer complex is also related to changing molal concentration of the thorium ion. In a 1-m solution of perchlorate, the monomer is the predominant species when the concentration of the thorium ion is 0.001-m. By the time the thorium concentration has increased to 0.1-m, the dimer, not the monomer, will be the main species formed initially.[11].

There are 26 known isotopes of thorium.[6] Because of its long half life (1.405E10 y), Th-232 is the most abundant radioactive element in the earth's crust.[7,17] Other thorium isotopes are present in much smaller quantities in the earth's surface as a result of contamination from the nuclear industry or as decay daughters in natural occurring decay chains. Th-227 (18.7 d), Th-228 (1.9 y), Th-230 (7.5E4 y), Th-231 (25.5 h), and Th-234 (24.1 d) are found as decay daughters in the natural radioactive decay series. (See Appendix A, Figure 20.) All other known isotopes of thorium have half lives of minutes or less except for Th-229 which has a half life of 7.3E3 years. Th-229 is a member of the neptunium decay series which is the 4n+1 series that is thought to be missing in nature.

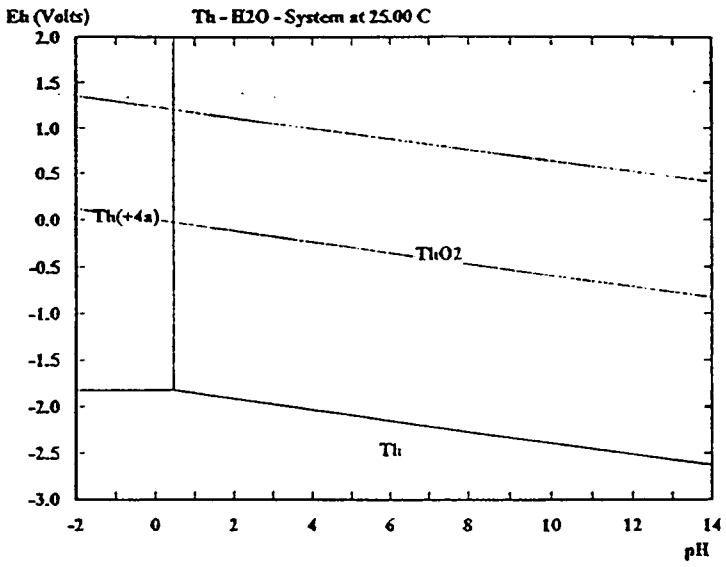
The primary decay mode of thorium-227, -228, -229, -230, and -232 is alpha emission.

Th-231 and -234 decay by beta emission. The energy of the alpha emissions are listed in Appendix B, Table IV.

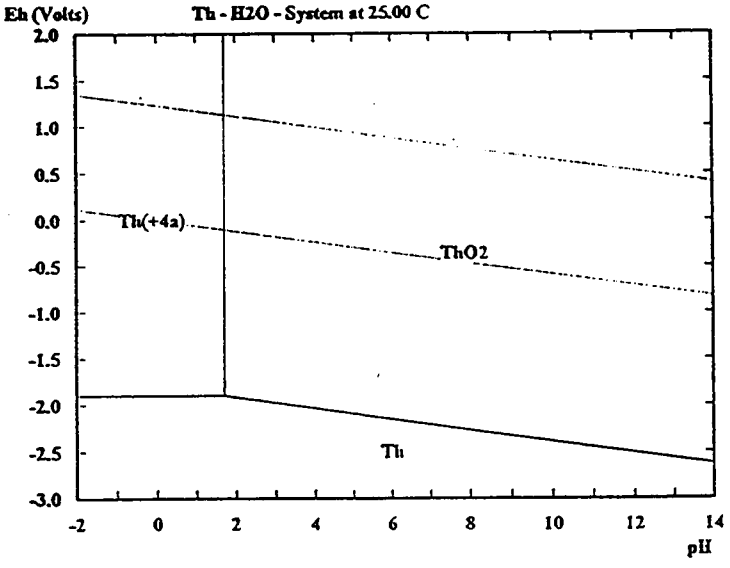
The aqueous solution equilibria for an element can be illustrated by making a plot of potential (E) versus pH. This type of plot is known as a Pourbaix diagram, having been named after Marcel Pourbaix, who was instrumental in its development.[18] It is also known as a potential-pH diagram or Eh-pH diagram. On the Pourbaix diagram, vertical lines represent a junction at which a change in pH produces a different species of the element, but no change in oxidation state. Horizontal lines indicate junctions at which changes in E result in oxidation state changes which are not influenced by pH changes. Sloping lines represent the junction at which the production of a different species is influenced by both E and pH changes. Areas in between the lines represent regions in which the indicated species are the predominant ones and all other species are present in smaller amounts.[9] The upper and lower dashed lines on a Pourbaix diagram represent the decompositions of water into O_2 and H_2 respectively. When the solution contains or contacts an oxidant with an E value above the upper water line, the species will oxidize OH^- or water to O_2 and will itself be reduced. When the solution contains or contacts a reductant with an E value below the lower water line, the species will reduce H^+ or water to H_2 and will itself be oxidized. Any species on the diagram that lies entirely outside the region of the water lines will be thermodynamically unstable in water solution, but may, however, be kinetically stable.[10]

The Pourbaix diagram will be used to illustrate the chemistry of the various actinide elements utilized in this research. The Pourbaix diagram for thorium is shown in Figure 1. The top Pourbaix diagram illustrates the species of thorium that exist in water solution when Th^{+4} is at a 1 molar concentration. However, the concentration of Th^{+4} in environmental samples is often much less. The typical level for which tests were performed in this project was 20 disintegrations per minute (dpm) of thorium. That amount of activity is produced by 3.4E-7 moles of Th-232 atoms. When it is dissolved in 25 ml of solution, the thorium ion concentration is 1.4E-5 Molar. The solution chemistry of Th-232 at that concentration is illustrated on the second Pourbaix

(a) Molality m mol / kg
Th 1.000E+00



(b) Molality m mol / kg
Th 1.000E-05



(c) Molality m mol / kg
Th 1.000E-12

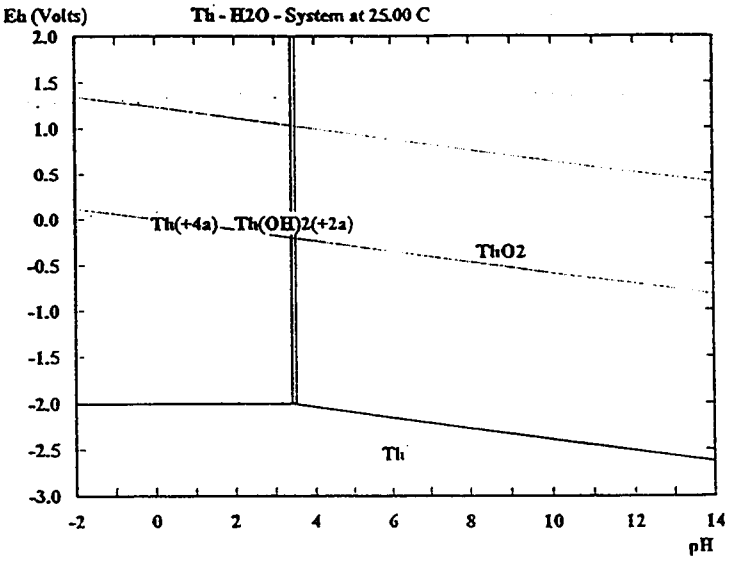


Figure 1: Pourbaix Diagrams of Th ions in Aqueous Solution: (a) 1.0-M , (b) 1.0E-5 M, (c) 1.0E-12 M.

diagram in Figure 1. Note the change in the pH value of the vertical line between the Th^{+4} species and the ThO_2. At the more dilute concentration, the Th^{+4} ion will continue to exist at a slightly higher pH as the predominant species in solution. If the detection of 20 dpm of Th-229 is desired, one is working 1.8E-13 moles of thorium atoms due to the shorter half-life of the isotope. In 25 ml of solution, the concentration of the thorium ions would be 7.4E-12 M. This is illustrated by the third Pourbaix diagram in Figure 1.

The thorium Pourbaix diagram shows only the one aqueous ion, Th^{+4}, one oxide, ThO_2, and the element, Th. The transition from Th metal to Th^{+4} occurs at approx. -1.7 volts between pH -2 and 2. The oxidation of Th to ThO_2 is a slanted line showing that the E value is dependant on pH. As the pH

increases, it becomes easier to remove the electrons from Th. The various complexes formed by the hydrolysis of Th^{+4} are not noted specifically on the Pourbaix diagram. They are included in the region designated by the predominant species Th^{+4} in order to simplify the overall diagram.

2. Uranium, U

Uranium has the atomic number 92. The electronic configuration of the neutral atom is $[Rn]5f^36d^17s^2$.[4] The known oxidation states for uranium are III, IV, V, and VI. By observing the Pourbaix diagram (Figure 2), it can been seen that the U^{+4} and UO_2^{+2}, representative of IV and VI oxidation states of uranium, exhibit stability in aqueous solution. The standard electromotive force (E^o) for the transition from UO_2^{+2} to U^{+4} in a 1-M H^+ solution is -0.333 volts.[18] U^{+3} is a strong reducing agent and therefore is difficult to stabilize as a simple aquated ion in water solution.[4] (Its region of prominent stability is below the lower water line on the Pourbaix diagram.) UO_2^+, the simple aqueous ion form of oxidation state V, is not present on the Pourbaix diagram because it is not stable in aqueous solution, rather UO_2^+ rapidly disproportionates to the U^{+4} and UO_2^{+2}.[15] Also notable on the Pourbaix diagram, is the difference in pH of the vertical line indicating the transition from UO_2^{+2} predominance to UO_3 predominance compared to the pH of the vertical line indicating the transition from U^{+4} predominance to UO_2 predominance. This

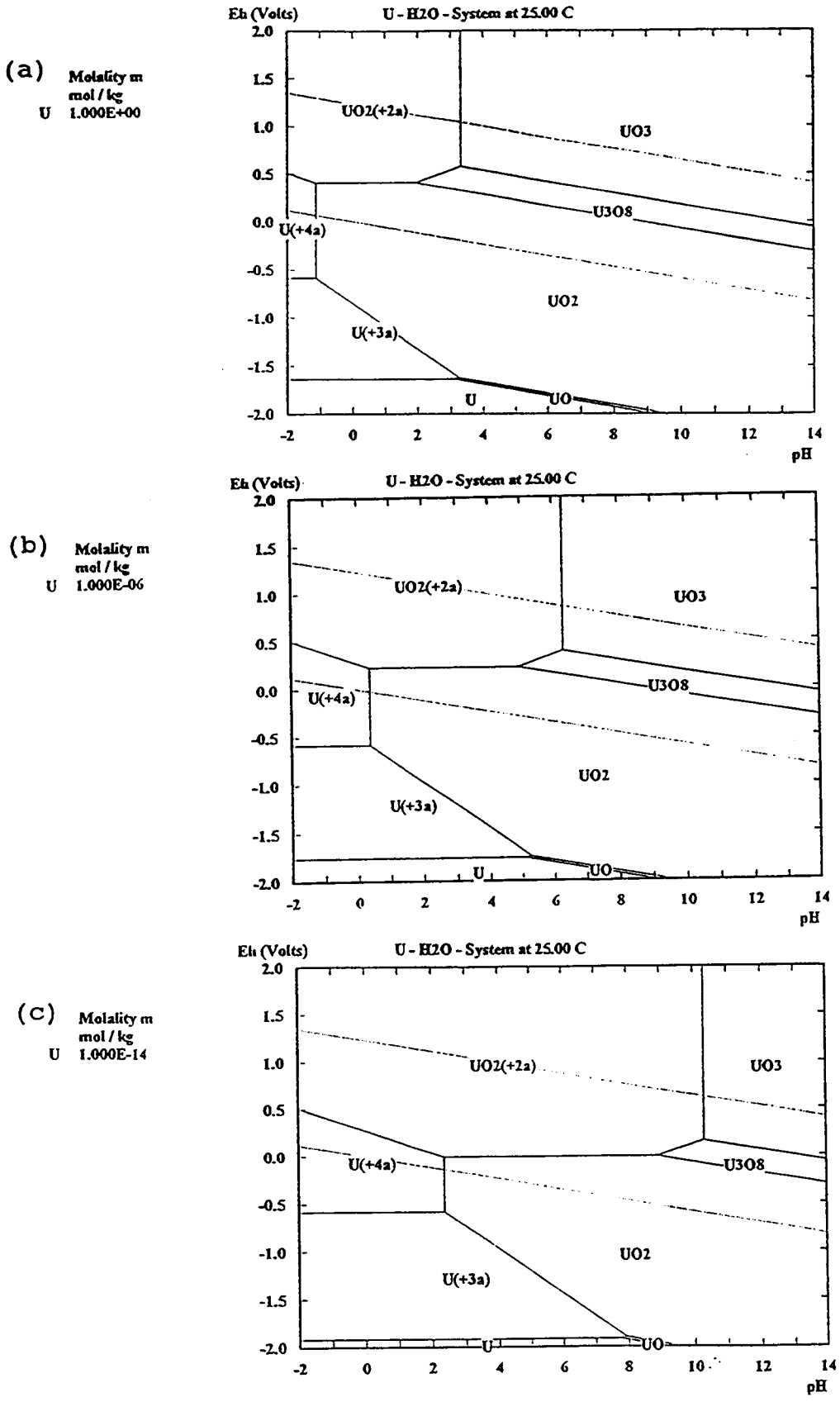


Figure 2: Pourbaix Diagrams of Uranium in Aqueous Solution: (a) 1.0-M, (b) 1.0E-6 M, (c) 1.0E-14 M.

indicates that the form and oxidation state of the uranium ion dictates at what pH one can safely maintain the ion in aqueous solution.

The uranyl ion, UO_2^{+2}, is the most common simple aqueous ion of U(VI) oxidation state in aqueous solution. The ion has a linear geometry and the axial O-U bonds are very strong.[15] Partial hydrolysis of the uranyl ions is important and occurs in dilute acid solution. This tendency for hydrolysis is fairly high in MO_2^{+2} ion, yet slightly less than in the M^{+4} ions. The loss of a proton from a coordinating HOH starts around pH 3 [11] and is followed by polymerization involving hydroxide bridges and yielding species such as $[(UO_2)_2(OH)_2]^{+2}$, and $[(UO_2)_3(OH)_5]^{+}$.[15] The tendency to hydrolyze is closely related to concentration. As the uranium becomes more dilute in solution, the formation of bridged species decrease. In chloride media the ion with three bridged uranyl groups is important, while the UO_2^{+2} seems to be well established in nitrate media.[11] Including the oxygens of the uranyl group, the coordination number of the metal ion often reaches 8, with 7 and 6 donor groups also having been observed.[4,11]

The simple aqueous U^{+4} resembles the chemical behavior of Th^{+4} in most ways except that U^{+4} can be oxidized by O_2.[4]

There are 18 known isotopes of uranium.[6] U-238 (4.468E9 y) is the most abundant (99.2745%). U-235 (7.038E8 y) (0.720%) and U-234 (2.45E5 y) (0.0055%) are also found in nature. All three of these isotopes are involved in natural occurring decay chains. U-233 (1.592E5 y) is a member of the 4n+1 series. (See Appendix A, Figure 20.) U-236 (3.34E7 y) and U-232 (68.9 y) are decay daughters of Pu-240 and Pu-236 respectively. The other isotopes of uranium have half lives of less than a day.

The primary decay mode for the isotopes of uranium listed above is alpha emission. The energies of the alpha emissions are listed in Appendix B, Table IV.

3. Neptunium, Np

Neptunium has the atomic number 93. The electronic configuration of the neutral atom is $[Rn]5f^4 6d^1 7s^2$.[4] The known oxidation states for neptunium are III, IV, V, VI, and VII. The simple

aqueous ions of neptunium are Np^{+3}, Np^{+4}, NpO_2^{+}, NpO_2^{+2}, and NpO_5^{-3}. Neptunium occurs predominantly as NpO_2^{+} in aqueous solutions that have been in contact with air for a long time. However, depending on the method of preparation, fresh solutions may contain Np^{+4} and NpO_2^{+2}. Because of the comparatively slow reaction with the dissolved O_2 from air, it takes considerable time to convert Np^{+4} to NpO_2^{+}. Therefore, neptunium ions of different oxidation states may exist in aqueous solution at the same time.

NpO_2^{+2} has some stability in water in very low pH ranges but can act as an oxidizing agent upon water in more dilute acid solutions. (See Figure 3 for neptunium Pourbaix diagram.)

NpO_2^{+} is generally considered to be the most stable aqueous ion of neptunium. However, its stability is very sensitive to pH changes, the presence of other oxidation states of neptunium (e.g. Np^{+3}), and mild reducing agents in solution. NpO_2^{+} disproportionates at pH values less than -1, but is stable in most dilute acids.[11]

Np^{+4} exists as a simple ion in aqueous solution in a low pH range of less than 2. Np^{+4} is a weaker acid than its neighbors, but still coordinates water molecules and splits off protons, similar to Th^{+4}, resulting in polymer formation.[11] In solutions of approximately 1-M Np^{+4} concentration, as the pH increases the Np^{+4} will hydrolyze to the point of precipitation, but in trace solution (10^{-10} M), the Np^{+4} disproportionates before precipitation occurs.

Np^{+3} is a fairly strong reducing agent although less so than U^{+3}. Np^{+3} does not decompose water as U^{+3} does.[11]

The highest aqueous oxidation state is Np(VII), which has the electron configuration of $[Rn]5f^0 6d^0 7s^0$. The aqueous ion, NpO_5^{-3}, is a strong oxidizing agent and is very unstable in water solution.[11]

There are 17 known isotopes of neptunium.[6] Although there is no important natural source of neptunium, very small amounts of Np-237 have been shown to exist in nature as a consequence of continuous formation by the n,2n reaction on U-238.[2] Np-237 is the longest lived member of the 4n+1 decay series with a half-life equal to 2.14E6 years, but this half-life is far too

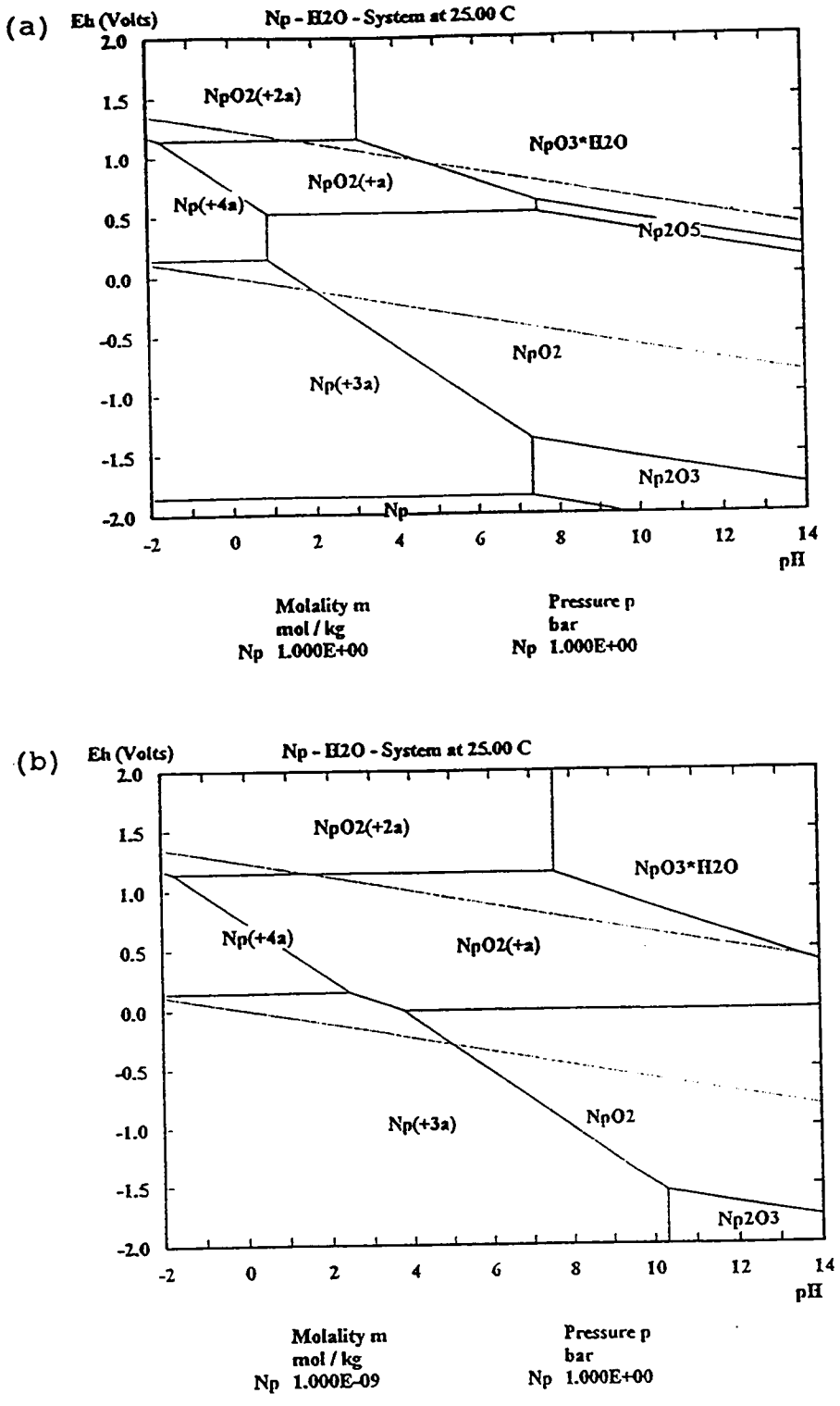


Figure 3: Pourbaix Diagrams of Neptunium in Aqueous Solution: (a) 1.0-M, (b) 1.0E-9 M.

short for the species to have survived since the time of the formation of the elements.[15] Large quantities of Np-237 have been produced as a by-product of the Pu-239 production from U-238. Np-237 is the only isotope of neptunium with a half-life greater than one hour that decays primarily by alpha emission. (Energies of the alpha emissions are listed in Appendix B, Table IV.) Np-236 (1.15E5 y) and Np-235 (396 d) decay primarily by electron capture. Np-238 (2.117 d) and Np-239 (2.355 d) decay by beta emission.

4. Plutonium, Pu

Plutonium has the atomic number 94. The electronic configuration of the neutral atom is $[Rn]5f^{6}7s^{2}$.[4] The known oxidation states for plutonium are III, IV, V, VI, and VII. The simple aqueous ions of plutonium are Pu^{+3}, Pu^{+4}, PuO_2^{+}, PuO_2^{+2}, and PuO_5^{-3}. (See Figure 4.)

The various aqueous plutonium ionic forms are often thought to resemble those of neptunium in many ways, however the ways that they differ become significant when attempting to separate them chemically. The PuO_2^{+2} is more stable in aqueous solution then its neptunium counterpart. PuO_2^{+} has a greater tendency todisproportionate than NpO_2^{+}.[19] Pu^{+4} is a stronger acid than Np^{+4} [11] and has a slightly higher tendency to partially hydrolyze and form soluble polymeric ions. The stability region of the Pu^{+4} is much less in water solution than Np^{+4}. Pu^{+3} is more easily produced and is a weaker reducing agent than Np^{+3}.[2]

The oxidation-reduction relationships for plutonium species are among some of the most intricate known to chemistry. Since all four simple aqueous ions show stability in acid solution, appreciable amounts of several aqueous ion forms, representing three or four different oxidation states, may exist together at equilibrium. For example, in 0.5-M HCl at 25°C, the equilibrium percentages of plutonium in the various ion forms in aqueous solution are approximately 27% Pu^{+3}, 58% Pu^{+4}, 1% PuO_2^{+}, and 14% PuO_2^{+2}.[4] Four significant equilibrium reactions taking place in an aqueous solution of plutonium ions are:[19]

$$2\ PuO_2^{+} + 4\ H^{+} \text{ <----> } Pu^{+4} + PuO_2^{+2} + 2\ HOH \qquad (2\text{-}1)$$

$$PuO_2^{+} + Pu^{+4} \text{ <----> } PuO_2^{+2} + Pu^{+3} \qquad (2\text{-}2)$$

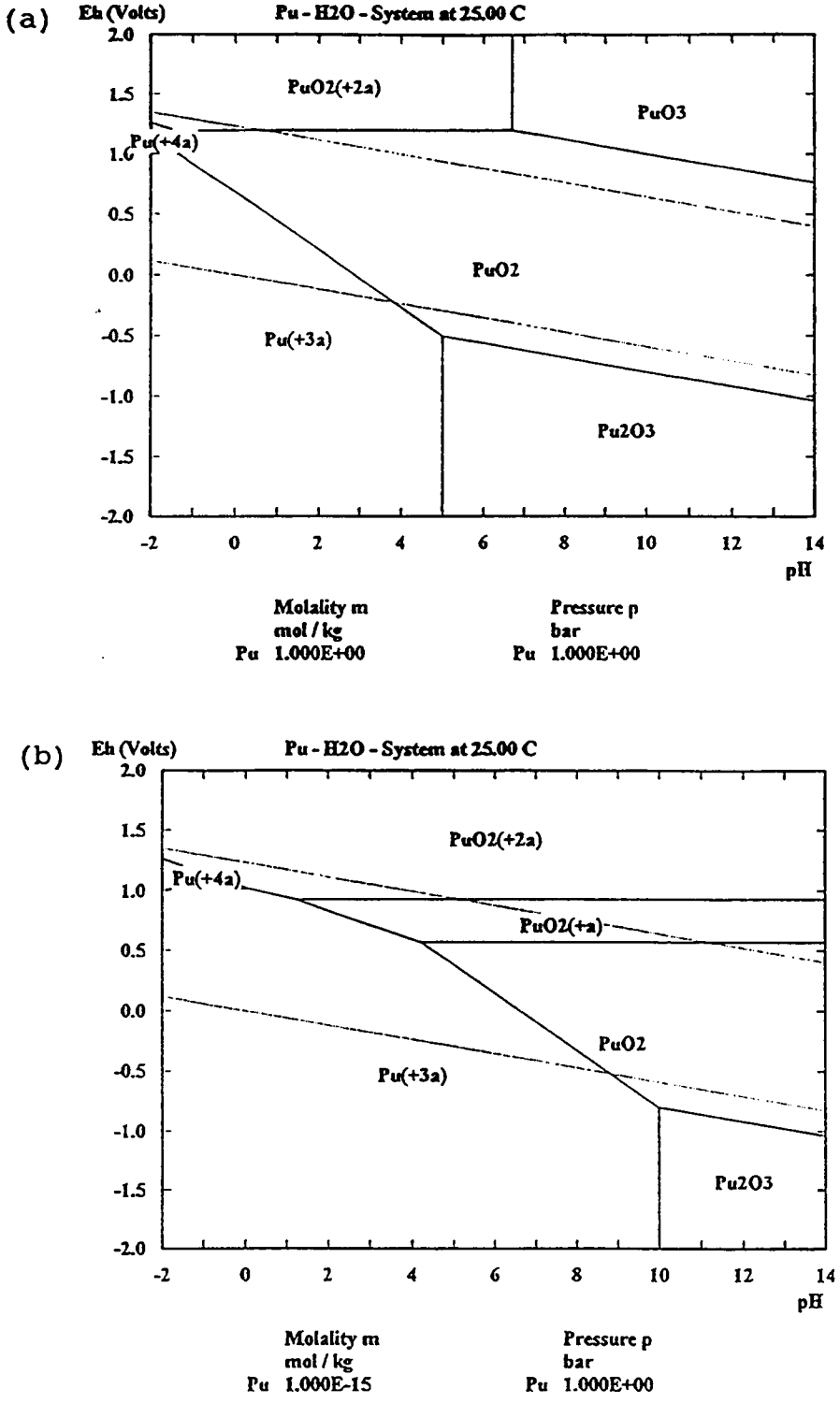


Figure 4: Pourbaix Diagrams of Plutonium in Aqueous Solution: (a) 1.0-M, (b) 1.0E-15 M.

$$PuO_2^{+} + Pu^{+3} + 4\,H^{+} \;<\!\!---\!\!>\; 2\,Pu^{+4} + 2\,HOH \qquad (2\text{-}3)$$

$$3\,Pu^{+4} + 2\,HOH \;<\!\!---\!\!>\; 2\,Pu^{+3} + PuO_2^{+2} + 4\,H^{+} \qquad (2\text{-}4)$$

All these reactions reach equilibrium quickly except for the disproportionation of Pu^{+4} ion, equation (2-4). This reaction involves partial hydrolysis of the Pu^{+4} ion and sequential the breaking of plutonium-oxygen bonds.

There are 17 known isotopes of plutonium.[6] Plutonium was the first synthetic element to be produced on a large scale. The isotope of interest was Pu-239 (2.4119E3 y) because of its abilityto fission. It was produced by the neutron bombardment of U-238.[2] Several other long lived isotopes of plutonium also were produced: Pu-236 (2.87 y), Pu-237 (45.2 d), Pu-238 (87.74 y), Pu-240 (6563 y), Pu-241 (14.35y), Pu-242 (3.733E5 y), Pu-244 (8.08E7 y), and Pu-246 (10.84 d). The plutonium isotopes that decay primarily by alpha emission are Pu-236, Pu-238, Pu-239, Pu-240, and Pu-242. The alpha emission energies are listed in Appendix B, Table IV.

5. Americium, Am

Americium has the atomic number 95. The electronic configuration of the neutral atom is [Rn] $5f^{7}7s^{2}$.[4] The known oxidation states for americium are II, III, IV, V, and VI. The simple stable aqueous ion forms of americium are Am^{+3}, AmO_2^{+}, and AmO_2^{+2}. An oxidation state of VII for americium has been claimed but not substantiated.[4]

Am^{+3} is the most stable aqueous ionic form of americium. This can be easily observed form the Pourbaix diagram in Figure 5. As the concentration of americium ions in solution decreases, the Am^{+3} form becomes even more predominant in aqueous solution. Americium(III) shows notable similarity to the lanthanide(III) ion chemistry. Coordination numbers of 6, 7, and 8 for the M^{+3} ions have been claimed.[11] The Am^{+3} ion forms a weak Brönsted acid with water. Hydrolysis of the trivalent ion takes place in a 1-M solution of americium near neutral pH values, while the more dilute solutions of americium do not hydrolyze until a much higher pH value is reached.

Americium has not been obtained in the divalent state in aqueous solution. Its presence

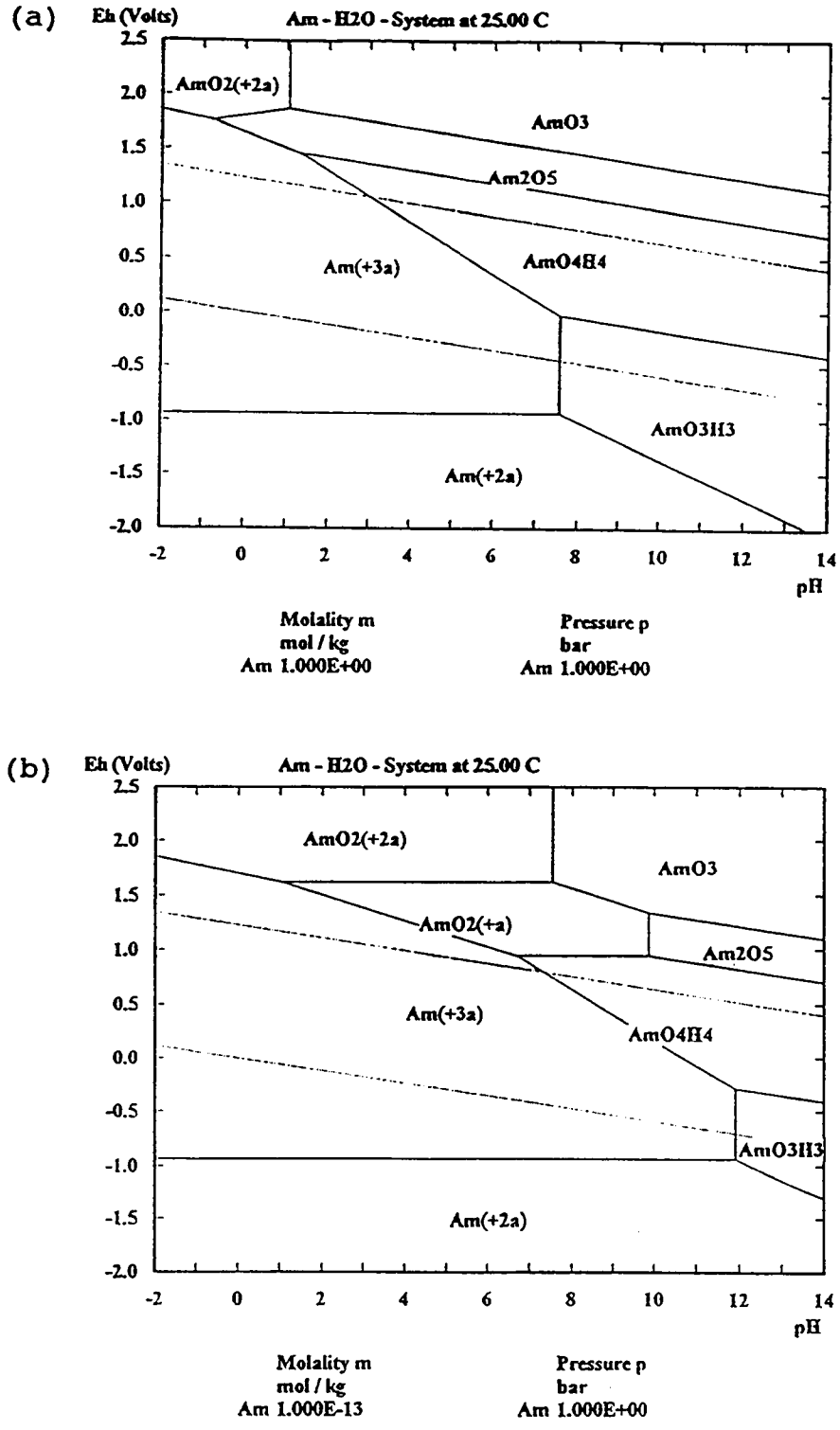


Figure 5: Pourbaix Diagrams of Americium in Aqueous Solution: (a) 1.0-M, (b) 1.0E-13 M.

has only been observed in solid compounds.[20]

Am^{+4} is present as solid americium dioxide but when dissolved in mild acid solutions, it disproportionates into Am^{+3} and AmO_2^{+2}. Therefore, Am^{+4} ion is not present as a simple aqueous ion. Small amounts of Am^{+4} complexes have been established in aqueous solution when the solid $Am(OH)_4$ is dissolved in a slightly basic solution of NH_4F.[20]

Americium can be oxidized to the pentavalent state in aqueous solution. However, AmO_2^{+} ion is unstable in an acid medium, and disproportionates into Am^{+3} and AmO_2^{+2}.

Hexavalent americium is formed from Am^{+3} by the oxidation in nitric or perchloric acid solution by ammonium persulfate or tetravalent cerium. However, AmO_2^{+2} has limited stability in water solution, in that it will slowly reduce in water to Am^{+3}. This reduction rate increases with increases in acidity and temperature of the solution. Hexavalent americium is a strong oxidant and is readily reduced in acid medium by many reducing agents to the trivalent state.

All tetravalent, pentavalent, and hexavalent simple aqueous ions of americium can be reduced in acid solution to the Am^{+3} ion on addition of hydroxylamine or hydrogen peroxide.[20]

There are 17 isotopes of americium.[6] Most of the isotopes have half lives of less than one day. Am-241(432.7 y), Am-242m (141 y), and Am-243(7380 y) are the only long lived isotopes. Am-241 and Am-243 decay primarily by alpha emission. (See Appendix B, Table IV.)

C. Tracer Level Chemistry

The term "tracer-level", when applied in the field of radiochemistry, generally implies unweighable quantities of material, less than 1E-8 grams. Undissolved quantities of this or lesser amounts are invisible, even under a microscope, and are generally below the sensitive level of normal chemical detection methods. In solution, tracer-level is usually assumed to mean less than 1E-7 M concentration.[19] Solutions of carrier-free radioactive isotopes often have tracer-level concentrations. Table V in Appendix B lists the concentrations of the different carrier-free isotopes used in this project when the activity of each isotope is 20 dpm per 25 ml of solution.

The behavior of substances at tracer-level concentrations may differ from that at macroscopic concentrations, and such differences are frequently predictable. An equilibrium that favors one set of products at normal or high concentrations may shift so that it lies in the opposite direction when the concentration of one of the reactants is greatly reduced. For example, a solution containing Ag^{+} and Cl^{-} ions at macroscopic concentration of the ions will result in the formation of a precipitate, AgCl. However, precipitation does not occur in tracer-level concentrations of 1E-11 M of the cation, even though the anion concentration may be 1-M. This is because the Ksp value of 1E-9.7 is not exceeded.

Another example of how a change in the relative concentrations can effect the species present is in the formation of complexes by a cation in the presence of an excess of anions. If the concentration of the cation is greatly reduced while the anion concentration remains high, a complex may be formed whose formation cannot be detected under macroscopic conditions. For example, in a mixture of macroscopic levels of Pb^{+2} and I^{-} ions, most of the Pb^{+2} will be in the form of a precipitate, PbI_2, because the Ksp value of 1E-8.1 is easily exceeded. The remaining Pb^{+2} will be in the form of Pb^{+2} or PbI^{+}, no notable percentage of negatively charged complexes will be formed. However when the Pb^{+2} ion concentration is lowered to about 1E-10 M and I^{-} is 1-M, no precipitates will be formed because the Ksp is not exceeded. Rather about 90% of the total Pb^{+2} ion concentration will be in the form of complex anions, $[PbI_n]^{-(n-2)}$. This may be shown by calculations using complexation constants and it has been demonstrated experimentally.[19]

The determination of electromotive potentials of metals in trace solutions has been difficult to do by traditional M/M^{+n} electrode methods. This is attributed to the inability of the electrode to function irreversibly at tracer level concentrations. However, approximate potential values for metal-metal ion couples have been obtained in two ways.[22] In the first, a solution of the tracer ion is mixed with a solution containing macro amounts of another metal ion in contact with its metal, and the deposition of the tracer measured. A series of such chemical deposition

experiments with various metals fixes the approximate position of the tracer couple in the electromotive series. In the second method, a tracer solution is electrolyzed between inert electrodes, and the deposition of the tracer is measured at various applied voltages. The potential of the cathode is also measured against a standard half-cell in contact with the tracer solution. The electrode potential at which appreciable deposition begins is called the critical deposition potential and has been used in the estimation of E^o values. The presence of a solid phase, adsorption, diffusion, and the slow rate at which some equilibria are established can make electrochemical phenomena very complex and difficult to predict especially in dilute solutions. Yet it is generally believed that the projection of the values and equations of macroscopic electrochemistry can be applied to very dilute solutions which do not contain an electrochemically active solid. For example, in a solution containing 0.1-M Cu^{+2} and 1E-12 M Fe^{+2}, the electron transfer reaction predicted by the Nernst equation does occur. Figure 21 in Appendix A shows a comparison of E values for some aqueous actinide elements and oxidizing and/or reducing agents. (See also Appendix B, Table VI.)

Radiocolloid is the name given by Paneth to tracer-level radioactive species which behave in solution as if they were colloidal.[19] Paneth noticed that in slightly acid, neutral, or ammonia solutions, some carrier-free radioisotopes did not diffuse through a parchment membrane and behaved somewhat like colloids. This behavior seems to be a common property of many radioactive elements. Radiocolloids have been shown to partially settle out of solution after standing for 24 hours.[22] Paneth observed that the extent of radiocolloid formation increased with pH and he attributed this to hydrolysis and the formation of basic salts or hydroxides. The addition of certain complexing agents or the increase of acid concentration has been shown to decrease radiocolloidal formation. The consequences of radiocolloid formation can be positive or negative depending on the chemical separation method chosen. For example, 1E-9 M solution of radioactive Sc^{+3} can be separated from 10^{-3} M Ca^{+2} at pH 8.5 by simple filtration.[19] Also, radiocolloids have been shown to have different distribution coefficients than the simple tracer

molecules between two immiscible solvents, sometimes even concentrating on the interface.[22]

The presence of impurities in solution strongly influence the relative extent of radiocolloid formation or, more accurately, pseudo-radiocolloid formation. It has been suggested that these radioelements, present as ions, are adsorbed on invisible impurities such as silica, dust, aluminum hydroxide, etc. The colloid formation seems to be easier for compounds with a tendency to form insoluble compounds. Therefore, the lower the solubility of a given compound on the macroscopic scale, the more strongly it will tend to be adsorbed on impurities at the tracer scale, consequently the more pronounced will be its pseudo-colloidal properties.[22]

Adsorption of cations onto container walls, stirring rods, ground glass surfaces and other equipment is a major concern when dealing with tracer-level solutions. It has been shown that the concentration of dilute acidic cation solutions stored in preleached glass bottles dropped to 20-40% of the original value in 15 days.[21] In some studies, quartz vessels have shown only minor changes in solution concentrations on storage. With polyethylene bottles, the results vary depending on the isotope. These results indicate that standard solutions must be checked before use and steps should be taken to prevent adsorption losses in the samples.

D. Rates of Reactions

Standard half-potential differences, standard free energy differences, and equilibrium constants reveal whether or not a reaction proceeds far enough toward completion to be useful in a particular chemical process. However, they provide no information about the rate at which the equilibrium state is approached. A reaction mechanism may have several steps, each proceeding at a different rate. The slowest step will be the rate-determining one and the others will be essentially at equilibrium. Controllable factors that affect the rates of reactions are temperature, the presence of a catalyst, and the concentrations of the reactants or products.[23] In general, redox reactions that involve the transfer of a single electron are fast. Conversely, reactions in which the electron changes in the oxidation numbers of the oxidizing and reducing agents are

unequal, may be slow.[24]

Relevant to actinide chemistry is the observation that reactions which do not involve the making and breaking of strong chemical bonds, that is, M^{+3} <--> M^{+4} and MO_2^{+} <--> MO_2^{+2}, seem to be fast and reversible. However, reactions that involve chemical bond formation, M^{+3} <---> MO_2^{+2} and M^{+4} <--> MO_2^{+}, tend to be slow and often not readily reversible.[4] For example, in the oxidation-reduction reaction between U^{+4} and Fe^{+3}, the rate-determining step involves a reaction between one U^{+4} and one Fe^{+3} to give the intermediate UO_2^{+}.[27]

Diffusion in solutions is quite rapid and most overall reaction rates are not limited by the rates at which the reactant species diffuse together. Some consideration has been given to the possibility that the high charge on actinide ions can effect the diffusion rate of the reactants and therefore the collision frequency when the other reactant is also a highly charged cation. A rate constant of approximately 1.2E2 $M^{-1}s^{-1}$ has been calculated for diffusion-controlled reactions between +3 and +4 cations.[27]

The terms fast and slow seem to be relative. Skoog and West regard a reaction to be fast if it proceeds to 50% completion in 10 seconds or less.[23] In separation processes for environmental samples, any reaction that reaches completion in 1 to 5 minutes is probably sufficiently fast.

In Appendix B, Table VII, the rates of several redox reactions relevant to this project are listed.

E. Separation Techniques

1. Precipitation

One of the oldest methods of chemical separation is precipitation. This method, when used in radiochemical separation procedures, is usually employed for the separation of macro-concentrations of matrix ions from the extremely small concentrations or trace amounts of the desired nuclides. This means setting the conditions so that a considerable amount of the major

constituent of the solution can be precipitated without causing losses from the solution of the trace constituents.

When attempting to use precipitation for the separation of tracer level ions, the concentration of the tracer is frequently so low that addition of large amounts of the precipitation anion does not exceed the solubility product constant (Ksp) of the tracer-anion compound. Therefore, a technique called coprecipitation or carrier precipitation is often utilized. Carrier precipitation is defined as the collection of the desired trace-level ions in a solution on a small quantity of another precipitate, which is called a collector, carrier, or gathering precipitate.[28] The carrier ion can be either isotopic or non-isotopic to the desired trace ion. Carrier precipitation involves two general types of carrying processes. The first is replacement of the carrier ion by a trace ion of the same sign and similar size to form a mixed crystal. The second is adsorption of the trace-level ion onto the oppositely charged surface of the precipitate.[22]

Coprecipitation or carrier precipitation is employed to concentrate and to separate the actinide elements from other elements and each other. The possibility of separation depends on maintaining the actinide element or elements in a desired oxidation state and selecting an appropriate precipitation reagent. Generally, precipitation of compounds with similar solubility behaviors to the actinides will usually carry the actinides. Carrying is ordinarily favored further if ionic species of the carrier cation and that of the actinide are the same. Examples of the coprecipitation behavior of actinide species and various carrier compounds are summarized in Appendix B, Table VIII.[26]

Many combinations of carrier precipitations can be used to isolate given actinide species and to obtain some separation from other ionic species. The process selected for isolation depends on desired yield, required time cycle, if changes in oxidation states can be utilized, and if the "reverse strike" can be either avoided or modified to give good yields. The reverse strike technique is the addition of the carrier cation to a solution already containing the anion of the insoluble compound. It is prone to poor yields because the cation may precipitate as it enters the

solution without incorporating the desired trace material that is dispersed in the solution.[26]

2. Solvent Extraction

In solvent extraction, the preference of a substance for a given solvent over a different immiscible solvent can be used to separate actinide ions. Often, the two immiscible solvents are water and an organic medium. The hydrated actinide ion will generally prefer the aqueous phase to the organic phase. For the metal ion to be extracted, the cation is usually complexed in order to form an electrically neutral and organophilic substance. Some of the ways in which this type of complex may be formed are: (1) coordinating the metal ion with organic anions to form neutral complexes, (2) replacing water in the coordination sphere of the metal ion with large neutral organic molecules and obtaining neutrality by additional coordination with simple anions, and (3) forming metal complexes with anionic ligands that then combine with large organic cations to form neutral ion pair complexes.[4]

Some polydentate organic anions can combine with metal cations to form neutral organophilic complexes. These polydentate anions usually form chelate ring structures with the actinides. Some of the complexing agents are acetylacetone, thenoyltrifluoroacetone (TTA), cupferron, derivatives of salicylic acid, 8-quinolinol, 1-nitroso-2-naphthol and sodium diethyldithiocarbamate.[2] This extraction process using polydentate organic anions to form neutral chelate complexes normally has a high acid dependence. In general, the more highly charged cations form stronger complexes and therefore usually extract more efficiently. Thus, these chelate systems ordinarily extract the M^{+4} species effectively, although the MO_2^{+2} species may be extracted with lower efficiency. The M^{+3} and MO_2^{+} species are usually not extracted well.[2]

Neutral organic extractants that may replace water in the coordinating sphere of the cation are alcohols, ester, ethers, and ketones. These organic extractants, in combination with simple anions (e.g., nitrate, chloride), combine with the actinide cations to form a neutral organophilic complexes that are extractable into organic solvents. In practice, nitrate solutions of the metal cations and the extractants, diethyl ether (Manhattan Project), methylisobutyl ketone

(hexone)(Redox process), and tributyl-phosphate (TBP)(Purex process), have been most commonly used.[2,4] With diethyl ether and hexone, the extractability order is usually $MO_2^{+2} > M^{+4} >> M^{+3} = MO_2^{+}$. With TBP the extractability order is usually $M^{+4} > MO_2^{+2} >> M^{+3} = MO_2^{+}$.[2] Other organophosphorus compounds can also be used as extractants. The extractability of the nitrates of M^{+4} and MO_2^{+2} with different aryl and alkyl substituted organophosphorus compounds was found to increase in the order: phosphate ester < phosphonate ester < phosphinate ester < phosphine oxide.[2]

Effective cationic extractants are usually high molecular weight amines, especially the tertiary or quaternary amines. These strong bases couple with anion-complexed actinide cations. The efficiency of the extraction is improved when the alkyl groups have long carbon chains. Two examples are trioctylamine and triisooctylamine.[4] Tri-n-octylamine in xylene extracts M^{+4} nitrates with distribution ratios about an order of magnitude greater than those obtained with 19% TBP under similar conditions. The coefficients for the MO_2^{+2} species are from 10 to 100 times smaller than for TBP and those for M^{+3} and MO_2^{+} are very low.[2]

3. Ion Exchange

Ion exchange is an important method of chemical separation for the actinide elements. It can be fast, efficient, and useful for chemically similar substances that are difficult to separate by other methods. In ion exchange, anions or cations are usually portioned between a mobile aqueous phase and a stationary solid phase. The difference in adsorbability on a given resin of two chemically similar ions is usually small, but when a solution is passed through a column packed with finely divided resin, the ions are adsorbed and desorbed many times. This small difference in adsorbability is amplified into an efficient separation. Efficiency of a given ion-exchange separation depends on a number of factors, e.g., type of ion-exchange resin, the elution solution, the flow rate, and the column dimensions.[22] Most commonly, the solid phase will be an organic polymer containing sulfonic, carboxylic, phosphoric, or phenolic acid groups (cation exchange resins) or quaternary ammonium groups (anion exchange resins).[22]

Actinide ions with oxidation states ranging from +3 to +6 can be adsorbed by cation exchange resins and eluted with complexing anions such as citrate, lactate, EDTA, glycolate, or -hydroxy isobutyrate. On most cation exchange resins in dilute acid solutions, the strengths of adsorption of the actinide ions are in the following order: $M^{+4} > M^{+3} > MO_2^{+2} > M^{+2} > MO_2^{+}$.[2,4] The differences in adsorption are sufficiently large to allow separation of the different types of ions. The flexibility of cation ion exchange systems is limited for Np and Pu because they usually need to be adsorbed from dilute acid solution. Under these conditions, their M^{+4} ions have limited stability. However, cation exchange resins have found considerable use in the concentration of trace amounts of the elements from large volumes of dilute acid by relatively small resin columns.[2]

When using cation exchange resins, the order of elution of aqueous ions with different oxidation states with 3-M HCl is PuO_2^{+2}, Pu^{+3}, then Pu^{+4} and NpO_2^{+}, NpO_2^{+2}, then Np^{+4}. The M^{+4} ions usually elute in the order, Pu, Np, then U. The MO_2^{+2} ions were usually found to elute in the few first column volume even in 6 M HCl. In 12 M HCl, the order of elution of M^{+3} ions is: Pu, Am, then Cm. In more dilute HCl, the elution sequence becomes Cm, Am, then Pu.

Separation of the actinides from the lanthanides has been accomplished by elution from cation exchange resin columns with concentrated HCl. Better separation of the two groups is obtained by elution with 20% ethanol solutions saturated with HCl.[2] In both cases, the actinides elute before the lanthanides because they are more strongly complexed by chloride ion. Americium of very high purity has been obtained by adsorption of Am^{+3} on a cation exchange resin column from dilute acid. After the column is washed with 1-M NH_4SCN to remove Fe^{+3}, the americium is eluted with 8-M NH_4SCN and adsorbed directly onto an anion column. Washing with 5 to 8-M NH_4SCN removes the last traces of the rare earths, and the americium may then be eluted with dilute acid from the anion column.[2]

In anion exchange, the actinides are complexed with ligands like Cl^- to form anionic complexes which can then be adsorbed on anion exchange resins. Anion exchange systems have

been favored for separation and purification due to the large number of anionic complexes associated with the various oxidation states.[2] The two most used complexing anions in anion exchange separations of the actinides are the nitrate and chloride ions. In Appendix B, Table IX lists the distribution coefficients of the actinide-anion complexes on a strong base anion exchange resin at various acid concentrations.[29]

Additional procedures using anion-exchange resin columns have been developed for the separation of actinides in various other acid media, e.g. sulfuric, phosphoric, and hydrofluoric acids. Some success has also been shown for the use of anion exchange using other than acid systems.[2] These include sodium carbonate, ammonium chloride and ammonium thiocyanate solution. [29] Limited work has been reported concerning ion exchange from other than aqueous media for the actinide separations.[2]

Separation based upon selective elution of one of the actinides by changing its oxidation state is sometimes considered superior to selective elutions using solutions of various anion concentrations. An example demonstrating this would be the following sequential separation of Am, Pu and Np. They can be removed individually from Dowex 1 anion exchange resin eluting Am^{+3} with 8-M nitric acid, Pu^{+3} with 0.02-M ferrous sulfamate in 4.5-M nitric acid, and then NpO_2^{+2} with 0.001-M cerium(IV) sulfate in 0.25-M nitric acid. If thorium is included in the mixture, Am^{+3} is eluted with 8-M nitric acid, Th^{+4} with 12-M hydrochloric acid, Pu^{+3} with 12-M hydrochloric acid containing 0.1-M ammonium iodide, and Np^{+4} by 4-M HCl. (UO_2^{+2} follows the Am^{+3} in both methods.)[29]

Strong base anion exchange resin columns have been used to adsorb the M^{+3} actinides from lithium chloride solutions, which provide sufficiently high chloride ion concentrations for the existence of the trivalent actinides predominantly as negative chloride complexes. Also, the adsorption of many trivalent actinides from ammonium thiocyanate solution has been investigated.[2]

4. Yield Tracer Technique

Often the success of a quantitative chemical analysis depends upon the ability of the process to recover 100 % of the analyte. Unfortunately, in many radiochemical testing processes, an elaborate separation scheme is often needed to extract small amounts of the actinides from other components of the matrix. This complex process tends to have losses of analyte at various stages and thus complete recovery is usually not achieved. Therefore, in order to get an accurate assessment of the amount of the actinide, a relationship is established between the total amount of actinide originally present in the sample and a similar radioactive substance added to the sample. This added substance is called a yield tracer.[30]

A yield tracer must behave chemically very similarly to the component it is required to track. So the best yield tracer is the element itself. This is usually accomplished by employing a different isotope of the element then the one or ones in the sample. Isotopes of the same element do possess distinctive properties from each other such as variation of mass and type of radioactive disintegration. For heavy elements, the relative mass difference between the isotopes is often small and the chemical behaviors are not significantly different.[31] The difference in radioactive decay is the property that allows the yield tracer process to work because it permits separate measurement of the different isotopes even though they are mixed together.

For a yield tracer to perform reliably in a separation procedure the following should be considered:

1. Chemical behavior of the yield tracer.
2. Early establishment of chemical equilibrium between tracer and analyte.
3. The oxidation state or states of both analyte and tracer.
4. Availability of analytical means for precise individual analysis of the yield tracer and analyte in the presence of each other.
5. Length of the half-life of the yield tracer.

6. Availability, cost, and purity of yield tracer.
7. Amount of yield tracer (to avoid degradation of spectrum yet provide good detection and good counting statistics.).
8. Possible interference from in-growth of decay daughters of the yield tracer. (e.g. Pu-236 --> U-232 --> Th-228)
9. Production of volatile intermediate decay products that may contaminate the detector.
10. The need for multiple yield tracers for the identification and measurement of more than one element in the sample.

See Appendix B, Table X for a listing of yield tracers for selected actinide elements.

F. Separation Schemes

Prior to alpha-spectrometric measurement several chemical manipulations are usually required to obtain alpha-emitting samples with the desired radiochemical and chemical purity. These include sampling, dissolution or leaching of the elements of interest, conditioning of the solution, chemical separation, and preparation of the alpha-emitting sample. In this section, there are presented some of the current schemes used for the chemical separation step, since that is the primary area of interest in this research project. Five sequential separation procedures (EPA, ITAS, Purex, Truex, and TRU-Resin™) are described in the following paragraphs.

The United States Environmental Protection Agency (EPA) has prescribed a procedure for the measurement of radioactivity in drinking water.[32] Figure 6 shows a flowchart of the EPA sequential separation procedure. In this procedure, actinide elements (Th, U, Np, Pu, Am, Cm) are initially concentrated by coprecipitation on $Fe(OH)_3$. The $Fe(OH)_3$ is then dissolved and Th, Np, Pu, Am, and Cm are coprecipitated on $BiPO_4$. This leaves U in solution for a subsequent solvent extraction using trioctylphosphine oxide (TOPO). The $BiPO_4$ is dissolved in concentrated HCl and Pu and Np are solvent extracted using triisooctylamine (TIOA). The Th is separated from

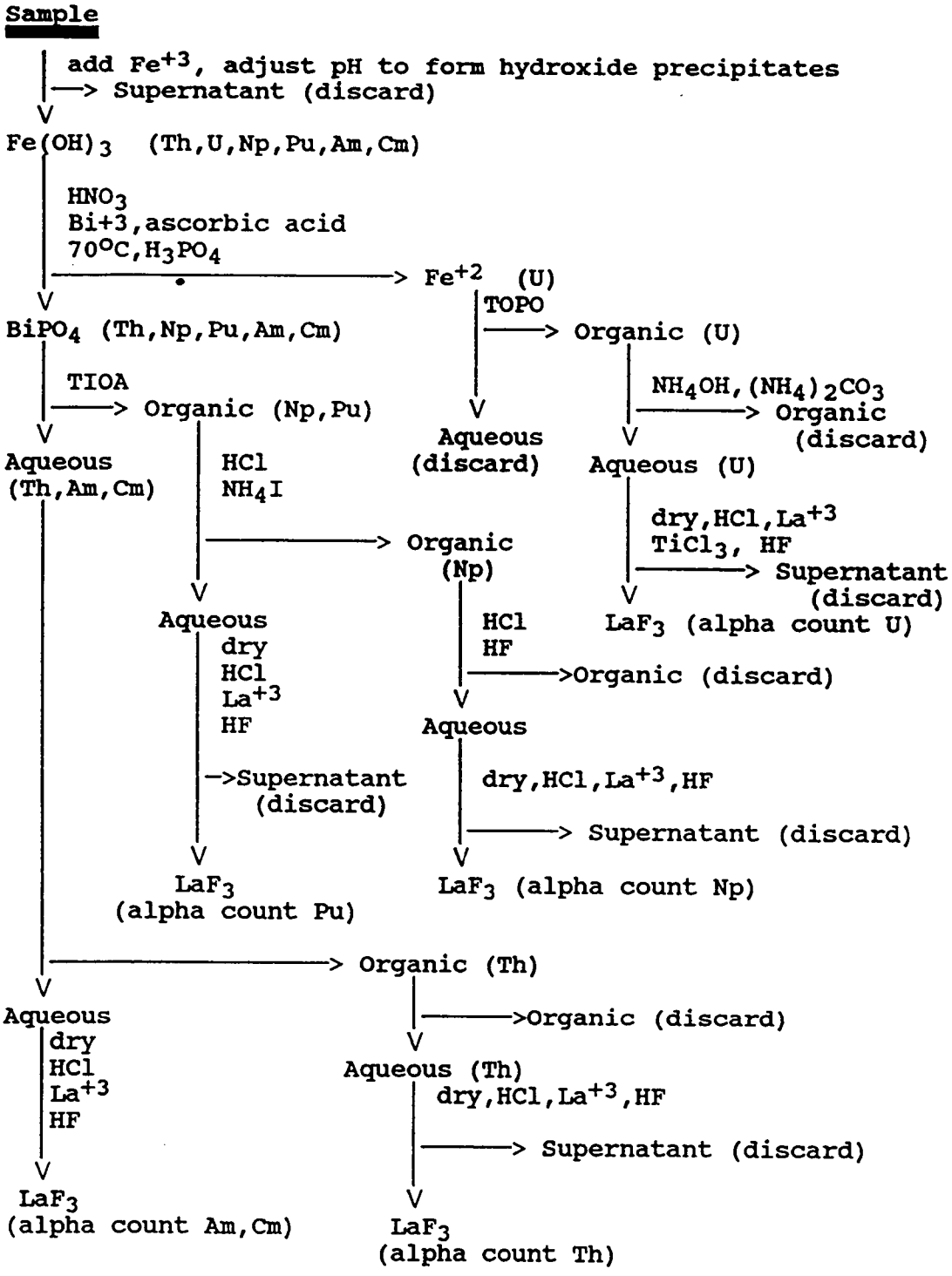


Figure 6: EPA Sequential Procedure for Measurement of Radioactivity in Drinking Water.[32]

Am and Cm by a solvent extraction with TOPO. The chemical yields in this method are stated to be 85% or better for clean water samples. This level of recovery is generally considered to be good. A disadvantage of this process is the number of separation steps required, these including three solvent extractions. This results in a time consuming and labor intensive process with the generation of mixed wastes.

An actinide separation procedure utilized by International Technology Analytical Services (ITAS) in the fall of 1992 reflects typical commercial laboratory processing. A detailed flow chart is shown in Figure 7. A soil or water sample, previously digested, is first treated with several redox agents, evaporated, and acidified until the desired elements are in aqueous ionic forms in a nitric acid solution. The desired ions are then separated from the solution by loading them onto a Bio-Rad-AG1-X8 resin using a batch technique. The resin is then placed in a column, and the actinide ions are subsequently eluted with various agents into three groups. U, Cm and Am are eluted first. The second fraction contains only Th. The last fraction contains Pu and Np. The first group is then passed through a second AG1 column where the Am/Cm are separated from the uranium. The Am/Cm fraction is then processed using coprecipitation with CaC_2O_4 and then $Fe(OH)_3$. After dissolution, the Am/Cm fraction is passed through two more columns before being ready for count preparation.

This procedure provides consistently good yields for Am and U. However when Th levels are elevated, there is often contamination of other fractions with Th. Since Pu and Np alpha emissions can be spectroscopically resolved, the elements are eluted together inorder to save separation steps. In the Pu/Np fraction a Pu yield tracer is used to determine any experimental losses. This can present a problem when quantitating Np because the Pu tracer and the Np behave the same chemically only under very specific conditions. Thus, this part of the procedure is sensitive to any slight change of conditions, such as pH, matrix, or redox agents.

ITAS Labs do not utilize this sequential method unless a limited amount of sample is submitted for analysis. They usually will split samples, if sufficient quantity is submitted, for use

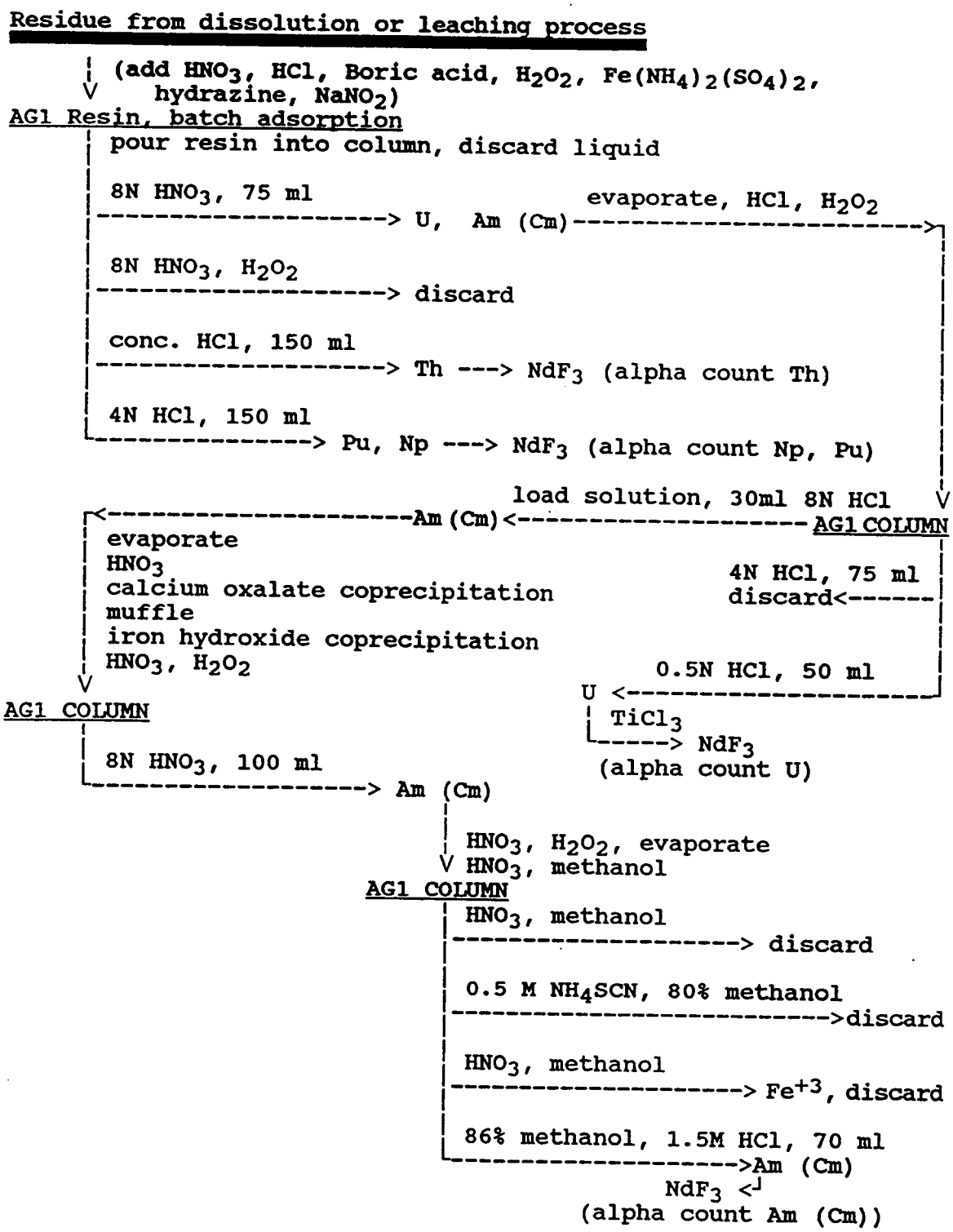


Figure 7: Typical Commercial Sequential Actinide Separation Procedure.

in single element separation procedures. Although split testing results in more samples needing to be handled, this often allows for more consistent results.

The Purex process is primarily a large volume commercial procedure used for the purification of U and Pu, but it can also be used for smaller scale separations.[33] Tributylphosphate (TBP) is applied as the extractant in the Purex process. The process solvent is obtained by diluting TBP with kerosene to a concentration of 5 to 30 volume percent. Figure 8 shows a flowsheet of the extraction cycles of the Purex process.[33] In the first extraction (extractor #1), the UO_2^{+2} and Pu^{+4} are extracted into the organic solvent phase in the form of the solvates, $UO_2(NO_3)_2 \cdot 2TBP$ and $Pu(NO_3)_4 \cdot 2TBP$. The Pu^{+4} is reduced (extractor #3) to Pu^{+3} which then moves into the aqueous phase while the UO_2^{+2} complex stays in the organic phase. In extractor #4, the U is removed from the organic layer with dilute HNO_3 and placed back into an aqueous solution. In the initial TBP extraction, significant amounts of Np and fission products generally are extracted into the organic phase along with the U and Pu complexes. Therefore, after each of the extraction steps, several wash steps are required to remove the contaminates and several cycles of this process need to be used to get the final purified products.

In the mid-1980's, an additional reagent was added to the basic purex TBP extractant solvent by Horwitz and this resulted in a new procedure called the Truex process.[34] This revised process used the actinide-lanthanide selective extractant called octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide (CMPO). CMPO has a unique combination of substituent groups which gives it the ability to extract trivalent actinides from various concentrations of HNO_3 or HCl. This extended the usage of the solvent extraction process to other actinides which were not efficiently extracted by TBP alone. Figure 9 shows a flowchart of the process.[35]

The last sequential separation procedure to be discussed uses EIChroM-TRU-Resin™ for the separation of the actinides. This actinide-sequential-separation procedure resulted from the development of a new type of extraction resin. These resins have an organic coating layered on the surface of a polymer bead. The removal of the ions from the solution by these resins is similar

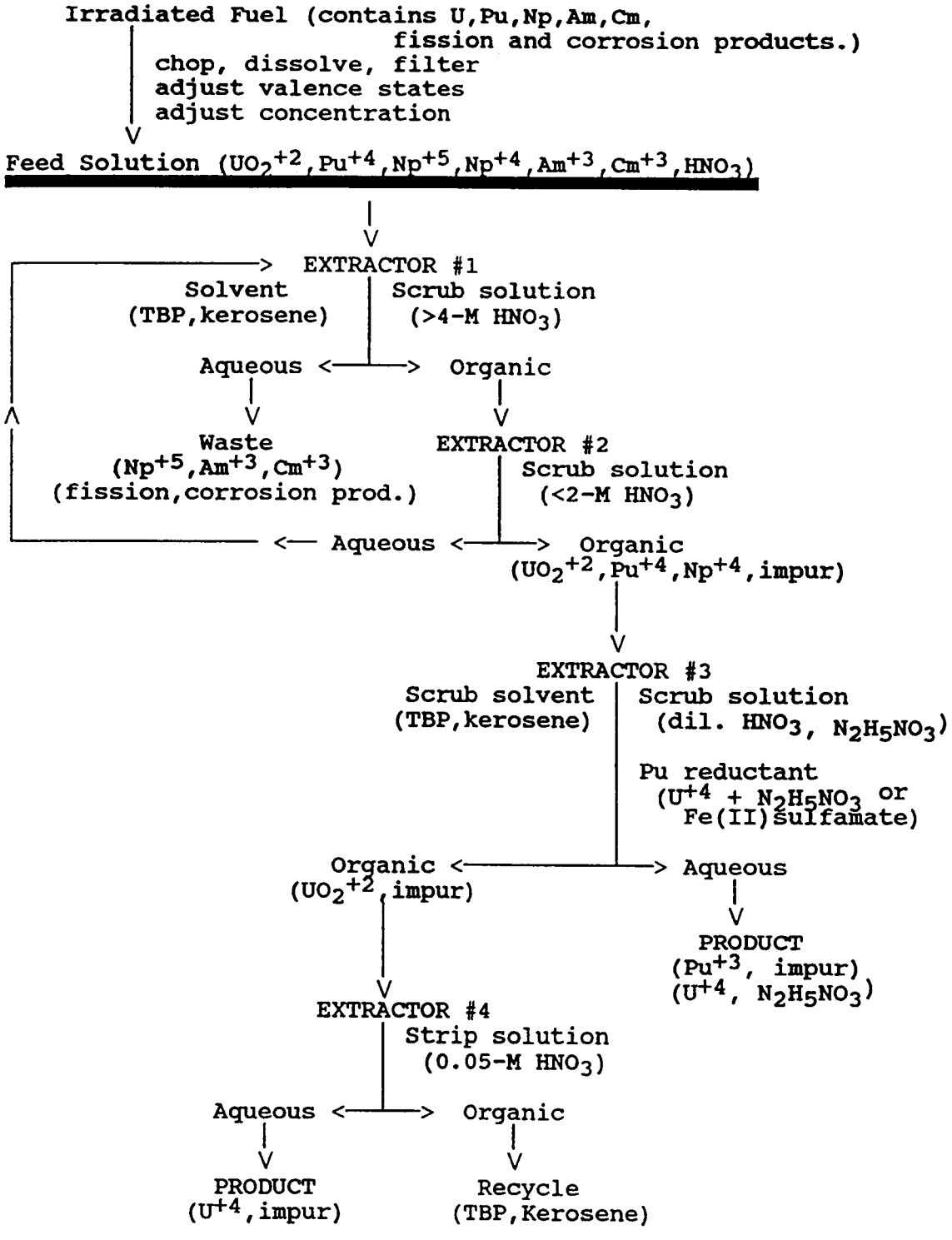


Figure 8: The Purex Process.[33]

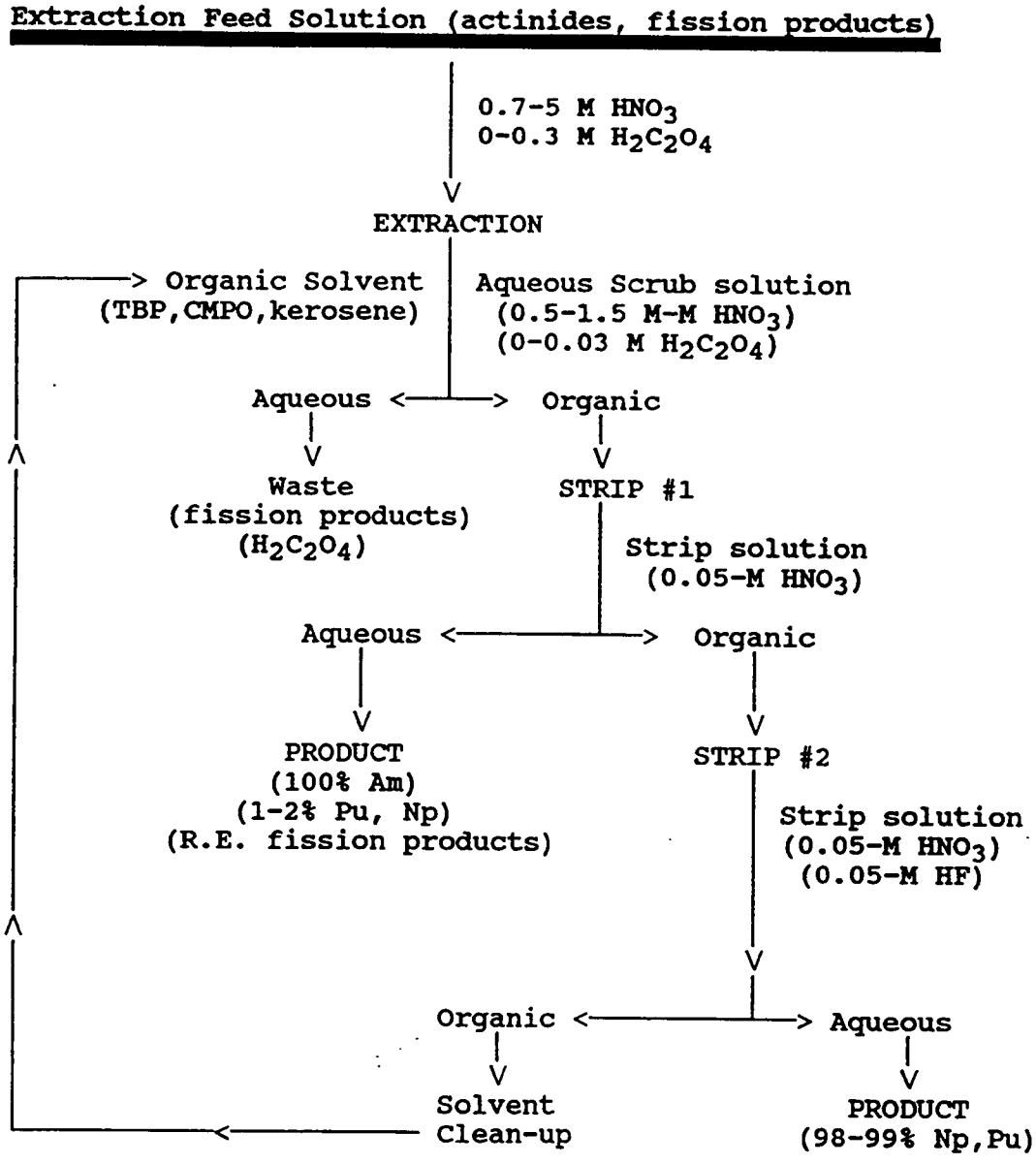


Figure 9: The Truex Process.[35]

to the chemistry of the extraction process but it is occurring between a supported organic layer and the aqueous solution rather than between two unsupported liquid solution layers. Horwitz has usedone of these extraction chromatographic resins (called TRU-Resin™) comprised of CMPO and TBP supported on an inert polymeric substrate to propose a sequential elution of individual actinides.[36] The proposed scheme was based on a series of one-element extraction studies. The procedure is shown in figure 10. The 50-100 μm diameter resin bead and high tracer levels were used in this experiment. Some overlaps of the different actinides were observed in the various fractions eluted. The amounts of overlaps seemed to depend, at least partly, on the total elution volume and flow rate of the column. Horwitz summarized that this sequential procedure was probably too tedious for routine analytical use. He suggests the use of TRU-Resin™ in conjunction with anion exchange columns to achieve an efficient separation with fewer manipulations.[36]

G. Alpha Sample Counting Preparations and Measuring Devices

Solid samples for alpha spectrometric measurements are usually prepared by direct evaporation, electrodeposition, or coprecipitation techniques. With solid samples, every effort must be made to avert self-absorption by producing samples as thin as possible. Direct evaporation is generally used for highly pure samples which contain very little non-volatile material. Low-activity samples usually require the concentration of the desired element from several milliliters of solution containing different salts remaining from the chemical separation and therefore are poor candidates for preparation by evaporation.

Electrodeposition is the preferred technique by many in the field of radiochemistry. It provides a very thin sample on the surface of a metal disc, thus minimizing self-absorption and providing the possibility for very good resolution. The actinide may be deposited on the cathode or the anode. Cathodic deposition involves reduction of the hydrogen ion concentration at the cathode with a corresponding increase in hydroxyl ion concentration thus resulting in the deposition

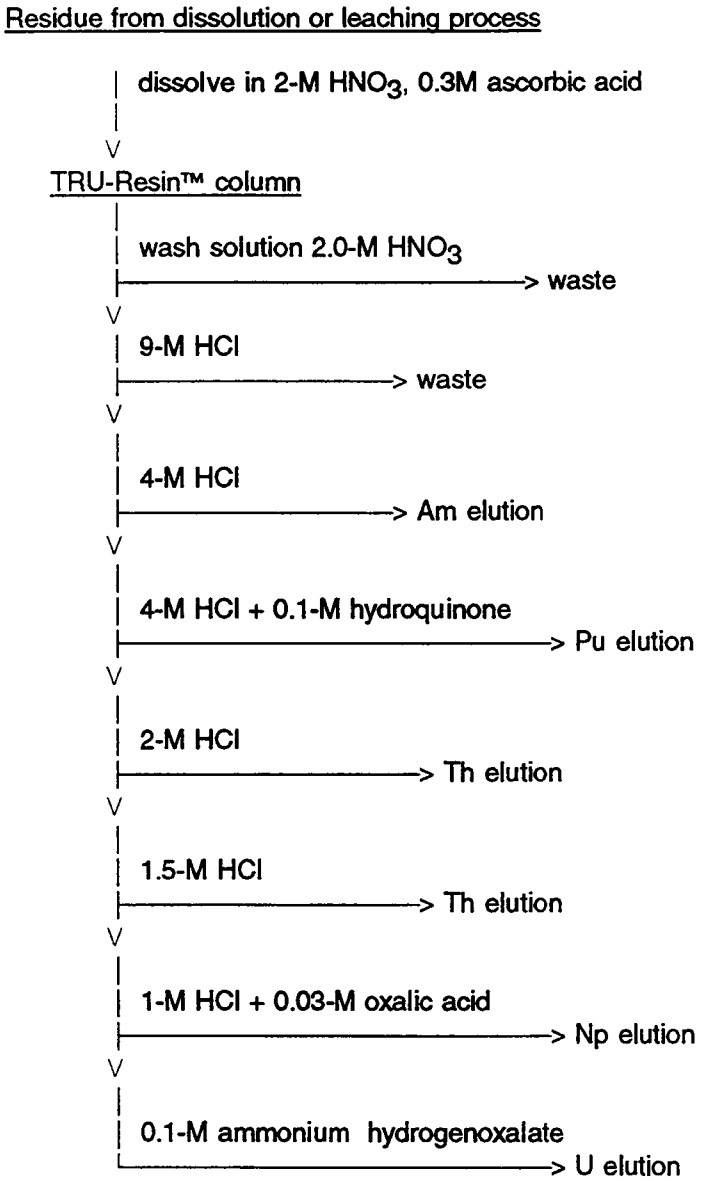


Figure 10: Separation of Actinides Using a TRU-Resin™ Column. Developed by Dr. Horwitz.[36]

of the actinide oxide or hydroxide.[38,39] Anodic deposition results in an actinide in solution being oxidized to a higher oxidation state and forming an insoluble compound, such as a higher-valent oxide, which then deposits on the anode.[22] The type of deposition depends on the actinide and the applied oxidizing potential. The rate of deposition depends on the applied voltage, rate of stirring, volume and nature of the solution, and area of the electrode. Electrodeposition is a very sensitive technique and requires careful preparative steps. Electroplating of actinides has been done from various solutions, such as ammonium formate, ammonium oxalate, ammonium chloride, ammonium sulfate, dimethylsulfoxide-nitric acid, or combinations of these electrolytes. The oxalate method is often preferred because it is generally faster and less sensitive to experimental variations and impurities in the electrolyte. The dimethylsulfoxide method is fast and yields nearly quantitative deposits in 5-10 minutes for most actinides but the high voltage used presents an operational hazard.[37]

Several coprecipitation procedures have been developed. Exemplary compounds that act as coprecipitating agents are cerium hydroxide, lanthanum fluoride, and neodymium fluoride. Tracer-level quantities of an actinide often do not precipitate because the corresponding anion concentration can not be raised high enough to exceed the Ksp of the actinide-anion compound. However, when a cation with similar precipitation properties to the actinide is added, coprecipitation will result. This precipitate has the actinide incorporated into the crystal structure or adsorbed on the surface of the cation-anion compound. The solid is then filtered out of solution or dispersed in a small volume of water and evaporated on the sample plate for counting.

Development of additional techniques for preparation of samples for alpha measurement is on going. One procedure under investigation is the use of cation exchange disks as a separation and measuring surface for alpha spectrometry.[37] The separation is performed by sulphonating polystyrene disks and then adsorbing the actinide from an aliquot of solution onto the disk. Another method involves solvent extraction into a scintillation solution and counting the light produced. A technique known by the acronym PEARLS uses a combined extracting-scintillation

cocktail as its sole sample preparation step.[72]

There are several instruments that can detect alpha particles. These instruments have a wide range of abilities from simple detection of the presence of alpha particles to the spectroscopic identification and quantitation of a mixture of different energy alpha particles. These instruments include Geiger counters, gas flow proportional counters, liquid scintillation counters, and surface barrier detectors. The instrument that was used in this project for the measurement of solid-alpha-sample counts was the surface barrier detector. It currently offers the best energy resolution. This instrument will be discussed in more detail in a later chapter.

CHAPTER III

MATERIALS AND APPARATUS

A. Tracers and Spikes

Isotopically specific actinide solutions of known concentrations were utilized as spikes and yield tracers in the testing procedures. These standards were purchased from various suppliers and diluted to the appropriate concentrations for use with low-level-radioactive samples.

The solutions used as spikes consisted of actinide nuclides that could be found in an environmental sample submitted for analysis by a customer. These nuclides usually have relatively long half-lives and are naturally-occurring or present in the environment due to nuclear waste contamination. Spikes were added to test samples in order to create customer-like samples of known concentration.

The yield tracer assists in the determination of any losses of the desired actinide due to the analytical procedure. A yield tracer is usually the same element, just a different isotope, as the desired actinide element for which the test is being performed. The tracer is an isotope that is rarely found in any environmental sample due to a short half live or the lack of its presence in any natural or radioactive-waste decay chain. The yield tracer technique has been discussed in chapter II, section E.4. The nuclides that were used in this project as yield tracers and/or spikes are listed in Appendix B, Table XI.

B. Resins

The different resins utilized in this project are AG-1, TRU-Resin™, and Diphonix. The Bio-Rad Laboratories Analytical Grade Anion Exchange Resin AG-1-X8, 100-200 mesh, is a basic anion exchange resin. It has a quaternary ammonium functional group attached to a styrene divinylbenzene copolymer lattice,$[R\text{-}CH_2\text{-}N^+\text{-}(CH_3)_3]Cl^-$. The AG-1-X8 is 8% cross linked and used

for exchange separations of inorganic anions, and in applications such as cyclic nucleotide assays and fractionation of organic acids.[12]

TRU-Resin™ is commercially available from EIChroM Industries, Darien, IL. The specific resin used in this project was product code TR-B50, lot # 100292, particle size 100-150 . This is an extraction chromatographic resin that is prepared by impregnating Amberchrom CG-71ms (Amberlite XAD-7, polyacrylate nonionic polymeric adsorbent) with a 0.75 M solution of octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide (CMPO) in tributyl phosphate (TBP). The water/methanol prewashed resin is combined with a mixture made by dissolving 0.75M CMPO in TBP and then mixing this with methanol. After gentle swirling, the methanol is then removed in vacuo leaving the CMPO-TBP-loaded resin.[36,40] The percent composition of the final product is 13% CMPO, 27% TBP, and 60% polyacrylate absorbent.[43]

The structure of CMPO is shown in Appendix A, Figure 22. CMPO is generally considered to behave as a neutral extractant. The oxygen of the P=O group is more basic than that of the C=O, suggesting a stronger binding capability of the former with the metal ion complexes. Measurements using NMR and IR have shown that the primary donor group is the more basic phosphoryl oxygen.[41,42] At trace metal ion levels, it has been reported that the coordination of CMPO to a metal anion complex is monodentate at the phosphoryl oxygen. However, under higher loading conditions, CMPO is reported to act as a bidentate ligand with both P=O and C=O binding to the metal anion complex.[42] The authors make no remarks indicating that they regard this as strange. Some difference in behavior has been noted when different anions are used in the matrix. The Am-nitrato-CMPO complex is much more extractable than the Am-chloro-CMPO complex at low to moderate acid concentrations. The difference in extractability is thought to be due to the softer donor properties and greater hydration energy of the chloride ion relative to the nitrate ion.[34] Another investigator noted that coordination to a metal ion extracted from a nitrate medium occurs primarily via the phosphoryl oxygen in a monodentate fashion while the phosphoryl and carbonyl groups are involved in a bidentate fashion from a chloride medium.[41]

The protonated form of the CMPO extractant, which is present at high acidities, is able to extract Am^{+3}. This occurs despite the competition between hydrogen ions and the Am^{+3} for the basic oxygen donor groups. This property is characteristic of extractants containing the carbamoylmethylphosphoryl moiety and one explanation is that the carbonyl group is protonated while the phosphoryl group bonds to the metal ion.[34] One interpretation is that the protonated carbonyl is acting as the more basic oxygen donor group. IR studies have indicated that the carbonyl group in a Nd^{+3} complex is protonated when extraction occurs.[34]

The nature of the binding of the extractant to the metal ion in the solvent extraction and the extraction chromatography absorbed complexes were found to be similar in IR studies.[42] CMPO forms complexes with tri-, tetra-, and hexavalent metal ions. The extraction equilibria may be represented in an oversimplified form as follows:

$$Nd^{+3}(aq) + 3\ NO_3^-(aq) + 3\ CMPO(org) \leftrightarrow Nd(NO_3)_3 \cdot 3CMPO(org)$$

$$Th^{+4}(aq) + 4\ NO_3^-(aq) + 2\ CMPO(org) \leftrightarrow Th(NO_3)_4 \cdot 2CMPO(org)$$

$$UO_2^{+2}(aq) + 2\ NO_3^-(aq) + 2\ CMPO(org) \leftrightarrow UO_2(NO_3)_2 \cdot 2CMPO(org)$$

Essentially, no extraction by CMPO of divalent metal ions has been observed. Only minimal extraction of the pentavalent metal ions seems to take place, e.g. K_d=0.30 for NpO_2^+.[44] However, some cation-cation complex formation between NpO_2^+-UO_2^{+2} with CMPO bridging ligands has been studied.[44]

In a series of original solvent extraction investigations, TBP was added to the CMPO extraction layer in the Truex solvent extraction procedure in order to reduce third-phase formation and to increase the loading capacity of the solvent.[34,46] However, TBP has long been used as the sole extractant in the Pruex-solvent-extraction process, as previously mentioned in chapter II, section F. TBP effectively extracts tetra- and hexavalent actinide ions, but only poorly extracts the tri- and pentavalent actinide ions.[2] The extraction by TBP in the Pruex process appears to result from the formation of a neutral unionized complex that is nonhydrated but solvated by a definite number of TBP molecules.[45]

The impact that TBP has in the CMPO-TBP mixed extractant in both solvent extraction and extraction chromatography is not clear. Chiarizia reports a positive effect by TBP on the CMPO extraction of Am^{+3} from aqueous nitric acid solutions. He suggests that the enhancement of the distribution coefficient for Am^{+3} at high acidity may be due to an acid buffering effect caused by the large concentration of basic P=O groups introduced by the presence of TBP.[46] Work by Mathur shows TBP as having generally little or no effect on the CMPO extraction of actinides, including Am^{+3}, from aqueous nitric acid solutions.[47]

Diphonix is a polyfunctional resin containing sulfonic and geminally substituted diphosphonic acid groups chemically bonded to a styrene-divinylbenzene polymeric network. The EIChroM product code for the resin sample used in this project was DP-B01, 60-100 mesh with lot # 93-002. Information from the manufacturer states that this resin is chemically stable, exhibits rapid metal exchange rates, and has the ability to retain tri- tetra-, and hexavalent actinides from highly acidic solutions, even in the presence of complexing anions such as sulfate, oxalate, phosphate, and fluoride. The diphonix resin binds the actinides by the formation of chelate complexes through the phosphoryl groups of the gem-diphosphonic acids.[48]

C. Reagents and Equipment

On the following list are some of the reagents and instruments that were used in this project. Standard laboratory apparatus and equipment that were utilized are not listed.

REAGENTS and MISC. EQUIPMENT	MANUFACTURER DATA
Ascorbic Acid	Baker Analyzed B581-07
Ethanol	EM Science 95% denatured
Nitric Acid	Baker Analyzed 69.0-71.0%
Sulfuric Acid	Baker Analyzed 96.2%
Hydrochloric Acid	Baker Analyzed 36.5-38.0%
Hydrofluoric Acid	Baker Analyzed 48.0-51.0 %
Hydrochloric Acid	Standardized 9.00±0.05 N, lot V328KLGN, UN1789
Hydrogen Peroxide	Mallinckrodt 30%, anal. reagent 5240
Oxalic Acid Dihydrate	Baker Analyzed, Lot F18160
Ammonium Oxalate monohydrate	Mallinckrodt, AR Lot 3452Kemp

Ammonium Iodide	Baker Analyzed 0708-01, Lot F47706
Potassium Carbonate, GR	EM Science, PX 1390-1, CAS 584-08-7, Lot32262249
Titanous Chloride	1.9-M Solution in 2.0-M HCl, stabilized w/$SnCl_2$, Aldrich cat 33,136-8, lot00409JZ
ColorpHast	pH range 0-2.5, EM Reagents, Cat 9580, US Patent No. 4.029.598
Chromatography Columns	Bio-Rad Poly-Prep,2ml bed volume, cat no. 731-1550
Prolabo-Microdigest	A-300, made in France, code06342012, date 09/90, N°;69880, Type F, 110v, 50/60 Hz, 600 W, Logicill/Software version 2.3, microwave 2450 mHz, Power output 200W,high voltage 3000v
Alpha Spectroscopy System	Units #1-16: Tennelec/Nucleus Inc., TC 256 Chamber, TB 4 Bin units #17-32: EG&G Ortec;model 676A, serial 396 chamber; 4001A bin NEC PowerMate 356/25 Canberra MCA Control, Hewlett Packard Laser Jet PCL 5 Vacuum Chamber, pressure ≤300 M of Hg Canberra Nuclear Data Nuclide Identification Software package (48-0201)Canberra Nuclear Data Spectroscopic Applications Software package (48-0198) Canberra Nuclear Data Alpha Spectroscopy Software package (48-0268)
Gelman Filters	0.1 m Metricel 25 mm polypropylene filter, product #M5PU025

D. Column Preparation

Each TRU-Resin™ column was prepared following the same procedure for each test. A 0.34 ± 0.02 gram sample of resin was soaked in approximately 8 ml of 0.50-N HCl for a minimum of 30 minutes. The resin slurry was then poured into a 2-ml-bed-volume Bio-Rad Poly-Prep chromatography column that had been pre-rinsed with distilled water. The resin column was then rinsed with 5 ml of 0.50-N HCl to help settle the resin. A glass wool plug was placed on top of the resin in the column. The resin was then ready for use. If it was not to be used immediately, the column was stoppered and stored with 0.50-N HCl on the resin. Prior to use, the resin would be preconditioned with approximately 15 ml of whatever reagent was going to be used to load the sample. Flow rates were generally 0.5-1.0 ml/min.

The above quantity of resin resulted in the production of a column that had approximately 1.0 ml bed volume. Such a small volume was selected in order to test the capacity of the resin, to determine what a small amount could accomplish, and to reduce cost.

E. Counting Sample Preparation (NdF_3 coprecipitation method)

After separation, most of the samples were counted by alpha spectroscopy. The following neodymium fluoride coprecipitation procedure was used for preparation of solid samples which were then counted using surface barrier detectors. The following is a variation of a procedure that was currently in place at IT (ITAS-OR-7124).

1. Reagents for NdF_3 coprecipitation procedure
   a. 0.73-M Na_2SO_4 in 18-M H_2SO_4
   b. 0.10 mg/ml Neodymium nitrate solution in 0.50-N HNO_3
   c. 29-M Hydrofluoric acid (Conc) (48% HF)
   d. Ethanol (70-100%, denatured)
   f. 20% Hydrofluoric acid solution (9.6% HF)
   g. Titanium trichloride, 20% solution in HCl
   h. Carbon substrate mixture - This is a mixture of 0.020 mg/ml neodymium nitrate, 0.50-N HCl, 0.60-N HF, and a small amount of finely suspended carbon particles.
2. Procedure for NdF_3 coprecipitation
   a. Dry down eluted fraction, do not bake (wet ash). Remove from hot plate. Cool slightly.
   b. Add 2.0 ml of 0.73-M Na_2SO_4 in 18-M H_2SO_4. Swirl gently to mix.
   c. Add 1.0 ml of H_2O_2. Swirl to mix. Warm slightly until bubbling stops, then heat strongly until the sulfate starts to fume (produce white vapors). Fume for 3-5 minutes. If dark color still present in solution, cool solution and repeat H_2O_2 treatment. Remove from heat. Allow to cool for about 5 min.
   d. Transfer to plastic centrifuge tube with 3.0-N HCl. Total volume after transfer is 15 ml.
   e. For Uranium only, add 0.30 ml of $TiCl_3$ solution. Allow to stand for 5 minutes. Proceed.
   f. Add 0.50 ml of 0.10 mg/ml Nd solution. Swirl gently.
   g. Add 5.0 ml of 29-M HF. Cap centrifuge tube and swirl solution. Allow sample to stand at room temperature for 20 to 30 minutes.
   h. Place Gelman filter into filtration unit and prewash using 5.0 ml Ethanol then 5.0 ml carbon substrate.
   i. Pour sample into filtration unit. Slowly pull solution through filter using a vacuum. Wash precipitate using 5.0 ml of 20% HF wash, then 5.0 ml DI water, then 5.0 ml ethanol. Air dry, mount on paper backing, and count.

This procedure was used for preparation of Th, U, Np, Pu, and Am samples. Some of the other current procedures use hot and/or cold baths during the precipitation process. The use of temperature baths did not show any improvement in the percent recovery of the actinides, so temperature baths have not been incorporated into this procedure.

For U, $TiCl_3$ must be added prior to the Nd solution in order to reduce UO_2^{+2} to U^{+4}. The hexavalent oxidation state does not coprecipitate with neodymium fluoride.

The IT precipitation procedure for uranium, ITAS-OR-7103, uses twice the amount of Nd as indicated in this procedure. No experimental data were determined for or against this amount. The above provides a uniform procedure for all the actinides but investigation may still need to be performed to determine the best Nd quantity for U precipitation.

F. Alpha Detectors

The initial alpha counts for this project were measured using silicon surface barrier (SSB) detectors. However, the measuring equipment was soon converted to passivated implanted planar silicon (PIPS) detectors for better resolution and efficiency capabilities.

The resolution of an alpha detector is usually defined as the FWHM (full width at half the maximum peak height) for the Am-241 5.486 MeV peak as measured at room temperature. Typical manufacturer values range from 15-50 keV for SSB detectors and 12-70 keV for PIPS detectors.[3] Major things that can effect this resolution are source to detector distance and the quality of the source itself. Samples consisting of thick deposits will exhibit self-absorption and the resulting resolution will be poor. In this project, the average resolution of a 800 count Am-241 peak for SSB detectors was 58±8 keV and for PIPS detectors was 46±8 keV. After March 1993, the sample to detector distance was increased and the resulting average resolution of the PIPS/lower shelf detectors arrangement was 36±7 keV.

Detection efficiency is the number of counts detected per unit time divided by the absolute activity (number of alpha particles emitted per unit time). In this project, the average of detection efficiency for the SSB detectors was 21±1% and 31±1% for the PIPS detectors. When the sample to detector distance was increased, the average detection efficiency for the PIPS detectors was decreased to 20±1%.

The SSB detectors used in this project were made by Ortec. It had an active surface area

of 450 mm^2. The SSB detectors are semiconductor junction detectors. They are typically made from a thin wafer of n-type silicon. To produce the junction, the silicon is etched on one surface with an acid vapor under conditions that promote oxidation. The resulting thin layer of silicon dioxide on the surface has the properties of a p-type material. The surface is then coated with a thin layer of gold to serve as the negatively charged cathode. The other side of the detector is coated with aluminum to serve as the positively charged anode. The SSB detectors were operated at room temperature, under vacuum, and in the dark. One disadvantage of SSB detectors is that the gold electrode is generally so thin that cleaning the detector face is almost impossible.[49]

The PIPS detectors used in this project were the partially depleted Canberra PIPS detectors (PD450-21-100AM). The active surface area is 450 mm^2, manufacturer's resolution is 21 keV for alpha detection with a silicon thickness of 100 m. The PIPS detectors were operated at room temperature, under vacuum, and in the dark. A PIPS detector is fabricated by the planar process using photolithographic techniques for defining device geometries. Proprietary techniques are used to provide precise control of the oxide passivation, and ion-implantation is used to form the accurately controlled junctions necessary for low reverse leakage currents and thin entrance windows. Low reverse leakage current translates into low noise contributions. Leakage current is typically 1/10 to 1/100 of that of SSB detectors. The face contact (entrance window) of the PIPS detectors is ion-implanted. This minimizes the window thickness while retaining the ruggedness, reliability and stability inherent in this type of contact. The PIPS detector has a window that is substantially thinner than conventional SSBs and far thinner than any detector approaching it in ruggedness. Because of this thin window the effective dead-layer thickness is less and the PIPS show improved resolution over that of comparable SSBs. Since PIPS detectors do not rely on delicate evaporated metallic contacts as do SSB detectors, but rather a passivated, implanted surface, they can be touched by hand and cleaned readily with a cotton ball dampened with acetone.[3]

G. Counting Statistics and Error

In order to determine the counting error in a radioactivity measurement the following assumptions are usually made. Nuclear transformations are random, independent, and occur at a constant rate. Another aspect and one that must be met is that the half-life of the isotope being measured be long compared to the counting time.

In radioactivity measurements, the Poisson distribution, derived from the binomial distribution, can be used since the total number of atoms (N_o) is usually very large and the observed gross counts (C_g) during the observing time (t_g) is very small compared to N_o. When using the Poisson distribution, the variance is equal to the mean and the estimated standard deviation (s) is equal to the square root of the mean.[49]

In order to determine the precision of a single measurement, some further assumptions need to be made: (1) that the single value has been drawn from a population whose theoretical distribution function is predicted by Poisson distribution statistics, and (2) that the mean of the distribution is equal to the single measurement. If these two assumptions are valid, the standard deviation is the square root of the single measurement.[49]

Since radioactivity measurements can be reported as gross count rates (R_g=gross count/observing time), the Poisson distribution is applied to count measurements such that the standard deviation of the count rate is equal to the square root of the count rate divided by the observing time. $[s_R=(R_g/t_g)^{1/2}]$

Most often, net count rates are reported rather than gross count rates. The net count rate is the rate for which the count rate due to background activity has been removed from the gross count rate $[R_{net} = R_g - R_b]$. The calculation of standard deviation must take into account the error from both measurements, therefore the standard deviation of the net count rate is shown in the following equation:

$$s_{net} = [(s_g)^2 + (s_b)^2]^{1/2} = [(R_g/t_g)^2 + (R_b/t_b)^2]^{1/2}$$

Sometimes error is expressed as a percentage. This is called coefficient of variation or

percent variation, and it is calculated by dividing the standard deviation by the count or count rate then multiplying by 100.

In this project, the designation most often used for error is the nine-tenths error (NTE) or reliable error calculation. NTE equals 1.645 times one on the standard deviation and represents a probability of 90% that a single measured value in the given set will not deviate from the average by more than ± 1.645(s). [38]

The error discussed above is solely due to the counting of random radioactive events. In low-level radioactivity work, the counting error is often the largest source of error, but other sources do exist. Some of the other sources of error in this project are measurements of volume and weight, losses resulting from chemical separations, losses resulting from transfer operations, variation in background radiation, instrument variations or contamination, self-absorption, and backscattering. These errors were usually minimized as much as possible. When calculating overall error, the square of the total error $[(s_{tot})^2]$ is equal to the sum of the squares of all the individual errors. An example of a calculation of overall error may include the following errors: Eppendorf pipet measurements 1% (100 ± 1 μL), balance measurements 0.3% (0.350 ± 0.001 grams), transfer losses 1%, self-absorption and scatter 1%, accuracy of tracer activity 4%, count error 10%.

$$(s_{tot})^2 = (1\%)^2 + (0.3\%)^2 + (1\%)^2 + (1\%)^2 + (4\%)^2 + (10\%)^2$$

$$s_{tot} = 11\%$$

Background determination of the surface barrier detectors was performed weekly. The count time was 1000 minutes [ITAS-OR-CR-7233]. Alpha background counts for all counting room instruments were generally from 0.01 to 0.1 cpm. The acceptable range was up to 0.5 cpm [ITAS-OR-CR-7223].

The efficiencies of the different surface barrier detectors were determined by the International Technology count room staff at quarterly intervals by using a primary calibration source which had at least one easily identifiable peak and had a National Institute of Standards

& Technology (NIST) traceable decay rate. Each detector was counted to a minimum of 20,000 count in the channels of interest. The efficiency was then determined to be the number of counts detected per unit time divided by the absolute activity (number of alpha particles emitted per unit time). The acceptable range for the efficiency of a detector was between 0.15 and 0.35 [ITAS-OR-CR-7235 and 7236].[54] The error in the efficiency values is a maximum of ±1.2% using NTE.

Samples within one experimental set were often measured simultaneously but on different detectors. This use of several different detectors is a possible source of error. A simple experimental test was performed in which one NdF_3 precipitated Am-241 sample was counted seven times under the same detector and then it was counted under 8 different detectors. The count time was 200 minutes with an average gross count of 900. For the seven samples counted under the same detector, the mean±NTE was 793±40 counts. The reliability factor was 0.863. If the reliability factor falls between 0.80 and 1.20, then the variability is generally due to radioactive decay. For the eight samples counted under different detectors, the gross counts were first converted to total net counts using the individual efficiency factors for each detector. The mean±NTE was 4342±362 counts and the reliability factor was 2.6. This reliability factor indicates that there is a high probability that the counts show a greater deviation from the Poisson distribution than would be expected from chance alone. In other words, something more than variability due to radioactive decay seems to be influencing the error.

Possible self-adsorption due to the thickness of the counting samples was evaluated by a series of NdF_3 precipitations of Am-241 using varying amounts of neodymium nitrate solution. The low concentrations of neodymium solution ($\leq$0.2 mg) showed poor recovery percentage and the high amounts of $Nd(NO_3)_3$ solution ($\geq$1.5 mg) resulted in spreading and degradation of the data peak. The optimum amount of added $Nd(NO_3)_3$ was 1.0 mg for nitrate acid solution (FWHM 44 keV) and 0.5 mg for sulfate-nitrate solution (FWHM 52 keV).

CHAPTER IV

EXPERIMENTAL APPROACH

A. Steps in Separations and Measurements

The overall process for the identification of actinides in low-level-radioactive environmental samples may be divided into five general steps. First, the sample must be solubilized. In the solubilization step, the submitted material is dissolved so that the desired actinides are put into aqueous ionic forms. The submitted samples can be in various forms, such as water, soil, concrete, sludge, building materials, biological tissue, etc. The second step is the elimination of any material in the solubilized matrix that may cause interference in subsequent steps of the actinide separation process. Matrix elimination may be accomplished by coprecipitation, complexation, reduction, ion exchange, etc. The third step is the separation of the desired actinide ions from each other into spectroscopically distinct groups. This is generally accomplished by solvent extraction or ion exchange. Fourth, the separated actinide ions must be put into some measurable form. Counting-sample preparation methods include direct evaporation, coprecipitation, electrodeposition, and cation exchange disks. The fifth and final step is the counting of the sample. Surface barrier detectors are usually used for spectroscopic measurement of alpha-emitting substances. Other detectors that could be used for gross counting are zinc sulfide scintillators and proportional counters.

In the experimental approach to this project, the final step of the separation process was investigated first. This approach was used to understand and/or control factors, as much as possible, in the last step that may effect the experimental results such that, when investigation of an earlier step is conducted, any change in experimental results may be more easily interpreted.

B. Counting of Samples

Surface barrier detectors were used to spectrometrically measure the alpha-particle energies.

The International Technology Analytical Services (ITAS) Count Room Staff determined the efficiencies of the surface barrier detectors by counting a NIST standard for 20,000 counts (see chapter III, section G). These efficiencies were used as determined by IT in the data calculations of this project.

Background determination of the surface barrier detectors was performed weekly by the IT Count Room Staff. The count time was 1000 minutes [ITAS-OR-CR-7233]. Alpha background counts for all counting-room instruments were generally from 0.01 to 0.1 cpm. The acceptable range was up to 0.5 cpm [ITAS-OR-CR-7223].

Counting variables which were investigated included (1) resolution of different energy alpha particles by use of split planchets, (2) precision check of counters, and (3) comparison of computer determined counts to the actual number of counts detected.

1. Resolution of different energy alpha particles.

One design consideration in the development of separation procedures for the light actinides (Th, U, Np, Pu, Am) was the capability of spectroscopically identifying mixed isotopes in one sample by the energies of their emitted alpha particles. This would be possible if resolution capabilities of the surface barrier detectors were sufficient to resolve to base line (or very close to base line) the energy peaks of the different alpha particles. This would allow identification of certain isotopes without requiring chemical separation procedures. In order to evaluate this identification capability (resolution capability) of the surface barrier detectors, a series of individual isotopic solutions was coprecipitated onto separate 25 mm filters using the NdF_3 procedure (chapter III, section E). The samples were spectroscopically measured as coprecipitated. Then, the filters were cut in half and different combinations of halves were measured spectroscopically. All the spectroscopic measurements of the above samples were performed on SSB detectors.

(See chapter III, section F, for a discussion about alpha detectors.)

A second resolution evaluation was performed using a solution containing Pu-242 (alpha energies: 4856(22%), 4900(78%) keV) and Np-237 (predominant alpha energies: 4640(6.2%), 4664(3.3%), 4766(8%), 4771(25%), 4788(47%), 4817(2.5%), 4873(2.6%) keV).[51] This mixture was measured both on the SSB detectors and the PIPS/lower-shelf detectors. Pu-242 was being considered as a yield tracer for measurements of Pu and Np isotopes.

A third resolution evaluation was conducted using the Th-227 isotope. This nuclide emits several alpha particles at different energies (predominant alpha energies: 5668(2.1%), 5700(3.6%), 5709(8.2%), 5713(4.9%), 5757(20), 5866(2.4%), 5959(3.0%), 5978(23%), 6009(2.9%), 6038(24%) keV). With complete resolution, the energies of the alpha particles could be separated into individual peaks. However, with ordinary spectroscopic instrumentation, that level of resolution cannot be achieved. Therefore, the intent of these measurements was to determine if two groups of similar-energy alpha particles could be resolved from each other. The first group ranges from 5668-5757 keV and the second group ranges from 5866-6038 keV. The lower energy band overlaps with some of the decay daughters of other Th isotopes that may be present, but if resolvable, the second band would be able to be measured quantitatively for the amount of Th-227 present. These samples were counted on the PIPS/lower-shelf detectors.

2. Precision check of counters.

Samples within one experimental set were often measured simultaneously but on different detectors. This use of several different detectors is a possible source of error. A simple experimental test was performed in which one NdF_3 precipitated Am-241 sample was counted seven times under the same SSB detector and them it was counted under 8 different detectors (4 SSB, 4 PIPS). The count time was 200 minutes with an average gross count of 900. The first part of the test was to evaluate the precision of repeated counts on one detector. The major source of error within these counts would be expected to be due primarily to the random decay characteristic of the radioactive material. The second part was to evaluate the precision of counts

measured on several different detectors. In this situation several sources of error are possible, such as random decay, efficiency measurements of different detectors, movement and realignment of the sample between counts.

3. Comparison of computer-determined counts to actual counts.

The computer program utilized in the evaluation of the resulting spectra had preset energy regions for determination of a given isotope's alpha particle energies. Sometimes these preset regions appeared not to include the entire area of the peak. Therefore, several series of measurements were tallied by hand and compared to the computer totals to evaluate any error that could be resulting due to computer-generated-lost peak area.

C. Preparation of Samples for Counting

The NdF_3 coprecipitation method was used for the preparation of actinide samples for alpha counting. The IT procedure included the following steps: First, the separated actinide fraction of the sample is converted by evaporation to the sulfate form, but only partially dried. The sulfates are dissolved in 15 ml of 3.0-M HCl and placed into a hot water bath (85°C) for 10 minutes. Second, the coprecipitate is formed by adding 0.50 ml of 0.10 mg/ml $Nd(NO_3)_3$ carrier, mixing, and then adding 5.0 ml 48% HF (29-M). This mixture is then placed into an ice bath for a minimum of 10 minutes. Third, a polypropylene membrane filter is prepared by filtering an aqueous, alpha-emitter-free carbon substrate suspension that contains a small, known amount of NdF_3 precipitate. (This is to improve spectral resolution by making the filter surface more nearly flat microscopically.) Finally, the sample is filtered through the prepared membrane using a moderate vacuum. The membrane with its precipitate is washed using dilute HF, then deionized HOH, and then ethanol. The membrane is dried, mounted onto a paper disk, and counted.[ITAS-OR-RS-7124][52]

The formation of a NdF_3 precipitate in a solution of very dilute actinides results in the +3 and +4 actinide ions being separated by the coprecipitate from the solution. These actinide ions

are coprecipitated with the NdF_3 by inclusion and/or adsorption/trapping processes. Inclusion is the situation in which an actinide ion of similar size and charge replaces the Nd^{+3} ion in the crystal lattice. In the adsorption process an ion adheres to the surface of the crystal and as the crystal continues to grow this adsorbed ion may become trapped within the crystal. Inclusions may be enhanced when the crystal anneals or ages while the adsorption/trapping may be lessened as the crystal ages.

Coprecipitation procedure variables which were evaluated are (1) the amount of $Nd(NO_3)_3$ being added with respect to self adsorption and optimum recovery, (2) the use of temperature controls to enhance coprecipitation, (3) the length of time allowed for precipitate formation, (4) the use of continuous agitation to enhance precipitate formation, (5) the effect of layering an actinide fluoride precipitate, without Nd^{+3} carrier, on the surface of a membrane coated with NdF_3 precipitate, (6) actinide ions remaining in solution after NdF_3 coprecipitation, and (7) the effect of Na_2SO_4/H_2SO_4 present in the solution matrix when NdF_3 coprecipitant is formed. The last investigation, relating to coprecipitation procedures, was a check for contamination in the reagents and equipment by setting up reagent blanks (number 8).

1. Amount of $Nd(NO_3)_3$ added.

A NdF_3 sample for alpha energy spectroscopy needs to be extremely thin in order to minimize the amount of degradation due to self-adsorption and self-scattering within the sample itself. Also, the amount of added Nd^{+3} should be sufficiently high in order to coprecipitate the desired actinides at consistently high percentage recovery levels. In order to evaluate the optimum quantity of Nd required, experiments were conducted in which Am^{+3} and Th^{+4} were separately coprecipitated using various amounts of $Nd(NO_3)_3$ reagent.

2. The use of temperature controls to enhance coprecipitation.

In the IT procedure, the sample is first heated in a hot water bath prior to the addition of the Nd^{+3} and HF. After addition of these coprecipitating reagents, the sample is then placed in an ice bath. Research had indicated that high temperature tends to help in the aging process of

the crystal while low temperature will slow down the aging process. Experiments were conducted to evaluate the impact of these temperature controls relative to coprecipitation at room temperature. Various actinide ions (An^{+3}, An^{+4}, and AnO_2^{+2}) were evaluated.

3. The length of time allowed for precipitate formation.

The IT procedure stated that the sample was required to stand for a minimum of 10 minutes in order to allow the coprecipitate to form. This time frame often varied in maximum length. An experiment was conducted to evaluate the impact of various lengths of time on the percentage recovery of the desired actinides.

4. Continuous agitation during coprecipitate formation.

After all reagents were added, the sample was mixed and allowed to stand until the sample was filtered. Some researchers have studied the effect of continual agitation relative to particle size formation and ion incorporation.[53] A series of samples was continuously agitated after the addition of the coprecipitating reagent and the results evaluated relative to non-agitated samples.

5. Filtration of actinide ions through a membrane whose surface had been previously coated with NdF_3 precipitate.

The thinner the layer of the fluoride precipitate on the membrane the better the possible resolution due to decreased self-adsorption. One proposed way to produce a thin actinide fluoride layer was by adsorbing it onto a previously filtered layer of NdF_3. This would possibly minimize the number of actinide ions which were included or trapped within the precipitate.

6. Actinide ions remaining in solution after NdF_3 coprecipitation.

At times, the percent recovery of actinide ions from solution was less than 100 ± NTE. It was possible that all the actinide ions were not being precipitated from the solution using the NdF_3 coprecipitation procedure. In order to check this, Am-241 samples were coprecipitated using the NdF_3 coprecipitation procedure at room temperature. The filtered solutions were saved and evaporated to reduce volume. The filtered solution samples were fumed and then diluted with 3.0-M HCl to 15 ml final volume. These again were coprecipitated using the NdF_3 procedure but

with the amount of added Nd^{+3} reagent doubled.

7. Na_2SO_4/H_2SO_4 in matrix of NdF_3 coprecipitation solutions.

In the first step of the NdF_3 coprecipitation procedure there is a conversion of the actinide by evaporation to the sulfate. This is primarily a final clean up step to remove any volatile acids (HNO_3, HCl) or organic material prior to the coprecipitation of the sample. The effect of SO_4^{-2} in the solution matrix on the percent recovery of Am^{+3} relative to no SO_4^{-2} ions in the solution matrix was evaluated.

8. Reagent Blank.

The final investigation of this step was an evaluation of reagent and/or equipment contamination. Several unspiked NdF_3 precipitated samples were counted to check for the presence of radioactivity due to contaminated glassware, reagents, filters, or filtration apparatus.

D. Separation of Actinide Ions into Spectroscopically Distinct Groups

In this series of experiments, the intention was to separate the Am, Th, Np, Pu, and U nuclides into groups that could be spectroscopically resolved using the surface barrier alpha detectors. Due to the usual commercial requirement for yield tracers and the resulting spectral overlaps, the experiments were designed to provide four separate fractions: Am nuclides fraction, Th nuclides fraction, U nuclides fraction, and Np/Pu nuclides fraction. In the following developmental steps, the actinide ions were placed in a deionized water matrix.

The first type of separation method investigated was the selective coprecipitation of a certain actinide(s) while leaving the other actinide(s) in solution. These were screening tests for a coprecipitation method that would separate and produce a sample of sufficient quality that it could be alpha counted without additional preparation in a surface barrier detection system. Coprecipitants tested as possible separation and counting media agents were: (1) zirconium phenylarsonate, (2) metal m-nitrobenzoate, (3) metal iodate, (4) yttrium-calcium-magnesium mixed-

fluorides, and (5) neodymium fluoride.

Next, TRU-Resin™ (EIChroM) was investigated as a possible tool for separation of the actinide ions. This series of experiments included: (6) separation of low activity levels of Am ions from low to high activity levels of U ions, (7) separation of low activity levels of Am ions from low to high activity levels of both Th and U ions, (8) separation of Np and Pu ions together in the same elution fraction from Am, Th, and U, (9) refinement of the Th elution step, (10) refinement of the U elution step, (11) some miscellaneous topics included Ce^{+4} as a carrier for Th-229 and the effect of various redox reagents on the elution pattern from the TRU-Resin™ column, and (12) independent separations performed by Mr. Tiam Xie.

1. Coprecipitation using zirconium phenylarsonate.

Phenylarsonic acid, $C_6H_5AsO(OH)_2$, forms an insoluble compound with the zirconium ion. This coprecipitant has been used for the separation of M^{+4} actinide ions from M^{+3} ions by several authors. [26,55,56,57] In this experiment, a solution of Th^{+4} and Am^{+3} ions in HCl solution was used. The $ZrO(NO_3)_2$, $Nd(NO_3)_3$, and $C_6H_5AsO(OH)_2$ were added in various concentrations. The phenylarsonate coprecipitate was measured directly using the surface barrier detectors and the filtered solution was also tested for the presence of any actinide ions remaining in solution.

2. Coprecipitation using m-nitrobenzoic acid.

Previous work describes the use of m-nitrobenzoic acid, $O_2N\text{-}C_6H_4\text{-}CO_2H$, for the precipitation of thorium from a solution of its salts.[58,59] The previous work discussed the use of this method as a means of removing Th from rare earth ores.

3. Coprecipitation using the iodate anion.

The precipitation of lanthanides and actinides cations with iodate anions is used in purifying Am and in removing Pu and Np from solution before proceeding with analyses.[56] In Gmelin, evidence is given for the selective coprecipitation of tetravalent cations of the transuranic elements with insoluble iodates.[26] Some relevant -log Ksp values are: $Ce(IO_3)_3$ 10.86, $Ce(IO_3)_4$ 9.50, $La(IO_3)_3$ 10.99, and $Th(IO_3)_4$ 14.62.[25]

4. Coprecipitation using Y-Ca-Mg mixed-fluoride.

Kallmann et. al. reports that the fluoride precipitation of a solution containing a mixture of yttrium, calcium, and magnesium cations will quantitatively collect rare earth fluorides in the presence of large amounts of uranium, beryllium, titanium and/or zirconium.[60] Since Am^{+3} can react similarly to the rare earth ions, this method was tested for its possible use in separating Am^{+3} ions from UO_2^{+2} ions.

5. Coprecipitation using NdF_3.

NdF_3 coprecipitation is routinely used to prepare a thin layer of solid material on a filtration membrane for counting purposes. However, all of these precipitations were performed after the desired actinide had been separated from other actinides and matrix ions. In Gmelin, it is indicated that a NdF_3 coprecipitation may be useful in coprecipitation of M^{+3}, M^{+4}, and/or MO_2^+ ions in the presence of MO_2^{+2}.[26] Some discussion is present concerning the effectiveness of a fluoride coprecipitation method for ions in the form of MO_2^+. The complexation constant, log $beta_1$, for NpO_2^+ with F^- has been measured to be 0.90-1.8.[61] Some relevant -log Ksp values are: CaF_2 10.41, CeF_3 15.1, ThF_4 28.3, UF_4 21.24. Some relevant log beta values with F^- are: La^{+3} $beta_1$ 3.6, Ce^{+3} $beta_1$ 4.0, Th^{+4} $beta_4$ 23.2, UO_2^{+2} $beta_4$ 21.24.[25]

Determinations using NdF_3 coprecipitation as a separation technique included: (a) separation of Am form U in a 1:1 mixture, (b) separation of Am from high levels of U, (c) the effect on the percent recovery of Np with a reductant ($TiCl_3$) in the solution, and (d) the effect on the percent recovery of Np, Pu, and Th with different oxidizing and/or reducing agent in solution ($TiCl_3$, H_2O_2, $K_2S_2O_8$).

The next set of experiments used the TRU-Resin™ extraction resin marketed by EIChroM. Previous research with this resin was discussed in chapter II, section F. For the various experiments discussed below, a 1.0 ml bed volume column of TRU-Resin™ was used. The preparation of this column is described in chapter III, section D.

6. Separation of low activity levels of Am ions from low to high activity levels of U ions.

Four experiments were conducted using a mixture of Am and U ions: (a) separation of low activity levels of both Am and U in 1.0-M HNO_3 load solution, (b) separation of a low activity level of Am from U activity levels that were 10, 100, and 1000 times higher than the Am level using a load solution of 3.0-M HNO_3, (c) separation of a low activity level of Am from U activity levels that were 10, 100, and 1000 times higher than the Am level using a load solution of 3.0-M HCl, (d) separation of a low activity level of Am from a 1000 times higher U activity level in 3.0-M HCl by comparing static resin loading with column resin loading.

The TRU-Resin™ material, particle size 50-100 m , has been characterized by previous researchers by measuring k' values as a function of HNO_3 or HCl concentration for tri-, tetra-, penta-, and hexavalent actinides and for selected non-actinide ions.[36] The constant k' is defined to be the number of free column volumes to peak maximum. It relates to the distribution ratio of the adsorbed ions to the unadsorbed ions ($D=[MR]^n/[M^{+n}]$) by the following equation: $k'= D(v_s/v_m)$ or k'= D(volume of stationary phase divided by the volume of the mobile phase). The applicable log k' values for these separations include: Am^{+3} in 1.0-M HNO_3=2.0, Am^{+3} in 3.0-M HNO_3=2.0, Am^{+3} in 3.0-M HCl=<0, UO_2^{+2} in 1.0-M HNO_3=3.1, UO_2^{+2} in 3.0-M HNO_3=3.6, UO_2^{+2} in 3.0-M HCl=3.3. EIChroM recommends that the TRU-Resin™ material be used at loadings of no more than 10-20% resin capacity. Resin capacity is defined as the maximum possible loading of the chromatographic column. The TRU-Resin™ 50-100 m particle size resin has a capacity of 9.1 mg of Am-241 per ml of resin bed, indicating a recommended useful loading of 1-2 mg/ml or approximately $1E^{10}$ dpm Am-241/ml of resin-bed volume.[40,62]

Complexation constants (log beta values) for different cations with NO_3^- are: $\underline{La}^{+3}$ $beta_1$=0.1; $\underline{Am}^{+3}$ $beta_1$=0.26; $\underline{UO_2}^{+2}$ $beta_1$=-0.3; $\underline{Th}^{+4}$ $beta_1$=0.67, $beta_2$=0.15; $\underline{U}^{+4}$ $beta_1$=0.20, $beta_2$=0.2, $beta_3$=0.0, $beta_4$=-0.5; $\underline{Np}^{+4}$ $beta_1$=0.38, $beta_2$=0.2, $beta_3$=-0.3; $\underline{NpO_2}^{+}$ $beta_1$=-0.25.[25] Complexation constants for different cations with Cl^- are: $\underline{La}^{+3}$ $beta_1$=-0.1, $beta_2$=-0.6; $\underline{Am}^{+3}$ $beta_1$=-0.6 $beta_2$=-0.4; $\underline{UO_2}^{+2}$ $beta_1$=-0.1; $\underline{Th}^{+4}$ $beta_1$=0.18, $beta_2$=-1.0

$beta_3$=-0.9, $beta_4$=-1.5, $beta_5$=-2.5; $\underline{U}^{+4}$ $beta_1$=0.3; $\underline{Np}^{+4}$ $beta_1$=-0.04, $beta_2$=-0.15, $beta_3$=-0.5; $\underline{NpO_2}^{+2}$ $beta_1$=-0.3, $beta_2$=-0.8; $\underline{Pu}^{+3}$ $beta_1$=-0.1; $\underline{Pu}^{+4}$ $beta_1$=0.14, $beta_2$=-0.17, $beta_3$=-1.0.[25]

7. Separation of low activity levels of Am ions from low to high activity levels of Th and/or U ions.

Two experiments were conducted using a mixture of Am and Th ions: (a) separation of a low activity level of Am from a Th activity level that was 4 times higher than the Am activity level. This experiment was conducted twice, first with a load solution of 4.0-M HCl and second with a load solution of 5.0-M HCl.

Two additional experiments were conducted using a mixture of Am, Th and U ions: (b) separation of low activity levels of Am and Th from U activity levels that were 100 and 300 times higher than the Am activity level using a load solution of 3.0-M HCl, (c) separation of a low activity level of Am from Th and U activity levels that were 4-6 times higher than the Am level using a load solution of 5.0-M HCl. In experiments (b) and (c), the Th ions were eluted from the TRU-Resin™ column using 1.0-M HCl solution.

The applicable log k' values for these separations include: Am^{+3} in 1.0-M HCl=<0, Am^{+3} in 3.0-M HCl=<0, Am^{+3} in 4.0-M HCl=<0, Am^{+3} in 5.0-M HCl=0.3, Th^{+4} in 1.0-M HCl=0.2, Th^{+4} in 3.0-M HCl=2.0, Th^{+4} in 4.0-M HCl=3.1, Th^{+4} in 5.0-M HCl=3.8, UO_2^{+2} in 1.0-M HCl=2.3, UO_2^{+2} in 3.0-M HCl=3.3, UO_2^{+2} in 5.0-M HCl=3.5.[36]

8. Separation of Np and Pu ions together in the same elution fraction from Am, Th, and U.

All of these experiments were conducted using a solution that contained a mixture of low activity levels (approx. 15 dpm) of Am, Np, Pu, U, and Th ions. The areas of investigation included: (a) loading Np and Pu ions onto the TRU-Resin™ with and without H_2O_2 in the load solution, (b) comparison of HCl, ammonium iodide, dilute oxalic acid, and a mixture of ammonium iodide and oxalic acid as possible eluting solutions for Np and Pu ions.

The applicable log k' values for these separations, in addition to the ones listed previously,

include: Am^{+3} in 0.5-M HCl=<0, Th^{+4} in 0.5-M HCl=<0.2, UO_2^{+2} in 0.5-M HCl=1.8, Np^{+4} in 0.5-M HCl=0.9, Np^{+4} in 1.0-M HCl=1.0, Np^{+4} in 4.0-M HCl=4.0, Np^{+4} in 5.0-M HCl=4.5, Pu^{+4} in 0.5-M HCl=1.0, Pu^{+4} in 1.0-M HCl=1.3, Pu^{+4} in 4.0-M HCl=5.0, Pu^{+4} in 5.0-M HCl=5.6.[36] No k' values are given for Np^{+3}, NpO_2^+, or Pu^{+3}. The Np^{+3} and the Pu^{+3} would probably behave similar to Am^{+3} on the TRU-Resin™.

The I^-, from the ammonium iodide, can be oxidized to I_2. The E^o value for the I_2/I^- couple is 0.54 volts.[18] Thus I^- has the potential to reduce Pu^{+4} to Pu^{+3}, but it does not have the potential to effect the oxidation state of the Np^{+4} ions. The oxalic acid, $H_2C_2O_4$, may also act as a redox agent. It can be reduced to formic acid, HCOOH, or oxidized to CO_2 or H_2CO_3, (E^o=0.08v for $H_2C_2O_4$/HCOOH, E^o=-0.38v for $H_2CO_3/H_2C_2O_4$, E^o=-0.49v for $CO_2/H_2C_2O_4$). [25]

Equilibrium constants (log values) for different cations with $C_2O_4^{-2}$ are: $\underline{H}^+$ K_{H1}=3.8, K_{H2}=1.0; $\underline{La}^{+3}$ $beta_1$=4.3, $beta_2$=7.9, $beta_3$=10.3; $\underline{Th}^{+4}$ $beta_1$=8.2, $beta_2$=16.8, $beta_3$=22.8; UO_2^{+2} $beta_1$=6.0, $beta_2$=10.6, $beta_3$=11.0. Complexation constants for different cations with I^- are: $\underline{U}^{+4}$ $beta_1$=0.2; $\underline{Bi}^{+3}$ $beta_1$=3.6, $beta_4$=15.0, $beta_5$=16.8, $beta_6$=18.8.[25] (Bi^{+3} is similar in ionic radius to Pu^{+3}, Np^{+3}, and Am^{+3}.)

A summary of the behavior of Np, Pu, U, and Am ions with H_2O_2 is shown in Appendix B, Table XII. Note that the NpO_2^+ ions are reduced to Np^{+4} and Pu^{+3} ions are oxidized to Pu^{+4}. The Pu^{+4} ions can combine with H_2O_2 to form a peroxyl complex.

9. Refinement of the Th elution step.

Some overlap between the Th eluted fraction and the Np/Pu eluted fraction became evident in the separation tests. Therefore additional steps were taken to further refine conditions to improve the separation between these two fractions. The goal of these tests was to determine a process that would minimize elution overlap of the two fractions, allow rapid elution of the Th fraction, and minimize the amount of reagent needed for elution. The experiments included: (a) adjusting the molar concentrations of the HCl eluting solution (2.0-M, 1.5-M, 1.4-M, 1.3-M, 1.1-M, 1.0-M), (b) adjusting the hydrogen ion concentration while keeping the chloride ion

concentration constant, (c) adjusting the chloride ion concentration while keeping the hydrogen ion concentration constant, and (d) miscellaneous observations relating to percent recovery of Th relative to (1) NH_4I in the solution and (2) sulfate fume clean up step.

10. Refinement of the U elution step.

In previous work, ammonium hydrogen oxalate ($NH_4HC_2O_4$) has been used to elute the U.[36] The percent recovery, when $NH_4HC_2O_4$ was used, was adequate but not 100%. The unrecovered U was not detected in any other eluted fraction so it was assumed that the U remained on the column. Therefore some additional tests were conducted to further refine conditions to improve the percent recovery of U. The experiments included: (a) elution of a previously eluted column using $(NH_4)_2C_2O_4$, (b) elution of U following the use of NH_4I in the previous eluting reagent (Np/Pu fraction), and (c) elution of U using HF. (The use of HF in dilute HCl for the elution of U has been tried.[63])

11. Miscellaneous topics.

(a) Using Ce^{+4} as a carrier for Th-229^{+4}.

One of the common commercial requirements for actinide separation work is to include a blank sample in each sample run. This blank is designed to test for the presence of any source of the desired actinides other than the sample itself. In the separation procedure, Th-229 is used as a tracer for Th analysis. Thus in the uncontaminated blank, Th-229 is the only Th isotope that would be present. In several sample runs, a pattern was noted in which the percent recovery of Th-229 from the blank was often low while in a sample that had other Th isotopes present, the Th-229 percent recovery was usually very good. This behavior of Th-229 may relate in part to the low molar concentration (1.4E-13 M) of the short-lived Th-229 in the solution and the strong hydrolyzing property of Th^{+4}. Since the percent recovery was good in the presence of other longer-lived Th isotopes, the use of a non-isotopic carrier ion for the Th-229 in the blank was proposed. An ion that is somewhat similar to Th^{+4} is Ce^{+4}. Peroxydisulfate was added to the solution to keep the Ce^{+4} from reducing to Ce^{+3}.

Some applicable data: E^o for $Ce(OH)^{+3}/Ce^{+3}$ = 1.7v, E^o for $S_2O_8^{-2}/SO_4^{-2}$ is 2.0v, ionic radius for Ce^{+4} is 101pm, ionic radius for Th^{+4} is 108pm, and complexation constants (log beta values) for SO_4^{-2}: $\underline{Am}^{+3}$ $beta_1$=1.57, $beta_2$=2.7; $\underline{Ce}^{+3}$ $beta_1$=1.77, $beta_2$=2.9; $\underline{Th}^{+4}$ $beta_1$=3.2, $beta_2$=5.5; $\underline{UO_2}^{+2}$ $beta_1$=1.8, $beta_2$=2.5, $beta_3$=3.7.[10,18,25] (The complexation characteristics of $S_2O_8^{-2}$ are probably similar to SO_4^{-2}.)

(b) The effect of various redox reagents on the elution pattern of the actinides from the TRU-Resin™ column.

The loading and eluting patterns of an ion on the TRU-Resin™ was assumed to relate in part to its oxidation state. In order to investigate this, different oxidizing and reducing agents were used to adjust the redox potential of the solution of actinide ions prior to or during loading of the TRU-Resin™ column. Redox agents used were: (1) Zn metal (E^o Zn^{+2}/Zn = 0.77v), (2) hydrazine sulfate, $H_2NNH_2 \cdot H_2SO_4$ (E^o $N_2/N_2H_5^+$ = -0.23v), and (3) H_2O_2 (E^o H_2O_2/H_2O = 1.77v, E^o O_2/H_2O_2 = 0.67v).[10,15,18]

12. Independent separations performed by Mr. Tiam Xie.

Mr. Xie, an experienced radiochemistry technician, was working for IT during my research. In this period, he conducted three experimental sample runs using a written copy of the TRU-Resin™ separation procedure. In each run, he made up 2 blanks and 4 or 6 test sample using a distilled water matrix. Mr. Xie tested for following elements: Am, Pu, Np, U, and Th. He used a 1.0 ml bed volume column prepared as previously described in chapter III, section D.

The separation procedures that Mr. Xie followed were slightly different each time reflecting the on going research. In the first separation run, he used 4.0 ml of 1.30-M HCl to elute the Th fraction, 20.0 ml of 0.10-M NH_4I + 0.030-M $H_2C_2O_4$ in 0.50-M HCl to elute the Np/Pu fraction, 40.0 ml of 0.10-M $(NH_4)_2C_2O_4$ to elute the U fraction, and 20 ml of 3.0-M HCl to elute the Am fraction. In the second and third separation runs, Mr. Xie used 6.0 ml of 1.30-M HCl to elute the Th fraction, 15.0 ml of 0.030-M $H_2C_2O_4$ in 0.50-M HCl to elute the Np/Pu fraction, 20.0-30.0 ml of 0.10-M $(NH_4)_2C_2O_4$ to elute the U fraction, and 15 ml of 3.0-M HCl to elute the Am fraction.

(In each run, a calcium oxalate coprecipitation procedure was used as part of the Am separation.) A detailed description of the separation procedure is presented in chapter VI, section A.

E. Separation of Actinide Ions in the Presence of Various Matrix Ions

In this series of experiments, the intention was to separate the Am, Th, Np, Pu, and U nuclides into spectroscopically resolvable fractions from samples that contained various metal ions in the matrix. The previous experiments in this project, that were used to develop a possible separating procedure for these actinides, used samples with a deionized water matrix. The two types of matrix samples that were investigated included soil samples and hard water samples.

The first type of matrix investigated was soil samples. One to two grams of ground and muffled soil were solubilized using the Prolabo digestion system (See chapter IV, section F.). These samples were from different parts of the country, such as Weston OH, Fernald OH, Denver CO, and Knoxville TN. Some of the samples contained radioactive elements, but other did not.

The composition of soil samples can vary significantly from one location to another. Common compositional ranges for soils are: 7.1-0.3% organic matter, 94.5-44.2% SiO_2, 28-2% Al_2O_3, 16-1% Fe_2O_3, 0.5-0% MnO, 1-0.5% K_2O, 1.2-0.7% TiO_2, 1.4-0.4% CaO, 2.0-0.1% MgO, 3.6-0.1 K_2O, and 1.4-0.1% Na_2O.[64]

After some initial soil matrix experiments, it became evident that the percent recovery of Am was low due to the presence of the soil matrix in the sample. An additional matrix cleanup step was added to improve the percent recovery of Am ions. This clean up step was a calcium oxalate coprecipitation of the load filtrate from the sample loaded in HCl.[26,56] The americium-calcium oxalate coprecipitate was then muffled and dissolved in nitric acid. Solid ascorbic acid was added to the HNO_3 solution to reduce any remaining Fe^{+3} to Fe^{+2}.[36] The HNO_3 sample was then loaded onto the same TRU-Resin™ column which had been used for the sample when it had been dissolved in HCl.

The steps in the calcium oxalate procedure are: (1) dissolve evaporated sample in

25-50 ml 8.0-M HNO_3, warm until dissolved, reduce volume to approx. 20 ml, add deionized water until total volume equals 100 ml, (2) add 80 ml saturated $H_2C_2O_4$, then 2.0 ml calcium carrier (50 mg/ml Ca^{+2} in 0.40-M nitric acid), (3) adjust pH to 1.6-1.7 by adding 30% NH_4OH dropwise, let mixture stand for at least 30 min., (4) centrifuge, decant, transfer solid to beaker and dry slowly over low heat, (5) heat to 475°C for 30 min. in a muffle furnace, and (6) wet ash with HNO_3 and H_2O_2.[ITAS-OR 7125]

The oxalate coprecipitation procedure will remove M^{+3} and M^{+4} actinide ions from solution. [26] Using calcium oxalate coprecipitation, Pu^{+3} and Am^{+3} were successfully separated from highly salted magnesium, aluminum, and calcium nitrate solutions and partially separated from rare earths solutions. [26,56] The oxalate anion can form complexes with some of the cations from the soil matrix, thus preventing them from significantly coprecipitating with the insoluble calcium oxalate. Some relevant complexation constants (log beta values) are: Ce^{+3} $beta_1$=4.5, $beta_2$=7.9, $beta_3$=14.3, $beta_4$=11.8; Al^{+3} $beta_1$=6.1, $beta_2$=11.1, $beta_3$=15.1; Fe^{+3} $beta_1$=7.5, $beta_2$=13.6, $beta_3$=18.5; Fe^{+2} $beta_1$=3.1, $beta_2$=5.2; Th^{+4} $beta_1$=8.2, $beta_2$=16.8, $beta_3$=22.8; UO_2^{+2} $beta_1$=6.0, $beta_2$=10.6, $beta_3$=11.0; Mn^{+2} $beta_1$=10.2, $beta_2$=16.6, $beta_3$=18.4; Ca^{+2} $beta_1$=1.7, $beta_2$=2.7; Mg^{+2} $beta_1$=2.7, $beta_2$=4.5. Some relevant -log K_{sp} values are: CaC_2O_4=7.9, $Ce_2(C_2O_4)_3$=25.5, MgC_2O_4=4.1, $Np(C_2O_4)_2$=22.1, $Pu(C_2O_4)_2$=21.3, $Th(C_2O_4)_2$=25.0, and $UO_2C_2O_4$=8.7.[25]

The initial soil matrix experiments included: (1) separation of Am, Th, and U ions from a soil matrix using HCl-load TRU-Resin™ separation, (2) separation of Am ions from a soil matrix using HNO_3-load TRU-Resin™ separation, and (3) separation of Am, Th, and U ions from a soil matrix using HCl-TRU-Resin™ separation followed by HNO_3-ascorbic acid-load TRU-Resin™ separation. From the results of these tests, the calcium oxalate coprecipitation step was added to the procedure. Some experiments that were conducted to evaluate the effectiveness of this new step were: (4) adding actinide spikes before and after the calcium oxalate coprecipitation procedure, (5) processing half of the eluted Am fraction with the calcium oxalate step and half

without, (6) processing soil samples for effective percent recovery of the actinide ions.

The second type of matrix investigated was simulated hard water samples (section 7). The matrix ions that were investigated included Al^{+3}, Fe^{+3}, Ca^{+2}, and Mg^{+2}. The samples consisted of varying amounts of the metal matrix ions dissolved in HCl solution and spiked with actinide ions.

1. Separation of Am, Th, and U ions from soil matrices using HCl-load TRU-Resin™ separation.

The initial experiments utilized dissolved soil samples with different combinations of Am, Th, and/or U ions added. The samples were then loaded onto the TRU-Resin™ column in 3.0-M HCl solution. The eluting solutions were the same as those that had worked for the deionized water matrix samples. The soil matrix presented problems for recovery of the Am ions.

2. Separation of Am ions from a soil matrix using HNO_3-load TRU-Resin™ separation.

When loaded in HNO_3 solution, the Am ions will attach to the TRU-Resin™. A soil sample containing Am ions was loaded in 3.0-M HNO_3 solution onto a TRU-Resin™ column. The Am was then eluted with 4.0-M HCl.

3. Separation of Am, Th, and U ions from a soil matrix using HCl-TRU-Resin™ separation followed by HNO_3-ascorbic acid-load TRU-Resin™ separation.

Three 2.0 gram soil samples were prepared. Each sample contained low activity levels of Am and 6 to 100 times higher activity levels of Th and/or U ions. The samples were loaded in 5.0-M HCl plus H_2O_2 solution. The eluted HCl-load solution (Am fraction) was wet ashed and reloaded in 3.0-M HNO_3 plus ascorbic acid onto the same TRU-Resin™ column.

4. Adding actinide spikes before and after the calcium oxalate coprecipitation procedure.

Two soil samples were solubilized. The first sample was spiked with Am, Np, Pu, U, and Th ions and then coprecipitated using the calcium oxalate procedure. The second sample was coprecipitated using the calcium oxalate procedure and then spiked with Am, Np, Pu, U, and Th ions. The percent recovery results were then compared to evaluate any loss of actinide ions

which could possibly be due to poor coprecipitation with calcium oxalate.

5. Processing half of the eluted Am fraction with the calcium oxalate coprecipitation step and half without.

Four soil samples and two deionized water samples were included in this experiment. The samples contained approximately 20 dpm each of Th, Np, Pu, Am and U ions. Each sample was loaded in 5.0-M HCl plus H_2O_2 solution onto a separate TRU-Resin™ column. The eluted HCl-load solution was divided into two equal volumes. One half was wet ashed only and the other half was coprecipitated using the calcium oxalate procedure. The two parts were then separately redissolved in 3.0-M HNO_3 plus ascorbic acid. The split fractions were loaded onto separate TRU-Resin™ columns and eluted with HCl to remove the Am ions. The percent recovery of the Am ions were compared to evaluate the effectiveness of the calcium oxalate step.

6. Processing soil samples for effective percent recovery of the actinide ions.

Some additional soil samples were processed using the TRU-Resin™ column separation procedure with the HCl-load steps followed by the calcium oxalate coprecipitation and the HNO_3-load steps.

7. Separation of Am, Th, Np, Pu, and U ions from hard water matrices.

These experiments included: (a) separation of actinide ions from solution matrices containing 10.0 g of Al, Fe, Ca, or Mg ions, (b) separation of actinide ions from solution matrices containing 1.0 g of the metal ions, and (c) separation of actinide ions from solution matrices containing 0.50 g, 0.10 g or 0.010 g of the metal ions.

F. Separation of the Actinide Ions as One Group from Soil Matrices.

In this series of experiments, the intention was to separate the Am, Th, Np, Pu, and U nuclides as one group from solubilized soil samples. This was investigated because the individual nuclides may be better separated if the sample matrix could be simplified prior to loading the sample onto the TRU-Resin™ column. Some characteristics of a good clean-up step are: simple

procedural steps, selectively separate actinide ions, inexpensive, and minimal labor requirements.

The soil samples were first solubilized, either by a manual microwave procedure or an automated microwave procedure (Prolabo). The microwave-digest methods are used to achieve sample dissolution by microwave heating of the sample with mixed acids. In the manual microwave procedure, the samples were wet ashed 3-4 times with HNO_3 plus HF using a hot plate. The samples were then transferred into a digestion vessel, combined with HNO_3 plus HF, and heated in a microwave. The samples were then removed and sequentially heated with $HClO_4$, aqua regia, and HNO_3 plus H_2O_2. In the Prolabo system, the following acids are added sequentially and automatically by computer control to the samples with microwave heating after each addition: HNO_3, HF, and aqua regia. The microwave power levels and times are designed to reflux or evolve the acids. The samples were then removed from the Prolabo vessel and heated sequentially with $Hclo_4$ and HNO_3 plus H_2O_2.

After solubilization of the soil samples, three processes were tried: (1) separation using barium sulfate coprecipitation, (2) separation using diphonix resin, and (3) separation using calcium fluoride coprecipitation. The tests conducted to evaluate these processes were only preliminary.

1. Separation using barium sulfate coprecipitation.

The precipitation of barium sulfate in a solution containing actinide ions has been shown to carry the M^{+3} and the M^{+4} ions. Uranium can also be coprecipitated with barium sulfate if it is reduced to the U^{+4} oxidation state.[65] The barium sulfate coprecipitant has been shown to be dissolved in alkaline EDTA and the actinides coprecipitated with the cerium hydroxide or dissolved in perchloric acid and extracted into suitable organic solvents. The strips can then be wet ashed and the actinides redissolved.[65,66] Also, when the barium sulfate is muffled with NaCl and carbon, it can be converted to barium chloride. Three tests were performed using the barium sulfate coprecipitation procedure: (a) separation from deionized water matrix, (b) samples with a reduction or oxidation reagent added, and (c) samples with Al, Ca, or Fe ions added. In each test the barium sulfate coprecipitant was filtered from the solution. An attempt was made to redissolve

the barium sulfate by the NaCl muffle method, however this was not very successful. The tests were evaluated by what actinide ions remained in the filtered solution.

2. Separation using Diphonix Resin.

Diphonix is a polyfunctional resin containing sulfonic and geminally substituted diphosphonic acid groups.[48,67] It has been described in chapter III, section B. The diphonix resin has the ability to adsorb most metal cations from acidic solution. This characteristic may make it a good scavenger for the actinide ions. The following tests looked at two areas. The first was to check the ability of the diphonix resin to adsorb various actinide ions from different matrices. The samples were placed into contact with the diphonix resin. The resin was filtered from the solution and the filtered solutions were then NdF_3 coprecipitated and alpha counted. The second area was the possibility of putting the adsorbed ions back into solution by muffling the diphonix resin. Two muffling procedures were tested: NaCl (m.p. 801°C) in a ceramic crucible and K_2CO_3 (m.p. 891°C) in a alumina crucible. Stripping methods for the diphonix resin have been published.[48,67] The tests conducted were: (a) loading Am-243, Np-337, Pu-236, U-238, Th-232 in 1.0-M HNO_3 and eluting with a large volume of 5.0-M HCl, (b) using different reduction or oxidation reagents, (c) comparing two muffle methods, (d) loading a small amount of Am-241 and Am-243 from a large volume of solution, and (e) separating actinide ions from soil matrices.

The diphonix resin has been characterized by previous researcher by measuring D(ml/g) values as a function of HNO_3 or HCl concentration for tri-, tetra-, and hexavalent actinides and for selected non-actinide ions. [48,67] The constant D(ml/g) is defined to be the dry weight distribution ratio. It is calculated as $D(ml/g) = [A_o-A_f/W]/[A_f/V]$ where A_o and A_f are the aqueous phase activities(cpm) before and after equilibration, respectively, W is the weight of the dry resin (g), and V is the volume of the aqueous phase (ml). Some log D values are:<u>1.0-M HNO_3</u>: Am^{+3} 3.5, Pu^{+4} 4.8, Np^{+4} 5.0, UO_2^{+2} 5.3, Th^{+4} 6.0, Fe^{+3} 5.5, Fe^{+2} --, Al^{+3} 2.6, Eu^{+3} 3.8, Bi^{+3} 3.7, Ca^{+4} 1.3, Zn^{+2} 1.0, Sb^{+3} 1.3, Ag^{+} 2.0; <u>5.0-M HCl</u>: Am^{+3} 2.5, Pu^{+4} 4.9, Np^{+4} 4.3, UO_2^{+2} 3.5, Th^{+4} 5.0, Fe^{+3} 3.3, Fe^{+2} 2.0, Al^{+3} 2.0, Eu^{+3} 2.7, Bi^{+3} --; <u>1.0-M HCl</u>: Am^{+3} 3.6, Pu^{+4} 5.0,

Np^{+4} 4.9, UO_2^{+2} 5.0, Th^{+4} 6.0, Fe^{+3} 5.0, Fe^{+2} 4.3, Al^{+3} 2.7, Eu^{+3}4.0, Bi^{+3} 1.0; 0.5-M HCl: Am^{+3} 4.5, Pu^{+4} 5.0, Np^{+4} 5.0, UO_2^{+2} 5.3, Th^{+4} 6.0, Fe^{+3} 5.3, Fe^{+2} 4.8, Al^{+3} 3.3, Eu^{+3} 4.5, Bi^{+3} 2.0; and 0.1-M HCl: Am^{+3} 6.3, Pu^{+4} 4.0, Np_{+4} 4.0, UO_2^{+2} 5.8, Th^{+4} 6.5, Fe^{+3} 5.3, Fe^{+2} 5.3, Al^{+3} 4.3, Eu^{+3} --, Bi^{+3} 5.0. [48,67]

3. Separation using Calcium Fluoride coprecipitation.

Coprecipitation of the actinide ions on insoluble neodymium fluoride is used for preparation of actinide samples for alpha counting. The M^{+3} and M^{+4} ions of the actinide and lanthanide ions form highly insoluble fluoride compounds. The plan, in this section, was to try to coprecipitate the actinide fluorides with some insoluble fluoride compound. Important considerations in coprecipitation processes are insolubility and crystal lattice fits. Calcium was chosen as the carrier ion because it forms an insoluble fluoride compound and it causes relatively little interference in the TRU-Resin™ separation process. Also taken into consideration was the elimination of some potential problematic matrix ions. The Fe^{+3} and Fe^{+2} ions do not form insoluble fluorides.[66] The Al^{+3} ion will form a complex ion, AlF_4^-, and therefore potentially not coprecipitate with the CaF_2. Some -log K_{sp} values are: BaF_2 5.8, CaF_2 10.4, CeF_3 15.1, LiF 2.8, MgF_2 8.2, SrF_2 8.5, ThF_4 28.3, and UF_4 21.2. Some complexation constants (log beta values) are: Ca^{+2} 1.1; Ln^{+3} 3.6; Th^{+4} 8.4, 15.1, 19.8, 23.2; UO_2^{+2} 4.3, 8.0, 10.6, 12.0; Np^{+4} 8.3, 14.5, 20.3, 25.1; Fe^{+3} 6.0, 9.1, 11.9; and Al^{+3} 7.0, 12.6, 16.7, 19.1, 19.4, 19.8.[25]

Two tests were conducted using a calcium fluoride coprecipitation process: (a) deionized water samples and (b) deionized water samples with Nd^{+3} and Ti^{+3} ions added. The CaF_2 was redissolved by wet ashing in HCl solution.

CHAPTER V

EXPERIMENTAL RESULTS

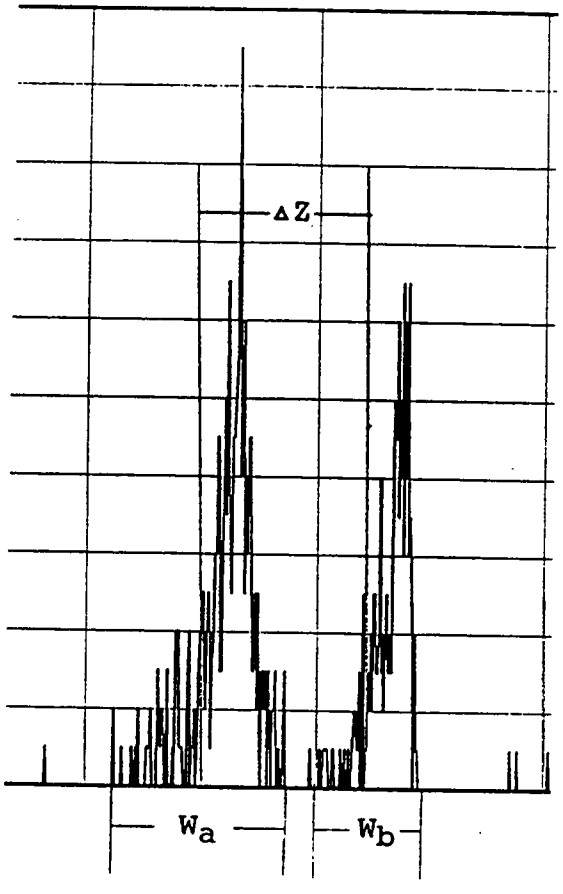
A. Counting of Samples

1. Resolution of different energy alpha particles.

Each alpha energy of a nuclide produces a spectral band located at a specific energy. Most alpha decaying nuclides emit alpha particles at more than one energy, and these energies are usually close. Therefore, the bands that are observed from these nuclides in a NdF_3 sample matrix measured on a surface barrier alpha detector are usually compound bands. These compound bands have various shapes due to the differing energies and the differing percentages of the several overlapping constituent bands. The shape of the compound band usually has a sharply-rising high energy trace, then a peak, and finally a broader tailing trace on the low-energy side. The low energy tail is due to self-adsorption and self-scatter in the NdF_3 matrix. For the purpose of estimating resolutions, the resolution formula, $R_S = 2\Delta Z/(W_a+W_b)$, has been used. ΔZ is the distance between the midpoints of band A and band B, W_a is the width of band A at the background baseline, and W_b is the width of band B at the background baseline. An example of a resolution determination is shown in Figure 11.

The resolution results from the split filter measurement experiment are listed in Appendix B, Table XIII. If the $R_S \geq 1.2$, the peaks were resolved to baseline sufficiently for an activity ofeach isotope to be determined with minimal error. The nuclides that could be resolved spectroscopically using SSB detectors are listed in Table I.

If yield tracers are used, then the overlapping of the yield tracer nuclides with the environmentally occurring nuclides would need to be considered. Possible nuclides that may be used for yield tracers are: U-232, Th-229, Pu-236, Am-243, Np-239. If these are added to the



Np-237 band, W_a = 29 mm

Pu-239 band, W_b = 16 mm

ΔZ = 27 mm

R_s = 2(27 mm)/(29 mm + 16 mm)

= 1.2

Figure 11: Resolution Estimate of Np-237 and Pu-239 Spectra.

TABLE I

NUCLIDES THAT COULD BE RESOLVED SPECTROSCOPICALLY
USING SSB DETECTORS.

	Th-232	Nat. U	Np-237	Pu-239	Am-241
Th-232	---	No	No	Yes	No
Nat. U	No	---	No	Yes	Yes
Np-237	No	No	---	Yes	Yes
Pu-239	Yes	Yes	Yes	---	Yes
Am-241	No	Yes	Yes	Yes	---

(Th-232 represents Th-232, Th-230, Th-228. Nat. U represents U-238, U-235, U-234.)

previous nuclide samples, the only nuclide combinations that can be resolved are: Pu-236/ Pu-239/Np-237/Np-239 and Am-241/Am-243/Np-237/Np-239. Therefore, if a separation procedure could be developed which could be used without a tracer, probably fewer separation steps would be required.

A second evaluation of detector resolution utilized a mixed sample of Pu-242 and Np-237. A projected line spectrum of the emitted alpha particle energies of these two nuclides is shown in Figure 12a. There is a 68 keV gap between the last significant high energy alpha particle of Np-237 (4788 keV) and the first significant low energy alpha particle of Pu-242 (4856 keV). The counting samples were prepared from spiked water samples by coprecipitation using the NdF_3 procedure. The first set was counted using the SSB detectors and the second set was counted using the PIPS/lower shelf detector/arrangement. Using a peak to valley ratio, the PIPS/lower shelf band shows a slight improvement in resolution over the SSB detector results (Examples shown in Fig.12b, 12c). However, neither showed sufficient resolution for quantitative determination based on the above criteria of resolution to baseline for minimal error.

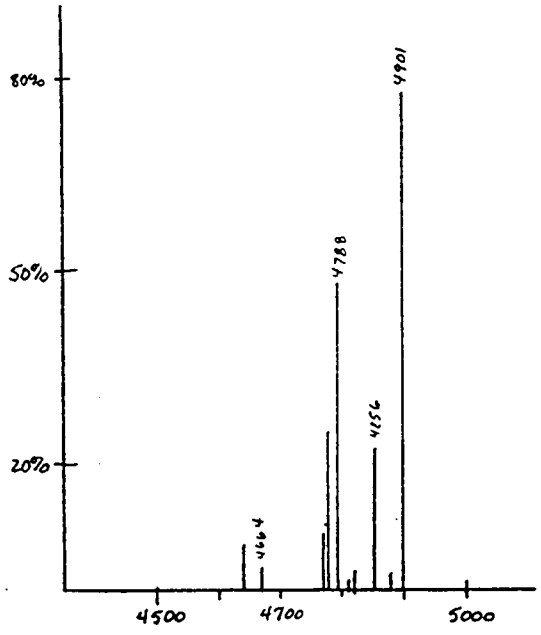
A third evaluation of detector resolution utilized a sample containing Th-230, Th-229, Th-227. The Th-227 isotope has the possibility of being resolved into two energy bands, 5668-5757 keV and 5866-6038 keV. The lower energy band often overlaps with decay daughters of other thorium isotopes but the higher-energy alpha band of Th-227, if resolvable, can be used to quantitate the amount of Th-227 present in the sample. Another separation concerned with resolution is between the Th-230 (4621-4687 keV) and the Th-229 (4814-5050 keV) energy bands. Figure 13 shows a typical spectrum obtained using PIPS/lower shelf detector/arrangement and the resolution values (R_S) calculated as previously discussed.

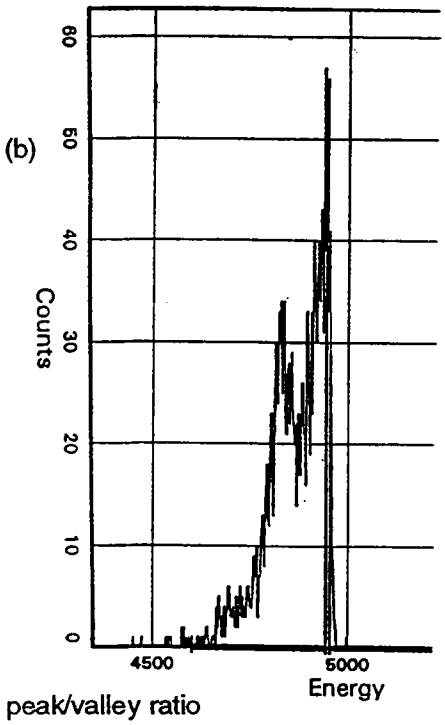
2. Precision check of counters.

a. A NdF_3 precipitated Am-241 sample was counted seven times, 200 minutes each time, under the same detector. The data are listed in Appendix B, Table XIV. The reliability factor (RF) value of 0.86 falls between 0.80 and 1.20. This indicates that the main source of error

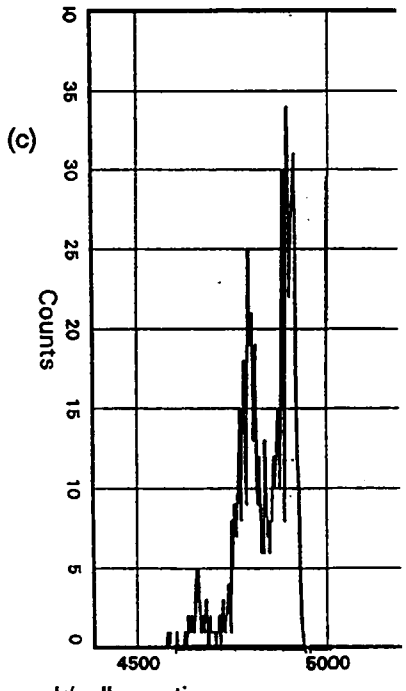
(a)

Np-237	4640 keV (6.2%)
Np-237	4664 keV (3.3%)
Np-237	4766 keV (8.0%)
Np-237	4771 keV (25%)
Np-237	4788 keV (47%)
Np-237	4803 keV (1.5%)
Np-237	4817 keV (2.5%)
Pu-242	4856 keV (22%)
Np-237	4873 keV (2.6%)
Pu-242	4901 keV (78%)





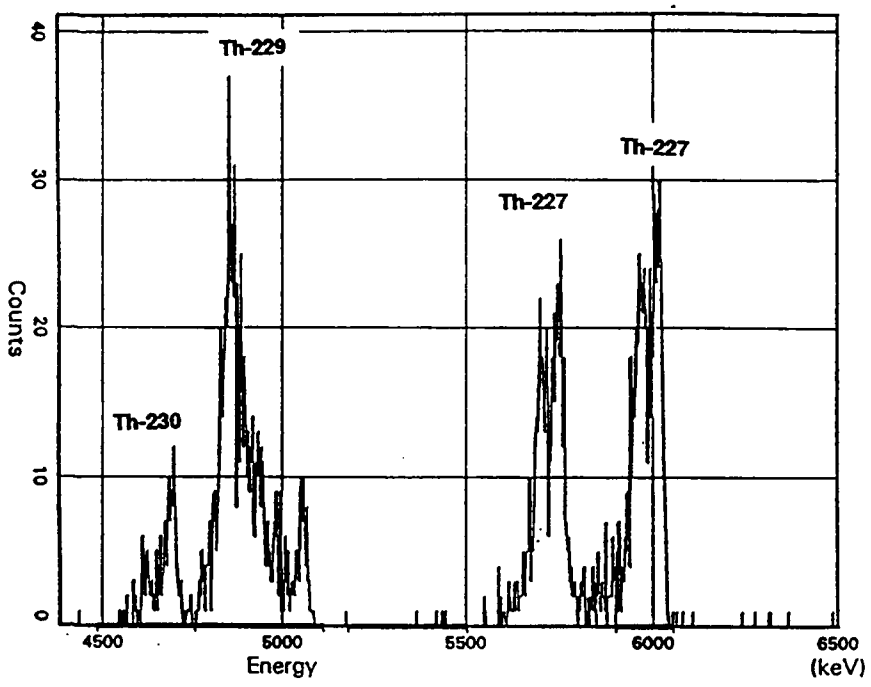
peak/valley ratio
Np-237 = 2.5
Pu-242 = 4.1



peak/valley ratio
Np-237 = 4.2
Pu-242 = 5.6

Figure 12: Alpha Particle Energy Resolution of Np-237 and Pu-242 (a) Energies of emitted alpha particle of Np-237 and Pu-242, (b) Resolution shown by SSB detectors, (c) Resolution shown by PIPS detectors/lower shelf arrangement.

	keV	%
Th-230	4621	23%
Th-230	4687	76%
Th-229	4814	9%
Th-229	4837	5%
Th-229	4845	56%
Th-229	4901	10%
Th-229	4968	6%
Th-229	4978	3%
Th-229	5050	5%
Th-227	5668	2%
Th-227	5700	4%
Th-227	5709	8%
Th-227	5713	5%
Th-227	5757	20%
Th-227	5866	2%
Th-227	5959	3%
Th-227	5978	23%
Th-227	6009	3%
Th-227	6038	24%



Average R_S values: Th-230/Th-229 = 1.3

Th-227(low E band)/Th-227(high E band) = 1.5

Figure 13: Typical SpectraResolution Using PIPS/lower-shelf Detectors.

under these conditions is counting error due to the random character of radioactive decay processes.[49]

b. A NdF_3 coprecipitated Am-241 sample was counted using 8 different surface barrier detectors. Each count time was 200 minutes. The data, in Appendix B, Table XV, indicate that the there is a high probability that the counts show a greater deviation than would be expected from chance alone. In other words, something more than variability due to radioactive decay seems to be influencing the error. Possible sources of this error could be efficiency determination of the detectors, handling of the sample between detectors, variation in the centering of sample under detector, variation in sample-to-detector distance, and variation in detector environment (e.g. vacuum, background, backscatter).

3. Comparison of computer-determined counts to actual counts.

For alpha counting of Am samples, the computer was programmed to quantify predetermined regions-of-interest for Am-241 [alpha energies 5388(1.4%) 5443(12.8%), 5486(85.2%) keV] and Am-243 [alpha energies (5185(1%), 5233(10.6%), 5275(87.9%) keV]. These regions-of-interest were located beside each other and the possibility existed for degraded Am-241 alpha counts to be included in the Am-243 counting region. Data were complied to determine the amount of degraded Am-241 counts that were observed in the Am-243 region by counting NdF_3 precipitated Am-241 samples with various activity levels and on different counters. (See Appendix B, Table XVI.) The average percentage overlap of the Am-241 into the Am-243 region was 4.0% using SSB detectors and 2.9% using PIPS detectors. This error is less than counting error (about 5.5% or greater) and thus would have only a small effect on the overall error. In Appendix A, Figure 23 two examples of Am spectra are shown. The first spectrum shows some degraded Am-241 alpha counts recorded in the Am-243 region of interest using an SSB detector and the second spectrum shows the Am-241 and Am-243 peaks resolved using a PIPS/lower shelf arrangement.

B. Preparation of Samples for Counting

1. Amount of $Nd(NO_3)_3$ added.

Three sets of data, RB101492, RB112592A1, and RB112592A2, were evaluated for the quantity of $Nd(NO_3)_3$ coprecipitating reagent that would allow for optimum recovery of the actinide and good resolution of the spectral bands. (See Appendix B, Tables XVII, XVIII, and XIX.) The NdF_3 coprecipitation procedure (ITAS-OR-RS-7124), described in chapter IV section C, was followed for each of the experiments. The mg amount of $Nd(NO_3)_3$ was varied from sample to sample. Some of the samples contained HCl solution as the initial matrix and the rest had a Na_2SO_4/H_2SO_4/HCl solution matrix. (The Na_2SO_4/H_2SO_4 is used in the final clean up step after actinide separation and is usually still present when the separated sample is coprecipitated.)

Figure 14 shows a plot of the percent recovery of the actinide verses the amount of $Nd(NO_3)_3$ used in the coprecipitation procedure. A high percent recovery is present from 0.030 mg to 0.30 mg $Nd(NO_3)_3$ for both Am^{+3} and Th^{+4} ions. However, as the amount of $Nd(NO_3)_3$ utilized increased, the observed spectral resolution began to deteriorate even though percent recovery remained high. A poorly resolved spectral band is usually broad with a smeared long low-energy tail that might be unresolvable in a mixed isotope spectrum. This deterioration in the high mass ranged samples is largely due to self-adsorption and self-scatter within the precipitated sample. Poor spectra were also observed in the low mass region (less than 0.030 mg). This may partially be due to uneven layering or clumpingof the very small amount of coprecipitate on the polypropylene membrane during filtration. Therefore, the overall range for optimum results, considering percent recovery and resolution of these data points, appears to be 0.030 mg to 0.10 mg $Nd(NO_3)_3$ added. One additional note is that the plot in figure 14 indicates that slight variations in the mg amount of $Nd(NO_3)_3$ within the acceptable range do not significantly effect the amount of actinide recovered.

Variation of the amount of Nd in NdF3 Coprecipitation Procedure

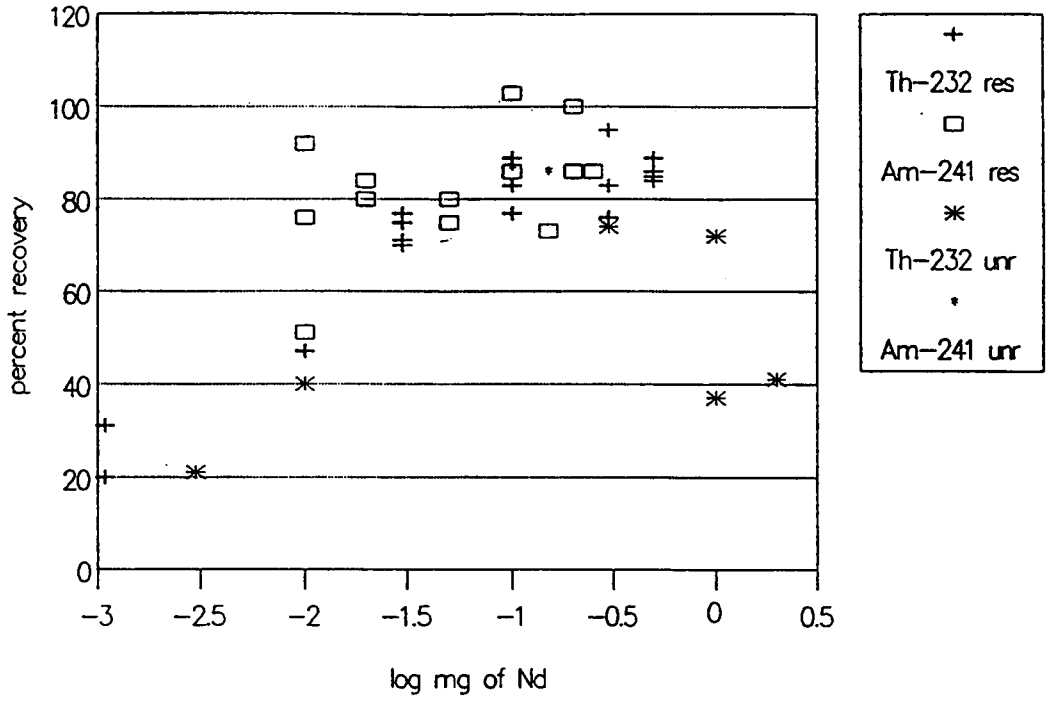


Figure 14: Variation of the Amount of $Nd(NO_3)_3$ Used as Coprecipitant in the NdF_3 Coprecipitation Procedure.

res = good resolution of spectral bands
unr = poor resolution of spectral bands

2. The use of temperature controls to enhance coprecipitation.

In the IT procedure, the samples are heated in a hot water bath (called a pre-water bath) prior to the addition of the Nd^{+3} and HF reagents. After addition of these coprecipitating reagents, the sample is mixed and placed into an ice bath (called a post-water bath). The following experiments were to evaluate the effects of the temperature changes relative to percent recovery of the actinide ions using the NdF_3 coprecipitation procedure.

Two experiments were conducted using Am^{+3}, RB102192 and RB112092. (See Appendix B, Tables XX and XXI.) No significant difference in percent recovery of Am^{+3} was observed between using room temperature (22°C) for pre- and post-water baths compared to using 85°C/0°C or 85°C/22°C temperature pre- and post-water baths.

The percent recovery of Th^{+4} was investigated in experiments DL061493, DL062593, DL072393, RB012894, and RB012293. (See Appendix B, Tables XXII, XXIII, XXIV, XXV, and XXVI.) Experiments RB012293 and RB012894 showed no statistically significant differences with variation in temperature. DL061493 showed statistically significant improvement in the recovery of Th^{+4} at a pre- and post-water bath temperature of 90°C in comparison to 22°C and 0°C. Results of experiment DL062593 indicate an average recovery of 105±8% for Th^{+4} when using pre- and post-90°C-water baths. The final experiment DL072393 compared the 90°C pre and post procedure with the original IT procedure using 90°C pre- and 0°C post-water baths. Results from this experiment showed statistically significant improvement in the percent recovery of Th^{+4} when the 90°C water bath is used both pre and post.

Experiments RB012293 and RB012694 were investigations of the percent recovery of both Pu and Np ions when the pre- and post-water bath temperature were varied. (See Appendix B, Tables XXVI and XXVII.) Percent recovery of the Pu ions was not significantly effected by changes in water bath temperatures. Percent recovery of Np ions was decreased greatly in Experiment RB012293 when the sample was placed in the high temperature water bath while good percent recovery was observed if only the 22°C or 0°C baths were used. This difference could be

attributed to a small concentration of NO_3^- ions in the solution which possibly oxidized the Np^{+4} to NpO_2^+ at the higher temperature. The NpO_2^+ does not coprecipitate well with NdF_3. In experiment RB012694, the nitrate from the standard solution was removed by a sulfate fume process prior to the NdF_3 coprecipitation procedure. In the later experiment, percent recovery of Np showed no statistically significant effects when the temperatures of the water baths were varied.

Experiment DL061693 (Appendix B, Table XXVIII) was an investigation of the percent recovery of U ions when the pre- and post-water bath temperatures were varied. In the coprecipitation of U ions, the UO_2^{+2} ions are reduced to U^{+4} by the addition of $TiCl_3$. The U^{+4} ions will coprecipitate with NdF_3 while the UO_2^{+2} ions do not. The results of this experiment showed no significant difference in the percent recovery of U ions at different pre- and post-temperature.

3. The length of time allowed for precipitate formation.

The impact of various lengths of precipitating time on the percentage recovery of Am^{+3} by the NdF_3 procedure was evaluated. Experiment RB112092 (Appendix B, Table XXI) indicated that a 30 min precipitation time at room temperature improved percent recovery of Am when compared with 15 minutes at room temperature. In the next experiment, RB010793C, the times varied from 15 minutes to 6.7 hours. (See Appendix B, Table XXIX.) Percentage recoveries ranged from 80±7% at 15 minutes to 94±7% at 4.4 hours. However, at an NTE deviation of ±7% this is not a significant difference. A final study, RB011593 (Appendix, Table XXX), was conducted using Am, Np, Th, and Pu. (It also included temperature variance which has been previously discussed.) The Am, Np, and Th data showed no statistically significant preference for any specific length of precipitation time. The Pu data (only three data points) indicated a possible preference for the shorter time frame at room temperature.

4. Continuous agitation during coprecipitate formation.

This investigation was conducted in order to determine if continual mixing of the solution would affect favorably the percent recovery of the desired actinide by the NdF_3 precipitation

procedure. Am-241 and U-238 were used in this experiment. A small amount of a standard solution was measured and diluted to 15 ml final volume with 3.0-M HCl. All steps were conducted at room temperature. All the samples were treated the same through the addition of the $NdCl_3$ and HF reagents. Then half of the samples were placed in a stationary rack and the other half were placed on a Burrell wrist-action shaker and continuously agitated for 15 minutes. All samples were then filtered and counted. The results of experiment DL062393 (Appendix B, Table XXXI) indicate no statistically significant effect on the percent of actinide recovery. The agitated samples did show slightly less scatter of data points then the non-agitated samples.

5. Filtration of actinide ions through a membrane whose surface had been previously coated with NdF_3 precipitate.

The first two samples were precipitated by the NdF_3 procedure at room temperature. The second two samples were treated identically to the first two except that an additional coating of NdF_3 was placed on the filter prior to sample filtration and no HF reagent was added to the sample. For the second two samples, the additional coating consisted of a mixture of 5.0 mg $Nd(NO_3)_3$ and 5.0 ml 48% HF. The results of experiment RB033193 (Appendix B, Table XXXII) show a comparison of the number of alpha counts recorded in the preset energy range for Am-241 compared to the alpha counts recorded in the preset energy range for Am-243. Since no Am-243 was added to the samples, the counts in the preset energy range for Am-243 are due to alpha particles being detected in a lower energy range than expected and this probably relates to degradation in the sample. This degradation is typical of self-absorption and self-scatter. The samples with the additional NdF_3 coating on the membrane showed a greater percentage of low energy degraded alpha particles than the samples without.

6. Actinide ions remaining in solution after NdF_3 coprecipitation.

Am-241 ions were separated from 3.0-M HCl solutions using the NdF_3 coprecipitation procedure. The solutions, remaining after filtration of the coprecipitant, were collected, evaporated, fumed with Na_2SO_4/H_2SO_4 solution, and rediluted with 3.0-M HCl to a total volume of 15 ml. The

treated filtered solutions were again coprecipitated using the NdF_3 procedure with one change. The amount of $Nd(NO_3)_3$ reagent added was doubled (1.0 ml, which gives 0.10 mg). The resulting alpha counts from the samples and the reprecipitated samples are shown in experiment RB022793 (Appendix B, Table XXXIII). The results showed essentially no additional recovery of Am by doing a second coprecipitation procedure.

7. Na_2SO_4/H_2SO_4 in matrix of precipitating solution.

In the first step of the NdF_3 coprecipitation procedure, 2.0 ml of 0.73-M Na_2SO_4 in 18-M H_2SO_4 solution was routinely added to the separated fraction. The fraction was than heated to near dryness. The proper state of near dryness was indicated by copious white SO_3 fumes being emitted by the fraction for at least 15 minutes [ITAS-OR-RS-7124]. The fraction was than cooled and diluted to 15 ml total volume with 3.0-M HCl. The following experiments were conducted to determine if the presence of the sulfate solution in the precipitating matrix had any effect on percent recovery. The only variation in the NdF_3 procedure was in the sulfate step. All other steps were performed the same. The data from experiments RB102392, RB120292, and RB010793B (Appendix B, Tables XXXIV, XXXV, and XXXVI) showed no significant difference in any of the variations. This indicates that the presence of the sulfate in the NdF_3 coprecipitating matrix has no statistically significant effect on the percent recovery of Am.

8. Reagent blank

Periodically, blanks were set up and counted along with the test samples. These blanks were treated the same as the rest of the samples in the experiment except no actinide was added. Thus, if the alpha spectra of the blanks contained any significant counts above background, it would be due to contamination in the reagents, glassware, filtration apparatuses, or the detectors. Table XXXVII in Appendix B lists results from the various blanks.

Experiment RB021293 tested various glass beakers for contamination. The glass beakers were taken from several sources: new unused beakers, cleaned beakers previously used for picocurie-level samples, cleaned beakers previously used for microcurie-level samples, cleaned

beakers with visible water spots, and cleaned beakers with visible scratches. The sides of the beakers were rinsed with concentrated HNO_3 and the sample was heated on a hot plate to partial dryness. This step was repeated 2 more times. Next 2.0 ml of Na_2SO_4/H_2SO_4 reagent was added and heated to partial dryness. The samples were then diluted to 15 ml with 3.0-N HCl and coprecipitated at room temperature using the NdF_3 procedure. Count results showed less than 0.02 pCi detected per sample.[54]

C. Separation of Actinide Ions into Spectroscopically Distinct Groups.

1. Coprecipitation using zirconium phenylarsonate.

Solutions containing 15-20 dpm each of Th^{+4} and Am^{+3} in HCl were used for the zirconium phenylarsonate coprecipitation experiments. The phenylarsonate ion was added in various amounts but always in excess of the metal cations. The amount of $ZrO(NO_3)_2$ was also varied. In the first test (RB051493, Appendix B, Table XXXVIII), Nd^{+3} was added as a possible hold back carrier for the Am^{+3} and the zirconium phenylarsonate coprecipitated samples were filtered and counted using PIPS detectors. In the second experiment (DL072093, Appendix B, Table XXXIX), the filtered coprecipitate was counted as well as any actinides remaining in the filtered solution. The NdF_3 precipitation of the filtered solution was to determine the percentage which was not precipitated with the zirconium phenylarsonate. In both tests, no coprecipitation of either the Am^{+3} and Th^{+4} ions with the zirconium phenylarsonate was detected. The ions were found in the filtered solution. The use of zirconium phenylarsonate coprecipitation of low levels of Th^{+4} in the presence of Am^{+3} as a separation method and a counting sample preparation method was not successful.

2. Coprecipitation using m-nitrobenzoic acid.

Six solutions, each containing 25 dpm of both Th^{+4} and Am^{+3} in 15 ml of 3.0-M HCl, were used for the m-nitrobenzoic acid coprecipitation experiment. To each of the solutions were added 0.5 ml of 0.1 mg/ml $Ce(SO_4)_2$ solution, 0.3 ml of 30% H_2O_2, and 5.0 ml of 0.0125 g/ml

m-nitrobenzoic acid, $O_2N\text{-}C_6H_4\text{-}CO_2H$, in 50% ethanol solution. The mixtures were allowed to stand for 15 minutes prior to filtration. The filtration discs containing the m-nitrobenzoate coprecipitant were dried and counted using surface barrier detectors. No counts were observed. Performance of test was assisted by D. Larkins.

3. Coprecipitation using the iodate anion.

Solutions containing approx. 20 dpm each of Am-241 and Am-243 in 15 ml of 3.0-M HNO_3 were used for the iodate coprecipitation experiment. To the americium solutions were added 0.50 ml of 0.10 mg/ml $Nd(NO_3)_3$ and then 5.0 ml of a saturated KIO_3 solution (15.0 g in 100 ml 1.0-N HNO_3). After standing for 15-20 minutes, the solution was then filtered using the standard NdF_3 procedure and the dried filters were sent to be counted. The results, listed in Appendix B, Table XL, indicate that essentially no Am was detected by this method. Performance of the test was assisted by D. Larkins.

4. Coprecipitation using Y-Ca-Mg mixed-fluorides.

Two separate tests, RB112592 and RB122392, were conducted to investigate the ability of a Y-Ca-Mg mixed-fluoride coprecipitation method to separate Am from U ions. The first test used Am activity and U activity in a 1:1 ratio. The second test had a 1:3 ratio of Am:U activity. In both tests, a comparison of the percent recovery of Am ions of three different cation carrier combinations was conducted. The three different cation carrier combinations were (a) Y^{+3}-Ca^{+2}-Mg^{+2} ions, (b) Ca^{+2}-Mg^{+2} ions and (c) Nd^{+3} ions. The steps used for the coprecipitation followed the previously described NdF_3 coprecipitation procedure except with the substitution of the appropriate cation solution. See Appendix B, Tables XLI and XLII for results. All three carriers did effectively coprecipitate Am in the presence of U ions. However, the Nd^{+3} was equal or better as a cation carrier for Am in comparison to the other two combinations.

5. Coprecipitation using NdF_3.

a. Separation of Am^{+3} from UO_2^{+2} in a solution of equal activities.

Three test samples contained Am-241 and U-238 at equal activity levels (approx. 25 dpm.

each) in 15 ml of 3.0-M HCl. Two similar test samples contained only the Am-241 nuclide. All five samples were coprecipitated at room temperature using the previously described NdF_3 procedure (0.50 ml of 0.10 mg/ml $Nd(NO_3)_3$ and 5.0 ml 48% HF). The Am percent recovery was good, range 80-94%. No U was detected on the NdF_3 coprecipitated sample. Good separation of the Am and the U was obtained in this test. See RB010793C in Appendix B, Table XLIII for results. (Also included in this test was variation of the length of time that the sample was allowed to stand after the reagents were added prior to the filtration step. The results of the time variation have previously been discussed in section B.3 of chapter V.)

b. Separation of Am^{+3} from UO_2^{+2} in a solution where the U activity is higher than the Am activity.

The first test sample contained 20 dpm Am-241 and 193 dpm U-238, 1:10 ratio. The second test sample contained 20 dpm Am-241 and 2011 dpm U-238, 1:100 ratio. The samples were coprecipitated at room temperature with a 20 minute stand time using the previously described NdF_3 procedure. The filtrate from each of the samples was saved and coprecipitated a second time. Results are listed in RB012293B, Appendix B, Table XLIV. Approximately 70% of the Am-241 was recovered by the first coprecipitation procedure and the remaining 30% was recovered in the second coprecipitation. Some uranium was coprecipitated in all the samples. The amount of U coprecipitated ranged from 6-8 dpm in both the 1:10 and the 1:100 ratio samples. (15% U recovered in 1:10 sample and 2% U recovered in 1:100 sample.)

c. The effect of $TiCl_3$ on the coprecipitation of Np and Pu.

Five test samples were prepared each containing approx. 20 dpm each of Np-237 and Pu-239. The first three samples were coprecipitated using the NdF_3 procedure as previously described (0.50 ml of 0.10 mg/ml $Nd(NO_3)_3$, 5.0 ml 48% HF). To the remaining samples, 0.30 ml of 20% $TiCl_3$ ($TiCl_6^{-3}$) was added prior to the HF reagent. The E^o value for TiO^{+2} to Ti^{+3} is 0.10 volts. Therefore $TiCl_3$ has the potential to reduce NpO_2^+ to Np^{+4} and Pu^{+4} to Pu^{+3}. The percent recovery of the Np without the $TiCl_3$ averaged 88±5% while the percent recovery of Np with the

$TiCl_3$ averaged 84±6%. The percent recovery of the Pu without the $TiCl_3$ averaged 85±5% while the percent recovery of Pu with the $TiCl_3$ averaged 83±6%. See RB021693 in Appendix B, Table XLV for complete results.

d. The effect of $TiCl_3$, H_2O_2, and $K_2S_2O_8$ reagents on the coprecipitation of Np, Pu, and Th.

Nine test samples were prepared each containing 21 dpm Th and approx. 5 dpm each of Np and Pu. The samples were diluted to 15 ml volume with 3.0-M HCl and placed in a warm water bath for 15 minutes. Then 0.50 ml of 0.10 mg/ml $Nd(NO_3)_3$ was added. Next, the following oxidizing/reducing agents were added to different samples: 0.4-0.6 ml 20% $TiCl_3$, 1.0 ml 30% H_2O_2, and 1.0 ml saturated $K_2S_2O_8$. After 5 minutes, 5.0 ml of 48% HF was added. The coprecipitated samples were then filtered and counted. The results of experiment RB080793 are listed in Appendix B, Table XLVI. The Th percent recovery had the following averages: $TiCl_3$ 83±6%, none 70±6%, H_2O_2 106±9%, $K_2S_2O_8$ 104±8%. The Pu percent recovery had the following averages: $TiCl_3$ 88±6%, none 87±7%, H_2O_2 105±8%, $K_2S_2O_8$ 104±8%. The Np percent recovery had the following averages: $TiCl_3$ 93±7%, none 78±7%, H_2O_2 106±9, $K_2S_2O_8$ 4±2%. In both the Th and Pu results, slightly higher percents recovery were observed when the H_2O_2 or the $K_2S_2O_8$ reagent was added to the coprecipitation solution. However for Np, the $TiCl_3$ and the H_2O_2 both showed high recoveries while the $K_2S_2O_8$ appeared to inhibit the coprecipitation of the Np ions.

6. Separation of low activity levels of Am ions from low to high activity levels of U ions using TRU-Resin™.

a. Separation of low activity levels of both Am and U in 1.0-M HNO_3 load solution.

This test was to evaluate a low activity level separation between U and Am ions using a 1.0-ml bed volume column of TRU-Resin™. Two samples were prepared. One contained only Am-241 ions and the other contained a mixture of Am-241 and U-238 ions (approx. 25 dpm each).

Each sample was diluted to 10ml with 1.0-M HNO_3. The Am only sample was loaded onto the TRU-Resin™ column first. The resin bed matrix was converted to HCl by washing with 9.0-M HCl and the Am was then eluted with 4.0-M HCl. After reconditioning the column to HNO_3, the second sample containing the mixture of Am and U ions was loaded. Again, the resin bed matrix was converted to HCl by washing with 9.0-M HCl and the Am was then eluted with 4.0-M HCl. The U was eluted with 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl. The Am percent recovery was approx. 72±8% in both columns. The U recovery was 67±8%. The Am and U fractions showed no overlap. See RB112192 in Appendix B, Table XLVII for results.

b. Separation of low activity levels of Am from U activity levels that were 10, 100, 1000 times higher than the Am level using a load solution of 3.0-M HNO_3.

Three test samples were prepared. The first was a 1:10 ratio sample containing 20 dpm Am to 200 dpm U. The second sample was a 1:100 ratio sample containing 20 dpm Am to 2000 dpm U. The third sample was a 1:1000 ratio sample containing 20 dpm Am to 20,000 dpm U. Each sample was wet ashed (evaporated to near-dryness) using HNO_3 and H_2O_2 and then diluted to 10 ml using 3.0-M HNO_3. The samples were loaded onto the same TRU-Resin™ column sequentially. After each sample was loaded, the column was converted to chloride form using 8.0-HCl solution. The Am was eluted with 4.0-M HCl and the U was eluted with 0.1-M $NH_4HC_2O_4$ in 0.1-M HCl. Then the column was reconditioned using 3.0-M HNO_3. Only the Am fractions were coprecipitated using the NdF_3 procedure and alpha counted. Results of the Am percent recovery were: 1:10 sample 82±7%, 1:100 sample 78±7%, and 1:1000 0%. In the third sample, U ions were present in the Am fraction. (See RB021793, Appendix B, Table XLVIII.)

The above test was repeated, using three samples with the same concentration ratios and the same separation procedure. This time more fractions were coprecipitated and counted. A recovery of 86±9% of the Am-241 without any U contamination resulted in the 1:10 ratio sample separation. Approximately 80% of the Am-241 was recovered in the 1:100 ratio sample but it was

spread throughout several fractions with the majority eluting in the U fraction. Almost no Am-241 was recovered in the 1:1000 ratio sample even though an extended Am wash step was used and an additional dilute acid step was added prior to the U fraction elution. (See RB031193, Appendix B, Table XLIX.)

In HNO_3, both the U and the Am ions appear to load onto the resin when the activity level of the U is low. However as the activity level of the U increases, the ability of the Am ions to load and/or elute appeared to be hampered.

c. Separation of low activity levels of Am from U activity levels that were 10, 100, and 1000 times higher than the Am level using a load solution of 3.0-M HCl.

When loading a solution mixture of Am and U ions onto the TRU-Resin™ column in HCl, the U was expected to attach to the resin, but the Am ions were not. This would result in separation of the two elements as long as the capacity of the resin was not exceeded by the U concentration. The test samples were made up in the same way as for section b above. The samples were loaded in 3.0-M HCl onto the same TRU-Resin™ column. After the 1:10 sample was loaded, the column was rinsed with 5.0 ml of 3.0-M HCl and then 5.0 ml of 1.0-M HCl. The load filtrate and rinse filtrate solutions were NdF_3 coprecipitated and counted. The U was eluted with 0.1-M $NH_4HC_2O_4$ in 0.1-M HCl. Then the column was reconditioned using 3.0-M HCl. The 1:100 and 1:1000 ratio samples were treated the same as the 1:10 sample. The load filtrates and the 3.0-M HCl rinse filtrates were NdF_3 coprecipitated and counted. In the 1:10 and the 1:100 ratio samples, the majority of the Am did not adhere to the column and was determined to be in the load filtrate. The filtrate from the 3.0-M rinse step contained an additional 6-10% of the Am ions. Neither fraction contained U ions. These Am ions were either unattached and in the column's liquid phase or very easily eluted from the column's resin. The two filtrates from the 1:1000 ratio sample were too radioactive to count using the surface barrier detectors without danger of contamination to the detectors. Therefore, only a gross alpha count was obtained for these on the

Tennelec LB4000, a gas-flow proportional counter. The measured activities of the 1:1000 sample's load filtrate and the rinse filtrate were much higher then the amount of Am ions present in the sample, thus indicating that U ions were present in these filtrates. (See RB040293, Appendix B, Table L.)

d. Separation of low activity levels of Am from a 1000 times higher U activity level in 3.0-M HCl by comparing static resin loading with column resin loading.

Two test samples were prepared. Each sample contained 15 dpm Am-241 and approximately 20,000 dpm U-238 in 3.0-M HCl. The first sample (A) was loaded onto a 1.0 ml bed volume TRU-Resin™™ column. The column was rinsed with 3.0-M HCl, and this rinse filtrate was added to the load filtrate. The combined filtrates from sample A were wet ashed and redissolved in 3.0-M HCl. The U ions were eluted from the column using 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl. Then, the column was reconditioned using 3.0-M HCl. The redissolved filtrate from sample A was loaded onto the column again. This procedure was repeated once more resulting in the fraction expected to contain the Am ions passing through the resin bed of the column a total of three times. The U ions were eluted from the column each time.

For the second sample (B), 0.34g of dry TRU-Resin™ was added to a beaker containing the combined U and Am ions solution. Total volume of the sample was 25 ml. The mixture was stirred periodically and at the end of two hours, the sample and resin were poured into an empty column. The beaker was rinsed with 3.0-M HCl which was also poured through the resin. The resulting combined filtrate of the load and rinse solutions was evaporated to approximately 10 ml. Then the resin was eluted with ammonium hydrogen oxalate to remove the U ions and then washed with 3.0-M HCl. The evaporated combined filtrate from sample B, expected to contain the Am ions, was poured through the resin a second time for additional U removal if necessary. The different fractions were then NdF_3 coprecipitated and counted.

The loading of the TRU-Resin™ appeared to be approximately the same whether

performed by the liquid being poured through a resin column or the resin being added to the solution and allowed to stand. The first elution of the U-238 from the column-loaded resin (A) resulted in a activity measurement of 8060±484 dpm and the first elution of the U-238 from the static-loaded resin (B) had a measured activity of 7070±424 dpm. From these results, the loading capacity appears to be in the range of 7000-8000 dpm U-238 ions from natural uranium for 1.0 ml of resin bed (0.34g of resin) of 100-150 m particle size. (20,500-23,500 dpm U-238(nat.U load)/g resin, or 0.010g U-238/ml resin bed) The second load and elution of U by both sample A and B yielded a count between 5000-7000 dpm U-238. A third U load and elution was performed with sample A only and it yielded 1391±140 dpm. The Am fraction from sample A (passed through the resin bed three times) had a percent recovery for Am-241 or 112±11%. The Am fraction from sample B (effectively passed through the resin twice) was still contaminated with U and unable to be counted. (See RB041393, Appendix B, Table LI.)

7. Separation of low activity levels of Am ions from low to high activity levels of Th and/or U ions.

   a. Separation of low activity levels of Am from Th activity levels that are 4 times higher than the Am activity level.

Two samples, each containing 15 dpm Am-241 and 60 dpm Th-232, were wet ashed. The first sample was dissolved in 4.0-M HCl and the second sample was dissolved in 5.0-M HCl. Both samples were then loaded onto separate TRU-Resin™ columns. The columns were sequentially eluted with dilute HCl and dilute ammonium hydrogen oxalate solutions. The columns were then washed with the appropriate HCl solution and the filtrates from the initial load solutions were reloaded onto the resins. Again the resins were sequentially eluted with dilute HCl and dilute ammonium hydrogen oxalate solutions. Each 5.0 ml of eluted solution was collected and saved as a separate fraction. All of the fractions were NdF_3 coprecipitated and alpha counted.

In both samples, approx. 82±7% of the Th activity was recovered in the first 10 ml of 1.0-M HCl elution solution. Less than 1% of Th activity was recovered in any of the other fractions

including the elutions after the second loading. The Am ions were recovered primarily in the eluate from the second load solution. (See RB043093 and RB050393 in Appendix B, Table LII and LIII.)

b. Separation of low activity levels of Am and Th from U activity levels that were 100 and 300 times higher than the Am level using a load solution of 3.0-M HCl.

Two samples were prepared. Both contained 15 dpm of each Am and Th. In addition, the first sample contained 2000 dpm U-238 and the second sample contained 6000 dpm U-238. The samples were wet ashed and dissolved in 10-15 ml of 3.0-M HCl. The samples were loaded onto separate TRU-Resin™ columns. After loading, the resin was rinsed with 3.0-M HCl, then 1.0-M HCl was added to attempt to elute the Th ions. Then $NH_4HC_2O_4$ was added to attempt to elute the U ions.

The results indicated U contamination in most fractions. Therefore the percents recovery of Th and Am in the appropriate fractions were unable to be determined. The U-238 ions that eluted in the first 5.0 ml of the $NH_4HC_2O_4$ eluting solution were approximately 2000 dpm in the first sample and 2500 dpm in the second sample (approximately 0.003 g U-238). (See RB042393, Appendix B, Table LIV.)

c. Separation of low activity levels of Am from Th and U activity levels that were 4-6 times higher than the Am level using a load solution of 5.0-M HCl.

The sample was a mixture of 15 dpm Am-241, 15 dpm Am-243, 60 dpm Th-232, and 97 dpm of U-238. The sample was wet ashed, dissolved in 5.0-M HCl, and loaded onto a TRU-Resin™ column. The eluate from the load and first rinse solution (10 ml 5.0-M HCl) was saved as the first fraction. The resin column was eluted with 1.0-M HCl solution, then by 0.1-M HCl solution, and then by 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl solution. The resin column was then rinsed with 5.0-M HCl solution and the saved first fraction was reloaded onto the resin. The same sequence of eluting solutions was used. The eluate was collected in 5.0 ml fraction. Each fraction

was NdF_3 coprecipitated and alpha counted. Results indicated that 90±9% of the Am activity was in the eluate of the second load, 88±8% of the Th was in the first 10 ml of the 1.0-M HCl solution, 34±5% of the U eluted in the 0.10-M HCl solution, the remaining 46±5% of the U eluted in the first 5 ml of the $NH_4HC_2O_4$ reagent. These fractions had very little or no overlap. (See RB050493, Appendix B, Table LV.)

8. Separation of Np and Pu ions together in the same elution fraction from Am, Th, and U ions.

a. Loading Np and Pu ions onto the TRU-Resin™ with and without H_2O_2 in the load solution.

In aqueous solution, the ions of Np and Pu may exist in more than one oxidation state. Hydrogen peroxide, H_2O_2, was added to some of the load solutions as one possible agent for controlling the redox environment. Comparison samples were loaded without H_2O_2 or any other specific reagent for redox control. The samples without H_2O_2 indicated mixed resin loading patterns. In one sample without H_2O_2, the load eluate contained 19±6% of the Np ions and 0% of the Pu ions. While in another sample without H_2O_2, the load eluate contained 76±7% of the Pu ions and 1±1% of the Np ions. (See RB061793A and RB061793B, Appendix B, Tables LVI and LVII.) In the other tests (Appendix B, Tables LVIII, LVIX, LX, LXI, LXII, LXIII, and LXIV), H_2O_2 was added to the load solution. The Np and Pu ions appeared to load consistently and relatively completely when H_2O_2 was in the load solution. The best loading results were obtained when the H_2O_2 was added just prior to loading the sample onto the resin and the sample was not heated after the H_2O_2 was added.

b. Comparison of HCl, NH_4I, $H_2C_2O_4$, and a combination of NH_4I and $H_2C_2O_4$ as possible eluting solutions for Np/Pu ions.

Several tests were conducted to compare the ability of these reagent as eluting solutions for Np/Pu ions from the TRU-Resin™ column. (1) In dilute HCl eluting solution, tests indicated that both Np and Pu ions will partially and slowly elute from the TRU-Resin™ column. In 10 ml of

1.0-M HCl, approximately 54% Np activity was recovered and approximately 10% of the Pu activity was recovered. (See RB062393A, RB062393B, and RB022294 in Appendix B, Tables LVII, LIX, and LX.) In an additional 10 ml of 1.0-M HCl elution solution, 20-30% more of both actinides eluted. Yet there still remained about 10% of the Np ions and 60% of the Pu ions on the resin. With 40 ml of 0.50-M HCl solution, approximately all of the Np ions eluted from the TRU-Resin™ column, however only approximately 40% of the Pu ions eluted with the same eluting solution. (See RB082093A, Appendix B, Table LXI.) When the HCl concentration was increased above 1.0-M HCl, both ions eluted at a slower rate (%recovery/volume eluted), such that with 20 ml of 1.5-M HCl eluting solution, approximately 10% of the Np ions and 5% of the Pu ions were detected in the eluate. (See RB022294, Appendix B, Table LX.) These results indicated that HCl alone may not be the best reagent for the elution of Np and Pu ions together in one fraction from the TRU-Resin™ column. (Also the ability of Np and Pu ions to elute in the 1.0-M HCl solution was seen as presenting potential overlap problems with the elution of the Th fraction using 1.0-M HCl. This is addressed in the next section (chapter V, section C.9).

(2) The ability of ammonium iodide, NH_4I, to elute either Np or Pu ions from the TRU-Resin™ was investigated in four separate tests. In each, 20 ml of 0.10-M NH_4I in either 1.0-M HCl or 0.5-M HCl was used to elute a mixture of Np and Pu ions which had been loaded onto a TRU-Resin™ column (and some partially eluted with 1.0-M HCl solution). In all four tests, approximately all the remaining Pu ions were eluted while less than 5% of the remaining Np ions were eluted. (See RB062393A, RB082093B, RB060993A, and RB060993B in Appendix B, Tables LXIII, LXI, LXII, and LXIII.)

(3) The ability of dilute oxalic acid, $H_2C_2O_4$, to elute Np and/or Pu ions from the TRU-Resin™ was investigated. The effect of a solution of 0.030-M $H_2C_2O_4$ in 1.0-M HCl solution was first observed when added to columns that had already been eluted with NH_4I solution. The $H_2C_2O_4$ solution eluted most of the Np ions that remained on the resin. (See RB062393A, RB060993A, and RB060993B in Appendix B, Tables LVIII, LXII, and LXIII.) Four other samples

were also processed. These samples contained a mixture of different actinides including Np and Pu ions. No NH_4I solution was used. The Np/Pu fractions were eluted with 15 ml of 0.030-M $H_2C_2O_4$ in 0.5-M HCl solution. The average percent recovery for Pu ions was 77±5% and for Np ions was 80±5%. (See RB110593E, RB110593F, RB110593G, and RB110593H in Appendix B, Table LXIV.)

(4) The ability of a combined reagent of ammonium iodide and oxalic acid to elute Np and Pu ions from the TRU-Resin™ was also investigated. In several tests, the elution solution used for the removal of Np/Pu ions in the same fraction was 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.0-M HCl or 0.5-M HCl. (See RB082093C,D, and RB110593A,B,C,D in Appendix B, Tables LXI and LXIV.) In test RB110593, the average percent recovery for Pu ions was 86±5% and for Np ions was 78±5%. When the concentration of HCl in the combined reagent was increased to 4.0-M, no Np eluted and Pu was not tested. (See RB061793B, Appendix B, Table LVII).

9. Refinement of the Th elution step.

a. Adjusting the molar concentration of the HCl eluting solution.

Initially, a 1.0-M HCl solution was used to elute Th ions from the TRU-Resin™ column. When Np and Pu ions were added to the sample mixtures, the Np and Pu ions began to eluted with the Th ions in the 1.0-M HCl solution. In 10 ml of 1.0-M HCl eluting solution, approximately 54% Np activity, 10% Pu activity, and 95% Th activity was recovered. (See RB062393A, RB062393B, RB022294, and RB080693A in Appendix B, Tables LVII, LIX, LX, and LXVI.) Therefore, additional investigation into the elution of Th was conducted. In RB071693, Th-232 ions were loaded onto a TRU-Resin™. (See Appendix B, Table LXV.) The molarity of the HCl eluting solution was decreased from 5.0-M to 4.0-M to 3.0-M to 2.0-M to 1.0-M to 0.10-M. The Th ions began to elute in the 2.0-M HCl solution and the remaining Th ions was completely eluted by the 1.0-M HCl solution. Next, six TRU-Resin™ columns were loaded with samples containing Np, Pu, and Th ions. For each column, a different molarity of HCl solution was used to elute the Th ions from the column. (See RB022294, Appendix B, Table LX.) The 1.30-M, 1.40-M and 1.50-M HCl

solutions resulted in the highest percent recovery of the Th ions with minimal overlap of Np and Pu ions. Percent recovery in 6.0 ml of 1.30-M HCl solution was: 92±10% Th, 1±1% Np, and 1±1% Pu. Percent recovery in 8.0 ml of 1.40-M HCl solution was: 92±10% Th, 2±1% Np, and 0±1% Pu. Percent recovery in 10.0 ml of 1.50-M HCl solution was: 89±9% Th, 1±1% Np, and 0±1% Pu. Some subsequent tests were conducted using 1.3-M HCl as the elution solution for Th ions. Results of 4 tests using 5.0 ml of 1.30-M HCl were: 89±9% Th, 0±1% Np, 1±1% Pu; 93±10% Th, 0±1% Np, 2±1% Pu; 94±10% Th, 1±1% Np, 2±1% Pu; and 86±9% Th, 1±1% Np, 2±1% Pu. (See RB082093A,B,C and RB081693A, Appendix 8, Tables LXI and LXIX.) Results of 4 tests using 6.0 ml of 1.30-M HCl were: 102±10% Th, 0±1% Np, 0±1% Pu; 81±9% Th, 0±1% Np, 0±1% Pu; 84±9% Th, 0±1% Np, 0±1% Pu; and 96±9% Th, 0±1% Np, 0±1% Pu. (See RB110593, Appendix B, Table LXIV.)

b. Adjusting the H^+ ion concentration in the Th eluting solution while keeping the Cl^- ion concentration constant.

A solution consisting of 1.0-M NaCl in 1.0-M HCl was used as an eluting solution for Th ions. This solution effectively provided a $[H^+]$=1.0-M and a $[Cl^-]$=2.0-M. The percent recovery results of duplicate tests, using 14.0 ml of eluting solution, were: 95±10% Th, 1±1% Np, 3±2% Pu; and 85±9% Th, 1±1% Np, 4±2% Pu. (See RB080693C and RB081693B,C in Appendix B, Tables LXVIII and LXX.) A second solution, consisting of 1.7-M NaCl in 0.3-M HCl, was used as an eluting solution for Th ions. This solution effectively provided a $[H^+]$=0.3-M and a $[Cl^-]$=2.0-M. The percent recovery result of this test, using 14.0 ml of eluting solution, was: 103±10% Th, 1±1% Np, 9±4% Pu.(See RB081693D, Appendix B, Table LXXI.)

c. Adjusting the Cl^- ion concentration in the Th eluting solution while keeping the H^+ ion concentration constant.

In this test, the solution consisted of 0.30-M HCl in 1.7-M $HClO_4$. This solution effectively provided a $[H^+]$ = 2.0-M and a $[Cl^-]$ = 0.3-M. The percent recovery result of this test, using

14.0 ml of eluting solution, was: 2±1% Th, 2±1% Np, 1±1% Pu. (See RB081693E, Appendix B, Table LXXII.)

d. Miscellaneous Observations

Because Th ions were included in several test that were designed to investigate other actinides, a couple additional observations about percent recovery of the Th ions were able to be noted. (1) A test sample, containing Np, Pu, and Th ions, was loaded onto a TRU-Resin™ column using 5.0-M HCl. The first eluting reagent added was 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl. After 20 ml of this solution, a large percentage of the Np and Pu ions had been eluted, but only 6±3% of the Th ions had been eluted. The next eluting solution to be added to this column was 0.030-M $H_2C_2O_4$ in 1.0-M HCl. In 10 ml of this solution, 88±9% of the Th ions was recovered. (See RB061793A, Appendix B, Table LVI.) (2) Since the Th ions were eluted with a solution of HCl only and since the NdF_3 coprecipitation procedure used the actinide ions dissolved in HCl, a few Th fractions were coprecipitated and alpha counted without first being evaporated and sulfate fumed. The average percent recovery of the Th ions in four fractions that were not evaporated and not sulfate fumed was 73±5%. The average percent recovery of the Th ions in four test fractions that were evaporated and sulfate fumed was 91±6%. (See RB110593, Appendix B, Table LXIV.)

10. Refinement of the U elution step.

a. Elution of pre-eluted TRU-Resin™ columns by $(NH_4)_2C_2O_4$ solution.

Two TRU-Resin™ columns were loaded with U-238 and washed with 20 ml of 0.10-M $NH_4HC_2O_4$ in 0.1-M HCl solution to elute the U ions. The percent recovery results of the U ions were: 63±7% and 60±7%. After two weeks of storage in 1.0-M HCl solution, the columns were eluted again. This time, it was with 20.0 ml of 0.10-M $(NH_4)_2C_2O_4$ solution. The repeat elution showed percent recovery values of 20±4% and 19±4%. (See RB062593, Appendix B, Table LXXIII.)

b. Elution of U following the use of NH_4I in the previous elution reagent (Np/Pu fraction).

In the first set of tests, one column was eluted with 1.3-M HCl, then 0.5-M HCl, and finally $(NH_4)_2C_2O_4$. There was some overlap of the actinides in the eluted fractions but 97±10% of the U ions were recovered. In the other three columns of this set, NH_4I was used as an eluting reagent prior to the $(NH_4)_2C_2O_4$ reagent. The U percent recovery results from these three columns were: 76±8, 62±7, and 71±8%. (See RB082093, Appendix B, Table LXI.)

In a second set of tests, 10 ml of 0.5-M HCl solution was used to rinse the TRU-Resin™ after the Np/Pu fractions were eluted. The combined reagent, 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.0-M HCl, was used to elute the Np/Pu fraction on two of the columns and a solution of 0.030-M $H_2C_2O_4$ (without NH_4I) was used as the Np/Pu eluting solution on the other two columns. The U percent recovery results after the combined reagent were: 78±13% and 73±12%. The U percent recovery results after the oxalic acid reagent (without NH_4I) were: 94±15% and 82±14%. (See RB110593, Appendix B, Table LXIV.)

c. Elution of U using HF.

A solution of 1.0-M HF in 0.10-M HCl was used to elute the U fraction from a set of four TRU-Resin™ columns. The U elutions followed Np/Pu elutions, half using the combined NH_4I and $H_2C_2O_4$ reagent and the other half using $H_2C_2O_4$ reagent. The U percent recovery results were: 56±9%, 56±9%, 61±10%, and 67±11%. (See RB110593, Appendix B, Table LXIV.)

11. Miscellaneous topics.

a. Using Ce^{+4} as a carrier for Th-229^{+4}.

Eight Th-229 samples were tested. For samples A and B, Ce ions were added as a carrier for Th-229 ions. For samples C and D, no carrier ions were added to the Th-229 ions solution. For samples E and F, Th-232 ions were added as a carrier for Th-229 ions. For samples G and H, Ce ions were added as a carrier for the Th-229 ions and additional actinide ions (Am-241, Np-237, Pu-239, and U-238) were added to the sample. The samples were dissolved and loaded onto

the columns with 0.020-M $K_2S_2O_8$ in 5.0-M HCl solution. The eluting solution for the Th fractions consisted of 1.30-M HCl containing 0.020-M $K_2S_2O_8$. For samples A-F, the columns were then eluted with 0.50-M HCl and 0.10-M HCl sequentially. For samples G and H, the columns were then eluted with 0.03-M $H_2C_2O_4$ in 0.50-M HCl to elute the Np/Pu fraction, then 0.50-M HCl, and then 0.10-M $(NH_4)_2C_2O_4$ to elute the U fraction.. The results showed almost no recovery of the Th-229 in samples A,B,C, and D. In samples E and F, results indicated equal recovery of both Th-229 and Th-232. (Sample E: 47±7% Th-229, 47±6% Th-232; Sample F: 104±14% Th-229, 99±11% Th-232) In samples G and H, the percents recovery of the Th-229 ions were 44±11% and 48±12%. Also, in samples E, F, G, and H, the Th ions did not elute with the 1.30-M HCl solution containing $K_2S_2O_4$. They eluted in the following 0.50-M HCl fraction that contained no $K_2S_2O_8$. In columns G and H, the 9 percent recovery of the other actinides (Am, Np, Pu, and U) was good and their elution patterns remained the same as previously observed. (See RB010494, Appendix B, Tables LXXIV and LXXV.)

b. The effect of various redox reagents on the elution pattern of the actinides from the TRU-Resin™ column.

(1) The first reducing agent tested was zinc metal. A wet-ashed sample, contained Am-241, Np-237, Pu-239, U-238, and Th-232 nuclides, was dissolved with 15 ml of 0.10-M HCl solution. To this solution, zinc metal was added and the combination was allowed to stand at room temperature for 20 minutes. The metal was then removed, rinsed with a minimal amount of distilled water, and the rinse was added to the sample. The sample was then adjusted with concentrated HCl until the HCl concentration of the solution was approximately 5.0-M. This sample was then loaded onto a TRU-Resin™ column. No H_2O_2 was added to the load solution. The eluting solutions were sequentially: 1.0-M HCl, 0.10-M NH_4I in 1.0-M HCl, 0.030-M $H_2C_2O_4$ in 1.0-M HCl, 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl, and 0.10-M $(NH_4)_2C_2O_4$. Results indicate that the most of the Pu did not load but was detected in the same fraction as the Am^{+3}. The Th and Np eluted in fractions where previously observed. About half of the U eluted in the same fraction as

the Np and the remaining U eluted in the ammonium hydrogen oxalate solution. (See RB063093, Appendix B, Table LXXVI.)

(2) Another reducing agent tested was hydrazine sulfate, $H_2NNH_2 \cdot H_2SO_4$. A 1.0 g amount of $H_2NNH_2 \cdot H_2SO_4$ was added to the load solution which contained Am, Np, Pu, U, and Th nuclides in 5.0-M HCl solution. This solution was loaded onto a TRU-Resin™ column and sequentially eluted using: 0.10-M hydrazine sulfate in 1.30-M HCl, 0.03-M $H_2C_2O_4$ in 0.50-M HCl, and 30.0 ml 0.10-M $(NH_4)_2C_2O_4$. This test procedure was ran in triplicate. Results indicate that the most of the Pu did not load but was detected in the same fraction as the Am^{+3}. The Th and Np eluted in fractions where previously observed. A small percentage of the U eluted in the same fraction as the Np and the most of the U eluted in the ammonium oxalate solution. (See RB012494-A,B,C, Appendix B, Tables LXXVII and LXXVIII.)

(3) Hydrogen peroxide, H_2O_2, was tested as a redox agent. A 1.0 ml amount of 30% H_2O_2 was added to the load solution which contained Am, Np, Pu, U, and Th nuclides in 5.0-M HCl solution. This solution was loaded onto a TRU-Resin™ column and sequentially eluted using: 0.15-M H_2O_2 in 1.30-M HCl, 0.03-M $H_2C_2O_4$ in 0.50-M HCl, and 30.0 ml 0.10-M $(NH_4)_2C_2O_4$. This test procedure was run in triplicate. Results indicate that the Pu loaded onto the TRU-Resin™. The Th eluted primarily in 1.30-M HCl fraction. The Np and Pu ions were largely detected in the fraction eluted using the 0.03-M $H_2C_2O_4$ in 0.50-M HCl reagent. The U eluted in the ammonium oxalate solution. One difficulty with this procedure was that bubbles formed in the column and slowed the flow rate. (See RB012494-D,E,F, Appendix B, Tables LXXIX and LXXX.)

12. Independent separations performed by Mr. Tiam Xie.

The data from the first set of samples processed by Mr. Xie are listed as TX111093 in Appendix B, Tables LXXXI and LXXXII. This was the first time Mr. Xie used the TRU-Resin™ separation procedure. He set up five test samples, all with distilled water matrices and containing approximately 15 dpm of each of the following nuclides: Th-232, Np-237, Pu-239, U-238, Am-241, Cm-244, Th-229, Am-243, Pu-236, U-232, and Pu-242. He also set up one blank sample, with a

distilled water matrix and containing approximately 15 dpm of each of the following nuclides: Am-243, Pu-242, Pu-236, U-232, and Th-229. The blank and test samples were processed the same. They were loaded onto TRU-Resin™ columns in 5.0-M HCl plus H_2O_2 solution. The columns were sequentially eluted using: 4.0 ml of 1.30-M HCl(Th fraction), 4.0 ml of 1.30-M HCl(wash), 20.0 ml of 0.10-M NH_4I plus 0.030-M $H_2C_2O_4$ in 0.50-M HCl(Np/Pu fraction), 40.0 ml of 0.10-M $(NH_4)_2C_2O_4$(U fraction). The eluate from the load solution was coprecipitated using a calcium oxalate coprecipitation procedure. The resulting oxalate coprecipitate was dissolved in nitric acid plus ascorbic acid and reloaded onto the TRU-Resin™ column. The Am ions were then eluted from the column with 20 ml of 3.0-M HCl solution. Considering this was the first time that Mr. Xie had ever used the procedure, the results were good. In his results, the various nuclides eluted in the expected fractions and the percents recovery of the yield tracers were in good agreement with the percents recovery of the spiked nuclides. In the five test samples, the following average percents recovery were: 88±5% Th-232, 98±5% Th 229, 56±6% Np-237/Pu-242, 67±4% Pu-239, 63±5% Pu-236, 66±4% U-238, 63±5% U-232, 94±6% Am-241, 107±7% Am-243, and 109±11% Cm-244. The Np-237 and Pu-242 were counted together due to alpha energy range overlap on the spectra. Cm-244 may contain some counts from Ac-225, a decay daughter of Th-229.

The data from the second set of samples processed by Mr. Xie are listed as TX012594 in Appendix B, Tables LXXXIII and LXXXIV. In this experiment, he set up four test samples, all with distilled water matrices and containing approximately 15 dpm of each of the following nuclides: Th-232, Np-237, Pu-239, U-238, Am-241, Th-229, Am-243, Pu-236, and U-232. He also set up two blank samples, with distilled water matrices and containing approximately 15 dpm of each of the following nuclides: Am-243, Pu-236, U-232, and Th-229. The columns were sequentially eluted using: 6.0 ml of 1.30-M HCl(Th fraction), 2.0 ml of 1.30-M HCl(wash), 15 ml of 0.030-M $H_2C_2O_4$ in 0.50-M HCl(Np/Pu fraction), 20.0 ml of 0.10-M $(NH_4)_2C_2O_4$(U fraction). The eluate from the load solution was coprecipitated using a calcium oxalate coprecipitation procedure. The resulting oxalate coprecipitate was dissolved in nitric acid plus ascorbic acid and reloaded onto the TRU-

Resin™ column. The Am ions were then eluted from the column with 15 ml of 3.0-M HCl solution. The steps of this procedure are similar to the ones used in the first experiment. The changes included: no ammonium iodide in Np/Pu elution solution, a slight increase in the elution volume for the Th fraction, and slight decreases in the elution volumes of the U and Am fractions. When preparing the loading solution for the second experiment, Mr. Xie added the necessary H_2O_2 reagent, however he then proceeded to heat the load solution until it stopped bubbling. This essentially destroyed the H_2O_2 and appears to have effected the recovery of the Pu ions. The percent recovery of Pu in the Np/Pu fraction was less than 10%. If the Pu was reduced to the +3 valence by the destruction of the Pu-H_2O_2 complex, it would not have loaded initially and would have remained in the load eluate with the Am fraction. The Pu was not detected in the Am fraction, but the process of dissolving the calcium oxalate coprecipitate in nitric acid may have oxidized the Pu to the +4 oxidation state. If this occurred, then the Pu would have remained on the column when the Am was eluted (if the Pu had coprecipitated with the calcium oxalate). [Step #11 (nitrate load eluate) and step #14 (1.0-M HCl rinse) had been combined. The combined samples were NdF_3 coprecipitated and alpha counted. A gross alpha count resulted in a range of 7 to 13 dpm per sample which may represent some of the lost Pu.] Except for the Pu ions, the various nuclides eluted in the expected fractions and the percents recovery of the yield tracers were in good agreement with the percents recovery of the spiked nuclides. In the four test samples, the following average percents recovery were: 92±5% Th-232, 93±5% Th 229, 103±6% Np-237, 9±2% Pu-239, 7±2% Pu-236, 93±5% U-238, 96±5% U-232, 63±4% Am-241, and 70±4% Am-243.

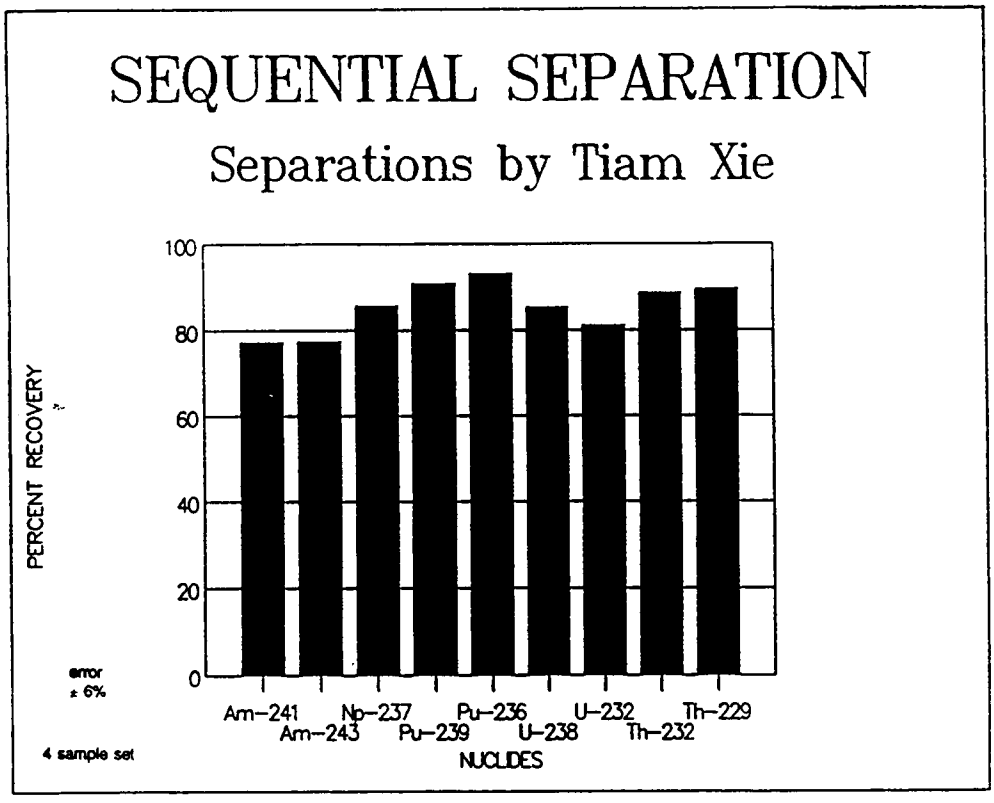
The data from the third set of samples processed by Mr. Xie are listed as TX020994 in Appendix B, Tables LXXXV and LXXXVI. In this experiment, he set up four test samples, all with distilled water matrices and containing approximately 15 dpm of each of the following nuclides: Th-232, Np-237, Pu-239, U-238, Am-241, Th-229, Am-243, Pu-236, and U-232. He also set up two blank samples, with a distilled water matrix and containing approximately 15 dpm of each of the

following nuclides: Am-243, Pu-236, U-232, and Th-229. The columns were sequentially eluted using: 6.0 ml of 1.30-M HCl(Th fraction), 3.0 ml of 1.30-M HCl(wash), 15 ml of 0.030-M $H_2C_2O_4$ in 0.50-M HCl(Np/Pu fraction), 30.0 ml of 0.10-M $(NH_4)_2C_2O_4$(U fraction). The eluate from the load solution was coprecipitated using a calcium oxalate coprecipitation procedure. The resulting oxalate coprecipitate was dissolved in nitric acid plus ascorbic acid and reloaded onto the TRU-Resin™ column. The Am ions were then eluted from the column with 15 ml of 3.0-M HCl solution. The steps of this procedure are similar to the ones used in the second experiment. The changes were small and included: slight increase in the elution volume for the rinse solution between the Th fraction elution and Np/Pu fraction elution and a slight increase in the elution volumes of the U fractions. The various nuclides eluted in the expected fractions with the percent recovery of the yield tracers in good agreement with the percent recovery to the other nuclides in the fraction. In the four test samples, the following average percents recovery were: 89±5% Th-232, 92±5% Th 229, 86±5% Np-237, 91±5% Pu-239, 96±8% Pu-236, 86±5% U-238, 81±5% U-232, 77±4% Am-241, and 78±4% Am-243. (See Figure 15.)

D. Separation of Actinide Ions in the Presence of Various Matrix Ions

1. Separation of Am, Th, and U ions from soil matrices using HCl-load TRU-Resin™ separation.

In the initial test, approx. 15 dpm of Am-241 and approx. 50 dpm of U-238 were added to a 1.0 g solubilized soil sample. (Soil sample was solubilized using a microwave digestion procedure with HCl, HNO_3, HF, H_3BO_3 and $HClO_4$.) The sample was loaded onto a 1.0-ml bed volume TRU-Resin™ column using 3.0-M HCl. The column was then eluted sequentially with 1.0-M HCl and 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl. The ammonium hydrogen oxalate fraction was wet ashed, NdF_3 coprecipitated, and counted. The percent recovery of the U ions was 83±10%. The Am ions were in the HCl-load eluate which still contained most of the soil matrix and was unable to be counted. (See RB051693, Appendix B, Table LXXXVII.)



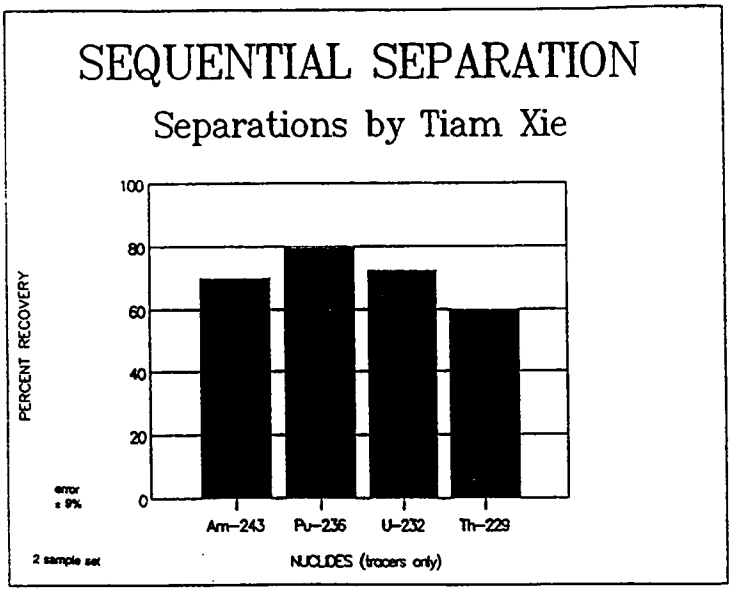


Figure 15: Separations by Mr. Tiam Xie. Experiment TX020994.

In a subsequent test, approx. 20 dpm each of Am-241, Th-232, and U-238 ions were added to a 2.0 g dissolved soil sample. The sample was loaded onto a TRU-Resin™ column in 5.0-M HCl plus H_2O_2 solution. The column was then eluted sequentially with 1.0-M HCl(Th fraction) and 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl(U fraction). The Th and U ions eluted in the expected fractions but with moderate percents recovery: 38±4% Th and 58±6% U. The Am ions were in the HCl-load eluate which also still contained most of the soil matrix and was unable to be counted. (See RB052793, Appendix B, Table LXXXVII.)

2. Separation of Am ions from a soil matrix using HNO_3-load TRU-Resin™ separation.

Approx. 15 dpm of Am-241 was added to a 2.0 g dissolved soil sample. The soil contained a small amount of natural uranium (2.5 dpm/g) previously measured by IT. The sample was loaded onto a 1.0-ml bed volume TRU-Resin™ column using 3.0-M HNO_3. The column was then eluted sequentially with 8.0-M HCl, 4.0-M HCl (Am fraction), and 0.10-M HCl. The percent recovery of the Am ions was 10±2%. The load solution was light yellow in color, possibly indicating the presence of Fe^{+3} ions. (See RB061893, Appendix B, Table LXXXIX.)

3. Separation of Am, Th, and U ions from a soil matrix using HCl-TRU-Resin™ separation followed by HNO_3-ascorbic acid-load TRU-Resin™ separation.

Three 2.0 g soil samples, from different geographical locations, were solubilized and numbered: samples #2, #7, and #12. Each sample originally contained some actinide ions and each was spiked with Am-241, Th-232, and U-238. The resulting amount of each actinide ion in the samples were approximately: Sample #2 15 dpm Am-241, 89 dpm Th-232, 198 dpm U-238; Sample #7 15 dpm Am-241, 86 dpm Th-232, 1550 dpm U-238; and Sample #12 15 dpm Am-241, 831 dpm Th-232, 100 dpm U-238. The samples were loaded in 5.0-M HCl plus H_2O_2. The column was then eluted sequentially with 1.0-M HCl (Th fraction) and 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl (U fraction). The HCl-load eluate was wet ashed and redissolved in 3.0-M HNO_3. Before the nitrate solution was loaded onto the column, ascorbic acid was added to reduce any Fe^{+3} ions to

Fe^{+2} ions. (Ammonium thiocyanate was used to check for complete reduction of the iron(III) ions.) The sample was then loaded onto the TRU-Resin™ column. The column was then eluted sequentially with 9.0-M HCl and 4.0-M HCl (Am fraction). The U, Th, and Am ions eluted in the expected fractions with no overlap into other fractions, except for a small amount of U ions in sample#7. The percents recovery of the actinide ions were: Sample #2 35±5% Am-241, 83±7% Th-232, 80±9% U-238; Sample #7 59±7% Am-241, 89±7% Th-232, 46±6% U-238, Sample #12 57±7% Am-241, 70±6% Th-232, 78±9% U-238. (See RB060893, Appendix B, Table XC.) Note that in all three sample, the Am ions were only moderately recovered.

4. Adding actinide spikes before and after the calcium oxalate coprecipitation procedure.

Two 2.0 g soil samples were solubilized and numbered: samples #13 and #14. Sample #13 was spiked with approximately 20 dpm each of Am-241, Np-237, Pu-239, U-238, and Th-232 ions after solubilization but before a calcium oxalate coprecipitation procedure was performed on the sample. Sample #14 was spiked with the same actinide ions after solubilization and after a calcium oxalate coprecipitation procedure was performed on the sample. The two samples were then loaded onto TRU-Resin™ columns in 5.0-M HCl plus H_2O_2. The columns were then eluted sequentially with 1.0-M HCl, 0.10-M NH_4I in 1.0-M HCl, 0.030-M $H_2C_2O_4$ in 1.0-M HCl, 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl, and 0.10-M $(NH_4)_2C_2O_4$. The HCl-load eluate was wet ashed and redissolved in 3.0-M HNO_3 plus ascorbic acid. This nitrate-dissolved fraction was then loaded onto the TRU-Resin™ column. The column was then eluted sequentially with 8.0-M HCl, 3.0-M HCl, and 0.10-M HCl. The percents recovery of the actinide ions were: Sample #13 - 51±6 Np-237, 80±9% Pu-239, 73±8% Th-232, 3±2% U-238; Sample #14 - 93±9% Am-241, 79±8 Np-237, 81±9% Pu-239, 82±9% Th-232, 70±7% U-238. (See RB070193, Appendix B, Table XCI.) These percents recovery seem to indicate that: (1) U-238 ions did not coprecipitate well with calcium oxalate, (2) Np-237 ions only partially coprecipitates with calcium oxalate, and (3) a high percentage of Pu-239 and Th-232 ions coprecipitate with calcium oxalate. There was no result for Am-241 from

sample #13 due to technical error, however several following experiments indicate that a high percentage of Am coprecipitates with calcium oxalate. (See RB082793 and RB072093, Appendix B, Tables XCII, XCIII, and XCV.)

5. Processing half of the eluted Am fraction with the calcium oxalate coprecipitation step and half without.

Four solubilized 2.0 g soil samples, a deionized water sample, and a deionized water blank were spiked with approximately 20 dpm each of Th-232, Np-237, Pu-239, Am-241 and U-238 ions. Each sample was loaded in 5.0-M HCl plus H_2O_2 solution onto a separate TRU-Resin™ column. The final HCl-load volumes of the samples were large, 50-60 ml, and required 2.5-4.0 hr. to completely pass through the resin bed. The columns were then eluted sequentially with 1.3-M HCl (Th fraction), 0.10-M NH_4I plus 0.030-M $H_2C_2O_4$ in 0.5-M HCl (Np/Pu fraction), 0.50-M HCl, and 0.10-M $(NH_4)_2C_2O_4$ (U fraction). The HCl-load eluate was divided into two equal volumes. One half was wet ashed and redissolved in 3.0-M HNO_3 plus ascorbic acid. This nitrate-dissolved fraction was then loaded onto the TRU-Resin™ column. The second half was coprecipitated using the calcium oxalate procedure. It was then redissolved in 3.0-M HNO_3 plus ascorbic acid and loaded onto the TRU-Resin™ column. The nitrate-columns were then eluted sequentially with 2.0-M HNO_3 plus 0.05 $H_2C_2O_4$, 8.0-M HCl, 3.0-M HCl (Am fraction), 1.0-M HCl, and 0.50-M HCl. The average percents recovery of the actinide ions from the four soil samples were: 91±5 Np-237, 86±5% Pu-239, 90±5% Pu-236, 68±7% Th-232, 66±7% Th-229, 37±3% U-238, 55±5% U-232, without CaC_2O_4 coppt 31±4% Am-241, with CaC_2O_4 coppt 84±8% Am-241, without CaC_2O_4 coppt 36±4% Am-243, and with CaC_2O_4 coppt 97±9% Am-243. The percents recovery of the actinide ions from the deionized water sample were: 84±9 Np-237, 78±8% Pu-239, 84±9% Pu-236, 88±16% Th-232, 83±15% Th-229, 63±8% U-238, 63±8% U-232, without CaC_2O_4 coppt 84±15% Am-241, with CaC_2O_4 coppt 94±17% Am-241, without CaC_2O_4 coppt 103±19% Am-243, and with CaC_2O_4 coppt 98±18% Am-243. (See RB082793, Appendix B, Tables XCII and XCIII.) The following may be noted from these results: (1) The Np-237 and Pu-239 ions eluted as expected

and had good percent recovery. (2) The total percent recovery of Th-232 was good. However the Th-232 was recovered partially from the HCl-load column and the HNO_3-load column. This may relate to the large load volume and long loading time of the HCl-column. (3) The recovery of the U-238 ions was low. This may relate to the large load volume and the long load time. Also, the U-238 percent recovery did not parallel the yield tracer's, U-232, recovery. This may relate to the soil sample originally containing some U-238 activity. and (4) The percents recovery of Am-241 ions with the use of the calcium oxalate coprecipitation procedure are significantly better than the percents recovery of Am-241 ions without the use of the calcium oxalate coprecipitation procedure.

6. Processing soil samples for effective percent recovery of the actinide ions.

Three 2.0 g soil samples were solubilized and spiked with approximately 15 dpm of Am-241, Np-237, Pu-239, Th-232, and U-238. The samples were loaded onto TRU-Resin™ columns in 5.0-M HCl plus H_2O_2. The columns were then eluted sequentially with 2.0-M HCl, 1.0-M HCl, 0.10-M NH_4I plus 0.030-M $H_2C_2O_4$ in 1.0-M HCl, 1.0-M HCl, and 0.10-M $(NH_4)_2C_2O_4$. The HCl-load eluates were calcium oxalate coprecipitated and redissolved in 3.0-M HNO_3. (The ascorbic acid was omitted.) These nitrate-dissolved fractions was then loaded onto TRU-Resin™ columns. The columns were then eluted sequentially with 8.0-M HCl, 3.0-M HCl, and 0.10-M HCl. The percents recovery of the actinide ions were: Np-237 103±18%, 65±12%, 82±14%; Pu-239 91±16%, 65±12%, 80±14%; U-238 71±10%, 71±10%, 61±9%; Th-232 95±13%, 74±10%, 85±9%; Am-241 69±9%, 59±7%, 85±10%. (See RB071993A-B and RB072093, Appendix B, Tables XCIV and XCV.)

7. Separation of Am-241, Np-237, Pu-239, Th-232, and U-238 ions from hard water matrices.

a. Separation of actinide ions from solutions matrices containing 10.0 g of Al^{+3}, $Fe^{+3/+2}$, Ca^{+2}, or Mg^{+2} ions.

The four matrix ion solutions were made by dissolving chloride compounds of each of the ions into separate beakers containing 200±50 ml of 5.0-M HCl solution. The amount of each

compound dissolved was calculated to yield 10 grams of the metal ion in solution. A blank sample consisted of 200 ml of 5.0-M HCl solution without any matrix ions added. All samples were spiked after matrix ions were fully dissolved. The samples were loaded onto TRU-Resin™ columns in 5.0-M HCl plus H_2O_2. With the large volumes of samples, the amount of time to load the HCl-column was very long (5-10 hr.). The columns were then eluted sequentially with 1.3-M HCl, 0.10-M NH_4I plus 0.030-M $H_2C_2O_4$ in 1.0-M HCl, 0.50-M HCl, and 0.10-M $(NH_4)_2C_2O_4$. The HCl-load eluates were calcium oxalate coprecipitated and redissolved in 3.0-M HNO_3 plus ascorbic acid. The HNO_3-column load volumes were 10-60 ml. These nitrate-dissolved fractions were then loaded onto TRU-Resin™ columns. The columns were then eluted sequentially with 8.0-M HCl, 3.0-M HCl, 1.30-M HCl, 0.03-M $H_2C_2O_4$, and 0.10-M $(NH_4)_2C_2O_4$. The percents recovery of the actinide ions in the expected elution fractions were: 10.0 g Al: Np-237 5±2%, Pu-239 7±3%, U-238 1±1%, Th-232 30±6%, Am-241 22±4%; 10.0 g Ca: Np-237 2±1%, Pu-239 73±7%, U-238 77±8%, Th-232 75±7%, Am-241 18±3%; 10.0 g Fe: Np-237 2±1%, Pu-239 54±6%, U-238 10±2%, Th-232 6±2%, Am-241 112±13%; 10.0 g Mg: Np-237 4±1%, Pu-239 60±6%, U-238 62±7%, Th-232 69±6%, Am-241 2±1%; and Blank: Np-237 81±8%, Pu-239 81±8%, U-238 72±7%, Th-232 76±6%, Am-241 121±14%. (See RB111993, Appendix B, Tables XCVI and XCVII.)

The following may be noted from these results. (1) In the blank sample: The sample had a 200 ml HCl-load volume and required 5.5 hr. to pass through the resin bed (flow rate = 0.6 ml/min.). This large volume could start to elute some of the actinide ions during the loading process. However, the percents recovery of the actinide ions in the blank sample were good and did not seem to show any significant negative effect from the large volume and the long load time of the sample. (2) In the Al sample: The Np-237 and Pu-239 ions did not elute in the expected fraction but 53% Np-237 and 69% Pu-239 eluted in the 1.30-M HCl solution (Th fraction). Only 29 % of the Th-232 ions eluted in the expected fraction, but an additional 22% eluted in the Am-241 fraction. (3) In the Ca sample: The Pu-239, Th-232, and U-238 ions showed good recovery in the expected fractions. The Np-237 ions were present in the Th-232 and Am-241 elution fractions.

The calcium oxalate coprecipitation procedure produced large amounts of precipitate. The processing of this large amount of precipitate was difficult and may have affected the recovery of the Am-241 ions. (4) In the Mg sample: The recovery pattern of the actinide ions was very similar to the recovery pattern of the ions in the Ca sample. (5) In the Fe sample: The percent recovery of the Am-241 ions was very good. The percent recovery of the Pu-239 was moderate in the expected fraction and an additional 22% was detected in other fractions. Although very little of the Th-232 and Np-237 ions were recovered in the expected fractions, approx. 30% Np-237 ions and approx. 60% Th-232 ions were recovered in an additional elution step after the Am fraction was eluted from the nitrate-column.

b. Separation of actinide ions from solution matrices containing 1.0 g of Al, Fe, or Ca ions.

Three matrix ion solutions were made by dissolving chloride compounds of each of the ions into separate beakers containing 20±5 ml of 5.0-M HCl solution. The amount of each compound dissolved was calculated to yield 1.0 gram of the metal ions in solution. A blank sample consisted of 20 ml of 5.0-M HCl solution without any matrix ions added. All samples were spiked after matrix ions were fully dissolved. These samples have approximately the same ion concentration per milliliter as the samples in section (a) above, however, these samples have a smaller total volume. The lengths of time required to load the samples varied from 20-150 minutes (blank 22 min., Al 152 min., Ca 35 min., Fe 55 min.). The samples were loaded onto TRU-Resin™ columns in 5.0-M HCl plus H_2O_2. The columns were then eluted sequentially with 1.3-M HCl, 0.030-M $H_2C_2O_4$ in 0.5-M HCl, and 0.10-M $(NH_4)_2C_2O_4$. The HCl-load eluates were calcium oxalate coprecipitated and redissolved in 3.0-M HNO_3 plus ascorbic acid. These nitrate-dissolved fractions were then loaded onto TRU-Resin™ columns. The columns were then eluted sequentially with 8.0-M HCl, 3.0-M HCl, and 0.10-M $(NH_4)_2C_2O_4$. The percents recovery of the actinide ions in the expected elution fractions were: 1.0 g Al: Np-237 81±13%, Pu-239 97±9%, U-238 56±7%, Th-232 80±8%, Am-241 88±8%; 1.0 g Ca: Np-237 91±14%, Pu-239 93±9%, U-238 64±8%, Th-232 89±9%, Am-

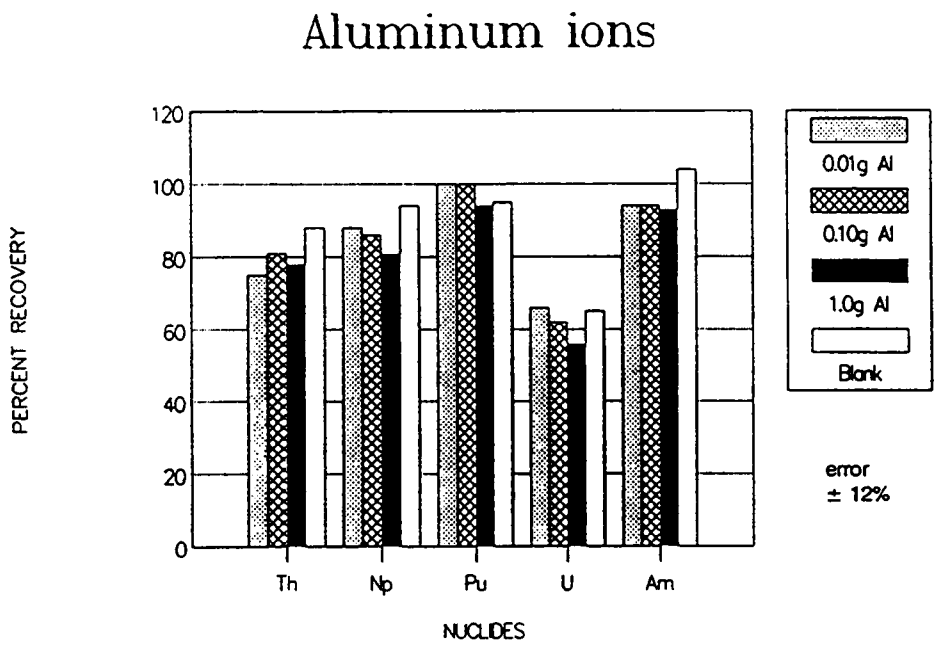
241 55±5%; 1.0 g Fe: Np-237 77±12%, Pu-239 26±3%, U-238 36±4%, Th-232 20±3%, Am-241 82±9%; and Blank: Np-237 84±13%, Pu-239 83±8%, U-238 54±7%, Th-232 82±8%, Am-241 104±10%. (See RB021594, Appendix B, Tables XCVIII and XCIX.)

c. Separation of actinide ions from solution matrices containing 0.50 g, 0.10 g, or 0.010 g of Al, Fe, or Ca ions.

Six matrix ion solutions were made by dissolving chloride compounds of each of the ions into separate beakers containing 20±5 ml of 5.0-M HCl solution. The following solutions were prepared: 0.10 g Al, 0.010 g Al, 0.10 g Ca, 0.50 g Ca, 0.10 g Fe, and 0.010 g Fe. A blank sample consisted of 25 ml of 5.0-M HCl solution without any matrix ions added. All samples were spiked after matrix ions were fully dissolved. The length of time to load the samples varied from 80-100 minutes. The samples were loaded onto TRU-Resin™ columns in 5.0-M HCl plus H_2O_2. The columns were then eluted sequentially with 1.3-M HCl, 0.030-M $H_2C_2O_4$ in 0.5-M HCl, and 0.10-M $(NH_4)_2C_2O_4$. The HCl-load eluates were calcium oxalate coprecipitated and redissolved in 3.0-M HNO_3 plus ascorbic acid. These nitrate-dissolved fractions were then loaded onto TRU-Resin™ columns. The columns were then eluted sequentially with 8.0-M HCl, 3.0-M HCl, and 0.10-M $(NH_4)_2C_2O_4$. The percents recovery of the actinide ions in the expected elution fractions were: 0.10 g Al: Np-238 86±13%, Pu-239 97±9%, U-238 65±7%, Th-232 85±8%, Am-241 88±8%; 0.010 g Al: Np-237 88±13%, Pu-239 96±9%, U-238 66±7%, Th-232 81±8%, Am-241 89±8%; 0.10 g Ca: Np-237 89±14%, Pu-239 92±9%, U-238 63±8%, Th-232 76±8%, Am-241 86±9%; 0.50 g Ca: Np-237 94±15%, Pu-239 98±9%, U-238 59±7%, Th-232 75±8%, Am-241 87±9%; 0.10 g Fe: Np-237 107±16%, Pu-239 80±9%, U-238 63±7%, Th-232 75±8%, Am-241 91±9%; 0.010 g Fe: Np-237 90±14%, Pu-239 83±9%, U-238 81±9%, Th-232 88±9%, Am-241 94±9%; and Blank: Np-237 104±15%, Pu-239 105±10%, U-238 82±9%, Th-232 93±9%, Am-241 88±9%. (See RB012994, Appendix B, Tables C and CI.)

The results discussed in sections 7b and 7c are shown as bar graphs in Figures 16 and 17. The Al samples (0.010 g, 0.10 g, and 1.0 g) showed good agreement with the results of the

(a)



(b)

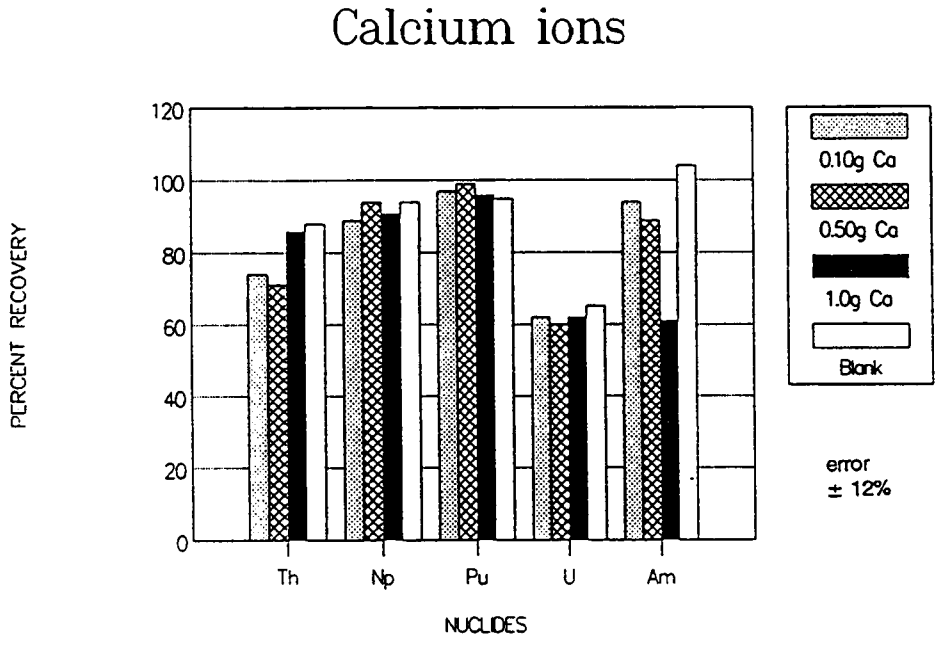
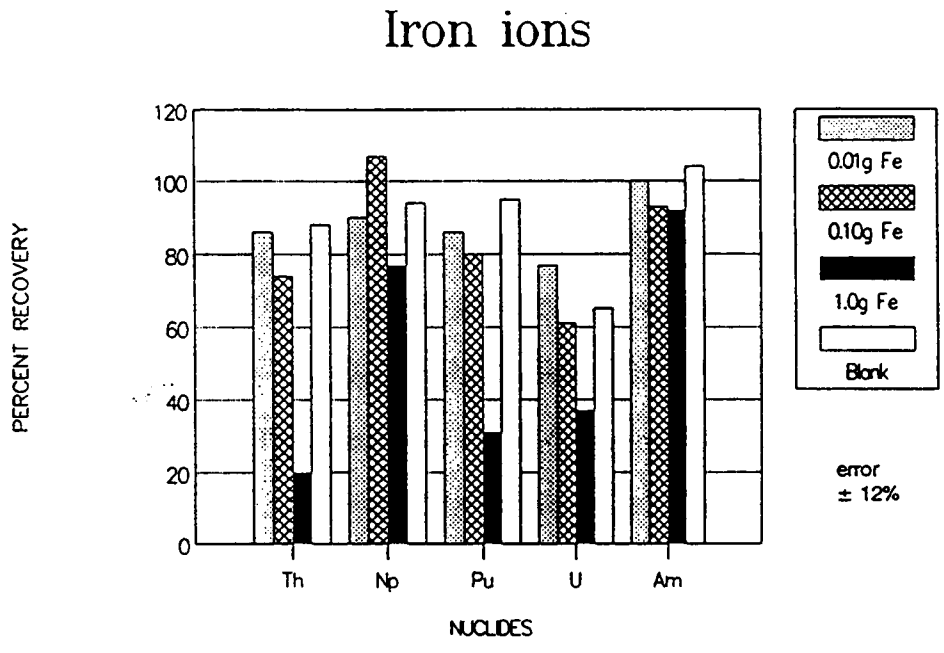


Figure 16: Separation of Actinide Ions from Hard Water Matrix: (a) Aluminum ions, (b) Calcium ions.

(a)



(b)

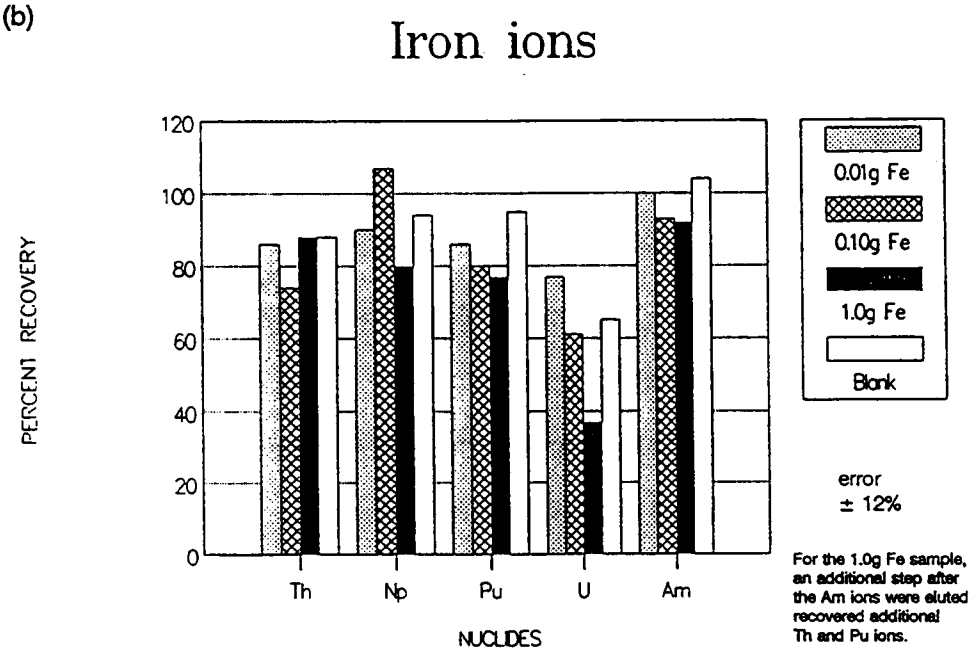


Figure 17: Separation of Actinide Ions from Hard Water Matrix: (a) Iron ions, (b) Iron ions, additional elutions.

blank samples (Figure 16a). The Ca samples (0.10 g, 0.50 g, and 1.0 g) also showed good agreement with the results of the blank samples except for the recovery of Am (55%) in the 1.0 g Ca sample (Figure 16b). In the test samples that had Ca ions, large amounts of precipitate would form during the calcium oxalate coprecipitation procedure. This may have effected the recovery of Am in the 1.0 g Ca sample. The Fe samples (0.010 g and 0.10 g) showed good agreement with the blank samples (Figure 17a). The 1.0 g Fe sample varied significantly from the blank samples in the recovery of Th, Pu, and U (Figure 17a). However, when additional elution steps were added after the Am ions elution step, additional Th and Pu were recovered: 55±6% Pu and 68±10% Th. This brought the total percents recovery of the Pu and Th into good agreement with the blank samples (Figure 17b).

E. Separation of the Actinide Ions as One Group from Soil Matrices

1. Separation using Barium Sulfate coprecipitation

a. Samples with deionized water matrix.

Three samples were prepared by adding approximately 25 dpm each of Am-243, Np-237, Pu-236, U-238, and Th-232 to 4.0 ml of conc. H_2SO_4 solution. Then 2.0-ml 0.45% $BaCl_2 \cdot 2HOH$ (3.7E-3 moles Ba) was added and the sample was heated on a hot plate until the sample fumed and the sulfate precipitate dissolved. The mixture was then cooled in a cold tap water bath for about 3 min. and 20 ml of deionized water was added. At this point, the sulfate precipitate formed. The sample was cooled for 20 more minutes. The solid was then filtered, dried, and alpha counted. An average alpha count of approximately 200 dpm was detected per coprecipitated sample. This would indicate that some of the nuclides coprecipitated with the barium sulfate. However the spectra were poorly resolved so individual percents recovery could not be determined. The filtrate was wet ashed, NdF_3 coprecipitated, and alpha counted. The average percents recovery in the filtrate were: 10±2% Am-243, 8±2 Np-237, 13±2% Pu-236, 8±2% U-238, 14±3% Th-232. After being counted, the solid sulfate coprecipitant and the filtration membranes

were placed into a ceramic crucible with NaCl and muffled at 850°C for 480 minutes. The muffled samples were wet ashed several times with mixtures of HCl, HNO_3, or H_2O_2. Then, the actinide ions in the muffled sample were separated using the TRU-Resin™ nitrate column. The average percents recovery were: 17±3% Am-243, 14±3 Np-237, 12±3% Pu-236, 0% U-238, 13±3% Th-232. (See RB012594, Appendix B, Tables CII and CIII.) Note that: (1) the UO_2^{+2} (U-238) was not expected to coprecipitate with the barium sulfate without a reducing agent in the solution and (2) the muffled sulfate may not have been successfully dissolved.

b. Samples with a reduction or oxidation reagent added.

Four samples were prepared by adding approximately 25 dpm each of Am-243, Np-237, Pu-239, U-238, and Th-232 to 4.0 ml of conc. H_2SO_4 solution. Each sample was fumed for about 3 minutes and diluted with 20 ml of deionized water. Then the following redox reagents were added, one to each sample: (A) 0.30 g $K_2S_2O_4$ plus 4 drops 1.0% $AgNO_3$, (B) 7 drops 30% H_2O_2, (C) 3 drops 20% $TiCl_3$, and (D) 0.15 g ascorbic acid plus 4 drops 0.004 g/ml Fe^{+2}/Fe^{+3} ions. After these mixtures sat for 10 min., 2.0-ml 0.45% $BaCl_2 \cdot 2HOH$ was added and the sample was heated briefly on a hot plate and the mixture was then cooled in a cold tap water bath. The solid was then filtered. The filtrate was wet ashed, NdF_3 coprecipitated, and alpha counted. The average percents recovery in the filtrate were: sample A 1±1% Am-243, 27±6 Np-237, 8±2% Pu-239, 49±8% U-238, 3±1% Th-232; sample B 35±7% Am-243, 20±4 Np-237, 84±13% Pu-239, 14±3% U-238, 53±8% Th-232; sample C 21±5% Am-243, 14±3 Np-237, 27±5% Pu-239, 12±2% U-238, 47±8% Th-232; and sample D 17±4% Am-243, 18±4 Np-237, 23±5% Pu-239, 15±3% U-238, 46±8% Th-232. The solid sulfate coprecipitates were placed into a ceramic crucible with NaCl plus carbon and muffled at 850°C for 480 minutes. The muffled sulfate coprecipitates were not successfully dissolved. (See RB020894, Appendix B, Table CIV.)

c. Samples with Al, Ca, or Fe ions added.

Three matrix ion solutions were made by dissolving chloride compounds of Al, Ca, or Fe ions into separate beakers containing 3.0-M HCl solution. The amount of each compound

dissolved was calculated to yield 0.10 grams of the metal ions in solution. A blank sample was also prepared without any matrix ions added. Approximately 25 dpm each of Am-243, Np-237, Pu-239, U-238, and Th-232 and 4.0 ml of conc. H_2SO_4 were added to each sample. Then 2.0-ml 0.45% $BaCl_2 \cdot 2HOH$ was added and the samples were fumed and diluted with 20 ml of deionized water. The solid was then filtered. The filtrate was wet ashed, NdF_3 coprecipitated, and alpha counted. The average percents recovery in the filtrate were: 0.10 g Al: 1±1% Am-243, 2±1 Np-237, 0% Pu-239, 1±1% U-238, 2±1% Th-232, 0.10 g Ca: 0% Am-243, 37±8 Np-237, 21±4% Pu-239, 58±9% U-238, 63±10% Th-232, 0.10 g Fe: 56±9% Am-243, 42±9 Np-237, 67±10% Pu-239, 50±9% U-238, 58±10% Th-232, Blank: 30±5% Am-243, 41±9 Np-237, 53±8% Pu-239, 39±6% U-238, 29±5% Th-232. (See RB021494, Appendix B, Table CV.)

2. Separation using Diphonix Resin.

The diphonix columns, which were used in the following experiments, consisted of 0.5 g of diphonix resin (Particle size 60-100 mesh, #DP-B01, lot 93-002) weighed as received from EIChroM (moist). The resin was soaked in the appropriate load solution and placed into a 2-ml-bed-volume Bio-Rad Poly-Prep chromatography column that had been pre-rinsed with distilled water. A glass wool plug was placed on the top of the resin. The resulting resin bed volume was 0.5 ml.

a. Loading Am-243, Np-237, Pu-236, U-238, Th-232 in 1.0-M HNO_3 and eluting with a large volume of 5.0-M HCl.

The loading in 1.0-M HNO_3 and the eluting with 5.0-M HCl was designed to minimize the attachment of possible matrix ions, such as Ca^{+2}, Fe^{+3}, Fe^{+2}, and Al^{+3}. No matrix ions were added to these samples so that the loading and eluting behaviors of certain actinide ions under these conditions could be observed.

Three samples (A,B, and C) containing approximately 25 dpm each of Am-243, Np-237, Pu-236, U-238, and Th-232 were wet ashed and dissolved in 10 ml 1.0-M HNO_3 solution. Samples A and B were loaded onto diphonix columns and rinsed with 2.5 ml of 1.0-M HNO_3

solution. The load eluate and rinse were collected, wet ashed and alpha counted. Next the columns were eluted with 10 ml of 5.0-M HCl solution. These eluates was also wet ashed and alpha counted. Then the columns were eluted with 50 ml of 5.0-M HCl solution. Again the eluates were wet ashed and alpha counted. Sample C was processed the same as samples A and B except that ascorbic acid was added to all loading and eluting solutions. The average percents recovery of the actinide ions were: (The results from samples A, B, and C are averaged together except for the Np results.) load and rinse eluate: 2±1% Am-243, 72±6%(A&B) 32±4%(C) Np-237, 1±1% Pu-236, 0% U-238, 0% Th-232; 10 ml 5.0-M HCl elution: 32±3% Am-243, 5±2%(A&B) 0%(C) Np-237, 6±2% Pu-236, 11±3% U-238, 0% Th-232; and 50 ml 5.0-M HCl elution: 39±4% Am-243, 0%(A&B) 0%(C) Np-237, 8±2% Pu-236, 8±2% U-238, 1±1% Th-232. (See RB012794, Appendix B, Table CVI.)

b. Using different reduction or oxidation reagents.

Four samples (A,B,C, and D) containing approximately 25 dpm each of Am-243, Np-237, Pu-236, U-238, and Th-232 were wet ashed and dissolved in 10 ml 1.0-M HCl solution. The following redox reagents were added to the samples: sample A 0.30 g $K_2S_2O_4$ plus 4 drops 1% $AgNO_3$, sample B 7 drops 30% H_2O_2, sample C 3 drops 20% $TiCl_3$ solution, and sample D 0.15 g ascorbic acid plus 4 drops 0.004 g/ml Fe^{+2}/Fe^{+3} ions. The mixed samples sat for 10 minutes and were loaded onto diphonix resin columns. (A small amount of AgCl formed in sample A. This was loaded onto the column as part of the sample.) The resin was rinsed with 2.5 ml 1.0-M HCl solution. The combined load and rinse eluates were wet ashed, NdF_3 coprecipitated, and alpha counted. The diphonix resins were removed from the columns and placed into ceramic crucibles with solid K_2CO_3 (m.p. 891°C). The resin samples were muffled at 900°C for 480 minutes. Dissolution of the muffled samples was unsuccessful. The percents recovery of the actinide ions in the load/rinse fractions were: sample A 1±1% Am-243, 7±2% Np-237, 0% Pu-236, 0% U-238, 0% Th-232; sample B 1±1% Am-243, 4±1% Np-237, 1±1% Pu-236, 0% U-238, 0% Th-232; sample C 58±7% Am-243, 13±3% Np-237, 52±7% Pu-236, 1±1% U-238,

1±1% Th-232; and sample D 1±1% Am-243, 0% Np-237, 1±1% Pu-236, 0% U-238, 0% Th-232. (See RB020794, Appendix B, Table CVII.)

c. Comparison of two muffle methods.

Four samples were prepared like sample D above. Each sample contained approximately 25 dpm each of Am-243, Np-237, Pu-236, U-238, and Th-232, then they were wet ashed and dissolved in 10 ml 1.0-M HCl solution. Ascorbic acid and Fe ions were added as redox reagents. The samples sat for 10 minutes and were loaded onto diphonix resin columns. The resin was rinsed with 2.5 ml 1.0-M HCl solution. The combined load and rinse eluates were wet ashed, NdF_3 coprecipitated, and alpha counted. The diphonix resins were removed from the columns, two were placed into ceramic crucibles with NaCl (m.p. 801°C) and two were placed into alumina crucibles with K_2CO_3 (m.p. 891°C). The resin samples were muffled at 700°C for 480 minutes. When the samples were cooled and removed from the muffle furnace, they appeared to be fused. The K_2CO_3 material appeared white and crumbly and the NaCl material had a light blue color. The muffled materials were dissolved with 5.0-M HCl and transferred to glass beakers. The K_2CO_3 material dissolved more easily than the NaCl material. The samples were wet ashed, fumed with Na_2SO_4, NdF_3 coprecipitated, and alpha counted. (The insoluble residue in the NaCl samples was filtered out prior to the NdF_3 coprecipitation. The filtered solid materials were alpha counted. They did not have significant activity.) The percents recovery of the actinide ions in the load/rinse fractions were: sample A 5±2% Am-243, 1±1% Np-237, 1±1% Pu-236, 1±1% U-238, 1±1% Th-232; sample B 2±1% Am-243, 0% Np-237, 1±1% Pu-236, 0% U-238, 0% Th-232; sample C 3±2% Am-243, 2±1% Np-237, 1±1% Pu-236, 0% U-238, 0% Th-232; and sample D 3±2% Am-243, 1±1% Np-237, 2±1% Pu-236, 1±1% U-238, 0% Th-232. The percents recovery of the actinide ions from the muffled resins were: sample A 19±4% Am-243, 38±8% Np-237, 25±4% Pu-236, 20±3% U-238, 97±12% Th-232; sample B 23±5% Am-243, 35±8% Np-237, 32±4% Pu-236, 17±3% U-238, 84±11% Th-232; sample C 62±13% Am-243, 80±16% Np-237, 87±13% Pu-236, 64±8% U-238, 98±12 Th-232; and sample D 68±13% Am-243, 95±19% Np-237, 97±15% Pu-236, 78±10% U-

238, 106±13 Th-232. (See RB021694, Appendix B, Table CVIII.) These results indicate that: (1) the tested actinide ions loaded in 1.0-M HCl plus ascorbic acid plus Fe, (2) the diphonix resin muffled with K_2CO_3 in an alumina crucible resulted in better percents recovery of the tested actinide ions than the NaCl in ceramic crucible method, and (3) percents recovery of Th-232 were good with either muffle method.

d. Loading a small amount of Am-241 and Am-243 ions from a large volume of solution.

Two samples containing approximately 50 dpm of Am-241 and 20 dpm of Am-243 each were wet ashed and dissolved in 1.0 liter of 0.10-M HCl solution. One sample was loaded onto a 2.0 g diphonix resin column by being poured through the resin. For the other sample, 2.0 g diphonix resin was placed into the solution and stirred using a magnetic stirrer (static-load). It took approximately 8 hours for the column-load sample to pass through the resin bed. The static-load sample was stirred for 3 hours and then the resin was filtered using Whatman 41 filter paper. In both samples, after the load process was completed, the 1.0 liter of solution was slowly evaporated, NdF_3 coprecipitated, and alpha counted. The resins from both samples were muffled using the K_2CO_3 in alumina crucible procedure previously discussed. The muffled sample were also alpha counted. The results showed that approximately 1% Am-241 and Am-243 remained in the solution of both samples. Approximately 35% of Am-241 and Am-243 was recovered from each of the muffled resins. Significant amount of Am ions were not recovered. (See RB022194, Appendix B, Table CIX.)

e. Separating actinide ions from soil matrices.

Two solubilized soil samples were diluted to 165 ml with 0.5-M HCl and approximately 25 dpm each of Th-232, U-238, and Am-241 was added. Diphonix resin (0.5 g) was added to each sample. The samples were stirred for 75 min. and the resin was removed by filtration with filter paper. The resins were muffled and redissolved. The filtered solutions and the redissolved resins were separated in Am, Th, and U fractions using the TRU-Resin™ procedure (chloride column,

calcium oxalate, nitrate column). The average percents recovery of the two samples were: dissolved resin 94±10% Am-241, 37±6% U-238, and 30±10% Th-232 and filtered solution 8±2% Am-241, 35±6% U-238, and 42±12% Th-232. (See RB030494, Appendix B, Table CX.) The percents recovery of the Th-232 ions listed above are the amount totaled from the Th-232 that eluted in several fractions from the columns.

3. Separation using Calcium Fluoride Coprecipitation.

a. Deionized water samples.

Two samples were prepared with each containing approximately 25 dpm of Th-232, Np-237, U-238, Am-241, and Pu-236 dissolved in 30 ml of 3.0-M HCl. Next, 100 mg of Ca^{+2} (2.0 ml of 1.25-M $Ca(NO_3)_2$) was added followed by 5.0 ml of 29-M HF. The mixed samples sat for 40 min. and were then centrifuged. The supernatant was decanted and the solid was washed with 15-M HF solution and recentrifuged. The calcium fluoride coprecipitate was slurried in 5-M HCl and heated slowly in a glass beaker until the mixture was almost dry. This wet ashing procedure was repeated twice more with 5.0 ml of 5.0-M HCl solution. The wet ashed samples (A-CaF_2, B-CaF_2) were dissolved in 5.0-M HCl plus H_2O_2 and loaded onto the TRU-Resin™ chloride column. The load eluate from the chloride column was wet ashed and diluted with HNO_3 plus ascorbic acid and loaded onto the TRU-Resin™ nitrate column. A calcium oxalate coprecipitation procedure was not performed. The decanted supernatant solutions were also wet ashed with HCl solution. These (A-supernatant, B-supernatant) were dissolved in HNO_3 plus ascorbic acid and loaded onto the TRU-Resin™ nitrate column. The percents recovery of the different fractions were: A-CaF_2 109±11% Am-241, 62±11% Np-237, 78±10% Pu-236, 25±4% U-238, 90±9% Th-232; B-CaF_2 97±10% Am-241, 83±12% Np-237, 96±11% Pu-236, 28±4% U-238, 95±9% Th-232; A-Supernatant 1±1% Am-241, 2±1% Np-237, 1±1% Pu-236, 40±6% U-238, 1±1% Th-232; and B-Supernatant 2±1% Am-241, 5±2% Np-237, 1±1% Pu-236, 48±7% U-238, 1±1% Th-232. (See Appendix B, Table CXI.)

b. Deionized water samples with Nd^{+3} and Ti^{+3} ions added.

Two samples were prepared with each containing approximately 25 dpm of Th-232, Np-237, U-238, Am-241, Pu-239, Th-229, Pu-236, U-232, and Am-243 dissolved in 30 ml of 3.0-M HCl. Next, 0.50 ml of 0.10 mg/dl of $Nd(NO_3)_3$ (7.0E-6 moles Nd^{+3}) and 2.0 ml of 1.25-M $Ca(NO_3)_2$ (2.5E-3 moles Ca^{+2}) were added followed by 5.0 ml of 29-M HF and 0.30 ml $TiCl_3$. The mixed samples sat for 20 min. and were then centrifuged. The supernatant was decanted and the solid was washed with 15-M HF solution and recentrifuged. The fluoride coprecipitate was slurried in HNO_3 plus HCl and was heated slowly in a glass beaker until the mixture was almost dry. This wet ashing procedure was repeated twice more with 5.0 ml of 5.0-M HCl solution. The wet ashed coprecipitated samples were dissolved in 5.0-M HCl plus H_2O_2 and loaded onto the TRU-Resin™ chloride column. A calcium oxalate coprecipitation procedure was not performed. The load eluate from the chloride column was wet ashed and diluted with HNO_3 plus ascorbic acid and loaded onto the TRU-Resin™ nitrate column. The average percents recovery of the dissolved fluoride coprecipitates were: 105±8% Am-241, 100±8 Am-243, 15±3% Np-237, 11±2% Pu-239, 10±2% Pu-236, 61±5% U-238, 57±4% U-232, 89±8% Th-232, and 93±8% Th-229. (See RB031094, Appendix B, Tables CXII and CXIII.) The $TiCl_3$ was added as a reductant for the UO_2^{+2} ion. The percent recovery of U was higher than in the previous experiment, however the recovery of Np and Pu was much lower.

CHAPTER VI

CONCLUSIONS

A. Overall Scheme

1. Sequential Separation Procedure for U, Th, Np, Pu, and Am.

The following is the proposed and tested sequential separation procedure for U, Th, Np, Pu, and Am ions that is the result of the experiments performed in this project. A flow chart of the separation procedure, Figure 18, follows the written description.

SEQUENTIAL SEPARATION PROCEDURE USING EIChroM TRU-Resin™ EXTRACTION RESIN

A. Required Supplies and Reagents

1. EiChrom TRU-Resin™ Resin (100-150 m particle size) 0.34 g/sample
2. 2 ml bed volume column, (Bio-Rad Poly-Prep chromatography columns, catalog #731-1550)
3. 12-M HCl (Conc.)
4. 8.0-M HCl (670 ml 12-M HCl/Liter)
5. 5.0-M HCl (420 ml 12-M HCl/Liter)
6. 1.30-M HCl (144 ml of standardized 9.00±0.05-M HCl/Liter)
7. 0.50-M HCl (42.0 ml 12-M HCl/Liter)
8. 0.50-M HCl plus 0.030 M $H_2C_2O_4$ (Oxalic Acid) (2.7 g $H_2C_2O_4$ per liter of 0.50-M HCl).
9. 0.10-M HCl (8.3 ml 12-M HCl/Liter)
10. 0.10-M$(NH_4)_2C_2O_4$ (AmmoniumOxalate)(14.2g $NH_4)_2C_2O_4$·HOH per liter deionized HOH)
11. 16-M HNO_3 (Conc.)
12. 4.0-M HNO_3 (250 ml 16-M HNO_3/liter)

13. 3.0-M HNO_3 (188 ml 16-M HNO_3/Liter)

14. Ascorbic Acid, solid powder

15. Reagents for Calcium Oxalate coprecipitation
    a. saturated CaC_2O_4 (10 g/ 100 ml)
    b. calcium carrier (50 mg/ml Ca^{+2} in 0.40-M HNO_3)

B. Column Preparation

1. For each sample soak 0.34 g TRU-Resin™ (regular particle size resin) in 8 ml of 0.50-M HCl for a minimum of 30 min.

2. Prepare empty column by rinsing it with 10 ml deionized HOH and 5 ml of 3.0-M HCl.

3. Load column with 0.34 g of HCl soaked TRU-Resin™ resin. Rinse column with 5 ml of 0.50-M HCl to settle resin beads. Place a plug of glass wool on top of the resin in the column.

4. Precondition column by adding 15 ml 5.0-M HCl. (Column is now ready for the sample.)

C. Actinide Separation

1. Dissolve digested and tracered sample in 5.0-M HCl. Use the minimum amount needed to completely dissolve the sample (approx. 15-30 ml). Warm if needed to dissolve.

2. After sample is dissolved and slightly cooled, add 7 drops H_2O_2 (30%). Swirl to mix. Let sample stand for minimum of 3 min. or maximum of 5 min. prior to loading onto column.

   * DO NOT heat the sample at this point. It is important that the H_2O_2 remains in the sample while it is being loaded onto the column.

3. Load Column with the above sample.
   (After the sample is loaded on the column, a few bubbles may appear in the liquid above the column. If this becomes a problem, tap the column to dislodge the bubbles. NOTE:If too much H_2O_2 is added, the bubbles can become a problem.) Note: flow rate approx. 0.5 ml/min

4. Wash column with 10 ml 5.0-M HCl.

   Collect the load eluate (from step C.3) and the 5.0-M HCl wash solution (from step C.4) together into one fraction to be used later in Step C.12 for a calcium oxalate precipitation. (Eluate contains Am^{+3}, possibly Cm^{+3}, and most M^{+3}, M^{+2}, and M^{+} matrix ions.)

5. Elute Th with 6.0 ml 1.30-M HCl. Save eluate.

6. Rinse with 3.0 ml 1.30-M HCl. Discard rinse eluate.

7. Elute Pu/Np with 15 ml of a solution which contains 0.030-M $H_2C_2O_4$ in 0.50-M HCl. Save eluate.

8. Elute U with 30 ml of 0.10-M $(NH_4)_2C_2O_4$. Save eluate.

9. Wash column with 10 ml 0.10-M HCl. (This eluate can be added to the U fraction.)

10. Add 3 ml of 0.10-M HCl to the column. Plug and cap column without allowing the solution to drain completely. Store the resin column with the HCl solution on the resin. The column will be used in the following Am separation.

11. Evaporate each of the last three eluted fractions (Th, Pu/Np, U) and proceed with NdF_3 precipitation procedures. (Or use any appropriate method to prepare the separated samples for alpha counting.)

12. On the first eluted fraction from step C.4.a, perform a calcium oxalate coprecipitation procedure.

13. Re-precondition the column by removing the plug and cap and allowing the HCl storage solution to drain. Then add 10 ml 4.0-M HNO_3 followed by 10 ml of 3.0-M HNO_3.

14. Dissolve the dried ashed residue from the calcium oxalate procedure in a small volume of 3.0-M HNO_3,(10-20 ml). Add a small amount of solid ascorbic acid (about 0.1 g). Stir gently. Allow sample to sit for about 10 min.

15. Load sample onto the column. Discard eluate.

16. Rinse column with 5 ml 3.0-M HNO_3. Discard eluate.

17. Add 1.0 ml 8.0-M HCl to the column. Discard eluate.

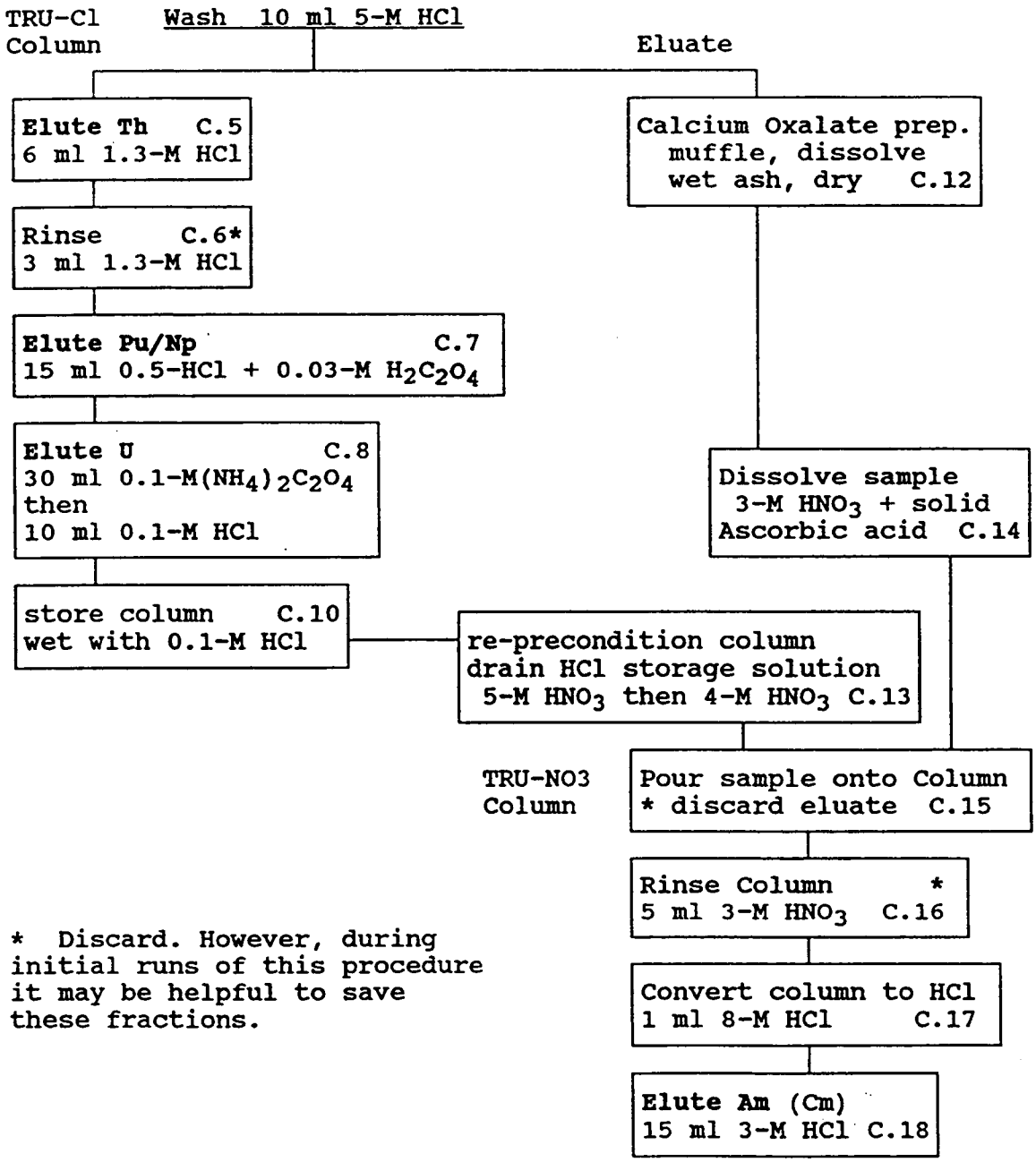
18. Elute Am from column with 15 ml 3.0-M HCl.
Note: If other +3 ions are present, a percentage of them will elute with this fraction (e.g. Cm).

19. Evaporate eluted Am fraction from step C.18 and proceed with NdF_3 precipitation procedures. (Or use any appropriate method to prepare the separated samples for alpha counting.)

TRU-Resin™ PROCEDURE Sequential Separation Flow Chart

Resin prep (step B.1-4): 34 g dry resin, soak 0.1-M HCl for 30 min., rinse column with deionized HOH and 1-M HCl, pour resin into column, rinse with 0.1-M HCl glass wool plug, precondition column 5.0-M HCl

Dried, digested, wet ashed sample, Add 5.0-M HCl plus H_2O_2
(DO NOT HEAT!) Pour onto column



* Discard. However, during initial runs of this procedure it may be helpful to save these fractions.

Figure 18: Tru-Spec Sequential Separation Procedure.

The selection of the steps for this separation process is the result of the following experimental observations:

Procedure Step C.1:

The samples are dissolved and loaded onto the TRU-Resin™ column in HCl solution. This allows low activity levels of Am ions to be separated from samples that contain high activity levels of U or Th ions. (See chapter V, sections C.6 and C.7.) The double pass through the column optimizes the amount of U and Th ions that can be separated.

Also, loading in HCl allows certain matrix ions, such as Fe^{+3} and Al^{+3}, to pass through the column with minimal attachment. This decreases interference due to matrix ions competing with the Np, Pu, Th, and U ions for attachment sites on the resin. (See chapter V, section D.7.) The presence of 1.0 grams of Al^{+3} ions and 0.10 grams of Fe^{+3} ions in the matrix did not affect the separation of the Am, Np, Pu, Th and U ions. Low levels of Am ions were successfully recovered from samples containing up to 10.0 g of Fe^{+3} ions.

Also, according to the literature, the Fe^{+3} needs to be reduced to Fe^{+2} with ascorbic acid prior to loading the TRU-Resin™ column with HNO_3.[36] Since the Fe^{+3} does not attach to the column when loading in HCl, a different reagent could be used to set the redox environment of the load solution. (See chapter V, sections C.8.a. and C.11.b.)

Procedure Step C.2:

Hydrogen peroxide, H_2O_2, was added to establish the redox environment of the load solution. The use of H_2O_2 established a solution environment in which the ions appeared to load and elute in the following oxidation states: UO_2^{+2}, Pu^{+4}, Np^{+4}, Th^{+4}. The Am^{+3} ions passed through the column when loaded in HCl. (See chapter IV, section D.8 and chapter V, sections C.8.a, C.11.b.) If the load solution is heated and the H_2O_2 decomposed, the load and elution patterns of some of the ions changed. For example, the Pu ions were detected in the same fraction as the Am^{+3} ions. (See chapter V, sections C.11.b and C.12.) H_2O_2 decomposes to water and oxygen when heated. This makes for easy clean-up of reducing agent in the eluted

fractions prior to the calcium oxalate coprecipitation procedure or the NdF_3 coprecipitation procedure.

Procedure Step C.5:

A decrease in the hydrogen ion concentration seemed to have the greatest impact on the elution of the Th ions. If the concentration of the Cl^- was reduced below 1.0-M but the H^+ ions were maintained higher then 2.0-M, very little Th activity could be detected in the eluate fractions. Initially the Th ions were eluted using 1.0-M HCl. This worked well for the Th ions but the Np and Pu ions would also partially elute in 1.0-M HCl. Additional tests showed that good recoveries of Th ions with minimal Np or Pu elution were possible with 6.0 ml of 1.30-M HCl, 8.0 ml of 1.40-M HCl, 10 ml of 1.50-M HCl, or 14 ml 1.0-M NaCl plus 1.0-M HCl. The 1.30-M HCl was incorporated into the separation procedure because of minimal volume requirements. (See chapter V, section C.9.)

Procedure Step C.6:

This rinse step is designed to remove any remaining Th ions from the column prior to the elution of the Np and Pu ions. In this fraction, less than 5% of Th, Np, or Pu ions was usually detected. The discarding of this fraction with a small amount of actinide ions was considered necessary to eliminate any contamination of the Th ions in the Np/Pu fraction or the Np/Pu ions in the Th fraction.

Procedure Step C.7:

This procedure is designed to elute the Np and Pu ions together. The alpha energies of these nuclides can be spectrally resolved.

The Np and Pu ions are eluted using 0.030-M $H_2C_2O_4$ in 0.50-M HCl solution. Possible eluting solutions that were tested included: 1.0-M HCl, 0.5-M HCl, 0.10-M NH_4I in HCl, 0.030-M $H_2C_2O_4$ in HCl, and 0.10-M NH_4I plus 0.030-M $H_2C_2O_4$ in HCl. The 0.50-M HCl eluted better than the 1.0-M HCl. It removed approximately all the Np ions and about 40% of the Pu ions from the column. The NH_4I worked well for removal of the Pu ions but it did not strip the Np ions from

the resin. The 0.030-M $H_2C_2O_4$ in HCl and the 0.10-M NH_4I plus 0.030-M $H_2C_2O_4$ in HCl resulted in good percents recovery for the Np and Pu ions. (See chapter V, section C.8.b.) However, the use of NH_4I in this step appeared to have a slight inhibitory effect on the elution of the U in the next fraction. (See chapter V, section 10.b.)

Procedure Step C.8:

The U ions are eluted using 0.10-M $(NH_4)_2C_2O_4$. The reagent suggested in the literature was ammonium hydrogen oxalate ($NH_4HC_2O_4$). Columns eluted with $NH_4HC_2O_4$ were subsequently eluted with $(NH_4)_2C_2O_4$ and additional U ions were recovered. (See chapter V, 10.a.) Hydrogen fluoride (HF) was also tested as a possible eluting reagent for U ions. HF did elute the U ions but did not appear to enhance the process and HF is a hazardous material.

Procedure Steps C.9,10,13:

The resin column is stored in dilute HCl and later it is preconditioned with HNO_3 and reused. Only a small amount, if any, actinide or matrix ions would still be attached after elution with 0.10-M HCl. Reuse of the column allows for minimal amount of resin usage.

Procedure Step C.12:

The calcium oxalate coprecipitation procedure is used for matrix clean-up of the sample prior to the separation of the Am ions. Am, Pu, and Th coprecipitated well with the calcium oxalate. Np only partially coprecipitated and U did not coprecipitate. (See chapter V, section D.4.) This procedure requires about one working day to perform (includes muffling time). It is not necessary to do a calcium oxalate procedure unless separation of Am is needed. The other four actinides (Np, Pu, Th, and U) were successfully separated from different matrices without using this additional clean-up step. (See chapter V, section D.5,6.) However in a sample containing 1.0 g Fe^{+3} ions, additional Pu and Th were recovered after the calcium oxalate coprecipitation. (See chapter V, section D.7.b.) In samples containing large amounts of Ca^{+2}, moderate recovery of Am resulted. This is probably due to the large amount of calcium oxalate precipitate that formed. (See chapter V, section D.7.a.) In samples containing large amount of Fe^{+3} (1.0 g, 10.0 g), the

recovery of Am was good following a calcium oxalate coprecipitation. (See chapter V, section 7.a,b.)

Procedure Step C.14:

The use of ascorbic acid was suggested in the literature to reduce Fe^{+3} to Fe^{+2}.[36] The addition of ascorbic acid seemed to improve the recovery of Am by about 20-30% from soil samples. (See chapter V, section D.2,3.)

Procedure Step C.17:

The addition of 8.0-M HCl rapidly converts the aqueous phase of the column to HCl. The Am is then eluted using 3.0-M HCl. These steps are suggested in the literature. [36]

Procedure Steps C.11,19

The eluted fractions were prepared for alpha counting by smoking with 0.73 M Na_2SO_4 in 18-M H_2SO_4. This removed any resin material that may have eluted in the actinide fractions. (See chapter V, section C.9.d.2.) The sample was then coprecipitated with NdF_3 and filtered. The filtration discs with the samples were then dried and alpha counted using surface barrier detectors.

Several tests were conducted to optimize the percents recovery of the actinide ions using the NdF_3 procedure. Some of the parameters that were investigated are: amount of Nd^{+3}, temperature variation of the pre- and post-precipitation solutions, agitation during precipitation, and different redox reagents in the precipitation solution. (See chapter V, sections B.1-7, C.4,5.)

The NdF_3 procedure used by IT is listed in chapter IV, section C. The NdF_3 procedure that was used in this project is listed below. The main differences are that in the procedure below all steps are performed at room temperature and that the same amount of Nd ions is added to all the actinide samples.

NdF_3 Coprecipitation Procedure:

1. Partially dry eluted fraction.
2. Add 2.0 ml 0.73 M Na_2SO_4 in 18-M H_2SO_4. Fume for 3 to 5 minutes.
3. Dilute to 15 ml with 3.0-M HCl.
4. For U only: Add 0.30 ml $TiCl_3$, stand 5 min.
5. Add 0.50 ml 0.10 mg/ml $Nd(NO_3)_3$ solution.
6. Add 5.0 ml 48% HF, let stand 20 min.

7. Preparation of polypropylene membrane: 5 ml ethanol then 5 ml carbon substrate (fine carbon suspension plus a small amount of NdF_3 precipitate), (use moderate vacuum)
8. Pour sample onto membrane, (use moderate vacuum)
9. Wash membrane: 5 ml 20% HF, then 5 ml deionized HOH, then 5 ml ethanol (use moderate vacuum)
10. Remove membrane, dry, mount, label, alpha count

2. Separation Procedure for Am ions.

A concern of IT was the length of time required to separate Am ions from a sample. The current procedure (methanol-HNO_3 elution) provided good results but required several days to complete. The following adapts the sequential separation procedure to a possible replacement for the Am-methanol-HNO_3 column procedure. This procedure would take approximately 1 to 1.5 working days to complete.

Note that the two column procedure is still maintained in order to prevent interference from other actinides, such as Pu as a +3 ion, high levels of U (up to approx. 8,000 dpm), and high levels of Th (up to approx. 1,000 dpm). (See chapter V, section C.6,7 and C.12.) If none of these interferants is present, the Am separation could be completed in one pass through the TRU-Resin™ column (Procedural steps C.13 - C.19). Note that the calcium oxalate coprecipitation is performed prior to the first load onto the column (TRU-Cl column). Then this load eluate from the TRU-Cl column can simply be wet ashed and loaded onto the reconditioned nitrate column (TRU-NO_3^- column) without repeating the calcium oxalate coprecipitation procedure.

Am SEPARATION PROCEDURE USING EIChroM TRU-Resin™ EXTRACTION RESIN

1. Proceed with current procedure through
    a. sample dissolution
    b. calcium oxalate coprecipitation procedure
    c. dissolution of calcium oxalate precipitate
2. Convert the sample from nitrate to chloride form.
    a. take to dryness
    b. add 5 ml of conc. HCl, wet ash
    c. add 5 ml of 5-M HCl, wet ash
3. Precondition column
    a. Steps B.1 -B.4
4. Load Sample and save eluate
    a. Steps C.1 - C.4

5. Strip column with 30-40 ml of 0.1 M $(NH_4)_2C_2O_4$. Discard.
   Then add 10 ml 0.10-M HCl to the column. Discard.
6. Re-precondition column
   a. Step C.13
7. Convert the saved eluate from chloride to nitrate from.
   a. evaporate sample to near-dryness
   b. wet ash with 5 ml of conc. HNO_3 and 1 ml H_2O_2
   c. repeat b.
   d. dissolve the sample in a small volume of 3.0-M HNO_3(15 ml).
   e. add solid ascorbic acid (about 0.1 g), dissolve, allow sample to sit for about 10 min.
   NOTE: Do not need to do the calcium oxalate procedure again.
8. Complete the remaining steps C.15 - C.19.

A flow chart of the Am separation procedure, Figure 19, follows on the next page.

B. Accomplishments

This research project has resulted in the development of a separation procedure that, when compared to a typical market procedure, requires smaller quantities of reagents, fewer types of reagents, fewer resin columns, fewer coprecipitation procedures, and fewer wet ashings (dry-downs). These changes have resulted in a procedure that requires less time, costs less and generates less waste. See Table II.

Some of the limitations of the 1.0 ml bed-volume column are: (1) The column capacity for U-238 was approximately 8,000 dpm and for Th-232 was approximately 1,000 dpm. (2) For a matrix containing Al^{+3} ions; good recoveries of the five actinide ions were observed for 0.01 g, 0.10 g, and 1.0 g samples but when 10.0 g of Al ions were in the solution, the actinide ions could not be separated cleanly. (3) For a matrix containing Ca^{+2} ions; good recoveries of the five actinide ions were observed for 0.01 g, 0.10 g, and 1.0 g samples. The Am recovery in the 1.0 g sample was 60% (20% lower than the blank) however this is a reportable level. When 10.0 g of Ca ions were in the solution, the Pu, Th, and U ions had good recoveries in the expected fractions but the Np and Am ions had poor recoveries. (4) For a matrix containing Fe_{+3} ions; good recovery of the five actinide ions was observed for 0.01 g and 0.10 g samples. The recoveries of Am and Np ions in the 1.0 g sample were good, however the recoveries of the Th, Pu, and U ions were between 20-35%. When 10.0 g of Fe ions were in solution, approximately 50% of the Pu and 100% of the Am were recovered in the expected fractions. About 60% of the Th was

TRU-Resin™ PROCEDURE Am Separation Flow Chart

Resin prep: 0.34 g dry resin, soak in 0.1-N HCl for 30 min.
rinse column with deionized HOH and 1-N HCl,
pour resin onto column, rinse with 0.1-N HCl,
plug with glass wool,
precondition column with 5-N HCl

Digested, wet ashed, dried sample
Calcium Oxalate prep.
muffle, dissolve, wet ash, dry

Add 5-M HCl plus H_2O_2 (DO NOT HEAT!)
Pour onto column

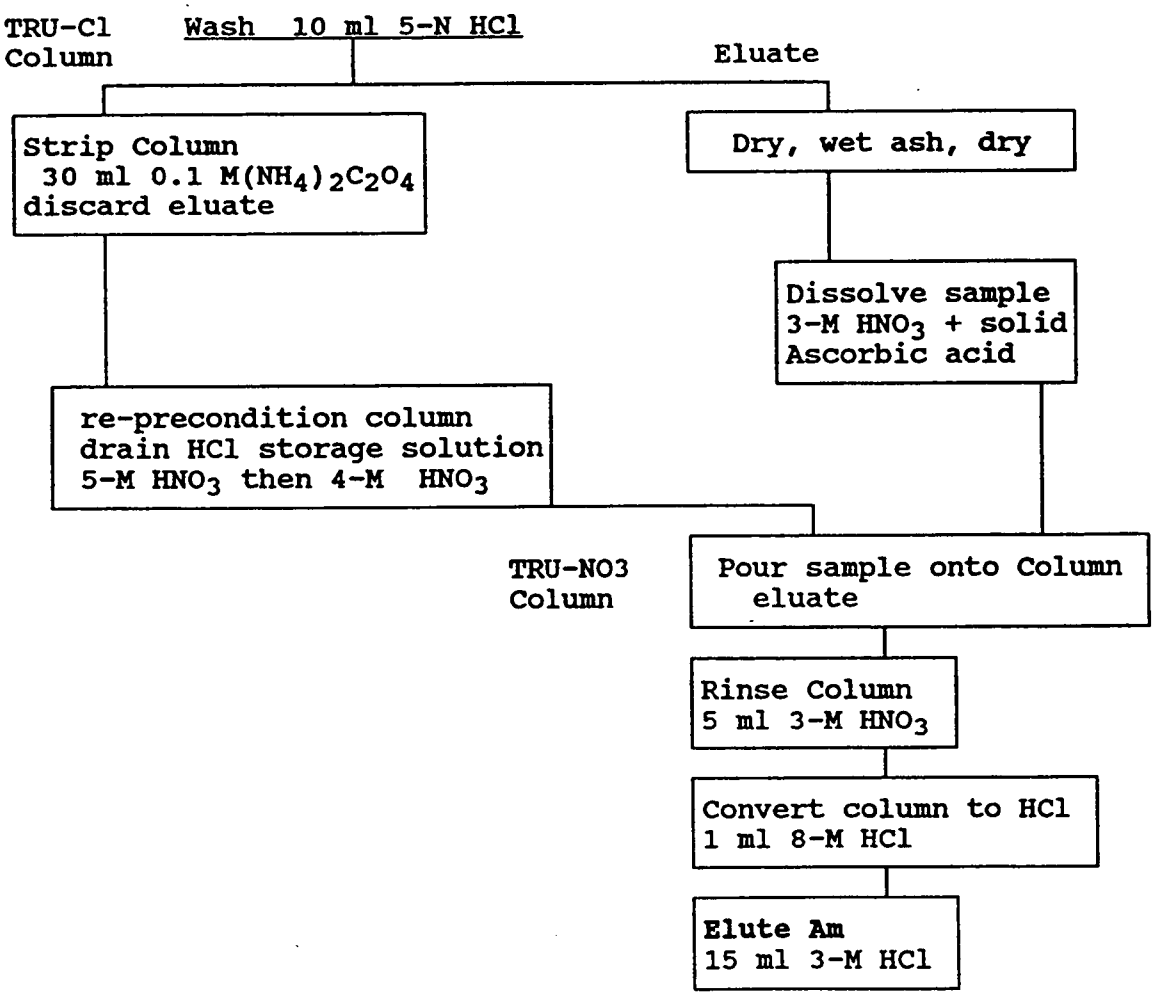


Figure 19: TRU-Resin™ Separation Procedure for Americium.

TABLE II

COMPARISON OF TYPICAL MARKET SEPARATION PROCEDURE AND TRU-Resin™ SEPARATION PROCEDURE

Typical Market Procedure	TRU-Resin™ Procedure
4 Resin columns AG1-X8(50-100 mesh) 10 ml AG1-X8(50-100 mesh) 10 ml AG1-X8(100-200 mesh) 8 ml AG1-X4(100-200 mesh) 5 ml	1 Resin column TRU-Resin™(100-150 m 1.0 ml
9 Redox or complexing reagents $NaNO_2$ $Fe(NH_4)_2(SO_4)_4$ H_3BO_3 $H_2NNH_2 \cdot H_2SO_4$ H_2O_2 NH_4I HF NH_4SCN	4 Redox or complexing reagents H_2O_2 $C_6H_8O_6$ (ascorbic acid) $H_2C_2O_4$ $(NH_4)_2C_2O_4$
HNO_3 8-M 900 ml HCl 12-M 175 ml 8-M 55 ml dilute 275 ml NH_4OH 30% 20 ml	HNO_3 8-M 50 ml dilute 40 ml HCl 8-M 1 ml dilute 100 ml NH_4OH 30% 10 ml
Organic reagents 300 ml methanol, methanol/HCl, and methanol/HCl/NH_4SCN	Organic reagents none
2 Coprecipitation procedures calcium oxalate iron hydroxide	1 Coprecipitation procedure calcium oxalate
9 Wet ashings(dry-downs)	3 Wet ashings(dry-downs)
Waste generated resin waste 45 g acid waste 480 ml base waste 50 ml organic waste 230 ml	Waste generated resin waste 0.34 g acid waste 200 ml
Time required 8 eight-hour-workdays	Time required 1.5 eight-hour-workdays

recovered, after a calcium oxalate coprecipitation, but the Np and U recoveries remained poor. (5) The flow rate of the column is approximately 0.5 ml/min, therefore large samples volumes could be a problem. (A 200 ml spiked deionized water sample required 5.5 hrs to load onto the column, yet the recoveries of the five actinides were very good.) (6) A difficulty of bubble in the column can develop due to the use of the H_2O_2 in the load solution. This can be minimized by using only small amounts of hydrogen peroxide. (7) A delicate point of the procedure is the separation between the Th and the Np/Pu fractions. High levels of Th, Np, and/or Pu in the sample may result in mixed recoveries.

C. Future Work

Listed below are items that may further improve the separation of the actinide ions but would require additional experimentation. These suggestions are the result of experimental observations made during this project.

1. If the prepared 2 ml bed volume TRU-Resin™ columns from EiChrom are used, the elution and rinse volumes will probably need to be doubled. Some testing will be needed to demonstrate this.
2. It also appears possible for the plutonium to be reduced to Pu^{+3} by heat decomposition of the peroxide complex just prior to loading the sample in step C.2. The Pu^{+3} would follow the Am^{+3} and not adhere to the resin when loaded in HCl. However, prior to loading onto the second (TRU-NO_3^-) column, the plutonium can be oxidized to Pu^{+4} and then it may be eluted after the Am by using the 0.10-M $H_2C_2O_4$ plus 0.50-M HCl reagent. (See chapter V, section 12.) This would allow individual separation of all 5 elements (Am, Np, Pu, Th, and U).
3. The elution of the Th nuclides was inhibited by the presenceof NH_4I in the eluting solution while Np and Pu nuclides eluted. (See chapter V, section C.9.d.1.) This may be used to change the elution order.

4. The continued elution of the TRU-NO_3^- column, after the elution of Am, may be useful to recovery additional Pu and Th ions if a sample has a complex matrix. (See chapter V, section D.7.)

5. The use of a Jones reductor (Zn metal) just prior to the TRU-Cl column load may reduce the oxidation states of Pu, Np and U in the load solution. This may be used to change the elution sequence and improve the percents recovery. (See chapter V, section C.11.b.)

6. When Cm^{+3} nuclides were added to the sample, they partially eluted with the Am^{+3} ions. (See chapter V, section C.12.)

7. Separation of other actinide elements by the TRU-Resin™ separation system, such as Ra, Ac, or Po ions, may be developed with further investigation.

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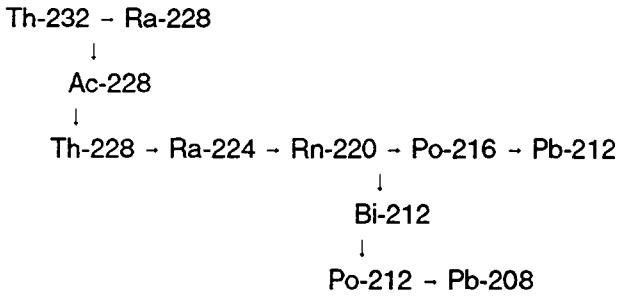
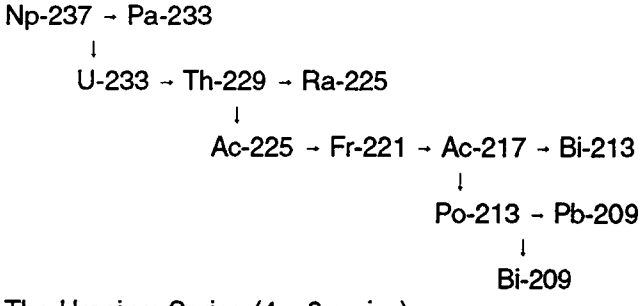
APPENDICES

APPENDIX A

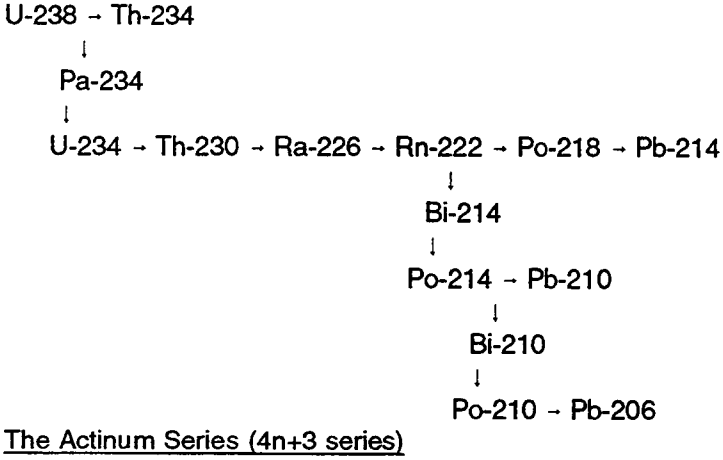
The Thorium Series (4n series)



The Neptunium Series (4n+1 series)



The Uranium Series (4n+2 series)



The Actinum Series (4n+3 series)

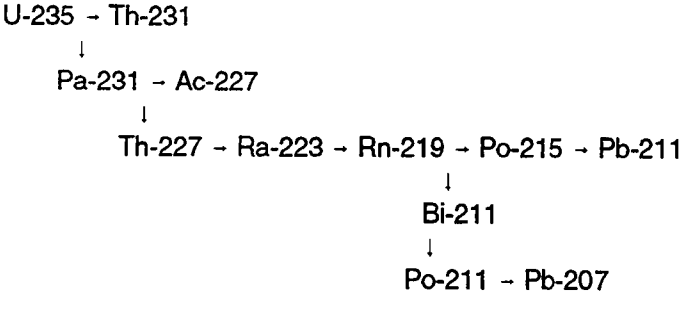


Figure 20: Decay Series. [KEY: → decay, ↓ beta decay]

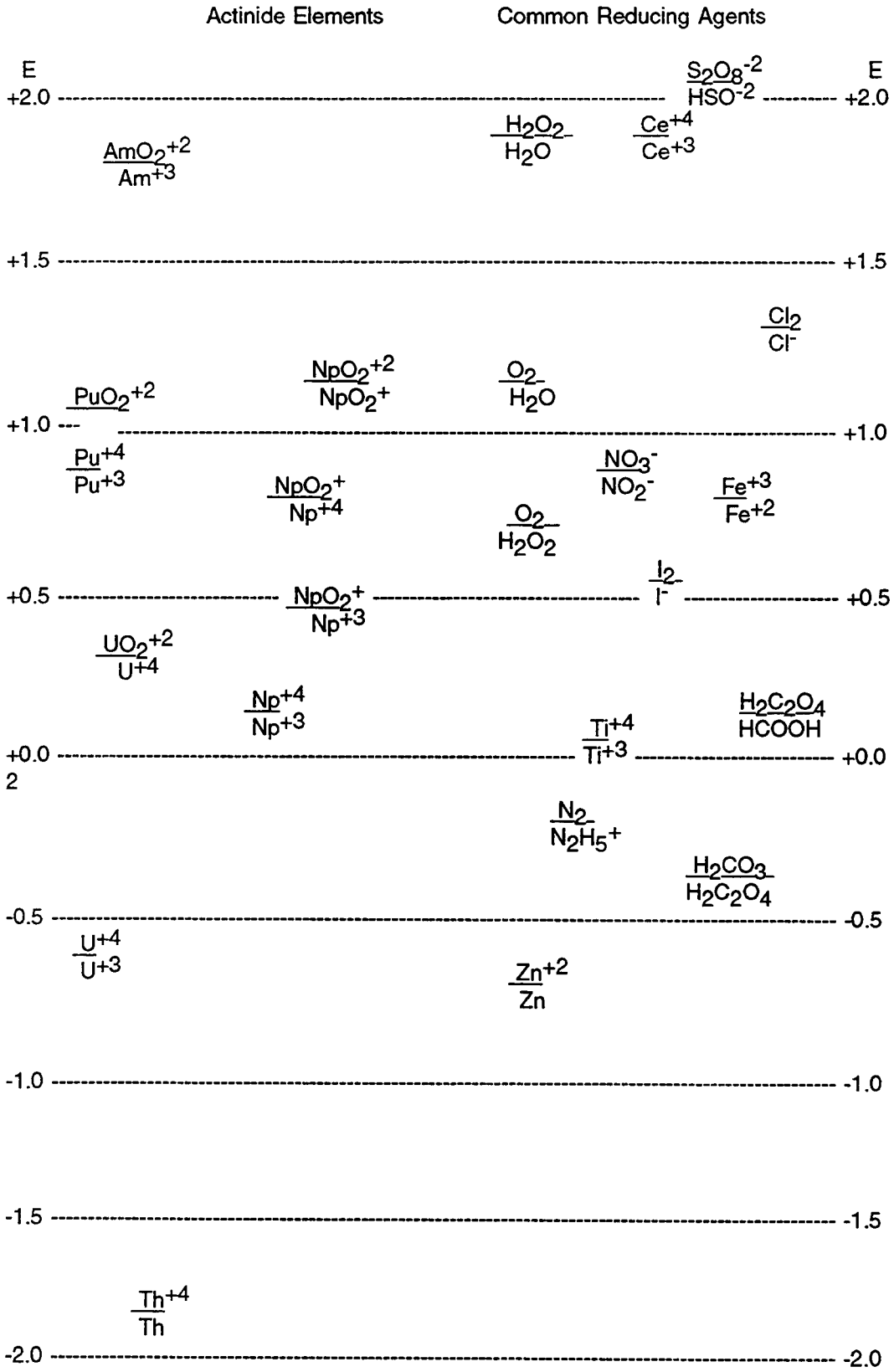
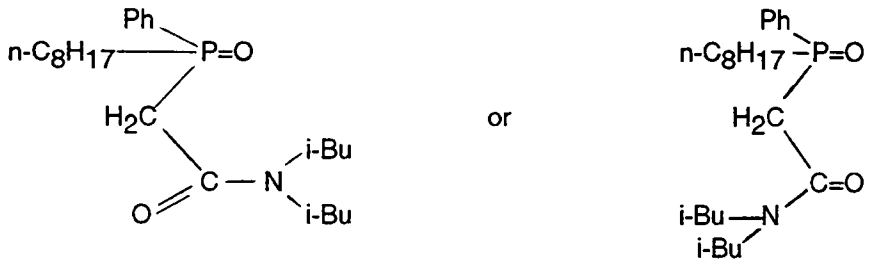
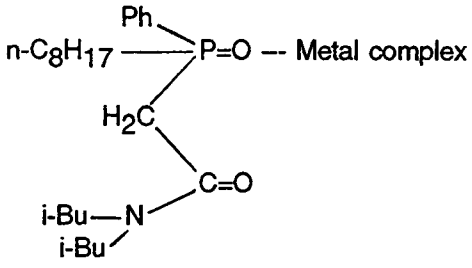


Figure 21: Comparison of Electromotive Potentials (volts) in 3-M H^+.

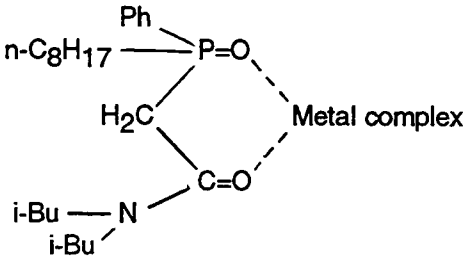
(a)



(b)



(c)



(d)

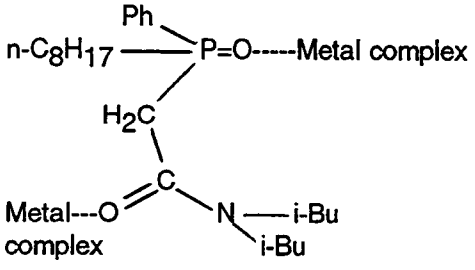


Figure 22: Structure of Octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine Oxide (CMPO): a) CMPO and (b-c) Projected attachment configurations of a metal complex with CMPO.[41,42]

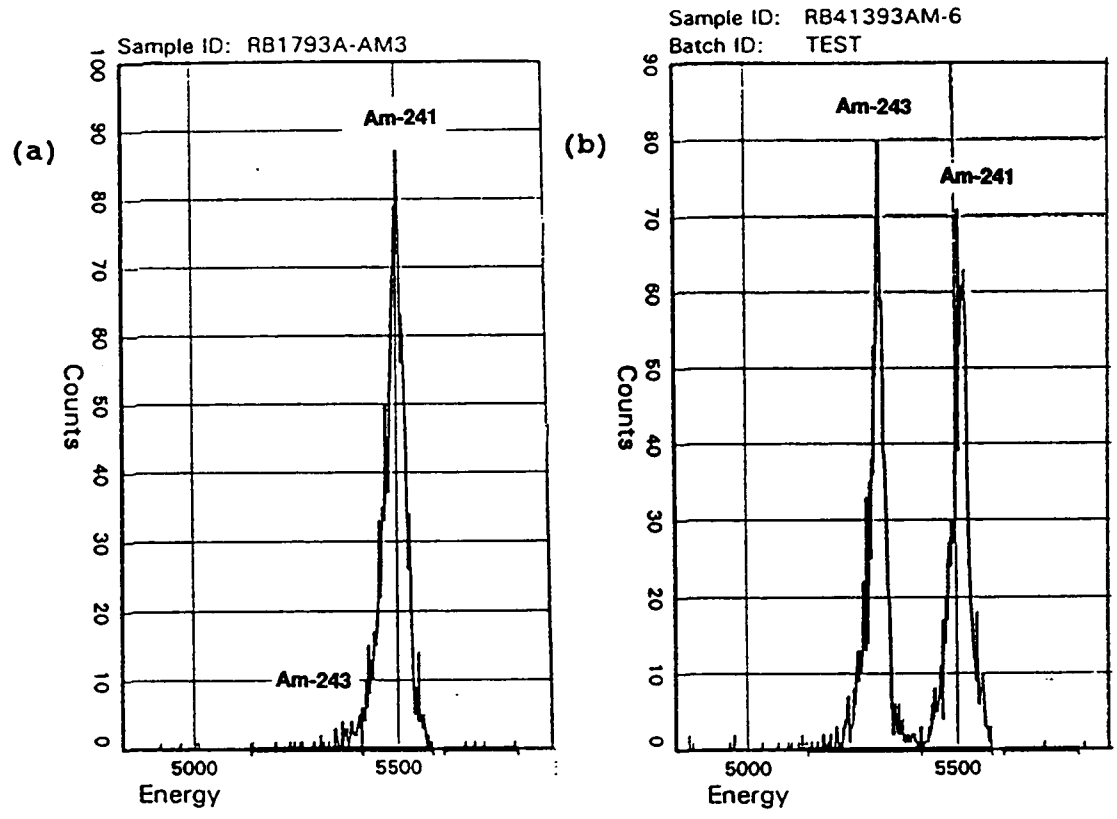


Figure 23: Computer Predetermined Regions-of-interest for Alpha Counting: (a) Spectrum of Am-241(using SSB),(b) Spectrum of Am-241 and Am-243 (using PIPS/ls).

APPENDIX B

TABLE III

COMPLEX FORMATION CONSTANTS

Thorium IV

Hydroxide ion: log $beta_1$ 9.6, log $beta_2$ 20, log $beta_3$ 28, log $beta_4$ 38

Nitrate ion: log $beta_1$ 0.7, log $beta_2$ 0.2

Fluoride ion: log $beta_1$ 8.4, log $beta_2$ 15, log $beta_3$ 20, log $beta_4$ 23

Chloride ion: log $beta_1$ 0.2, log $beta_2$ -1.0, log $beta_3$ -0.9, log $beta_4$ -1.5, log $beta_5$ -2.5

Sulfate ion: log $beta_1$ 3.2, log $beta_2$ 5.5

Phosphate ion: log MHL/M.HL 10.8

Uranium VI

Hydroxide ion: log $beta_1$ 9.6, log $beta_{12}$ 9.6, log $beta_{22}$ 21.7, log $beta_{43}$ 42.2, log $beta_{53}$ 52.6

Nitrate ion: log $beta_1$ -0.3

Fluoride ion: log $beta_1$ 4.3, log $beta_2$ 8.0, log $beta_3$ 10.6, log $beta_4$ 12

Chloride ion: log $beta_1$ -0.1

Sulfate ion: log $beta_1$ 3.0, log $beta_2$ 4.0, log $beta_3$ 3.7

Phosphate ion: log MHL/M.HL 7.2

Americium III

Hydroxide ion: log $beta_1$ 8.0

Nitrate ion: log $beta_1$ 0.3

Fluoride ion: log $beta_1$ 3.4

Chloride ion: log $beta_1$ -0.6, log $beta_2$ -0.4

Sulfate ion: log $beta_1$ 3.2, log $beta_2$ 5.1

Phosphate ion: log $MH_2L/M.H_2L$ 1.7

TABLE IV
ACTINIDE ALPHA ENERGIES

Main Alpha Lines keV	(% intensity)	Weighted Average Energies keV	
3953	Th-232 (23.0%)	3996	Th-232
4101	Th-232 (77.0%)		
4147	U-238 (23.0%)	4185	U-238
4196	U-238 (77.0%)		
4364	U-235 (11.0%)	4390	U-235
4396	U-235 (55.0%)		
4621	Th-230 (23.4%)	4672	Th-230
4688	Th-230 (76.3%)		
4724	U-234 (15.0%)	4767	U-234
4766	Np-237 (8.0%)		
4771	Np-237 (25.0%)	4782	Np-237
4776	U-234 (72.4%)		
4785	Ra-226 (94.5%)		
4788	Np-237 (47.0%)	4839	Th-229
4845	Th-229 (56.2%)		
4856	Pu-242 (22.4%)	4890	Pu-242
4901	Th-229 (10.2%)		
4901	Pu-242 (77.6%)		
5104	Pu-239 (11.5%)		
5114	Po-208 (100%)	5147	Pu-239
5143	Pu-239 (15.1%)		
5155	Pu-239 73.3%)	5270	Am-243
5234	Am-243 (10.6%)		
5264	U-232 (31.2%)	5302	U-232
5275	Am-243 (87.9%)		
5304	Po-210 (100%)		
5320	U-232 (68.6%)		
5340	Th-228 (26.7%)	5401	Th-228
5423	Th-228 (72.7%)		
5443	Am-241 (12.8%)	5480	Am-241
5457	Pu-238 (28.3%)		
5486	Am-241 (85.2%)	5487	Pu-238
5489	Rn-222 (100%)		
5499	Pu-238 (71.6%)		
5684	Ra-224 (95%)		
5722	Pu-236 (31.8%)	5754	Pu-236
5757	Th-227 (20.3%)		
5763	Cm-244 (23.6%)	5795	Cm-244
5770	Pu-236 (68.1%)		
5792	Ac-225 (27%)		
5805	Cm-244 (76.4%)		
5829	Ac-225 (52%)		
5977	Th-227 (23.4%)	5933	Th-227

TABLE IV (CONTINUED)

Main Alpha Lines keV	(% intensity)	Weighted Average Energies keV
6002	Po-218(100%)	
6038	Th-227 (24.5%)	
6051	Bi-212 (25%)	
6090	Bi-212 (10%)	
6125	Fr-221 (15%)	
6288	Rn-220 (100%)	
6339	Fr-221 (83%)	
6778	Po-216 (100%)	
7066	Ac-217 (100%)	
7687	Po-214 (100%)	

[7,8,51]

TABLE V

MOLARITY EQUIVALENT TO 20 dpm/25 ml

Nuclide	Activity	Half-life	Molarity (M^{n+})
U-238	0.80 dpm/ml	4.47E9 y	4.5E-6 M
U-232	0.80 dpm/ml	6.9E1 y	7.0E-14 M
Th-232	0.80 dpm/ml	1.4E10 y	1.4E-5 M
Th-229	0.80 dpm/ml	7.3E3 y	7.4E-12 M
Np-237	0.80 dpm/ml	2.14E6 y	2.2E-9 M
Pu-236	0.80 dpm/ml	2.87 y	2.9E-15 M
Pu-239	0.80 dpm/ml	2.41E4 y	2.4E-11 M
Am-241	0.80 dpm/ml	4.33E2 y	4.3E-13 M
Am-243	0.80 dpm/ml	7.4E3 y	7.5E-12 M

[1.01E-15(t) = M]

TABLE VI

REDUCTION EQUATIONS AND ELECTROMOTIVE POTENTIALS
FOR AQUEOUS SPECIES

Reduction Equations	Electromotive Potentials
$U^{+4} + e \rightarrow U^{+3}$	$E = -0.607+0.0591 \log[U^{+3}/U^{+4}]$
$UO_2^{+2} + 4H^+ + 2e \rightarrow U^{+4} + 2\ HOH$	$E = 0.333-0.1182pH+0.0295 \log[U^{+4}/UO_2^{+2}]$
$Th^{+4} + 4e \rightarrow Th$	$E = -1.899+0.0148 \log[Th^{+4}]$
$Np^{+4} + e \rightarrow Np^{+3}$	$E = 0.152+0.0591 \log[Np^{+3}/Np^{+4}]$
$NpO_2^+ + 4H^+ + 2e \rightarrow Np^{+3} + 2\ HOH$	$E = 0.451-0.1182pH+0.0295 \log[Np^{+3}/NpO_2^+]$
$NpO_2^+ + 4H^+ + e \rightarrow Np^{+4} + 2\ HOH$	$E = 0.749-0.2364pH+0.0591 \log[Np^{+4}/NpO_2^+]$
$NpO_2^{+2} + e \rightarrow NpO_2^+$	$E = 1.149+0.0591 \log[NpO_2^{+2}/NpO_2^+]$
$Pu^{+4} + e \rightarrow Pu^{+3}$	$E = 0.967+0.0591 \log[Pu^{+3}/Pu^{+4}]$
$PuO_2^{+2} + 4H^+ + 2e \rightarrow Pu^{+4} + 2\ HOH$	$E = 1.042-0.1182pH+0.0295 \log[Pu^{+4}/PuO_2^{+2}]$
$AmO_2^{+2} + 4H^+ + 3e \rightarrow Am^{+3} + 2\ HOH$	$E = 1.694-0.0788pH+0.0197 \log[Am^{+3}/AmO_2^{+2}]$
$2H_2CO_3 + 2H^+ + 2e \rightarrow H_2C_2O_4 + 2\ HOH$	$E^o = -0.39$
$H_2C_2O_4 + 2H^+ + 2e \rightarrow 2HCOOH$	$E^o = 0.25$
$N_2 + 5H^+ + 4e \rightarrow N_2H_5^+$	$E^o = -0.23$
$H_2O_2 + 2H^+ + 2e \rightarrow 2HOH$	$E^o = 1.763$
$O_2 + 2H^+ + 2e \rightarrow H_2O_2$	$E^o = 0.695$
$O_2 + 4H^+ + 4e \rightarrow 2HOH$	$E^o = 1.229$
$Cl_2 + 2e \rightarrow 2Cl^-$	$E^o = 1.37$
$I_2 + 2e \rightarrow 2I^-$	$E^o = 0.621$
$NO_3^- + 3H^+ + 2e \rightarrow HNO_2^- + HOH$	$E^o = 0.94$
$Zn^{+2} + 2e \rightarrow Zn$	$E^o = -0.763$
$Fe^{+3} + e \rightarrow Fe^{+2}$	$E^o = 0.771$
$TiO^{+2} + 2H^+ + e \rightarrow Ti^{+3} + HOH$	$E^o = 0.100$
$Ce^{+4} + e \rightarrow Ce^{+3}$	$E^o = 1.75$ (HCl 1.28, H_2SO_4 1.44, HNO_3 1.61)
$S_2O_8^{-2} + 2H^+ + 2e \rightarrow 2HSO_4^-$	$E^o = 2.08$

[18,68]

TABLE VII

REACTION RATES

Reaction	Reactant	Conditions solvent	temp.(°C)	Remarks
Np^{+3}-->Np^{+4}	O2	HClO4	RT	slow
	Fe^{+3}	HClO4	25	very rapid, catalyze by Cl-
	UO_2^{+2}	HClO4, HCl	25	very slow, partial
Np^{+4}-->Np^{+3}	U^{+4}	$HClO_4$, dil	25	rapid, partial
Np^{+3}-->NpO_2^+	H2O2	HClO4	25	rapid, transforms to Np^{+3} + NpO_2^+<-->2 Np^{+4}
Np^{+4}-->NpO_2^+	NO3-	HNO3,7M	25	very slow,(stable 9 days)
			100	slow, (stable 2 hrs)
NpO_2^+-->Np^{+4}	Fe^{+2}	$HClO_4$	25	slow and partial
	Fe^{+2}	H_2SO_4	25	very rapid
	I-	HCl 5M	25	complete in 40 min.
	H_2O_2	HNO_3 0.5-M	25	very slow
	H_2O_2	HNO_3 6-8 M	25	moderately rapid
	U^{+4}	HClO4, HCl	25	moderately rapid
	Pu^{+4}	HClO4	25	very slow
NpO_2^+-->NpO_2^{+2}	Ce^{+4}	HNO3	25	very rapid
NpO_2^{+2}-->NpO_2^+	H_2H_4	H^+ 1-M	RT	very rapid, then slow to Np^{+4}
	H2O2	HClO4	25	very rapid, then reduce to Np^{+4}
	Fe^{+2}	H^+	RT	very rapid, then reduce to Np^{+4}
	U^{+4}	$HClO_4$	25	rapid
	Pu^{+3}	$HClO_4$	25	rapid
Pu^{+3}-->Pu^{+4}	Ce^{+4}	$HClO_4$,0.5-M	RT	t1/2 = 5 sec.
	O_2	HCl 0.5-M	RT	Pu^{+3} stable at least 42 hrs
			100	2.5% oxidized in 4 hrs.
Pu^{+4}-->Pu^{+3}	Ascorbic acid	HNO_3 5M	RT	very rapid with H_2SO_4 unless react with HNO_3
	N_2H_4 0.2-M	HNO_3	25	slow, catalyze with Fe_{+2}
	H_2O_2	$HClO_4$	28	slow, forms brown peroxy complex
	I^- 0.1-M	HCl 0.4-M	RT	Rapid
	Fe^{+2}	H^+	RT	Rapid, catalyzed by Cl^-
	U^{+4}	$HClO_4$, HNO_3	25	Rapid
	Zn^o	HCl, 0.5-M	RT	Rapid

TABLE VII (CONTINUED)

Reaction	Reactant	Conditions solvent	temp.(°C)	Remarks
Pu^{+4}-->PuO_2^{+2}	Ce^{+4} 0.02-M	HNO_3 1-M	27	t1/2 = <60 min.
PuO_2^{+2}-->Pu^{+4}	$C_2O_4^{-2}$	HNO_3 5-M	50	complete in 45 min.
	Fe^{+2}	$HClO_4$, HCl	25	Rapid, PuO_2^{+} intermediate likely
	Zn^{o}	HCl 0.5-M	RT	Gives Pu^{+4} or Pu^{+3}
	I^- 2.3-M	HNO_3 0.5-M	--	Rapid, PuO_2^{+} intermediate likely
	U^{+4}	$HClO_4$	25	Rapid, PuO_2^{+} intermediate likely
PuO_2^{+2}-->PuO_2^{+}	I^-	pH 2	RT	very rapid
	Fe^{+2}	$HClO_4$, HCl	25	Rapid
	U^{+4}	$HClO_4$	25	Rapid
	N_2H_4	HCl 0.5-M	25	t = 3 hrs
PuO_2^{+}-->Pu^{+4}	N_2H_4	HCl 0.5-M	25	slow
	I^-	pH 2	RT	slow
	Fe^{+2}	HCl dilute	25	Rapid
	U^{+4}	$HClO_4$	25	slow
Am^{+3}-->AmO_2^{+2}	$S_2O_8^{-2}$	HNO_3 0.2-M	88	80% AmO_2^{+2} in 10 or 15 min. then slowly reduces to Am(V)
			TA	no detectable oxidation
	Ce^{+4}	$HClO_4$	--	partial
AmO_2^{+2}-->Am^{+3}	H_2O	H_2SO_4		RT rapid
	I^-	NH_4F 13-M	RT	intermediate
	Cl^-	NH_4F 13-M	RT	slow
AmO_2^{+}-->Am^{+3}	H_2O_2	H^+	25	complete and rapid
	Fe^{+2}	H^+	RT	very rapid

TABLE VIII

COPRECIPITATION OF ELEMENTS 93 TO 103 WITH VARIOUS CARRIERS

Carrier Compound	M^{+2}	M^{+3}	M^{+4}	MO_2^{+}	MO_2^{+2}
Lanthanum fluoride	NC	C	C	C	NC
Zirconium phosphate	-	NC	C	NC	NC
Bismuth phosphate	-	C	C	NC	NC
Thorium diphosphate	-	NC	C	-	NC
Thorium hypophosphate	-	-	C	-	NC
Lanthanum oxalate	-	C	C	-	NC
Thorium oxalate	-	C	C	-	NC
Uranium(IV) oxalate	-	C	C	-	NC
Bismuth oxalate	-	C	C	-	NC
Zirconium iodate	-	NC	C	-	NC
Thorium iodate	-	NC	C	-	NC
Cerium(IV) iodate	-	NC	C	-	NC
Zirconium phenylarsonate	-	-	C	P	NC
Bismuth arsonate	-	-	C	-	NC
Zirconium benzenesulfinate	-	-	C	NC	NC
Sodium uranyl acetate	-	-	NC	P	C
Thorium peroxide	-	NC	C	NC	NC
Uranium(IV) peroxide	-	NC	C	NC	NC
Barium sulfate	C	C	C	NC	NC
Potassium lithium sulfate	-	C	C	NC	NC
Insoluble hydroxides	P	-	C	C	C
Potassium uranyl carbonate	-	NC	NC	C	C

NC = coprecipitation less than a few percent under proper conditions;
C = coprecipitation nearly complete under proper conditions;
P = partial coprecipitation. [26]

TABLE IX

DISTRIBUTION COEFFICIENTS ON STRONG BASE ANION EXCHANGE RESIN AT VARIOUS ACID CONCENTRATIONS

	Nitric Acid, M					Hydrochloric acid, M					
ION	2	4	6	8	10	2	4	6	8	10	12
Th^{+4}	15	50	200	300	200		not absorbed				
Pa(V)	2	8	10	10	4		1	20	300		4000
U^{+4}								1	30	100	
UO_2^{+2}	0.1	3	7	8	4	8		300			1000
Np^{+4}	300	600	1800	2500	2500	1	1	1		100	
NpO_2^{+2}						40	250	1000			
Pu^{+3}		not absorbed				not absorbed					
Pu^{+4}		700	2100	3500		1	1		100		
PuO_2^{+2}						40	250	1000		4000	
Am^{+3}		not absorbed				not absorbed					
Cm^{+3}		not absorbed									

[29]

TABLE X

YIELD TRACERS OF SELECTED ACTINIDES

Environmental Important Isotopes	Yield Tracer Isotopes	Comments
Thorium		
232	Th-229	- emitted alpha energy close to Th-228
230		- can have slight Th-228 contamination
228	Th-234	- beta emitter, separate assay needed
227		- readily available
		- daughter of U-238
Uranium		
238	U-232	- half-life 72 years
234		- good alpha energy separation
235		- decay daughter Th-228,close alpha energy
	U-230	- short half-life, 20.8 days
	U-233	- alpha energy overlap with U-234
	U-236	- alpha energy overlap with U-235
Neptunium		
237	Np-239	- beta or gamma decay, no alpha
		- short half-life, 2.36 days
		- ready supply, decay daughter of Am-243
	Np-238	- beta decay
		- short half-life, 2.1 days
		- difficult to obtain
	Pu-236	- alpha emitter
		- different element, different chemistry
	Np-235	- longer half-life, 396 days
		- electron capture decay, measure uranium L x-rays using Si(Li) detector
		- possible Np-237 contamination
Plutonium		
238	Pu-236	- half-life 2.85 yrs.
239		- alpha energy reasonable well separated
240		from Pu-238, overlap if spectra degrades
	Pu-242	- long half-life, 3.87E5 yrs.
		- high purity, moderate cost
		- alpha energy separation good
Americium		
241	Am-243	- alpha decay
		- possible overlap due to spectral degradation
		- decay daughter is Pu-239

[30]

TABLE XI

NUCLIDES USED FOR YIELD TRACERS AND/OR SPIKES

ID# (nuclide)	Activity* (dpm/ml)	Assay Date (mm-dd-yy)	Matrix	Half-life*	Supplier (NWC1990)
Th2-186-A6(Th232)	3.0E1	05-01-79	1N HNO_3	1.4E10y	NBS (N1A)
Th-07D(Th232)	8.7E1	05-01-79	1N HNO_3	1.4E10y	NBS (N1A)
Th-07E(Th232)	3.0E1	05-01-79	1N HNO_3	1.4E10y	NBS (N1A)
Th-07F(Th232)	3.0E1	05-01-79	1N HNO_3	1.4E10y	NBS (N1A)
Th-08B(Th229)	4.2E1	05-07-84	0.5N HNO_3	7.3E3y	NIST (4328)
Th-09D(Th229)	4.0E1	05-07-84	1N HNO_3	7.3E3y	NIST (4328)
Th-09F(Th229)	4.0E1	05-07-84	1N HNO_3	7.3E3y	NIST (4328)
Th-09E(Th229)	2.3E1	05-07-84	1N HNO_3	7.3E3y	NIST (4328)
Th-10(Th232)	8.3E2	05-01-79	1N HNO_3	1.4E10y	NBS (N1A)
U-10(U238)	2.4E2	05-01-89	2N HNO_3	4.47E9y	EPA(1843-1)
U-11(U238)	4.8E1	05-01-89	2N HNO_3	4.47E9y	EPA(1843-1)
U-12(U238)	4.8E1	05-15-89	1N HNO_3	4.47E9y	EPA(1842-1)
U2-088-A1(U232)	2.9E1	02-09-84	1N HNO_3	6.9E1y	EPA(946-1)
U-05AA3(U232)	4.9E1	01-28-86	**	6.9E1y	EPA
U8-282-A(U232)	4.8E1	05-01-86	1N HNO_3	6.9E1y	EPA
Np-01B(Np237)	9.5E1	12-01-80	0.1N HNO_3	2.14E6y	Amer.(P28568)
Np-020-A3(Np237)	9.3E1	12-01-80	0.1N HNO_3	2.14E6y	Amer.(P28568)
Pu-13D(Pu239)	8.3E1	12-13-90	1N HNO_3	2.41E4y	Amersham
Pu-19A(Pu236)	3.4E1	06-17-92	2N HNO_3	2.87y	Amer.(AE5758)
Pu-19B(Pu236)	3.3E1	06-17-92	2N HNO_3	2.87y	Amer.(AE5758)
Pu-236A1(Pu239)	2.9E1	12-13-90	1N HNO_3	2.41E4y	EPA (2510-1)
Pu-9-236A2(Pu239)	2.9E1	12-13-90	1N HNO_3	2.41E4y	EPA (2510-1)
Pu-22(Pu242)	4.5E1	12-18-89	1N HNO_3	3.73E5y	NIST(92-242511)
(Pu-12(Pu241)	1.0E4	07-01-86	1N HNO_3	1.44E1y)	Amersham
Pu-18(Pu241)	3.6E1	12-18-89	1N HNO_3	1.44E1y	Amersham
Am-06A(Am241)	2.6E2	02-27-92	0.5N HCl	4.33E2y	EPA(2461-2)
Am-06D(Am241)	1.0E2	02-27-92	1N HCl	4.33E2y	EPA(2461-2)
Am-07(Am241)	1.3E3	02-27-92	1N HCl	4.33E2y	EPA(2461-2)
Am-08A(Am243)	5.0E2	07-11-90	1N HNO_3	7.4E3y	NIST(4332C)
Am-09(Am243)	3.8E1	07-11-90	1N HNO_3	7.4E3y	NIST (4332C)
Cm-02(Cm244)	3.2E2	04-01-89	1N HNO_3	1.81E1y	NIST (SRM4320)

* uncertainty in last digit
** H_2SO_4, K_2SO_4, $Pb(NO_3)_2$

EPA - U. S. Environmental Protection Agency, Environmental Monitoring Systems Laboratory, Las Vegas, NV.
NBS - U. S. Department of Commerce, National Bureau of Standards, Washington, D.C.
NIST - U. S. Department of Commerce, National Institute of Standards & Technology, Gaithersbure, MD.
Amer. or Amersham - Amersham Corporation, Arlington Heights, IL.

TABLE XII

REACTIONS WITH H_2O_2

Reaction	Comments
* Oxidize U^{+4}:	$U^{+4} + H_2O_2$ ----> UO_2^{+2} (a)
* Reduce NpO_2^{+2}:	$NpO_2^{+2} + H_2O_2$ <---> NpO_2^{+} inhibited by presence of NpO_2^{+} (a) favored by an increase in $[H^+]$ in HCl (b) instantaneously, H_2O_2 in 0.5-M HNO_3, 25°C (f,h)
* Reduce NpO_2^{+}:	$NpO_2^{+} + H_2O_2$ ---> Np^{+4}, as $[H^+]$ increases in HNO_3, rate increases (c) complete in 1-2 min, H_2O_2, 0.5-M HNO_3, 100°C (f) in 0.5-M HNO_3, 25°C, very slow (h) in 6-8 M HNO_3, 20°C, moderately rapid (h)
* Oxidize Pu^{+3}:	$Pu^{+3} + H_2O_2$ -----> Pu^{+4} (a) in $HClO_4$, 28°C, slow, forms proxyl complex (h)
* Reduce Pu^{+4}:	$Pu^{+4} + H_2O_2$ -----> Pu^{+3} (a)
* Forms peroxy complexes with Pu^{+4}:	$Pu^{+4} + H_2O_2$ -----> [Pu^{+4}--HOOH]complex (a) 2 Pu^{+4} + 2 H_2O_2 <---> $[Pu(O_2)_2]^{+4}$ + $4H^+$ (red complex) (g) 2 Pu^{+4} + H_2O_2 + HOH <---> $[PuO_2PuOH]^{+5}$ + $3H^+$ (brn complex) (e,g) reaction proceeds rapidly to the right, complex decomposes to Pu^{+3} (approx 1 %/min at room temp), Decomposition rate = K_2[brown complex]$[H^+]$ = 1.4E-4/Msec, rate increases slightly with temp. and/or $[H^+]$ increase. (e)
* Stabilize Pu^{+4}:	H_2O_2 worked well (b)
* Reduce AmO_2^{+2}:	2 AmO_2^{+2} + H_2O_2 ---> 2 AmO_2^{+} + $2H^+$ + O_2 (a,h) $AmO_2^{+2} + H_2O_2$ ----> Am^{+3}, rapidly (d)
* Reduce AmO_2^{+}:	$AmO_2^{+} + H_2O_2$ ----> Am^{+3}, warmed, rapidly (d) in acid, 25°C, complete and rapid (h)

a. [33]
b. [69]
c. [70]
d. [20]
e. [71]
f. [2]
g. [61]
h. [26]

TABLE XIII

RESOLUTION OF SPLIT FILTERS EXPERIMENT

Isotopes	R_S*	Visually distinct
U-238/Am-241	4.5	yes
U-234/Am-241	3.0	yes
Np-237/Am-241	1.3	yes
Pu-239/Am-241	1.3	yes
Th-232/Am-241	6.8	yes
Th-230/Am-241	4.5	yes
Th-228/Am-241	0.7	no
Th-232/U-238	1.0	no
Th-232/U-235	2.3	yes
Th-232/U-234	3.1	yes
Th-230/U-238	2.3	yes
Th-230/U-235	1.3	yes
Th-230/U-234	0.8	no
Th-228/U-238	5.5	yes
Th-228/U-235	5.7	yes
Th-228/U-234	2.0	yes
Np-237/Th-232	3.0	yes
Np-237/Th-230	0.8	no
NP-237/Th-228	2.1	yes
Np-237/U-238	2.2	yes
Np-237/U-234	0.1	no
NP-237/Pu-239	1.2	yes
Pu-239/Th-232	5.0	yes
Pu-239/Th-230	2.7	yes
Pu-239/Th-228	1.3	yes
Pu-239/U-238	3.5	yes
Pu-239/U-234	1.5	yes

* $R_S = 2\ \Delta Z / (W_a + W_b)$
If the $R_S \geq 1.2$, the peaks were resolved to baseline sufficiently for an activity of each isotope to be determined with minimal error.

TABLE XIV

PRECISION CHECK OF SURFACE BARRIER DETECTOR SYSTEM
REPEATED COUNTS USING SAME DETECTOR

Counter number	Counter type	Am-241 C(net)	FWHM	Counter efficiency(±1.2%)
32	SSB	822	63.03	0.1948
32	SSB	764	57.68	0.1948
32	SSB	766	51.76	0.1948
32	SSB	805	65.08	0.1948
32	SSB	783	52.82	0.1948
32	SSB	823	45.47	0.1948
32	SSB	793	55.01	0.1948

Mean = 793 counts Standard deviation(s) = ± 25 counts
NTE deviation = ± 40 counts Reliability factor (RF) = s / √mean = 0.86

TABLE XV

PRECISION CHECK OF SURFACE BARRIER DETECTORS
MULTIPLE DETECTOR USAGE

Counter number	Counter type	Am-241 C(net)	FWHM	Counter efficiency	C(net) / efficiency
32	SSB	793	55.01	0.1948	4071
17	SSB	761	51.72	0.1739	4376
16	SSB	1149	70.39	0.2557	4494
15	SSB	1052	54.97	0.2529	4161
37	PIPS	1379	56.80	0.3053	4516
40	PIPS	1355	42.24	0.3008	4503
50	PIPS	1374	43.73	0.3192	4304
49	PIPS	1300	40.20	0.2913	4463

Mean of total counts (c_{net}/eff) = 4361 counts Standard deviation(s) = ± 169 counts
NTE deviation = ± 278 counts
Reliability Factor (RF) = s / √mean = 2.6
RF for SSB only = 3.0 RF for PIPS only = 1.5

TABLE XVI

PERCENTAGE DEGRADATION OF Am-241 INTO Am-243 COMPUTER PREDETERMINED REGION-OF-INTEREST

Am-241 range C(net)	Am-243 range C(net)	Percentage degraded	Visually Accepted	Counter Type
409	13	3.2	yes	SSB
404	28	6.9	yes	SSB
1685	58	3.5	yes	SSB
1263	256	20.3	no	SSB
803	19	2.4	yes	SSB
749	18	2.5	yes	SSB
738	28	3.8	yes	SSB
770	35	4.6	yes	SSB
752	31	4.2	yes	SSB
794	29	3.7	yes	SSB
760	33	4.4	yes	SSB
1353	26	2.0	yes	PIPS
1318	37	2.9	yes	PIPS
1333	41	3.1	yes	PIPS
1255	45	3.6	yes	PIPS

Average percentage degradation of visually accepted spectra:
overall = 3.9%
for SSB = 4.0%
for PIPS = 2.9%

TABLE XVII

VARIATION IN THE AMOUNT OF $Nd(NO_3)_3$ ADDED
EXPERIMENT RB101492

$Nd(NO_3)_3$ Added (mg)	Counts (total) ($1.65 \pm \sqrt{c}$)	Counter type,#	Counter efficiency (±0.01)	C(net) (eff) (±6%)	% Am recovery (±8%)	Visually Resolved (Am-241)
A. In 15 ml 3-M HCl solution						
0.01	1029	PIPS(35)	0.30	3428	76	yes
0.02	1199	PIPS(36)	0.31	3866	84	yes
0.05	1065	PIPS(37)	0.31	3435	75	yes
0.10	1228	PIPS(38)	0.26	4723	103	yes
0.15	2167*	PIPS(39)	0.32	3386	73	yes
0.20	1188	PIPS(40)	0.30	3960	86	yes
0.25	1072	PIPS(41)	0.27	3967	86	yes
0.01	1303	PIPS(42)	0.31	4203	92	yes
0.05	1138	PIPS(43)	0.31	3671	80	yes
0.10	1267	PIPS(44)	0.32	3959	86	yes
0.15	contaminated spectrum					no
0.20	1335	PIPS(46)	0.29	4603	100	yes
B. In 2.0 ml 0.73-M Na_2SO_4/18-M H_2SO_4 + 13 ml 3-M HCl solution						
0.01	743	PIPS(47)	0.32	2322	51	yes
0.05	1138	PIPS(48)	0.31	3671	80	yes
0.10	1193	PIPS(49)	0.30	3977	87	poor
0.15	1231	PIPS(50)	0.31	3971	86	poor
0.20	1263	PIPS(51)	0.32	3947	86	yes

* sample was spiked twice

RB101492
Count time: 120 min.

Am-241 (Am-06A)(2.55E2 ± 0.10 dpm/ml)
100% yield = 4590 ± 180 counts

TABLE XVIII

VARIATION IN THE AMOUNT OF $Nd(NO_3)_3$ ADDED
EXPERIMENT RB112592A1

$Nd(NO_3)_3$ Added (mg)	Counts (total) ($1.65 \pm \sqrt{c}$)	Counter type,#	Counter efficiency (± 0.01)	C(net) (eff) ($\pm 6\%$)	% Am recovery ($\pm 8\%$)	Visually Resolved (Am-241)
A. In 15 ml 3-M HCl solution						
0.001	235	SSB(17)	0.17	1382	31	yes
0.003	filter damaged					no
0.010	308	SSB(19)	0.17	1806	40	poor
0.030	594	SSB(20)	0.17	3476	77	yes
0.10	767	SSB(21)	0.19	4026	89	yes
0.30	679	SSB(23)	0.18	3767	83	yes
0.50	685	SSB(24)	0.18	3800	84	yes
1.00	587	SSB(25)	0.18	3256	72	poor
2.00	318	SSB(27)	0.17	1859	41	poor
3.00	unresolvable					no
B. In 2.0 ml 0.73-M Na_2SO_4/18-M H_2SO_4 + 13 ml 3-M HCl solution						
0.001	171	SSB(29)	0.19	895	20	yes
0.003	195	SSB(31)	0.20	970	21	poor
0.010	405	SSB(32)	0.19	2116	47	yes
0.030	575	SSB(17)	0.17	3382	75	yes
0.10	721	SSB(18)	0.18	4006	89	yes
0.30	568	SSB(19)	0.17	3335	74	poor
0.50	665	SSB(20)	0.17	3894	86	yes
1.00	317	SSB(21)	0.19	1658	37	poor
2.00	unresolvable					no
3.00	unresolvable					no

RB112592A1
count time 400 min.

Th-232 (Th-07D) (87.02 ± 1.0 dpm/ml)
100% yield = 4525 ± 91 counts

TABLE XIX

VARIATION IN THE AMOUNT OF $Nd(NO_3)_3$ ADDED
EXPERIMENT RB112592A2

$Nd(NO_3)_3$ Added (mg)	Counts (total) ($1.65 \pm \sqrt{c}$)	Counter type,#	Counter efficiency (±0.01)	C(net) (eff) (±6%)	% Am recovery (±8%)	Visually Resolved (Am-241)
A. In 15 ml 3-M HCl solution						
0.030	141	SSB(21)	0.175	806	71	yes
0.10	178	SSB(23)	0.189	942	83	yes
0.30	176	SSB(24)	0.164	1073	95	yes
0.50	173	SSB(25)	0.171	1012	89	yes
B. In 2.0 ml 0.73-M Na_2SO_4/18-M H_2SO_4 + 13 ml 3-M HCl solution						
0.030	148	SSB(27)	0.188	787	70	yes
0.10	162	SSB(28)	0.186	871	77	yes
0.30	154	SSB(29)	0.178	865	76	yes
0.50	175	SSB(31)	0.183	956	85	yes

RB112592A2
count time 100 min.

Th-232 (Th-07D) (87.02 ± 1.0 dpm/ml)
100% yield = 1131 ± 23 counts

TABLE XX

PRE-WATER BATH AND POST-WATER BATH TEMPERATURE VARIATION
EXPERIMENT RB102192

Counts (total) ($\pm 1.65\sqrt{c}$)	Counter PIPS	Counter Efficiency (± 0.01)	C(net) (eff) ($\pm 7\%$)	%Am Recovery ($\pm 8\%$) pre	Temperature Baths, °C post	
1036	45	0.32	3228	70	85	0
841	46	0.29	2896	63	85	0
904	47	0.32	2822	63	85	0
908	48	0.31	2929	64	85	0
874	46	0.29	3003	65	85	0
904	47	0.32	2822	61	85	0
957	48	0.31	3087	67	85	0
1018	33	0.28	3632	79	85	0
903	37	0.31	2906	63	85	0
847	48	0.31	2732	59	85	0
989	33	0.28	3529	77	22	22
973	37	0.31	3132	68	22	22
826	44	0.32	2578	56	22	22
960	45	0.32	3000	65	22	22
975	33	0.28	3479	76	22	22
998	37	0.31	3212	70	22	22
893	44	0.32	2787	61	22	22
996	45	0.32	3113	68	22	22
846	46	0.29	2906	63	22	22
912	47	0.32	2847	62	22	22

RB102192
count time: 400 ± 1 min.
(sulfate fume)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4592 ± 230 counts

TABLE XXI

PRE-WATER BATH AND POST-WATER BATH TEMPERATURE VARIATION
EXPERIMENT RB112092

Counts (total) (±1.65√c)	Counter #,SSB	Counter Efficiency (±0.01)	C(net) (eff) (±8%)	%Am Recovery (±9%)	Temp Baths, °C pre	post	Precipitation Time (min)
632	20	0.17	3718	91	85	0	15
692	21	0.19	3642	89	85	0	15
495	23	0.18	2750	67	85	0	15
623	24	0.18	3461	85	85	0	15
501	25	0.18	2783	68	85	0	15
	(sum C(net)/eff: 16354 ± 591 counts, sum % yield 400 ± 21)						
575	27	0.17	3382	83	22	22	15
635	28	0.18	3578	86	22	22	15
677	29	0.19	3563	87	22	22	15
603	31	0.20	3015	74	22	22	15
579	32	0.19	3047	75	22	22	15
	(sum C(net)/eff: 16585 ± 597 counts, sum % yield 405 ± 21)						
903	1	0.24	3763	92	22	22	30
894	2	0.26	3438	84	22	22	30
861	3	0.24	3588	88	22	22	30
864	4	0.23	3757	92	22	22	30
873	5	0.25	3492	86	22	22	30
	(sum C(net)/eff: 18038 ± 648 counts, sum % yield 442 ± 21)						
893	6	0.22	4059	99	85	22	15
872	7	0.26	3354	82	85	22	15
813	8	0.23	3535	87	85	22	15
797	9	0.22	3623	89	85	22	15
734	10	0.26	2823	69	85	22	15
	(sum C(net)/eff: 17394 ± 628 counts, sum % yield 426 ± 21)						

RB112092
count time: 200 ± 1 min.
(sulfate fume)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4082 ± 205 counts

TABLE XXII

PRE-WATER BATH AND POST-WATER BATH TEMPERATURE VARIATION
EXPERIMENT DL061493

Counts (total) (±1.65√c)	Counter #,PIPS/Is	Counter Efficiency (±2%)	C(net) (eff) (±8%)	%Th Recovery (±9%)	Temperature Baths, °C pre	post
599	17	0.207	2894	70	0	0
626	18	0.218	2879	69	0	0
934	21	0.242	3867	93	90	90
872	22	0.196	4457	107	90	90
661	23	0.214	3092	75	21	21
652	24	0.209	3115	75	21	21

DL061493
count time: 200 ± 1 min.
Assisted by D. Larkins

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
(no sulfate fume, 15 min in baths)

TABLE XXIII

PRE-WATER BATH AND POST-WATER BATH TEMPERATURE VARIATION
EXPERIMENT DL062593

Counts (total) (±1.65√c)	Counter #,PIPS/Is	Counter Efficiency (±2%)	C(net) (eff) (±8%)	%Th Recovery (±9%)	Temperature Baths, °C pre	post
575	1	0.1726	3331	100	90	90
591	2	0.1827	3235	97	90	90
630	3	0.1802	3496	105	90	90
673	4	0.1890	3561	107	90	90
620	5	0.1807	3431	103	90	90
638	6	0.1622	3933	118	90	90

DL062593
count time: 200 ± 1 min.
Assisted by D. Larkins

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 3324 ± 200 counts
(no sulfate fume, 15 min in baths)

TABLE XXIV

PRE-WATER BATH AND POST-WATER BATH TEMPERATURE VARIATION
EXPERIMENT DL072393

Counts (total) ($\pm 1.65\sqrt{c}$)	Counter #,PIPS/Is	Counter Efficiency ($\pm$2%)	C(net) (eff) ($\pm$8%)	%Th Recovery ($\pm$9%)	Temperature Baths, °C pre	post
587	1	0.1712	3429	83	90	0
582	2	0.1721	3382	81	90	0
521	3	0.1794	2904	70	90	0
766	4	0.1945	3938	95	90	90
814	5	0.1765	4612	111	90	90
707	6	0.1569	4506	108	90	90

DL072393
count time: 200 $\pm$ 1 min.
Assisted by D. Larkins

Th-232 (Th-10) 8.31E2 $\pm$ 0.10 dpm/ml
100% yield: 4155 $\pm$ 208 counts
(no sulfate fume, 15 min in baths)

TABLE XXV

PRE-WATER BATH AND POST-WATER BATH TEMPERATURE VARIATION
EXPERIMENT RB012894

Th-232/Th-229 Count(total) (±1.65√c)	Detector #,PIPS/Is	Counter Efficiency (±2%)	Th-232/Th-229 Recovery (8%)	Temp. Baths, °C pre	post	Na_2SO_4 added *
935/727	01	0.161	117/109	80	80	yes
718/602	02	0.166	87/ 87	80	80	yes
694/534	03	0.170	82/ 76	80	80	no
780/610	04	0.187	84/ 78	80	80	no
738/588	05	0.180	83/ 79	22	22	yes
735/638	07	0.188	79/ 82	22	22	yes
630/476	08	0.169	75/ 67	22	22	no
547/470	09	0.170	64/ 67	22	22	no
718/523	10	0.172	84/ 73	0	0	yes
653/543	11	0.173	76/ 76	0	0	yes
520/409	12	0.182	57/ 54	0	0	no
473/368	13	0.162	59/ 55	0	0	no
814/634	15	0.181	90/ 84	80	0	yes
814/631	16	0.175	93/ 87	80	0	yes
791/608	17	0.200	79/ 73	80	0	no
767/660	18	0.211	73/ 75	80	0	no

(*all samples heated for 10 minutes with or without sulfate)

RB012894
count time: 200 ± 1 min.
(pre bath 10 min, post bath 15 min)

Th-229 (Th-08B) 4.15E1 ± 0.10 dpm/ml
100% yield: 4152 ± 125 counts
Th-232 (Th-10D) 8.315E1 ± 0.10 dpm/ml
100% yield: 4986 ± 150 counts

TABLE XXVI

PRE-WATER BATH AND POST-WATER BATH TEMPERATURE VARIATION
EXPERIMENT RB012293

Nuclide	Counts (total) (±1.65√c)	Counter Efficiency (±2%)	C(net) (eff) (±9%)	Percent Recovery (±10%)	Temperature Baths, °C pre	post
Th-232	640	0.175	3660	87	85	0
Th-232	558	0.189	2953	71	85	0
Th-232	910	0.164	3357	80	85	0
Th-232	615	0.171	3576	85	85	22
Th-232	613	0.188	3249	78	85	22
Th-232	530	0.176	2845	68	85	22
Th-232	427	0.178	2367	57	22	0
Th-232	440	0.183	2393	57	22	0
Th-232	524	0.195	2675	64	22	0
Th-232	648	0.243	2671	64	22	22
Th-232	638	0.246	2596	62	22	22
Th-232	594	0.223	2646	63	22	22
Np-237	118	0.218	529	14	85	0
Np-237	8	0.234	35	1	85	0
Np-237	61	0.247	223	6	85	0
Np-237	65	0.236	263	7	85	22
Np-237	23	0.249	89	2	85	22
Np-237	22	0.266	60	2	85	22
Np-237	765	0.240	3184	84	22	0
Np-237	773	0.235	3265	86	22	0
Np-237	866	0.253	3420	90	22	0
Np-237	801	0.256	3097	82	22	22
Np-237	597	0.174	3388	89	22	22
Np-237	633	0.182	3400	89	22	22
Pu-239	823	0.218	3762	90	85	0
Pu-239	644	0.234	2740	66	85	0
Pu-239	910	0.247	3654	88	85	0
Pu-239	788	0.236	3309	80	85	22
Pu-239	866	0.249	3468	83	85	22
Pu-239	834	0.266	3109	75	85	22
Pu-239	862	0.240	3580	86	22	0
Pu-239	814	0.235	3435	83	22	0
Pu-239	914	0.253	3611	87	22	0
Pu-239	786	0.256	3039	73	22	22
Pu-239	682	0.174	3894	94	22	22
Pu-239	667	0.182	3637	87	22	22

RB012293
count time: 200 ± 1 min.
(no sulfate fume)
(precipitation time 20 min)
detectors: SSB

Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 125 counts
Np-237 (Np-01B) 9.505E1 ± 0.10 dpm/ml
100% yield: 3800 ± 115 counts
Th-232 (Th-07E) 2.99E1 ± 0.10 dpm/ml
100% yield: 4185 ± 168 counts

TABLE XXVII

PRE-WATER BATH AND POST-WATER BATH TEMPERATURE VARIATION
EXPERIMENT RB012694

Np/Pu239/Pu236 Count(total) ($\pm 1.65\sqrt{c}$)	Counter #,SSB	Counter Efficiency ($\pm 2\%$)	Np/Pu239/Pu236 Recovery ($\pm 7\%/\pm 7\%/\pm 12\%$)	Temperature Baths, °C pre	post
2310/2010/380	23	0.230	88/ 88/ 92	80	80
2141/1791/318	24	0.200	94/ 90/ 88	80	80
2162/1826/343	25	0.205	93/ 89/ 93	80	0
2362/2133/360	26	0.218	95/ 98/ 92	80	0
2569/2296/390	27	0.212	107/109/103	0	0
2623/2267/382	28	0.224	103/101/ 95	0	0

RB012694
count time: 399 ± 1 min.
(sulfate fume)
(pre bath 10 min, post bath 15 min)

Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 9959 ± 299 counts
Np-237 (Np-01B) 9.505E1 ± 0.10 dpm/ml
100% yield: 11377 ± 342 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed):1791 ± 90 counts
(half-life = 2.87 yrs)

TABLE XXVIII

PRE-WATER BATH AND POST-WATER BATH TEMPERATURE VARIATION
EXPERIMENT DL061693

U-238 Count(total) (±1.65√c)	U-232 Count(total) (±1.65√c)	Counter Efficiency (±2%)	U-238 Recovery (±9%)	U-232 Recovery (±9%)	Temperature Baths, °C pre	Temperature Baths, °C post
1054	905	0.183	85	73	0	0
1206	1037	0.201	89	76	0	0
1132	904	0.192	87	69	87	87
854	743	0.178	71	61	87	87
1091	947	0.189	85	74	21	21

Assisted by D. Larkins
count time: 399 ± 1 min.
(no sulfate fume)
(15 min. in baths)

U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 6762 ± 203 counts
U-232 (U-05AA2) 4.9E1 ± 0.10 dpm/ml
100% yield(decayed): 6809 ± 206 counts
(half-life = 69 yrs.)

TABLE XXIX

PRECIPITATION TIME VARIANCE
EXPERIMENT RB010793C

Nuclide Added	Time (min)	Counts (total) (±1.65√c)	Counter Efficiency (±2%)	C(net) (eff) (±7)	%Am recovery (±9)
Am-241	15	1092	0.266	4071	80
Am-241	60	1055	0.240	4384	86
Am-241	185	1043	0.235	4400	86
Am-241	265	1091	0.226	4810	94
Am-241	400	1092	0.253	4314	85

RB010793C
count time: 200 ± 1 min.
detectors: SSB

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 5100 ± 215 counts

TABLE XXX

PRECIPITATION TIME VARIANCE
EXPERIMENT RB011593

Nuclides added	Temp (°C)	Time (min)	Counts (total) (±1.65√c)	Counter Efficiency (±2%)	C(net) (eff) (±8%)	%Am Recovery (±9%)	%Np Recovery (±9%)
Am/Np	22	15	921/844*	0.246	3747/3429	92	90
Am/Np	22	60	771/784	0.241	3198/3247	78	85
Am/Np	22	180	780/749	0.223	3491/3344	86	88
Am/Np	85	15	843/757	0.229	3688/3312	90	87
Am/Np	85	60	966/823*	0.218	4437/3776	109	99
Am/Np	85	180	844/814	0.234	3601/3469	88	91
Am/Th	22	15	679/623	0.174	3880/3572	95	92
Am/Th	22	60	621/582	0.182	3409/3191	84	82
Am/Th	22	180	651/606	0.165	3930/3654	96	94
Am/Th	85	15	648/628	0.176	3674/3566	90	92
Am/Th	85	60	681/601	0.175	3896/3432	95	88
Am/Th	85	180	672/668	0.189	3550/3514	87	91
Am/Pu	22	15	643/685	0.164	3899/4160	96	105
Am/Pu	22	60	628/646	0.171	3652/3774	89	96
Am/Pu	22	180	578/619	0.188	3063/3286	75	83
Am/Pu	85	15	611/559	0.186	3263/2995	80	76
Am/Pu	85	60	608/633	0.178	3393/3544	83	90
Am/Pu	85	180	617/585*	0.183	3354/3191	82	81

* poorly resolved spectrum

RB011593
count time: 200 ± 1 min.
detectors: SSB

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4082 ± 204 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 115 counts
Th-229 (Th-09E) 2.28E1 ± 0.10 dpm/ml
100% yield: 3876 ± 194 counts
Pu-241 (Pu-18) 3.59E1 ± 0.10 dpm/ml
100% yield: 3950 ± 158 counts

TABLE XXXI

AGITATION DURING COPRECIPITANT FORMATION
EXPERIMENT DL062393

Nuclide	Count(tot) (±1.65√c)	Counter Efficiency (±2%)	C(net) eff. (8%)	Percent Recovery (9%)	Agitation Time(min)
Am-241	649	0.1802	3602	94	15
Am-241	648	0.1890	3429	90	15
Am-241	648	0.1807	3586	94	15
Am-241	712	0.1622	4390	115	0
Am-241	697	0.2072	3364	88	0
Am-241	701	0.1811	3871	101	0
U-238	539	0.1834	2939	87	15
U-238	526	0.2005	2623	78	15
U-238	564	0.1920	2938	87	15
U-238	570	0.1777	3208	95	0
U-238	569	0.1894	3004	89	0
U-238	451	0.1858	2427	72	0

count time: 200 ± 1 min., Set up by D. Larkins, detectors: PIPS/ls
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml, 100% yield: 3360 ± 101 counts
Am-241 (Am-06A) 2.55E1 ± 0.10 dpm/ml, 100% yield: 3826 ± 192 counts

TABLE XXXII

ADDITIONAL NdF_3 COATING ON FILTER MEMBRANE PRIOR TO FILTRATION
EXPERIMENT RB033193

Am-241 Counts (total) (±1.65√c)	Degraded Counts (Am-243 area)	Counter PIPS/ls*	Counter Efficiency (±2%)	C(net) (eff) Am/degraded (±5%)/(±12%)	% Am Recovery Am/degraded (±7%)/(±13%)
A. Regular procedure, Nd and HF added to sample					
1755	24	15	0.186	9435/129	92/ 1
1730	22	16	0.188	9202/117	90/ 1
B. Filter coated with additional NdF_3, no Nd added to sample, HF added to sample					
1704	204	17	0.206	8272/990	81/10
1372	216	18	0.225	6098/960	60/ 9

*PIPS detectors/lowered shelf arrangement, count time: 400 ± 1 min.
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml, 100% yield: 10204 ± 510 counts

TABLE XXXIII

REPRECIPITATION OF THE FILTERED SOLUTION
EXPERIMENT RB022793

	Counts (total) (±1.65√c)	Counter SSB# (±2%)	Counter Efficiency (±7%)	C(net) (eff) (±8%)	%Am Recovery
Sample #1	790	13	0.235	3329	82
Reprecipitate #1	2	15	0.253	4	0
Sample #2	900	16	0.256	3504	86
Reprecipitate #2	10	17	0.174	34	1

RB022793
count time: 200 ± 1 min.

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4082 ± 205 counts

TABLE XXXIV

SULFATE AS PART OF NdF_3 PRECIPITATING MATRIX
EXPERIMENT RB102392

Na_2SO_4 Added/ min.fumed	Counts (total) (1.65±√c)	Counter type,#	Counter efficiency (±0.01)	C(net) (eff) (±7%)	%Am R0ecovery (±9%)
none/none	871	PIPS/45	0.32	2722	59
none/none	830	PIPS/33	0.28	2961	64
2.0ml/none	903	PIPS/33	0.28	3221	70
2.0ml/none	887	PIPS/46	0.29	3048	66
2.0ml/10 min	859	PIPS/37	0.31	2765	60
2.0ml/10 min	924	PIPS/36	0.31	2971	65
2.0ml/40 min	957	PIPS/32	0.32	2988	65
2.0ml/40 min	912	PIPS/44	0.32	2847	62

RB102392
count time: 120 min.

Am-241 (Am-06A) (2.55E2±0.10 dpm/ml)
100% yield = 4592 ± 230 dpm

TABLE XXXV

SULFATE AS PART OF NdF_3 PRECIPITATING MATRIX
EXPERIMENT RB120292

Na_2SO_4 Added/ min.fumed	Counts (total) ($1.65 \pm \sqrt{c}$)	Counter Type,#	Counter Efficiency (±2%)	C(net) (eff) (±8%)	% Am Recovery (±9%)
2.0ml/none	819	SSB/17	0.174	4667	91
2.0ml/none	845	SSB/18	0.182	4615	90
2.0ml/none	743	SSB/19	0.165	4467	88
2.0ml/none	802	SSB/20	0.176	4523	87
2.0ml/10 min	819	SSB/21	0.175	4651	91
2.0ml/10 min	891	SSB/23	0.189	4698	92
2.0ml/10 min	810	SSB/24	0.164	4915	96
2.0ml/10 min	743	SSB/25	0.171	4316	85
2.0ml/10 min	799	SSB/27	0.188	4239	83
2.0ml/10 min	829	SSB/17	0.174	4563	89
2.0ml/10 min	798	SSB/18	0.182	4363	86
2.0ml/10 min	768	SSB/19	0.165	4642	91

RB120292
count time: 200 min.

Am-241 (Am-06A) (2.55E2 ± 0.10 dpm/ml)
100% yield = 5102 ± 255 dpm

TABLE XXXVI

SULFATE AS PART OF NdF_3 PRECIPITATING MATRIX
EXPERIMENT RB010793B

Na_2SO_4 Added/ min.fumed	Counts (total) ($1.65 \pm \sqrt{c}$)	Counter type,#	Counter efficiency (±2%)	C(net) (eff) (±6%)	% Am Recovery (±7%)
none/none	1074	SSB/1	0.243	4420	87
none/none	1128	SSB/2	0.246	4555	89
none/none	1009	SSB/3	0.241	4183	82
2.0 ml/none	1031	SSB/4	0.223	4582	90
2.0 ml/none	957	SSB/5	0.229	4179	82
2.0 ml/none	1109	SSB/6	0.218	5069	99
2.0 ml/15 min	899	SSB/7	0.234	3842	75
2.0 ml/15 min	1152	SSB/8	0.247	4619	91
2.0 ml/15 min	1055	SSB/9	0.236	4451	87

RB010793B
count time: 200 min.

Am-241 (Am-06A) (2.55E2±0.10 dpm/ml)
100% yield = 5102 ± 255 dpm

TABLE XXXVII

BLANKS

Count Date	Count Time(min)	Detector # / type	Detector Efficiency	Total Count	Background count*
10-26-92	120	48 / PIPS	0.31	1±2	1±1
10-28-92	120	44 / PIPS	0.32	0	0
11-12-92	120	33 / PIPS	0.28	2±3	1±1
11-16-92	200	1 / SSB	0.24	72±15	16±3
12-01-92	399	25 / SSB	0.18	20±8	18±5
01-13-93	200	10 / SSB	0.249	0	1±1
01-19-93	200	20 / SSB	0.176	1±2	6±2
01-20-93	200	1 / SSB	0.243	0	1±1
07-07-93	200	11 / PIPS/ls	0.183	4±4	6±2
07-19-93	200	4 / PIPS/ls	0.189	0	1±1
02-01-94	200	19 / PIPS/ls	0.217	8±5	6±2
02-01-94	200	21 / PIPS/ls	0.223	7±5	6±2

* 1000 min. counts

TABLE XXXVIII

ZIRCONIUM PHENYLARSONATE COPRECIPITATION TESTS
EXPERIMENT RB051493

$Nd(NO_3)_3$ added (ml)	$ZrO(NO_3)_2$ added (ml)	Phenyl arsonate added (ml)	Am-241 Counts* (total) ($\pm 1.65\sqrt{c}$)	Th-232 Counts (total) ($\pm 1.65\sqrt{c}$)	Detector PIPS/ls Effeciency ($\pm 2\%$)	Am/Th 241/232 Recovery %
0.10	0.10	0.10	0	1	0.183	0 / 0
0.10	0.10	0.10	4	1	0.183	0 / 0
0.10	0.10	1.0	6	17	0.183	0 / 0
0.10	0.10	1.0	4	29	0.183	0 / 0
0.10	0.30	0.30	5	6	0.183	0 / 0
0.10	0.30	0.30	10	15	0.183	0 / 0
0.10	0.30	1.0	unable to count due to excessive particulate matter on the filter			
0.10	0.30	1.0				
0.10	1.0	1.0	unable to count due to excessive particulate matter on the filter			
0.10	1.0	1.0				
0	0.30	1.0	unable to count due to excessive particulate matter on the filter			
0	0.30	1.0				

*estimated due to alpha energy spectral overlap with Th-228

RB051493
count time: 200 ± 1 min.
(22°C)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 5102 ± 256 counts
Th-232 (Th-07E) 2.99E1 ± 0.10 dpm/ml
100% yield: 2990 ± 150 counts
Th-229 (Th-09D) 4.0E1 ± 0.10 dpm/ml
100% yield: 2808 ± 141 counts

Solutions used:
8.74 mg/ml phenylarsonic acid ($C_6H_5AsO_3H_2$) (0.0433-M)
1.02 mg/ml $Nd(NO_3)_3$ (0.00309-M)
2.54 mg/ml $ZrO(NO_3)_2$ (0.0110-M)
(final pH of solution mixtures between 0.1 and 0.3)

General Procedure:
-combined standards, diluted to 15.0 ml volume with 0.50-M HCl
-added Nd and /or Zr
-added phenylarsonate ion
-precoated filters with 3 ml of slightly cloudy Zr phenylarsonate mixture wash ETOH
-pour samples, rinse 0.5 M HCl, HOH, ETOH
-mount, count

TABLE XXXIX

ZIRCONIUM PHENYLARSONATE COPRECIPITATION TESTS
EXPERIMENT DL072093

Test #	Counts (total) ($\pm 1.65\sqrt{c}$)	Detector #, PIPS/Is	Counter efficiency ($\pm 2\%$)	C(net) (eff) (± 8)	% Th Recovery (± 9)
1A	766	1	0.173	4438	107
1B	668	2	0.183	3650	88
2A	677	3	0.180	3761	91
2B	774	4	0.189	4095	99
3A	717	5	0.181	3961	95
3B	733	6	0.162	4525	109

DL072093
count time: 200 ± 1 min.
Assisted by D. Larkins

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
Am-241 (Am-06A) 2.55E1 ± 0.10 dpm/ml
100% yield: 3827 ± 192 counts

Solution used:
samples #1(A,B) used 0.5 ml of 0.1 mg/ml $ZrO(NO_3)_2$ (0.00043 M)
samples #2(A,B) used 1.5 ml of 0.1 mg/ml $ZrO(NO_3)_2$
samples #3(A,B) used 0.5 ml of 1.0 mg/ml $ZrO(NO_3)_2$ (0.0045 M)
In all samples, 1.0 ml of 8.74 mg/ml phenylarsonic acid (0.0433M)

The filtered coprecipitated zirconium phenylarsonate showed not alpha radioactivity when counted on the PIPS detectors.
Above are the results from the NdF_3 coprecipitated in the remaining solutions after the zirconium phenylarsonate had been filtered.
The Am-241 alpha count was unable to be determined due to energy overlap with Th-228 but Am-241 was significantly present in all the spectra of the filtered solutions.

TABLE XL

IODATE COPRECIPITATION TESTS
EXPERIMENT DL070193

Sample #	Am-241 Counts (total) ($\pm 1.65\sqrt{c}$)	Am-243 Counts (total) ($\pm 1.65\sqrt{c}$)
#1, #2, #3, #4	2, 3, 3, 1	2, 2, 2, 1

Assisted by D. Larkins, count time: 200 ± 1 min.
Am-241 (Am-06A) 2.55E1 ± 0.10 dpm/ml, 100% yield: 3827 ± 192 counts
Am-243 (Am-08A) 4.97E1 ± 0.10 dpm/ml, 100% yield: 3973 ± 199 counts

TABLE XLI

COPRECIPITATION TESTS USING Y-Ca-Mg MIXED-FLUORIDES
EXPERIMENT RB112592B

Coppt Cation	Am-241 Counts (total) ($\pm 1.65\sqrt{c}$)	U-238 Counts (total) ($\pm 1.65\sqrt{c}$)	Counter #,SSB	Counterr Efficiency (±0.1)	%Am Recovery (±9%)
Nd^{+3} *	1204	0	2	0.26	91
	1100	0	5	0.25	89
Y^{+3}, Ca^{+2},Mg^{+2}**	858	0	3	0.24	70
	819	0	6	0.22	73
Ca^{+2}, Mg^{+2} ***	946	0	4	0.23	81
	934	0	7	0.26	70

* 0.900 ml added of cation solution, ** 0.300 ml added of each cation solution, *** 0.500 ml added of each cation solution. count time: 200 ± 1 min.
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml, 100% yield: 5102 ± 256 counts
U-238 (U-10) 2.41E2 ± 0.10 dpm/ml, 100% yield: 4820 ± 241 count
Reagents: $Nd(NO_3)_3$ 3.0E-4 M (0.10 mg/ml) in 1% HNO_3, $Y(NO_3)_3$ 1.0E-3 M in HOH
$Ca(NO_3)_3$ 1.0E-3 M in HOH, $MgSO_4$ 1.0E-3 M in HOH
Procedure: Standards were combined with the coprecipitating cation(s) solutions. 5.0 ml of 48% HF was added. After 1.5 hrs. the precipitates were filtered and counted.

TABLE XLII

COPRECIPITATION TESTS USING Y-Ca-Mg MIXED-FLUORIDES
EXPERIMENT RB122392

Coppt Cation	Am-241 Counts(tot) ($\pm 1.65\sqrt{c}$)	U-238 Counts(tot) ($\pm 1.65\sqrt{c}$)	Counter #,SSB	Counter Efficiency (± 0.1)	%Am Recovery ($\pm 11\%$)
Nd^{+3}*	676	5	23	0.18	91
Y^{+3}, Ca^{+2}, Mg^{+2} **	508	3	24	0.18	69
Ca^{+2}, Mg^{+2} ***	601	0	25	0.18	81

* 0.900 ml added of cation solution, ** 0.300 ml added of each cation solution
*** 0.500 ml added of each cation solution, count time: 200 ± 1 min.
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml, 100% yield: 4082 ± 205 counts
U-238 (U-10) 2.41E2 ± 0.10 dpm/ml, 100% yield: 12050 ± 603 count
Reagents: $Nd(NO_3)_3$ 3.0E-4 M (0.10 mg/ml) in 1% HNO_3, $Y(NO_3)_3$ 1.0E-3 M in HOH
$Ca(NO_3)_3$ 1.0E-3 M in HOH, $MgSO_4$ 1.0E-3 M in HOH
PROCEDURE SAME AS Table XLI (RB112592B), except ppt time is longer
Procedure: Standards were combined with the coprecipitating cation(s) solutions. 5.0 ml of 48% HF was added. After 3 hrs and 20 min (200 min) the precipitates were filtered and counted.

TABLE XLIII

COPRECIPITATION TESTS USING NdF_3
EXPERIMENT RB010793C

Nuclides added	Time (min)	Am Count(tot)($\pm 1.65\sqrt{c}$)	U	Counter Efficiency($\pm 2\%$)	%Am Recovery($\pm 9\%$)	%U
Am/U	15	1092	3	0.266	80	0
Am	60	1055		0.240	86	--
Am/U	185	1043	3	0.235	86	0
Am/U	265	1091	2	0.226	94	0
Am	400	1092		0.253	85	--

count time: 200 ± 1 min., no sulfate fume
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml, 100% yield: 5100 ± 215 counts
U-238 (U-12) 4.8E1 ± 0.10 dpm/ml, 100% yield: 4800 ± 192 count

TABLE XLIV

COPRECIPITATION TESTS USING NdF_3
EXPERIMENT RB012293B

Nuclide used	Am-241 Counts (total) ($\pm 1.65\sqrt{c}$)	U-238 Counts (total) ($\pm 1.65\sqrt{c}$)	Counter Efficiency ($\pm 0.2\%$)	Am %Recovery	U %Recovery
Am/U#1 (1st ppt)	218	506	0.2426	73 ± 10	18 ± 2
Am/U#1 (2nd ppt)	90	355	0.2458	29 ± 6	12 ± 2
Am/U#2 (1st ppt)	190	455	0.2234	69 ± 10	2 ± 1
Am/U#2 (2nd ppt)	60	396	0.2286	21 ± 5	1 ± 1

count time: 60 ± 1 min., (sulfate fume)
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml, 100% yield: 1224 ± 62 counts
U#1 U-238 (U-11) 4.83E1 ± 0.10 dpm/ml, 100% yield: 11592 ± 464 counts
U#2 U-238 (U-11) 4.83E1 ± 0.10 dpm/ml and (U-06) 4.88E2 ± 0.10 dpm/ml
100% yield:120704 ± 9657 counts

TABLE XLV

COPRECIPITATION TESTS USING NdF_3
EXPERIMENT RB021693

$TiCl_3$ added/ temp	Am-241 Counts(tot) ($\pm 1.65\sqrt{c}$)	Pu-239 Counts(tot) ($\pm 1.65\sqrt{c}$)	Counter Efficiency ($\pm 0.2\%$)	%Np Recovery ($\pm 8\%$)	%Pu Recovery ($\pm 8\%$)
no/22°C	805	889	0.2344	89	90
no/85°C	779	847	0.2469	83	82
no/85°C	814	811	0.2357	91	83
yes/22°C	842	944	0.2494	89	91
yes/22°C	803	829	0.2663	79	75

count time: 60 ± 1 min., (sulfate fume)
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml, 100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml, 100% yield: 4160 ± 167 counts

TABLE XLVI

COPRECIPITATION TESTS USING NdF_3
EXPERIMENT RB080793

Redox Reagent	Np* 237 Counts (total) (±1.65√c)	Pu 239	Th 232	Detector PIPS/Is #/Eff (±2%)	Np 237 % Recovery (±13%)	Pu 239 (±18%)	Th 232 (±10%)
TiCl3 (0.4 ml)	245	117	580	0.171	90	82	82
TiCl3 (0.4 ml)	238	122	541	0.172	89	85	76
TiCl3 (0.6 ml)^	377	195	899	0.239	99	98	91
none	217	130	532	0.179	74	87	72
none	241	140	543	0.194	81	87	67
H_2O_2	309	152	816	0.176	101	104	111
H_2O_2	278	138	662	0.157	111	106	101
$K_2S_2O_8$^	152	181	832	0.206	6±3	106	97
$K_2S_2O_8$^	162	173	937	0.205	2±1	101	110

* corrected for the presence of Th-230 by subtracting [0.17(#counts of Th-232)]
^ spikes not wet ashed, some nitrate may be present in sample

RB080793
count time: 200 ± 1 min.
(sulfate fume)
(88°C)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 950 ± 40 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 832 ± 35 counts

TABLE XLVII

SEPARATION OF Am AND U IONS USING TRU-Resin™
EXPERIMENT RB112192

Step	Am-241 Counts (total) ($\pm 1.65\sqrt{c}$)	U-238 Counts (total) ($\pm 1.65\sqrt{c}$)	Counter #,SSB	Counter Efficiency (± 0.1)	%Am Recovery ($\pm 11\%$)	%U Recovery ($\pm 11\%$)
3	609	--	17	0.17	70	--
4	14	--	27	0.17	2	--
6	672	--	18	0.18	73	--
7	--	581	28	0.18	--	67

RB112192
count time: 200 ± 1 min.

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 5102 ± 256 counts
U-238 (U-10) 2.41E2 ± 0.10 dpm/ml
100% yield: 4820 ± 241 count

Procedure:
Column A: Am sample only
Step 1. Am in 1.0-M HNO_3, load volume 8 ml, wash 6.5 ml 1.0-M HNO_3
Step 2. Rinse 6.5 ml 9.0 M HCl
Step 3. Elute Am: 13 ml 4.0-M HCl
Column B: Am and U sample
Step 4. Load Am/U sample in 10 ml 1.0-M HNO_3, wash 7.5 ml 1.0-M HNO_3
Step 5. 6.5 ml 9.0-M HCl
Step 6. 13 ml 4.0-M HCl
Step 7. 20 ml 0.1-M $NH_4HC_2O_4$

TABLE XLVIII

SEPARATION OF Am AND U IONS
EXPERIMENT RB021793

EXP Step	Am/U sample	Am-241 Counts (total) ($\pm 1.65\sqrt{c}$)	U-238 Counts (total) ($\pm 1.65\sqrt{c}$)	Counter #,SSB	Counter Efficiency ($\pm 2\%$)	%Am Recovery
3	1:10	818	1	1	0.2427	82 ± 7
4	1:10	26	1	2	0.2458	2 ± 1
10	1:100	714	2	4	0.2234	78 ± 7
11	1:100	15	0	5	0.2286	2 ± 1
17	1:1000	7	15	6	0.2177	0 ± 1
18	1:1000	1	292	7	0.2344	0 ± 1

count time: 200 ± 1 min.
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml, 100% yield: 4082 ± 205 counts
U-238 made from solid $UO_2(NO_3)_2 \cdot 6H_2O$ using 0.0628 grams diluted to 22.20 ml
1:1000 = 20.4 dpm Am:20000 dpm U, 1:100 = 20.4 dpm Am: 2000 dpm U,
1:10 = 20.4 dpm Am: 200 dpm U

Procedure:

Step 1	prep column 3.0 M HNO_3, loaded 1:10 mixture, Rinse 18 ml 3.0 M HNO_3
Step 2	convert column to Cl^-, 2 ml 8.0 M HCl
Step 3	Elute Am: 6.0 ml 4.0-M HCl,
Step 4	Elute Am: 5.0 ml 4.0-M HCl,
Step 5	10 ml 1.0-M HCl
Step 6	15 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl,
Step 7	2 ml 8 M HNO_3, 15ml 3-M HNO_3 (to switch column back to HNO_3)
Step 8	Load 1:100, rinse 20ml 3.0-M HNO_3
Step 9	2 ml 8.0-M HCl
Step 10,11	Am elution: 5.0 ml 4.0-M HCl
Step 12	10.0 ml 1.0-M HCl
Step 13	30ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 14	rinse column 0.1 M HNO_3, 3.0-M HNO_3, sat overnight, 15ml 3.0-M HNO_3
Step 15	load 1:1000, rinse 15 ml 3.0-M HNO_3
Step 16	2 ml 8.0-M HCl
Step 17,18,19	Am elution: 5.0 ml 4.0-M HCl
Step 20	Am elution: 10.0 ml 1.0-M HCl
Step 21	30 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl

TABLE XLIX

SEPARATION OF Am AND U IONS
EXPERIMENT RB031193

EXP Step	Am:U sample	Am-243 Counts (total) (±1.65√c)	Am-241 Counts (total) (±1.65√c)	U-238 Counts (total) (±1.65√c)	Counter Type/Eff (±2%)	Am 243/241/238 %Recovery (±10%)
3	1:10	822	743	0	PIPS/0.320	86 76 0
11	1:100	7	8	0	SSB /0.266	
12	1:100	4	2	0	PIPS/0.293	
13	1:100	24	22	1	PIPS/0.315	3 2 0
14	1:100	38	28	0	SSB /0.235	5 4 0
15*	1:100	558	451	1320	SSB /0.253	74 58 1
22	1:1000	28	18	142	SSB /0.192	5 3 0
23	1:1000	4	5	3	SSB /0.182	
24	1:1000	3	5	4	SSB /0.165	
25	1:1000	2	2	4	SSB /0.176	
26	1:1000	1	0	4	SSB /0.164	
27	1:1000	0	4	3	SSB /0.171	
28	1:1000	2	1	173	SSB /0.186	
29	1:1000	2	0	314	SSB /0.178	
30	1:1000	3	0	220	SSB /0.183	0 0 0

* spectrum poorly resolved

RB031193
count time: 200 ± 1 min.
(sulfate fume)
(22°C) (no $TiCl_3$)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 2980 ± 149 counts
U-238 made from solid $UO_2(NO_3)_2 \cdot 6H_2O$, 0.0639 grams diluted to 22.20 ml

1:1000 = 15 dpm Am-241 : 15 dpm Am-243 : 20000 dpm U (20.0 ml)
1:100 = 15 dpm Am-241 : 15 dpm Am-243 : 2000 dpm U (2.00 ml)
1:10 = 15 dpm Am-241 : 15 dpm Am-243 : 200 dpm U (0.200 ml)

Procedure:
Step 1. loaded 1:10 mixture(15ml), rinse 10ml 3.0-M HNO_3
Step 2. 2.0 ml 8.0-M HCl
Step 3,4. Elute Am: 5.0 ml 4.0-M HCl
Step 5. Elute U: 30 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 6. rinse 10 ml 1.0 M HCl, stored overnight, prep 30 ml 3.0-M HNO_3
Step 11. loaded 1:100 mixture(15ml), rinse 10ml 3.0-M HNO_3
Step 12. 2.0 ml 8.0-M HCl
Step 13,14. Elute Am: 5.0 ml 4.0-M HCl
Step 15. Elute U: 30 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Used new column for 1:1000 sample: 0.35 g TRU-Spec resin
soaked 1 hr in 3N HNO_3, glass plug, 10 ml x 5 3.0-M HNO_3
Step 21. loaded 1:1000 mixture(22ml)
Step 22. rinse 5.0 ml 3.0 M HNO_3
Step 23. 2.0 ml 8.0 M HCl
Step 24,25,26,27. Elute Am: 5.0 ml 4.0-M HCl
Step 28,29,30. Wash: 5.0 ml 1.0 M HCl
Step 31. Elute U 25 ml 0.1 M $NH_4HC_2O_4$ in 0.1 HCl

TABLE L

SEPARATION OF Am AND U IONS
EXPERIMENT RB040293

EXP Step	Am:U sample	Am-243 Counts (total) ($\pm 1.65\sqrt{c}$)	Am-241 Counts (total) ($\pm 1.65\sqrt{c}$)	U-238 Counts (total)	Detector PIPS/ls Eff ($\pm 2\%$)	Am/Am/U 243/241/238 Recovery %		
1	1:10	727	720	0	0.185	64 ± 6	66 ± 6	0
2	1:10	92	100	2	0.192	9 ± 2	8 ± 2	0
3	1:10	1	6	1	0.182	0	0	0
6	1:100	853	881	4	0.170	85 ± 7	84 ± 7	0
7	1:100	95	65	0	0.167	6 ± 2	10 ± 2	0
11	1:1000	gross alpha count = 630 dpm*				-	-	-
12	1:1000	gross alpha count = 124 dpm**				-	-	-

*measured on tennelec LB4000, 776 counts, count time 4.0 min, efficiency 0.308
**measured on tennelec LB4000, 160 counts, count time 4.0 min, efficiency 0.322
***measured on PIPS/ls, 50 microliter volume, 1814 counts, count time 399 min., efficiency 0.234.

RB040293
count time: 399 ± 1 min.
(sulfate fume)
(22°C)
(no $TiCl_3$)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 6107 ± 306 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 5945 ± 298 counts
U-238 made from solid $UO_2(NO_3)_2 \cdot 6H_2O$
0.06236 grams diluted to 44.40 ml***

1:1000 = 15 dpm Am-241 : 15 dpm Am-243 : 17200 ± 870 dpm U (40.0 ml)
1:100 = 15 dpm Am-241 : 15 dpm Am-243 : 1720 ± 87 dpm U (4.00 ml)
1:10 = 15 dpm Am-241 : 15 dpm Am-243 : 172 ± 9 dpm U (0.400 ml)

Procedure:

Step 1.	loaded 1:10 mixture in 3.0-M HCl (10ml)
Step 2.	5.0 ml 3.0-M HCl
Step 3.	5.0 ml 1.0-M HCl,
Step 4.	15 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 5.	10 ml 3.0 M HCl
Step 6.	loaded 1:100 mixture in 3.0-M HCl (10ml)
Step 7,8.	5.0 ml 3.0-M HCl
Step 9.	15.0 ml 0.1 M $NH_4HC_2O_4$ in).10-M HCl
Step 10.	20 ml 3.0 M HCl
Step 11.	loaded 1:1000 mixture in 3.0-M HCl (15ml)
Step 12,13.	5.0 ml 3.0-M HCl
Step 14.	20 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 15.	5ml 3M HCl, 5 ml 8M HCl, 15 ml 1.0 M HCl, stored

TABLE LI

SEPARATION OF Am AND U IONS
EXPERIMENT RB041393

EXP Step	Am-243 Counts (total) (±1.65√c)	Am-241 Counts (total) (±1.65√c)	U-238 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Am 243 dpm	Am 241 dpm	U 238 dpm	Am 243 %Recovery	Am 241 %Recovery	U 238 %Recovery
2	0	0	2298*	1/0.179	0	0	8060±403	0	0	41±4
3	0	0	79	2/0.186	0	0	1±1	0	0	0
5	0	0	1514*	3/0.181	0	0	5175±259	0	0	26±3
6	1108	1094	315	6/0.162	17±2	17±2	5±1	112±11	113±11	0
7	gross alpha count***				-	-	1391±140	-	-	7±2
12	0	0	2051*	4/0.182	0	0	7070±354	0	0	36±4
13	gross alpha count****				?	?	1135±114	?	?	6±2
14	0	0	2036	5/0.183	0	0	6987±350	0	0	35±4

*the portion of sample alpha counted was 1/250 of sample volume
** measured on PIPS/ls, 20 microL of the 40.0 ml volume, 1500 counts, count time 399 min., efficiency 0.191.
***measured on tennelec LB4000, 504 counts, quantity counted was 1/5 of sample volume, count time 3.0 min, efficiency 0.302, U-238 estimated to be 1/2 of gross count
****measured on tennelec LB4000, 2118 counts, count time 3.0 min, efficiency 0.311, U-238 estimated to be 1/2 of gross count, unable to estimate Am

count time: 399 ± 1 min., sulfate fume, 22°C, $TiCl_3$ except 6 & 13
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml, 100% yield: 6107 ± 306 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml, 100% yield: 5945 ± 298 counts
U-238 made from solid $UO_2(NO_3)_2 \cdot 6H_2O$, 0.12 grams diluted to 40.0 ml**

1:1000 = 15 dpm Am-241 : 15 dpm Am-243 : 19700 ± 1400 dpm U (20.0 ml)

Procedure:
Sample A: resin placed into a column and sampled poured over beads
Step 1. loaded 1:1000 mixture in 3.0-M HCl(12ml), 15.0 ml 3.0-M HCl,
Step 2. 30 ml 0.10 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 3. Wash: 25 ml 3.0 M HCl
Step 4. loaded eluate from Step 1 in 3.0-M HCl(20ml), 20 ml 3.0-M HCl,
Step 5. 30 ml 0.10 M $NH_4HC_2O_4$ in 0.10-M HCl, 10 ml 3.0-M HCl
Wash: 10ml 3.0-M HCl, 3 ml 8.0-M HCl, 5 ml 3.0-M HCl (discarded)
Step 6. loaded eluate from Step 4 (10ml) in 3.0-M HCl, 10 ml 3.0 M HCl
Step 7. 20 ml 0.10 M $NH_4HC_2O_4$ in 0.10-M HCl
Sample B: resin placed into solution containing ions for static extraction: Added 0.34 g dry TRU-Reg resin to sample, total volume approx. 25 ml in 3.0-M HCl, sample sat at RT for 2.0 hours, swrilled periodically.
Step 11. poured sample B with resin into an empty column to filter off solution, rinsed beaker with 25 ml 3.0-M HCl and poured over resin beads, reduced eluted volume to 10 ml by evaporation
Step 12. 25 ml 0.10 M $NH_4HC_2O_4$ in 0.10-M HCl, 5 ml 8.0-M HCl, 10 ml 3.0-M HCl
Step 13. loaded eluate from step 11, 12 ml 3.0-M HCl
Step 14. 20 ml 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl

TABLE LII

SEPARATION OF Am FROM Th AND/OR U IONS
EXPERIMENT RB043093

EXP Step	Am-241 Counts (total) (±1.65√c)	Th-232 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff	Am 241 %Recovery (±2%)	Th 232
4	0	1658	0.173	0	80±7
5	0	15	0.183	0	1
21	501	0	0.224	73±8	0
22	11	0	0.242	1	0

Steps 2,3,6,7,8,9,10,11,23,24,25,26,27,28,29,30,and 31 showed less than 1% recovery of Th or Am nuclides.

RB043093
count time: 200 ± 1 min.
(sulfate fume)
(85°C)
(no $TiCl_3$)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Th-232 (Th-07E) 2.99E1 ± 0.10 dpm/ml
100% yield: 11960 ± 539 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 2980 ± 149 counts
Th-229 (Th-09D) 4.0E1 ± 0.10 dpm/ml
100% yield: 3208 ± 129 counts

Procedure:

Step 1. loaded sample mixture in 4.0-M HCl (6ml)
Step 2. 5.0 ml 4.0-M HCl,
Step 3. 5.0 ml 4.0-M HCl,
Step 4,5,6,7,. 5.0 ml 1.0-M HCl,
Step 8. 5.0 ml 0.10-M HCl,
Step 9. 5.0 ml 0.10-M HCl,
Step 10. 5.0 ml 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl
Step 11. 10.0 ml 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl, 18.0 ml 3.0-M HCl, sat overnight
Step 21. loaded eluate from step 1,(wet ashed, dissolved in 4.0-M HCl(6ml))
Step 22. 6.0 ml 4.0-M HCl,
Step 23. 5.0 ml 4.0-M HCl,
Step 24. 5.0 ml 1.0-M HCl,
Step 25,26,27. 5.0 ml 1.0-M HCl,
Step 28. 5.0 ml 0.10-M HCl,
Step 29. 5.0 ml 0.10-M HCl,
Step 30. 5.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 31. 10.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl, 18.0 ml 3.0-M HCl,

TABLE LIII

SEPARATION OF Am FROM Th AND/OR U IONS
EXPERIMENT RB050393

EXP Step	Am-241 Counts (total) ($\pm 1.65\sqrt{c}$)	Th-232 Counts (total) ($\pm 1.65\sqrt{c}$)	Detector PIPS/ls #/Eff	Am 241 %Recovery ($\pm 2\%$)	Th 232
54	0	1788	4/0.189	0	79 ± 7
55	0	63	5/0.181	0	3 ± 1
56	0	7	6/0.162	0	0
71	476	0	5/0.181	86 ± 9	0
72	8	0	6/0.162	2 ± 2	0

Steps 52,53,57,58,59,60,61,73,74,75,76,77,78, 79,80,and 81 showed less than 1% recovery of Th or Am nuclides.

RB050393
count time: 200 ± 1 min.
(sulfate fume)
(85°C)
(no $TiCl_3$)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Th-232 (Th-07E) 2.99E1 ± 0.10 dpm/ml
100% yield: 11960 ± 539 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 2980 ± 149 counts
Th-229 (Th-09D) 4.0E1 ± 0.10 dpm/ml
100% yield: 3208 ± 129 counts

Procedure:

Step 51. loaded sample mixture in 5.0-M HCl (6ml)
Step 52. 5.0 ml 5.0-M HCl,
Step 53. 5.0 ml 4.0-M HCl,
Step 54. 5.0 ml 1.0-M HCl,
Step 55,56,57. 5.0 ml 1.0-M HCl,
Step 58. 5.0 ml 0.10-M HCl,
Step 59. 5.0 ml 0.10-M HCl,
Step 60. 5.0 ml 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl
Step 61. 15.0 ml 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl, 15.0 ml 3.0-M HCl
Step 71. loaded eluate from step 51, (wet ashed, dissolved in 5.0-M HCl (6ml))
Step 72. 5.0 ml 5.0-M HCl,
Step 73. 5.0 ml 4.0-M HCl,
Step 74. 5.0 ml 1.0-M HCl,
Step 75. 5.0 ml 4.0-M HCl,(wrong solution)
Step 76,77,78. 5.0 ml 1.0-M HCl,
Step 79. 5.0 ml 0.10-M HCl,
Step 80. 5.0 ml 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl
Step 81. 10.0 ml 0.10-M $NH_4HC_2O_4$ in 0.10-M HCl, 20.0 ml 3.0-M HCl

TABLE LIV

SEPARATION OF Am FROM Th AND/OR U IONS
EXPERIMENT RB042393

EXP Step	Am-241 Counts (total)	Th-232 Counts(tot) (±1.65√c)	U-238 Counts(tot) (±1.65√c)	Detector PIPS/ls Eff	Am 241 Recovery(±2%)	Th 232	U 238
1		gross alpha count 162 dpm*			-	-	-
2	8	4	12	0.178	0	0	0
3		gross alpha count 170 dpm**			-	-	-
4	0	8	359	0.186	0	0	1±1
5		gross alpha count 2087 dpm***			-	-	104±25
6	0	0	261	0.173	0	0	0
7	2	2	148	0.180	0	0	0
11		gross alpha count 167 dpm^			-	-	-
12	82	118	166	0.189	14±4	21±5	0
13		gross alpha count 252 dpm^^			-	-	-
14	0	4	1347	0.173	0	0	1±1
15	0	0	651^^^	0.189	-	-	38±12
16	0	0	628	0.181	0	0	0
17	0	0	445	0.162	0	0	0

* measured on tennelec LB4000, 100 counts, count time 2.0 min, efficiency 0.308 ** measured on tennelec LB4000, 105 counts, count time 2.0 min, efficiency 0.308
***counted 1/40 of sample volume, measured on tennelec LB4000, 67 counts, count time 2.0 min, efficiency 0.321, assume 1/2 of the gross count to be U-238
^ measured on tennelec LB4000, 105 counts, count time 2.0 min, efficiency 0.314 ^^ measured on tennelec LB4000, 158 counts, count time 2.0 min, efficiency 0.314
^^^counted 1/133 of sample volume

count time: 200 ± 1 min., sulfate fume, 22°C, $TiCl_3$ with 5,6,7,15,16,17,
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml, 100% yield: 3061 ± 154 counts
Th-232 (Th-07E) 3.0E1 ± 0.10 dpm/ml, 100% yield: 2990 ± 120 counts
U-238 made from solid $UO_2(NO_3)_2 \cdot 6H_2O$, 1:100 0.0054 grams, 1:300 0.0173 grams

1:100 = 15 dpm Am-241 : 15 dpm Th-232 : approx. 2000 U-238
1:300 = 15 dpm Am-241 : 15 dpm Th-232 : approx. 6000 U-238

Procedure:
First sample: 1 Am: 1 Th: 100 U
Step 1. loaded 1:1:100 mixture in 3.0-M HCl(7ml), rinse 8 ml 3.0-M HCl
Step 2. 5.0 ml 3.0-M HCl,
Step 3,4. 5.0 ml 1.0-M HCl,
Step 5,6. 5ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 7. 10ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl, 15 ml 3.0-M HCl
Second sample: 1 Am: 1 Th: 300 U
Step 11. loaded 1:1:300 mixture in 3.0-M HCl(10ml), rinse 6 ml 3.0-M HCl
Step 12, 13,14. 5.0 ml 3.0-M HCl,
Step 15,16. 5.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 17. 20.0 ml 0.1 M $NH_4HC_2O_4$ in 1.0-M HCl

TABLE LV

SEPARATION OF Am FROM Th AND/OR U IONS
EXPERIMENT RB050493

EXP Step	Am-241 Counts (total) (±1.65√c)	Th-232 Counts (total) (±1.65√c)	U-238 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff	Am 241 %Recovery (±2%)	Th 232	U 238
2	24	1	0	0.214	4±2	0	0
4	0	1535	0	0.173	0	74±6	0
5	0	307	0	0.183	0	14±2	0
6	0	14	0	0.180	0	1±1	0
8	0	7	338	0.223	0	0	8±1
9	0	4	1136	0.227	0	0	26±2
10	0	17	1952	0.219	0	1±1	46±4
11	0	1	11	0.235	0	0	0
21	565	3	0	0.206	90±9	0	0
22	5	0	0	0.215	1±1	0	0

Steps #3,7,23,24,25,26,27,28,29,30,31,32 showed less than 1% recovery of Am, Th, and/or U nuclides.

RB050493
count time: 200 ± 1 min.
(sulfate fume)(85°C)
($TiCl_3$)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 2980 ± 149 counts
Th-232 (Th-07E) 2.99E1 ± 0.10 dpm/ml
100% yield: 11960 ± 539 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 19320 ± 589 counts

Procedure:
Step 1. loaded sample in 5.0-M HCl, rinse 5.0-M HCl(10ml)
Step 2. 5.0 ml 5.0-M HCl,
Step 3. 5.0 ml 4.0-M HCl,
Step 4,5. 5.0 ml 1.0-M HCl,
Step 6.7. 5.0 ml 1.0-M HCl,
Step 8,9. 5.0 ml 0.10-M HCl,
Step 10. 5.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 11. 5.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 12. 10.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl, 10.0 ml 3.0-M HCl, 10 ml 5-M HCl
Step 21. loaded eluted solution from step 1, wet ashed and dissolved in 5.0-M HCl, rinse 5.0-M HCl(6ml)
Step 22. 5.0 ml 5.0-M HCl,
Step 23. 5.0 ml 4.0-M HCl,
Step 24,25,26. 5.0 ml 1.0-M HCl,
Step 27. 5.0 ml 1.0-M HCl,
Step 28,29. 5.0 ml 0.10-M HCl,
Step 30. 5.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 31. 5.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 32. 10.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl, 10.0 ml 3.0-M HCl, 10 ml 5-M HCl

TABLE LVI

ELUTION OF Np/Pu IN THE SAME FRACTION
EXPERIMENT RB061793A

EXP Step	Am 241	Np 237 Counts (total) (±1.65√c)	Pu 239	U 238	Th 232	Detector PIPS/ls Eff (±2%)	Am 241 (±9%)	Np 237 (±10%)	Pu 239 % Recovery (±8%)	U 238 (±8%)	Th 232 (±9%)
1	1067	266	7	0	0	0.186	94	19	0	0	0
2	0	32	11	0	0	0.217	0	2	1	0	0
3	0	822	1049	0	10	0.218	0	50	58	0	1
4	0	51*	13	0	83	0.242	0	2	1	0	5
5	0	0	0	0	1143	0.196	0	0	0	0	88
6	0	0	111	781	85	0.214	0	0	6	63	6

* adjusted for Th-230 (17% of Th-232 counts). Alpha energy range of the Th-230 and Np-237 overlap on spectrum.

RB061793A
count time: 399 ± 1 min.
(sulfate fume)(90°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 6107 ± 306 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 7585 ± 228 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 8299 ± 332 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 5782 ± 232 counts
Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 6631 ± 398 counts

Procedure:

Step 1.	loaded sample mixture in 5.0-M HCl, rinse 5.0-M HCl(15 ml) ** No H2O2**, 5 ml 5.0-M HCl
Step 2.	5.0 ml 5.0-M HCl,
Step 3-A.	10.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 4-A.	10.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 5.	10 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl (no NH_4I)
Step 6.	20.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl

TABLE LVIII

ELUTION OF Np/Pu IN THE SAME FRACTION
EXPERIMENT RB062393A

EXP Step	Am 241	Np 237	Pu 239	U 238	Th 232	Detector PIPS/ls Eff(±2%)	Am 241 (±10%)	Np 237 (±9%)	Pu 239 (±9%)	U 238 (±9%)	Th 232 (±10%)
		Counts (total) (±1.65√c)					% Recovery				
1	848	0	4	0	0	0.218	96	0	0	0	0
2	0	0	0	0	0	0.242	0	0	0	0	0
3	0	164*	24	0	681	0.196	0	4	3	0	90
4	0	409	71	0	24	0.214	0	50	8	0	3
5	0	255	219	0	7	0.206	0	33	27	0	1
6A	0	11	290	0	0	0.215	0	1	32	0	0
7A	0	0	117	0	0	0.223	0	0	13	0	0
8A	0	6	39	0	0	0.227	0	0	4	0	0
9A	0	60	15	0	0	0.219	0	7	2	0	0
10A	0	7	4	0	0	0.235	0	0	0	0	0
11A	0	4	3	0	2	0.173	0	0	0	0	0
12A	0	0	0	0	0	0.183	0	0	0	0	0
13A	0	0	0	0	0	0.180	0	0	0	0	0
14	0	0	0	539	0	0.189	0	0	0	74	0
15	0	0	0	64	2	0.181	0	0	0	9	0
16	0	0	0	26	2	0.162	0	0	0	4	0

* Np-237 and Th-230 have the same alpha energy range. Estimated Th-230 (0.17 of Th-232 count) and assigned the rest to Np-237.

RB062393A
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4082 ± 205 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 3864 ± 155 counts
Th-232 (Th-07E) 3.0E1 ± 0.10 dpm/ml
100% yield: 3888 ± 195 counts

Procedure: Sample A

Step 1.	loaded sample mixture in 5.0-M HCl, 3 drops H2O2(15 ml), 5 ml 5.0-M HCl
Step 2.	5.0 ml 5.0-M HCl,
Step 3,4.	5.0 ml 1.0-M HCl,
Step 5.	10.0 ml 1.0-M HCl,
Step 6A.	5.0 ml 0.10-M NH_4I in 1.0-M HCl
Step 7A.	5.0 ml 0.10-M NH_4I in 1.0-M HCl
Step 8A.	10.0 ml 0.10-M NH_4I in 1.0-M HCl
Step 9A.	5.0 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 10A.	5.0 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 11A.	10.0 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 12A.	5.0 ml 1.0-M HCl
Step 13A.	5.0 ml 1.0-M HCl
Step 14,15.	5.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 16.	10.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl

TABLE LVII

ELUTION OF Np/Pu IN THE SAME FRACTION
EXPERIMENT RB061793B

EXP Step	Am 241	Np 237	Pu 239	U 238	Th 232	Detector PIPS/ls #/Eff (±2%)	Am 241 (±9%)	Np 237 (±12%)	Pu 239 (±8%)	U 238 (±8%)	Th 232 (±9%)
		Counts (total) (±1.65√c)							% Recovery		
1	1287	14	1325	0	0	24/0.209	101	1	76	0	0
2	0	0	0	0	0	25/0.206	0	0	0	0	0
3	0	0	4	0	0	26/0.215	0	0	0	0	0
4	0	0	2	0	1	28/0.223	0	0	0	0	0
5	0	741*	5	27	153	29/0.227	0	43	0	2	10
6	0	678*	0	160	1148	30/0.219	0	41	0	13	79

* counts adjusted: alpha energy spectral range overlap from Th-230 (approx. equivalent to 17% of Th-232 counts) and U-234 (approx. equivalent to U-238 counts).

RB061793B
count time: 399 ± 1 min.
(sulfate fume)(90°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 6107 ± 306 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 7585 ± 228 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 8299 ± 332 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 5782 ± 232 counts
Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 6631 ± 398 counts

Procedure:

Step 1.	loaded sample mixture in 5.0-M HCl(15 ml), ** No H2O2**, 5 ml 5.0-M HCl
Step 2.	5.0 ml 5.0-M HCl,
Step 3-B.	10.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 4.0-M HCl
Step 4-B.	10.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 4.0-M HCl
Step 5.	10 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl (no NH_4I)
Step 6.	20.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl

TABLE LIX

ELUTION OF Np/Pu IN THE SAME FRACTION
EXPERIMENT RB062393B

EXP Step	Am 241	Np 237	Pu 239	U 238	Th 232	Detector PIPS/ls Eff (±2%)	Am 241	Np 237	Pu 239	U 238	Th 232
		Counts (total) (±1.65√c)						% Recovery (±10%)	(±9%)	(±9%)	(±9%) (±10%)
1	692	0	0	0	0	0.201	85	0	0	0	0
2	0	0	0	0	0	0.192	0	0	0	0	0
3	0	112*	13	0	542	0.178	0	2	2	0	79
4	0	282	43	0	17	0.189	0	39	5	0	2
5	0	196	141	0	12	0.186	0	28	18	0	2
6B	0	54	390	0	2	0.218	0	7	43	0	0
7B	0	32	142	0	0	0.242	0	3	14	0	0
8B	0	4	5	0	0	0.196	0	0	0	0	0
9B	0	3	0	0	0	0.214	0	0	0	0	0
10B	0	1	1	0	2	0.206	0	0	0	0	0
11B	0	3	2	0	1	0.215	0	0	0	0	0
12B	0	0	0	0	0	0.223	0	0	0	0	0
13B	0	0	0	0	0	0.227	0	0	0	0	0
14	0	0	0	655	0	0.235	0	0	0	72	0
15	0	0	0	49	0	0.173	0	0	0	7	0
16	0	0	0	16	0	0.183	0	0	0	2	0

* Np-237 and Th-230 have the same alpha energy range. Estimated Th-230 (0.17 of Th-232 count) and assigned the rest to Np-237.

RB062393B
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4082 ± 205 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 3864 ± 155 counts
Th-232 (Th-07E) 3.0E1 ± 0.10 dpm/ml
100% yield: 3888 ± 195 counts

Procedure: Sample B
Step 1. loaded sample mixture in 5.0-M HCl and 3 drops H2O2, 5 ml 5.0-M HCl
Step 2. 5.0 ml 5.0-M HCl,
Step 3.4. 5.0 ml 1.0-M HCl,
Step 5. 10.0 ml 1.0-M HCl,
Step 6B. 5.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 7B. 5.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 8B. 10.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 9B. 5.0 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 10B. 5.0 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 11B,12B. 5.0 ml 1.0-M HCl
Step 13B. 7.0 ml 1.0-M HCl with 7 d H_2O_2
Step 14,15. 5.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 16. 10.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl

TABLE LX

ELUTION OF Np/Pu IN THE SAME FRACTION
EXPERIMENT RB022294

Exp. Step	Np 237 Counts(total) (±1.65√c)	Pu 239	Th 232	Detector PIPS/ls #/Eff (±2%)	Np 237 (±18%)	Pu 239 % recovery (±12%)	Th 232 (±10%)
RB022294 1.XX = 1.00-M HCl							
2^	275*	0	1352	1/0.164	3	0	83
3^	285*	12	45	2/0.171	14	1	3
4^	493	14	24	4/0.188	23	1	1
5^	359	22	18	5/0.174	18	2	1
6^	247	26	21	7/0.205	10	2	1
7^	144	24	8	8/0.160	8	3	0
8^	104	24	3	9/0.173	5	2	0
9^	62	33	7	10/0.181	3	3	0
10^	85	10	8	11/0.177	4	1	0
11^	196	783	37	12/0.182	9	74	2
RB022294 1.XX = 1.10-M HCl							
2^	222*	0	1280	13/0.164	1	0	78
3^	211*	10	50	15/0.178	10	1	3
4^	334	17	18	16/0.177	16	2	1
5	208	6	8	1/0.164	22	1	1
6	140	7	0	2/0.171	14	1	0
7	108	15	6	4/0.188	10	3	0
8	63	15	3	5/0.174	6	3	0
9	41	14	3	7/0.205	7	2	0
10	32	12	4	8/0.160	3	3	0
11	101	398	15	12/0.182	10	77	2
RB022294 1.XX = 1.20-M HCl							
2	104*	3	686	10/0.181	0	0	76
3	40*	25	8	11/0.177	3	2	3
4	76	1	7	12/0.182	7	0	1
5	153	12	4	13/0.164	16	3	0
6	168	9	3	15/0.178	16	2	0
7	119	6	2	16/0.177	12	1	0
8	124	12	4	23/0.227	10	2	0
9	61	17	1	24/0.195	5	3	0
10	57	15	4	25/0.200	5	2	0
11	210	505	24	26/0.225	16	77	2
RB022294 1.XX = 1.30-M HCl							
2	142*	0	821	27/0.192	0	0	86
3	29*	9	63	28/0.222	1	1	6
4	44	3	19	29/0.218	3	0	2
5	107	5	6	30/0.211	7	0	0
6	156	9	5	31/0.243	11	1	0
7	133	6	5	32/0.213	11	0	0
8	148	6	4	23/0.227	11	0	0
9	103	10	3	24/0.195	9	2	0
10	93	17	2	25/0.200	8	3	0
11	344	444	18	26/0.225	27	68	2

TABLE LX (CONTINUED)

Exp. Step	Np 237 Counts(total) (±1.65√c)	Pu 239	Th 232	Detector PIPS/ls #/Eff (±2%)	Np 237 (±18%)	Pu 239 % recovery (±12%)	Th 232 (±10%)
RB022294 1.XX = 1.40-M HCl							
2	119*	0	670	1/0.164	0	0	82
3	26*	3	75	2/0.172	1	0	9
4	18	4	11	4/0.188	1	0	1
5	37	2	5	5/0.174	4	0	0
6	49	5	5	6/0.153	5	1	0
7	85	2	4	7/0.205	7	0	0
8	90	3	3	8/0.160	10	0	0
9	101	4	2	9/0.173	10	0	0
10	63	2	2	10/0.181	2	0	0
11	623	470	13	11/0.177	62	91	1
RB022294 1.XX = 1.50-M HCl							
2	102*	0	624	12/0.182	0	0	69
3	30*	2	142	13/0.164	0	0	17
4	6	2	15	15/0.178	0	0	2
5	9	2	12	16/0.177	1	0	1
6	14	2	7	17/0.196	1	0	1
7	27	6	8	18/0.210	2	1	1
8	26	6	4	19/0.221	2	1	0
9	46	2	3	20/0.207	4	0	0
10	26	22	3	21/0.210	2	4	0
11	1025*	546	26	22/0.180	99	103	3

^ count time 399 ± 1 min.

* Np-237 and Th-230 have the same alpha energy range. Estimated Th-230 (0.17 of Th-232 count) and assigned the rest to Np-237.

RB022294
count time: 200 ± 1 min.
(sulfate fume)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 5703 ± 229 counts
Pu-239 (Pu-236A1) 2.93E1 ± 0.10 dpm/ml
100% yield: 2930 ± 147 counts

Procedure:

Step 1.	loaded sample mixture in 5.0-M HCl plus H2O2 (10 ml), 10.0 ml 5.0-M HCl,
Step 2.	4.0 ml 1.XX-M HCl,
Step 3.	2.0 ml 1.XX-M HCl,
Step 4.	2.0 ml 1.XX-M HCl,
Step 5.	2.0 ml 1.XX-M HCl,
Step 6.	2.0 ml 1.XX-M HCl,
Step 7.	2.0 ml 1.XX-M HCl,
Step 8.	2.0 ml 1.XX-M HCl,
Step 9.	2.0 ml 1.XX-M HCl,
Step 10.	2.0 ml 1.XX-M HCl,
Step 11.	20.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl

TABLE LXI

ELUTION OF Np/Pu IN THE SAME FRACTION
EXPERIMENT RB082093

EXP Step	Np	Pu	U	Th	Detector #/Eff (±2%)		Np (±10%)	Pu % Recovery (±10%)	U (±10%)	Th (10%)
		Counts (total) (±1.65√c)								
RB082093A										
1 load	6	10	0	2	1/0.171		0	0	0	0
2 1.3M HCl	0*	30	0	1333	2/0.172		0	2	0	93
3 0.5M HCl	942	249	0	15	3/0.179		69	17	0	1
4	56	202	0	10	4/0.194		4	12	0	1
5	40** 177		125	20	5/0.176		3	12	9	1
6	0	36	405	0	6/0.157		0	3	33	0
7 $(NH_4)_2C_2O_4$	0	118	722	0	1/0.171		0	8	55	0
						Total%	76	54	97	96
RB082093B										
1 load	9	4	0	0	7/0.184		1	0	0	0
2 1.3M HCl	10* 32		0	1459	8/0.186		1	2	0	94
3 (.5M HCl	57	1104	0	18	9/0.178		4	75	0	1
4 + NH_4I)	14	40	0	10	10/0.173		1	3	0	1
5	20	11	0	20	11/0.194		1	1	0	1
6 0.5M HCl	775	15	0	26	12/0.182		56	1	0	2
7 AmOx 288**	30	1010	0		2/0.172		22	2	76	0
						Total%	86	84	76	99
RB082093C										
1 load	1	5	0	0	13/0.166		0	0	0	0
2 1.3M HCl	15* 30		0	1180	14/0.165		1	2	0	86
3 (.5M HCl	1091	1094	0	45	15/0.185		78	71	0	3
4 + NH_4I	10	2	0	6	16/0.185		1	0	0	0
5 + $H_2C_2O_4$)	14	1	0	3	17/0.205		1	0	0	0
6 0.5M HCl	4	9	0	3	19/0.220		0	1	0	0
7 AmOx	40** 44		858	0	3/0.179		3	3	62	0
						Total%	84	77	62	89
RB082093D (tracers only)										
1 load N/A		0^	0	41	21/0.228		N/A	0	0	2
2 1.3M HCl		0	0	733	22/0.192			0	0	48
3 (.5M HCl		1224	0	136	24/0.208			96	0	8
4 + NH_4I		3	0	0	25/0.206			0	0	0
5 + $H_2C_2O_4$)		3	0	0	26/0.205			0	0	0
6 0.5M HCl		1	0	0	29/0.220			0	0	0
7 AmOx		82	986	89	4/0.194			7	71	6
						Total%		103	71	64

* estimated due to same energy range for Th-230 and Np-237
** estimated due to same energy range for U-234 and Np-237
^ estimated due to same energy range for Ac-225 (Th-229 daughter) and Pu-236
NOTE: In RB082093D, 95±9% of Am-243 was recovered. Raw counts = 1719.

Reagent summary for each column:
A: 1.3-M HCl/0.50-M HCl/ammonium oxalate
B: 1.3-M HCl/0.50-M HCl with NH_4I/ 0.50-M HCl/ammonium oxalate
C: 1.3-M HCl/0.50-M HCl with NH_4I plus $H_2C_2O_4$/0.50-M HCl/ammonium oxalate
D: 1.3-M HCl/0.50-M HCl with NH_4I plus $H_2C_2O_4$/0.50-M HCl/ammonium oxalate

TABLE LXI (CONTINUED)

RB082093A,B,C	Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
count time: 399 ± 1 min.	100% yield: 8291 ± 415 counts
(sulfate fume)(22°C)	Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
($TiCl_3$ in all)	100% yield: 7585 ± 304 counts
detectors: PIPS/ls	Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
	100% yield: 8299 ± 332 counts
	U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
	100% yield: 7709 ± 309 counts
RB082093D (yield tracers only)	Th-229 (Th-09D) 4.01E1 ± 0.10 dpm/ml
count time: 399 ± 1 min.	100% yield: 8000 ± 400 counts
(sulfate fume)(22°C)	Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
($TiCl_3$ in all)	100% yield: 7926 ± 397 counts
	Pu-236 (Pu-19A) 3.44E1 ± 0.10 dpm/ml
	100% yield(decayed): 6149 ± 308 counts
	U-232 (U-05AA2) 4.86E1 ± 0.10 dpm/ml
	100% yield(decayed): 7182 ± 360 counts

Procedure: Column A
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2(15 ml), 4.0 ml 3.0-M HCl,
Step 2. 5.0 ml 1.30-M HCl,
Step 3. 10.0 ml 0.50-M HCl,
Step 4. 10.0 ml 0.50-M HCl,
Step 5. 20.0 ml 0.50-M HCl,
Step 6. 20.0 ml 0.50-M HCl,
Step 7. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$

Procedure: Column B
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2(15 ml), 4.0 ml 3.0-M HCl,
Step 2. 5.0 ml 1.30-M HCl,
Step 3. 10.0 ml 0.10-M NH_4I in 0.50-M HCl
Step 4. 10.0 ml 0.10-M NH_4I in 0.50-M HCl
Step 5. 20.0 ml 0.10-M NH_4I in 0.50-M HCl
Step 6. 20.0 ml 0.50-M HCl,
Step 7. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$

Procedure: Column C and D
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2(15 ml), 4.0 ml 3.0-M HCl,
Step 2. 5.0 ml 1.30-M HCl,
Step 3. 10.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 4. 10.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 5. 20.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 6. 20.0 ml 0.50-M HCl,
Step 7. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$

TABLE LXII

ELUTION OF Np/Pu IN THE SAME FRACTION
EXPERIMENT RB060993A

EXP Step	Am 241	Np 237	Pu 239	U 238	Detect. PIPS/ls	Am 241	Np 237	Pu 239	U 238	Eluting Reagent
		Counts (total) ($\pm 1.65\sqrt{c}$)			Eff($\pm$2%)	($\pm$9%)	% Recovery ($\pm$8%)	($\pm$8%)	($\pm$8%)	
1	675	11	57	0	0.207	106	1	7	0	5.0-M HCl
2	0	63	350	0	0.233	0	7	36	0	1.0-M HCl
3	0	8	356	0	0.224	0	1	38	0	0.10-M NH_4I
4	0	4	13	0	0.242	0	0	1	0	0.10-M NH_4I
5	0	590	6	0	0.196	0	79	0	0	0.030-M $H_2C_2O_4$
6	0	532*	35	392	0.214	0	17	4	63	0.10-M $NH_4HC_2O_4$

* Np-237 and U-234 alpha energy range overlaps, estimated U-234 (equal U-238 count) and assigned the rest to Np-237.

RB060993A
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 114 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 2898 ± 116 counts

Procedure:
Step 1. loaded sample mixture in 5.0-M HCl and H2O2(10ml), 5 ml 5.0-M HCl
Step 2. 10.0 ml 1.0-M HCl,
Step 3. 10.0 ml 0.10-M NH_4I in 1.0-M HCl
Step 4. 10.0 ml 0.10-M NH_4I in 1.0-M HCl
Step 5. 10 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 6. 20.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl

TABLE LXIII

ELUTION OF Np/Pu IN THE SAME FRACTION
EXPERIMENT RB060993B

EXP Step	Am 241	Np 237 Counts (total) (±1.65√c)	Pu 239	U 238	Th 229	Detector PIPS/ls #/Eff (±2%)	Am 241 (±9%)	Np 237 (±10%)	Pu 239 (±8%)	U 238 (±8%)	/Th /229 (±10%)
								% Recovery			
1	705	19^	68	0	0	25/0.206	112	2	8	0	0
2	0	42^	408	0	812	26/0.215	0	5	46	0	94
3	0	21^	357	0	0	27/0.236	0	2	36	0	0
4	0	0	17	0	0	28/0.223	0	0	2	0	0
5	0	464	0	0	0	29/0.227	0	54	0	0	0
6	0	438*	17	390	0	30/0.219	0	6*	2	60	0

* Np-237 and U-234 alpha energy range overlaps, estimated U-234 (equal U-238 count) and assigned the rest to Np-237.
^ Np-237 and Th-229 alpha energy range overlaps, estimated the assignment of counts.

RB060993B
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 114 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 2898 ± 116 counts
Th-229 (Th-09D) 4.01E1 ± 0.10 dpm/ml
100% yield: 4010 ± 201 counts
Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 33 ± 11 counts

Procedure:
Step 1. loaded sample mixture in 5.0-M HCl and H2O2,(10ml), 5 ml 5.0-M HCl
Step 2. 10.0 ml 1.0-M HCl,
Step 3. 10.0 ml 0.10-M NH_4I in 1.0-M HCl
Step 4. 10.0 ml 0.10-M NH_4I in 1.0-M HCl
Step 5. 10 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 6. 20.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl

TABLE LXIV

ELUTION OF Np/Pu IN THE SAME FRACTION
EXPERIMENT RB110593

EXP Step	Am 241	Np 237	Pu 239	U 238	Th 232	Cm 244	Det.	Am 241	Np 237	Pu 239	U 238	Th 232	Cm 244
		Counts (total) (±1.65√c)				Eff.	(±2%)	(±10%)	(±10%)	Percent Recovery (±10%)	(±16%)	(±9%)	(±10%)
RB110593A													
1	528	0	0	0	0	491	0.186	111	0	0	0	0	91
2	0	0	0	0	646	0	0.167	0	0	0	0	78	0
3	0	48	9	0	11	0	0.186	0	7	1	0	1	0
4	0	488	679	0	6	0	0.169	0	76	81	0	1	0
5	0	10	42	0	5	0	0.170	0	2	5	0	0	0
6	0	4	7	0	4	0	0.212	0	0	0	0	0	0
7	0	3	13	0	9	0	0.221	0	0	1	0	1	0
8	0	0	0	150	0	0	0.193	0	0	0	27	0	0
9	0	0	0	120	0	0	0.215	0	0	0	19	0	0
10	0	0	0	68	0	0	0.227	0	0	0	10	0	0
RB110593B													
1	453	0	0	0	0	448	0.184	97	0	0	0	0	91
2	0	0	0	0	612*	0	0.160	0	0	0	0	77	0
3	0	44	25	0	18	0	0.170	0	7	3	0	2	0
4	0	525	704	0	16	0	0.174	0	80	81	0	2	0
5	0	9	59	0	3	0	0.193	0	1	6	0	0	0
6	0	3	3	0	3	0	0.215	0	0	0	0	0	0
7	0	0	3	0	5	0	0.221	0	0	0	0	0	0
8	0	0	0	179	0	0	0.185	0	0	0	34	0	0
9	0	0	0	74	0	0	0.202	0	0	0	13	0	0
10	0	0	0	53	0	0	0.201	0	0	0	9	0	0
RB110593C									* poor spectral resolution				
1	592	0	0	0	0	477	0.188	124	0	0	0	0	88
2	0	0	0	0	599	0	0.190	0	0	0	0	63	0
3	0	39	22	0	15	0	0.186	0	6	2	0	2	0
4	0	556	784	0	9	0	0.170	0	86	93	0	1	0
5	0	7	10	0	6	0	0.185	0	0	1	0	0	0
6	0	7	13	0	4	0	0.222	0	0	1	0	0	0
7	0	1	6	0	4	0	0.201	0	0	0	0	0	0
8	0	0	0	309	0	0	0.167	0	0	0	64	0	0
9	0	0	0	30	0	0	0.221	0	0	0	5	0	0
10	0	0	0	28	0	0	0.216	0	0	0	4	0	0
RB110593D													
1	575	0	0	0	0	497	0.180	126	0	0	0	0	103
2	0	0	0	0	630	0	0.175	0	0	0	0	72	0
3	0	39	29	0	10	0	0.184	0	6	3	0	1	0
4	0	476	689	0	8	0	0.190	0	66	73	0	1	0
5	0	5	16	0	5	0	0.167	0	0	2	0	0	0
6	0	2	4	0	3	0	0.221	0	0	0	0	0	0
7	0	2	6	0	1	0	0.201	0	0	0	0	0	0
8	0	0	0	315	0	0	0.160	0	0	0	68	0	0
9	0	0	0	32	0	0	0.172	0	0	0	6	0	0
10	0	0	0	26	0	0	0.215	0	0	0	4	0	0
RB110593E													
1	434	0	0	0	0	503	0.178	107	0	0	0	0	98
2	0	0	0	0	1013	0	0.201	0	0	0	0	102	0
3	0	34	2	0	21	0	0.188	0	5	0	0	2	0
4	0	545	685	0	14	0	0.184	0	78	75	0	2	0

TABLE LXIV (CONTINUED)

EXP Step	Am 241	Np 237	Pu 239 Counts (total) (±1.65√c)	U 238	Th 232	Cm 244 Eff.	Det. (±2%)	Am 241	Np 237	Pu 239	U 238	Th 232	Cm 244 Percent Recovery (±10%)	(±10%)	(±10%)	(±16%)	(±9%)	(±10%)
5	0	10	13	0	4	0	0.160	0	2	2	0	0	0					
6	0	6	5	0	3	0	0.172	0	0	0	0	0	0					
7	0	8	5	0	2	0	0.235	0	1	0	0	0	0					
8	0	0	0	303	0	0	0.190	0	0	0	55	0	0					
9	0	0	0	51	0	0	0.201	0	0	0	9	0	0					
10	0	0	0	20	0	0	0.202	0	0	0	3	0	0					
RB110593F																		
1	447	0	0	0	0	433	0.170	103	0	0	0	0	95					
2	0	0	0	0	864	0	0.216	0	0	0	0	81	0					
3	0	34	25	0	16	0	0.180	0	7	3	0	2	0					
4	0	457	679	0	15	0	0.188	0	64	73	0	2	0					
5	0	22	11	0	5	0	0.190	0	3	1	0	0	0					
6	0	4	8	0	3	0	0.201	0	0	0	0	0	0					
7	0	4	1	0	2	0	0.232	0	0	0	0	0	0					
8	0	0	0	258	0	0	0.175	0	0	0	51	0	0					
9	0	0	0	51	0	0	0.201	0	0	0	9	0	0					
10	0	0	0	9	0	0	0.221	0	0	0	1	0	0					
RB110593G																		
1	512	0	0	0	0	510	0.193	104	0	0	0	0	92					
2	0	0	0	0	904	0	0.215	0	0	0	0	84	0					
3	0	43	19	0	24	0	0.178	0	6	2	0	3	0					
4	0	599	838	0	16	0	0.180	0	88	94	0	2	0					
5	0	8	9	0	2	0	0.175	0	1	1	0	0	0					
6	0	7	3	0	2	0	0.201	0	0	0	0	0	0					
7	0	5	1	0	2	0	0.227	0	0	0	0	0	0					
8	0	0	0	531	0	0	0.201	0	0	0	92	0	0					
9	0	0	0	7	0	0	0.235	0	0	0	1	0	0					
10	0	0	0	5	0	0	0.172	0	0	0	1	0	0					
RB110593H																		
1	492	0	0	0	0	467	0.185	105	0	0	0	0	95					
2	0	0	0	0	960	0	0.202	0	0	0	0	96	0					
3	0	19	22	0	19	0	0.170	0	3	3	0	2	0					
4	0	536	525	0	19	0	0.178	0	79	59	0	2	0					
5	0	20	16	0	4	0	0.201	0	3	2	0	0	0					
6	0	4	4	0	4	0	0.235	0	0	0	0	0	0					
7	0	4	2	0	0	0	0.169	0	0	0	0	0	0					
8	0	0	0	483	0	0	0.216	0	0	0	77	0	0					
9	0	0	0	14	0	0	0.232	0	0	0	2	0	0					
10	0	0	0	18	0	0	0.201	0	0	0	3	0	0					

RESULTS Summary:

Th: Columns A-D: NdF_3 without sulfate fume ave: 73 ± 5% additive error
Columns E-H: NdF_3 with sulfate fume ave: 91 ± 6% additive error

Am: Ave 110 ± 5% additive error

Cm: Ave 95 ± 5% additive error

	1.0-M NH_4I	0.5-M NH_4I	no NH_4I
Np:	79 ± 6%	76 ± 6%	80 ± 5%
Pu:	86 ± 7%	84 ± 6%	77 ± 5%

	HF after NH_4I	HF after no NH_4I	$NH_4C_2O_4$ after NH_4I	$NH_4C_2O_4$ after no NH_4I
U:	51 ± 4%	64 ± 5%	76 ± 6%	88 ± 7%

TABLE LXIV (CONTINUED)

RB110593
count time: 200 ± 1 min.
(80°C for #2,3 only)
($TiCl_3$ in #8,9,10 only)
(sulfate fume except #2-A,B,C,D)
detector: PIPS/ls

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 2892 ± 116 counts
Am-241 (Am-06A) 2.55E1 ± 0.10 dpm/ml
100% yield: 2551 ± 128 counts
Cm-244 (Cm-02) 3.20E1 ± 0.10 dpm/ml
100% yield(decayed): 2682 ± 135 counts

Procedure: Columns A and B
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2(10 ml), 3.0 ml 5.0-M HCl,
Step 2. 6.0 ml 1.30-M HCl,
Step 3. 2.0 ml 1.30-M HCl,
Step 4. 5.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 5. 5.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 6. 5.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 7. 10.0 ml 0.50-M HCl,
Step 8. 5.0 ml 1.0-M HF in 0.10-M HCl
Step 9. 5.0 ml 1.0-M HF in 0.10-M HCl
Step 10. 20.0 ml 1.0-M HF in 0.10-M HCl
Procedure: Columns C and D
Step 1.-3. Same as for Columns A and B.
Step 4. 5.0 ml 0.05-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 5. 5.0 ml 0.05-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 6. 5.0 ml 0.05-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 7. 10.0 ml 0.50-M HCl,
Step 8. 5.0 ml 0.10-M $(NH_4)_2C_2O_4$
Step 9. 5.0 ml 0.10-M $(NH_4)_2C_2O_4$
Step 10. 20.0 ml 0.10-M $(NH_4)_2C_2O_4$
Procedure: Columns E and F
Step 1.-3. Same as for Columns A and B
Step 4. 5.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 5. 5.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 6. 5.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 7. 10.0 ml 0.50-M HCl,
Step 8. 5.0 ml 1.0-M HF in 0.10-M HCl
Step 9. 5.0 ml 1.0-M HF in 0.10-M HCl
Step 10. 20.0 ml 1.0-M HF in 0.10-M HCl
Procedure: Columns G and H
Step 1.-3. same as for Columns A and B.
Step 4. 5.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 5. 5.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 6. 5.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 7. 10.0 ml 0.50-M HCl,
Step 8. 5.0 ml 0.10-M $(NH_4)_2C_2O_4$
Step 9. 5.0 ml 0.10-M $(NH_4)_2C_2O_4$
Step 10. 20.0 ml 0.10-M $(NH_4)_2C_2O_4$
Note: eluted fractions from step 8 and 9 were left stored in glass beakers overnight, Step 10 was stored in teflon beakers overnight. Some cloudiness resulted in HF eluted samples B-8, E-8, F-8, A-9, B-9, E-9, F-9, and F-10

TABLE LXV

REFINEMENT OF Th ELUTION STEP
EXPERIMENT RB071693

EXP Step	Procedure	Th-232 Counts (total) (±1.65√c)	Detector PIPS/Is #/Eff (±2%)	Th-232 %Recovery (±10%)
Step 1.	loaded sample in 5.0-M HCl,(10 ml) 2.0 ml 5.0-M HCl,	0	7/0.207	0
Step 2.	5.0 ml 4.0-M HCl,	0	08/0.181	0
Step 3.	5.0 ml 3.0-M HCl,	0	10/0.183	0
Step 4.	5.0 ml 2.0-M HCl,	271	11/0.201	16
Step 5.	5.0 ml 2.0-M HCl,	629	12/0.192	39
Step 6.	5.0 ml 1.0-M HCl,	597	13/0.178	40
Step 7.	5.0 ml 0.10-M HCl,	16	31/0.232	1

RB071693
count time: 200 ± 1 min.
(77°C) (no sulfate fume)
(no $TiCl_3$)

Th-232 (Th-06A) 8.31E2 ± 0.10 dpm/ml
100% yield: 8310 ± 333 counts

TABLE LXVI

REFINEMENT OF Th ELUTION STEP
EXPERIMENT RB080693A

EXP Step	Np 237	Pu 239	Th 232	Detector PIPS/ls #/Eff (±2%)	Np 237	Pu 239	Th 232
	Counts (total) (±1.65√c)				% Recovery (±20%)	(±20%)	(±10%)
1 load	9	6	1	1/0.171	1	1	0
2 1M HCl	7	1	14	2/0.172	1	0	2
3	0	4	431**	3/0.179	0	0	58
4	24*	3	65	4/0.194	3	0	8
5	64	2	9	5/0.176	10	0	1
6	74	13	5	6/0.157	12	2	0
7	54	4	5	7/0.184	8	0	0
8	83	10	1	8/0.186	12	1	0
9	55	7	1	9/0.178	8	1	0
10	33	4	4	10/0.173	5	0	0
11	43	18	1	11/0.194	6	2	0
12 combined	30	39	0	12/0.182	4	5	0
13 reagent	29	111	0	13/0.166	5	16	0
14	58	182	0	15/0.185	8	24	0
15	40	97	0	16/0.189	6	12	0
16	14	28	1	17/0.205	2	3	0
17	28	25	2	19/0.220	3	3	0
				TOTAL %	94	70	69

* estimated due to energy region overlap of Np-237 and Th-230.
** small amount of sample spilled during NdF_3 precipitation procedure

RB080693A
count time: 200 ± 1 min.
(sulfate fume)(80°C)
($TiCl_3$ in all)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts

Procedure: Column A
Step 1. loaded sample mixture in 5.0-M HCl, 6d H2O2(15 ml), 5.0 ml 3.0-M HCl,
Step 2,3,4,5,6. 1.0 ml 1.00-M HCl,
Step 7,8,9. 1.0 ml 1.00-M HCl,
Step 10. 1.0 ml 1.00-M HCl,
Step 11. 1.0 ml 1.00-M HCl,
Step 12,13,14. 1.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.00-M HCl
Step 15,16. 1.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.00-M HCl
Step 17. 20.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.00-M HCl,10.0 ml 1.0-M HCl

TABLE LXVII

REFINEMENT OF Th ELUTION STEP
EXPERIMENT RB080693B

EXP Step	Np 237	Pu 239	Th 232	Detector PIPS/Is #/Eff (±2%)	Np 237	Pu 239	Th 232
	Counts (total) (±1.65√c)				% Recovery (±20%)	(±20%)	(±10%)
1 load	14	7	0	1/0.171	2	2	0
2 1.3M HCl	0	0	513	2/0.172	0	0	54
3	4	6	409	3/0.179	0	0	51
4	42	4	10	4/0.194	5	0	1
5 1.2M HCl	64	5	8	5/0.176	8	0	1
6	88	14	5	6/0.157	11	2	0
7	113	6	5	7/0.184	14	1	0
8 1.0M HCl	130	14	8	8/0.186	16	2	1
9	97	26	4	9/0.178	11	3	0
10	56	25	2	1/0.171	9	3	0
11	46	60	0	11/0.194	7	8	0
12 combined reagent	80	432	2	12/0.182	12	58	0
				TOTAL %	95	79	108

RB080693B
count time: 200 ± 1 min.
(sulfate fume)(80°C)
($TiCl_3$ in all)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts

Procedure: Column B
Step 1. loaded sample in 5.0-M HCl, 6d H2O2(15 ml), 5.0 ml 3.0-M HCl,
Step 2. 2.0 ml 1.30-M HCl,
Step 3. 2.0 ml 1.30-M HCl,
Step 4. 2.0 ml 1.30-M HCl,
Step 5. 2.0 ml 1.20-M HCl,
Step 6. 2.0 ml 1.20-M HCl,
Step 7. 2.0 ml 1.20-M HCl,
Step 8. 2.0 ml 1.00-M HCl,
Step 9. 2.0 ml 1.00-M HCl,
Step 10. 2.0 ml 1.00-M HCl,
Step 11. 5.0 ml 1.00-M HCl,
Step 12. 20.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.0-M HCl, 10.0 ml 1.0-M HCl

TABLE LXVIII

REFINEMENT OF Th ELUTION STEP
EXPERIMENT RB080693C

EXP Step	Np 237 Counts (total) (±1.65√c)	Pu 239 Counts (total) (±1.65√c)	Th 232 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Np 237 % Recovery (±20%)	Pu 239 % Recovery (±20%)	Th 232 % Recovery (±10%)
1 load	11	2	0	1/0.171	1	0	0
2 1M HCl	6	3	7	2/0.172	1	0	1
3 + 1M NaCl	3	7	152	3/0.179	0	1	23
4	4	2	250	4/0.194	0	2	33
5 1.0M HCl	0	6	248	5/0.176	0	1	32
6 +0.3MNaCl	16	8	30	6/0.157	2	1	4
7	36	8	3	7/0.184	5	1	0
8 0.3M HCl	53	9	3	8/0.186	7	1	0
9 +1M NaCl	55	9	0	9/0.178	8	1	0
10	33	17	0	1/0.171	5	2	0
11 1M HCl	77	13	2	1/0.171	11	2	0
12	101	27	2	11/0.194	14	4	0
13	144	78	5	11/0.194	18	9	0
14 combined reagent	133	526	4	12/0.182	16	58	0
				TOTAL %	88	83	93

RB080693C
count time: 200 ± 1 min.
(sulfate fume)(80°C)
($TiCl_3$ in all)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts

Procedure: Column C
Step 1. loaded sample in 5.0-M HCl, 6d H2O2(15 ml), 5.0 ml 3.0-M HCl,
Step 2. 2.0 ml 1.00-M HCl and 1.0-M NaCl,
Step 3. 2.0 ml 1.00-M HCl and 1.0-M NaCl,
Step 4. 2.0 ml 1.00-M HCl and 1.0-M NaCl,
Step 5. 2.0 ml 1.00-M HCl and 0.3-M NaCl,
Step 6. 2.0 ml 1.00-M HCl and 0.3-M NaCl,
Step 7. 2.0 ml 1.00-M HCl and 0.3-M NaCl,
Step 8. 2.0 ml 0.30-M HCl and 1.0-M NaCl,
Step 9. 2.0 ml 0.30-M HCl and 1.0-M NaCl,
Step 10. 2.0 ml 0.30-M HCl and 1.0-M NaCl,
Step 11,12. 2.0 ml 1.00-M HCl,
Step 13. 5.0 ml 1.00-M HCl
Step 14. 20.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.0-M HCl, 10.0 ml 1.0-M HCl

TABLE LXIX

REFINEMENT OF Th ELUTION STEP
EXPERIMENT RB081693A

EXP Step	Np 237 Counts (total) (±1.65√c)	Pu 239 Counts (total) (±1.65√c)	Th 232 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Np 237 % Recovery (±20%)	Pu 239 % Recovery (±20%)	Th 232 % Recovery (±10%)
1 load	0	0	0	1/0.171	0	0	0
2 1.3M HCl	0	3	616	2/0.172	0	0	86
3 1.3M HCl	0	9	21	3/0.179	0	1	3
4 combined	678	581	20	4/0.194	92	72	3

Reagent summary:
A: 1.3-M HCl

RB081693A
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts

Procedure: Column A
Step 1. loaded sample mixture in 5.0-M HCl, 6d H2O2(15 ml), 5.0 ml 3.0-M HCl,
Step 2. 4.0 ml 1.30-M HCl,
Step 3. 1.0 ml 1.30-M HCl,
Step 4. 30.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.00-M HCl, 10.0ml 1.0-M HCl

TABLE LXX

REFINEMENT OF Th ELUTION STEP
EXPERIMENT RB081693B,C

EXP Step	Np 237 Counts (total) (±1.65√c)	Pu 239 Counts (total) (±1.65√c)	Th 232 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Np 237 % Recovery (±20%)	Pu 239 % Recovery (±20%)	Th 232 % Recovery (±10%)
RB081693B							
1	3	9	2	5/0.176	0	1	0
2	2	3	5	6/0.157	0	0	0
3	0*	4	381	7/0.184	0	0	50
4	0*	6	223	8/0.186	0	0	29
5	3*	10	80	9/0.178	0	1	11
6	3*	9	27	10/0.173	0	1	4
7	8	6	12	11/0.194	1	0	1
8	508	503	13	12/0.182	73	66	1
RB081693C							
1	1	2	0	13/0.166	0	0	0
2	1	7	10	14/0.165	0	1	1
3	7*	9	282	15/0.185	1	1	37
4	4*	7	231	16/0.185	0	1	30
5	4*	0	76	1/0.171	0	0	11
6	4*	5	37	2/0.172	0	0	5
7	2	9	9	3/0.179	0	1	1
8	540	474	12	2/0.194	73	59	1

*estimated due to energy region overlap of Np-237 and Th-230
Reagent summary: B and C: 1.0-M HCl plus 1.0-M NaCl
RB081693B,C
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts

Procedure: Column B and C
Step 1. loaded sample mixture in 5.0-M HCl, 6d H2O2(15 ml), 5.0 ml 3.0-M HCl,
Step 2. 3.0 ml 1.00-M HCl and 1.0-M NaCl,
Step 3. 3.0 ml 1.00-M HCl and 1.0-M NaCl,
Step 4. 2.0 ml 1.00-M HCl and 1.0-M NaCl,
Step 5. 2.0 ml 1.00-M HCl and 1.0-M NaCl,
Step 6. 2.0 ml 1.00-M HCl and 1.0-M NaCl,
Step 7. 2.0 ml 1.00-M HCl and 1.0-M NaCl,
Step 8. 20.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.0-M HCl, 10.0 ml 1.0-M HCl

TABLE LXXI

REFINEMENT OF Th ELUTION STEP
EXPERIMENT RB081693D

EXP Step	Np 237 Counts (total) (±1.65√c)	Pu 239 Counts (total) (±1.65√c)	Th 232 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Np 237 % Recovery (±20%)	Pu 239 % Recovery (±20%)	Th 232 % Recovery (±10%)
1	4	7	3	5/0.176	0	1	2
2	5	10	28	6/0.157	0	1	4
3	0*	0	273	7/0.184	0	0	36
4	0*	8	214	8/0.186	0	1	28
5	0*	8	146	9/0.178	0	1	20
6	9*	17	52	10/0.173	1	2	7
7	0	21	29	11/0.194	0	3	4
8	560	479	4	12/0.182	81	63	4

*estimated due to energy region overlap of Np-237 and Th-230

Reagent summary:
D: 0.30-M HCl plus 1.7-M NaCl

RB081693D
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts

Procedure: Column D
Step 1. loaded sample mixture in 5.0-M HCl, 6d H2O2(15 ml), 5.0 ml 3.0-M HCl,
Step 2. 3.0 ml 0.30-M HCl and 1.7-M NaCl,
Step 3. 3.0 ml 0.30-M HCl and 1.7-M NaCl,
Step 4. 2.0 ml 0.30-M HCl and 1.7-M NaCl,
Step 5. 2.0 ml 0.30-M HCl and 1.7-M NaCl,
Step 6. 2.0 ml 0.30-M HCl and 1.7-M NaCl,
Step 7. 2.0 ml 0.30-M HCl and 1.7-M NaCl,
Step 8. 20.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.0-M HCl, 10.0 ml 1.0-M HCl

TABLE LXXII

REFINEMENT OF Th ELUTION STEP
EXPERIMENT RB081693E

EXP Step	Np 237	Pu 239	Th 232	Detector PIPS/ls #/Eff (±2%)	Np 237	Pu 239	Th 232
	Counts (total) (±1.65√c)				% Recovery (±20%)	(±20%)	(±10%)
1	0	3	0	13/0.166	0	0	0
2	2	1	2	14/0.165	0	0	0
3	0	1	1	15/0.184	0	0	0
4	0	0	1	16/0.165	0	0	0
5	3	0	1	17/0.203	0	0	0
6	3	0	0	19/0.220	0	0	0
7	20	10	17	20/0.212	2	1	2
8	621*	680	773	21/0.228	72	72	82

*estimated due to energy region overlap of Np-237 and Th-230

Reagent summary: E: 0.30-M HCl plus 1.7-M $HClO_4$
count time: 200 ± 1 min., sulfate fume, 22°C, $TiCl_3$ in all

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml, 100% yield: 4155 ± 208 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml, 100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml, 100% yield: 4160 ± 167 counts

Procedure: Column D
Step 1. loaded sample mixture in 5.0-M HCl, 6d H2O2(15 ml), 5.0 ml 3.0-M HCl,
Step 2. 3.0 ml 0.30-M HCl and 1.7-M $HClO_4$,
Step 3. 3.0 ml 0.30-M HCl and 1.7-M $HClO_4$,
Step 4. 2.0 ml 0.30-M HCl and 1.7-M $HClO_4$,
Step 5. 2.0 ml 0.30-M HCl and 1.7-M $HClO_4$,
Step 6. 2.0 ml 0.30-M HCl and 1.7-M $HClO_4$,
Step 7. 2.0 ml 0.30-M HCl and 1.7-M $HClO_4$,
Step 8. 20.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 1.0-M HCl, 10.0 ml 1.0-M HCl

TABLE LXXIII

REFINEMENT OF U ELUTION STEP
EXPERIMENT RB062593

Second Elution of Column for U Procedural Steps		U-238 Counts (total) ($\pm 1.65\sqrt{c}$)	Detector PIPS/ls #/Eff ($\pm$2%)	Additional U-238 Recovery ($\pm$17%)	Previous U-238 Recovery ($\pm$9%)
Column # 060993A					
1.	drained 1.0-N HCl storage solution	0	7/0.207	0	-
2.	eluted with 20.0 ml 0.10-M $(NH_4)_2C_2O_4$ (pH=6)	110	8/0.181	21	63
Column # 060993B					
1.	storage solution - discarded				
2.	eluted with 20.0 ml 0.10-M $(NH_4)_2C_2O_4$ (pH=6)	99	10/0.183	19	60

Second elution of column for U

RB062593
(sulfate fume, 22°C)
count time: 200 $\pm$ 1 min.
($TiCl_3$ in all)

U-238 (U-11) 4.83E1 $\pm$ 0.10 dpm/ml
100% yield: 2898 $\pm$ 116 counts

TABLE LXXIV

USING Ce IONS AS COPRECIPITATION CARRIERS FOR Th-229 IONS
EXPERIMENT RB010494 A,B,B,D,E,F

Column/ Step	Th 232 Counts(total) (±1.65√c)	Th 229 Counts(total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Th 232 % Recovery (±11%)	Th 229 % Recovery (±13%)
A / 2	-	1	19/0.217	-	0
A / 3	-	2	20/0.215	-	0
A / 4	-	9	31/0.239	-	1
A / 6	-	8	8/0.160	-	1
B / 2	-	0	21/0.223	-	0
B / 3	-	0	22/0.177	-	0
B / 4	-	11	2/0.166	-	2
B / 6	-	14	9/0.173	-	2
C / 2	-	5	23/0.229	-	1
C / 3	-	3	24/0.200	-	0
C / 4	-	13	3/0.170	-	2
C / 6	-	17	10/0.181	-	2
D / 2	-	3	25/0.205	-	0
D / 3	-	3	26/0.121	-	0
D / 4	-	16	4/0.187	-	2
D / 6	-	19	11/0.177	-	3
E / 2	0	2	27/0.212	0	0
E / 3	0	1	28/0.224	0	0
E / 4	425	332	5/0.180	47	47
E / 6	29	14	12/0.182	3	2
F / 2	2	4	30/0.200	0	0
F / 3	0	1	31/0.248	0	0
F / 4	752	629	6/0.151	99	104
F / 6	96	82	13/0.164	12	13

Reagent used: Ce solution: 0.50 ml of 0.0007-M $Ce(SO_4)_2$ in 3.0-M HCl
Samples: A & B: Th-229 with Ce and $K_2S_2O_8$
C & D: Th-229 without Ce but with $K_2S_2O_8$
E & F: Th-229 with Th-232 without Ce but with $K_2S_2O_8$
G & H: Am, Pu, Np, U, Th-229 with Ce and $K_2S_2O_8$ (See Table LXXV)

RB010494 A,B,C,D,E,F
count time: 200 ± 1 min.
(sulfate fume)
(88_0C)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 300 counts
Th-229 (Th-09F) 3.96E1 ± 0.10 dpm/ml
100% yield: 3960 ± 198 counts

Procedure: Column A-F, CHLORIDE Column:
Step 1. loaded sample mixture in 5.0-M HCl plus $K_2S_2O_8$ (15 ml), 5.0 ml 0.020-M $K_2S_2O_8$ 4.6-M HCl
Step 2. 6.0 ml 0.020-M $K_2S_2O_8$ in 1.30-M HCl,
Step 3. 2.0 ml 0.020-M $K_2S_2O_8$ in 1.30-M HCl,
Step 4. 10.0 ml 0.50-M HCl,
Step 6. two weeks later, 10.0 ml 0.10-M HCl

TABLE LXXV

USING Ce IONS AS COPRECIPITATION CARRIERS FOR Th-229 IONS
EXPERIMENT RB010494 G,H

EXP Step	Am 241	Np 237	Pu 239	U 238	Th 229	Detector PIPS/ls Eff (±2%)	Am 241 (±8%)	Np 237 (±25%)	Pu 239 (±8%)	U 238 (±9%)	Th 229 (±25%)
		Counts (total) (±1.65√c)						Recovery			
G-1	1294	24*	0	0	24*	0.215	118	1*	0	0	1*
G-2	0	0	0	0	4	0.224	0	0	0	0	0
G-3	0	0	0	0	1	0.200	0	0	0	0	0
G-4	0	978*	956	73	978*	0.223	0	80*	86	7	30*
G-5	0	0	0	747	90**	0.177	0	0	0	88	10**
G-6	0	0	0	0	27	0.178	0	0	0	0	4
H-1	1292	16*	0	0	16*	0.229	111	1*	0	0	1*
H-2	50	0	0	0	3	0.211	5	0	0	0	0
H-3	0	0	0	0	4	0.217	0	0	0	0	0
H-4	0	888*	911	21	888*	0.200	0	80*	91	2	30*
H-5	0	0	0	707	130**	0.205	0	0	0	72	13**
H-6	0	0	0	0	33	0.177	0	0	0	0	5

* estimated percentage due to Np-237 and Th-229 energy range overlap
** estimated percentage due to U-234 and Th-229 energy range overlap

Reagent used:
Ce solution: 0.50 ml of 0.0007-M $Ce(SO_4)_2$ in 3.0-M HCl

Samples:
A & B: Th-229 with Ce and $K_2S_2O_8$ (See Table LXXIV)
C & D: Th-229 without Ce but with $K_2S_2O_8$ (See Table LXXIV)
E & F: Th-229 with Th-232 without Ce but with $K_2S_2O_8$ (See Table LXXIV)
G & H: Am, Pu, Np, U, Th-229 with Ce and $K_2S_2O_8$

RB10494 G,H
count time: 200 ± 1 min.
(sulfate fume)
(88_oC)

Th-229 (Th-09F) 3.96E1 ± 0.10 dpm/ml
100% yield: 3960 ± 198 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
U-238 (U-12) 4.83E1 ± 0.10 dpm/ml
100% yield: 4830 ± 194 counts
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 5102 ± 256 counts

Procedure: Column G-H
CHLORIDE Column:
Step 1. loaded sample in 5.0-M HCl plus $K_2S_2O_8$(15 ml),
5.0 ml 0.020-M $K_2S_2O_8$ 4.6-M HCl
Step 2. 6.0 ml 0.020-M $K_2S_2O_8$ in 1.30-M HCl,
Step 3. 2.0 ml 0.020-M $K_2S_2O_8$ in 1.30-M HCl,
Step 4. 30.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
10.0 ml 0.50-M HCl,
Step 5. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$
Step 6. two weeks later, 10.0 ml 0.10-M HCl

TABLE LXXVI

THE REDOX EFFECT OF ZINC ON THE ELUTION PATTERN OF THE ACTINIDES EXPERIMENT RB063093

EXP Step	Am 241	Np 237	Pu 239	U 238	Th 232	Detector PIPS/ls	Am 241	Np 237	Pu 239	U 238	Th 232
		Counts (total) (±1.65√c)			#/Eff (±2%)		(±9%)	% Recovery (±9%)	(±9%)	(±9%)	(±10%)
1	608	0	506	0	0	11/0.201	74	0	61	0	0
2	0	31*	94	0	558	12/0.192	0	4	12	0	75
3	0	595**	0	322	30	13/0.178	0	88	0	47	4
4	0	6	0	0	0	15/0.189	0	0	0	0	0
5	0	6	0	0	0	16/0.186	0	0	0	0	0
6	0	0	0	353	0	18/0.218	0	0	0	42	0
7	0	0	0	8	0	21/0.242	0	0	0	1	0

* estimated due to energy region overlap of Np-237 and Th-230.
** estimated due to energy region overlap of Np-237 and U-234.

RB063093
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4082 ± 205 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 3864 ± 155 counts
Th-232 (Th-07E) 3.0E1 ± 0.10 dpm/ml
100% yield: 3888 ± 195 counts

Procedure: Sample A
Step 1. loaded sample mixture in 5.0-M HCl(60 ml), 6 ml 5.0-M HCl
Step 2. 6.0 ml 1.0-M HCl,
Step 3. 24.0 ml 1.0-M HCl,
Step 4. 24.0 ml 0.10-M NH_4I in 1.0-M HCl
Step 5. 24.0 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 6. 12.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 7. 24.0 ml 0.1 M $(NH_4)_2C_2O_4$ in deionized HOH

TABLE LXXVII

THE REDOX EFFECT OF HYDRAZINE SULFATE ON
THE ELUTION PATTERN OF THE ACTINIDES
EXPERIMENT RB012494 A,B,C
RAW DATA

Column-Exp step	Am 241	Am 243	Np 237	Pu 239 Counts ($\pm 1.65\sqrt{c}$)	Pu 236 (total)	U 238	U 232	Th 232	Th 229 Eff	Detector PIPS/ls ($\pm 2\%$)
A-1	959	805	0	862	705*	0	0	0	0	0.161
B-1	1016	787	0	563	626*	0	0	0	0	0.166
C-1	1036	876	0	528	581*	0	0	0	0	0.170
A-2	0	0	0	0	0	0	0	688	574	0.182
B-2	0	0	0	0	0	0	0	796	579	0.182
C-2	0	0	0	0	0	0	0	633	536	0.162
A-3	0	0	0	0	0	0	0	11	20	0.223
B-3	0	0	0	0	0	0	0	8	8	0.177
C-3	0	0	0	0	0	0	0	7	11	0.229
A-4^	0	0	2003**	143	94	101	93	0	0	0.161
B-4^	0	0	1931**	383	196	80	86	16	0	0.166
C-4^	0	0	1704**	477	258	108	79	20	0	0.170
A-6^	0	0	0	0	0	1415	1157	0	0	0.211
B-6^	0	0	0	0	0	1460	1240	0	0	0.217
C-6^	0	0	0	0	0	1086	926	0	0	0.215

RB012494 A,B,C
count time: 200 ± 1 min.
(sulfate fume)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 5703 ± 229 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 193 counts
Am-241 (Am-06D) 1.05E2 ± 0.10 dpm/ml
100% yield: 6300 ± 378 counts
Th-229 (Th-09F) 3.96E1 ± 0.10 dpm/ml
100% yield: 3960 ± 198 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 2230 ± 134 counts
U-232 (U-05AA3) 4.86E1 ± 0.10 dpm/ml
100% yield(decayed): 4505 ± 271 counts
Am-243 (Am-08A) 4.97E1 ± 0.10 dpm/ml
100% yield: 4966 ± 298 counts

TABLE LXXVIII

THE REDOX EFFECT OF HYDRAZINE SULFATE ON
THE ELUTION PATTERN OF THE ACTINIDES
EXPERIMENT RB012494 A,B,C
PERCENT RECOVERY

Column-Exp step	Am 241 (±9%)	Am 243 (±10%)	Np 237 (±8%)	Pu 239 (±12%)	Pu 236 (±14%)	U 238 (±9%)	U 232 (±10%)	Th 232 (±9%)	Th 229 (±11%)
	% Recovery								
A-1	95	100	0	108	*	0	0	0	0
B-1	98	96	0	68	*	0	0	0	0
C-1	97	104	0	62	*	0	0	0	0
A-2	0	0	0	0	0	0	0	80	84
B-2	0	0	0	0	0	0	0	88	80
C-2	0	0	0	0	0	0	0	79	84
A-3	0	0	0	0	0	0	0	1	2
B-3	0	0	0	0	0	0	0	1	1
C-3	0	0	0	0	0	0	0	1	1
A-4	0	0	104	9	12	7	6	0	0
B-4	0	0	98	23	26	5	6	1	0
C-4	0	0	88	28	33	7	5	1	0
A-6	0	0	0	0	0	70	61	0	0
B-6	0	0	0	0	0	70	64	0	0
C-6	0	0	0	0	0	53	48	0	0

* contains counts from Ac-227, Ra-224 and Pu-236, unable to determine the percentage that is Pu-236.
^ count time 399 ± 1 min.
** contains U-234 and Np-237, used the amount of U-238 to estimate the amount of U-234 counts.

Procedure: Column A-C
CHLORIDE Column:
Step 1. loaded sample in 5.0-M HCl+1.0g hydrazine sulfate$(NH_2)_2 \cdot H_2SO_4$ (15ml),
5.0 ml 0.10-M hydrazine sulfate in 5.0-M HCl
Step 2. 6.0 ml 0.10-M hydrazine sulfate in 1.30-M HCl,
Step 3. 2.0 ml 0.10-M hydrazine sulfate in 1.30-M HCl,
Step 4. 20.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl,
Step 6. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$
note: load problems with the peroxide (bubbles in column)

TABLE LXXIX

THE REDOX EFFECT OF HYDROGEN PEROXIDE ON
THE ELUTION PATTERN OF THE ACTINIDES
EXPERIMENT RB012494 D,E,F
RAW DATA

Column-Exp step	Am 241	Am 243	Np 237	Pu 239	Pu 236	U 238	U 232	Th 232	Th 229	Detector PIPS/ls Eff(±2%)
				Counts (total)(±1.65√c)						
D-1	1045	902	0	0	0	0	0	0	0	0.187
E-1	1004	797	0	0	0	0	0	0	0	0.188
F-1	1070	940	0	0	0	0	0	0	0	0.170
D-2	0	0	691*	80	**	0	0	565	691*	0.181
E-2	0	0	931*	127	**	0	0	777	931*	0.200
F-2	0	0	996*	77	**	0	0	861	996*	0.217
D-3	0	0	271*	86	**	0	0	52	271*	0.200
E-3	0	0	264*	84	**	0	0	116	264*	0.218
F-3	0	0	235*	78	**	0	0	91	235*	0.224
D-4^	0	0	1397*	1139	733**	0	0	248	1397*	0.187
E-4^	0	0	1609*	1185	665**	0	0	224	1609*	0.188
F-4^	0	0	1554*	1296	217**	0	0	127	1554*	0.170
D-5^	0	0	127*	95	148**	0	0	125	127*	0.173
E-5^	0	0	44*	40	35**	0	0	42	44*	0.162
F-5^	0	0	60*	62	**	0	0	27	60*	0.175
D-6^	0	0	0	0	0	1646	1385	0	0	0.223
E-6^	0	0	0	0	0	1473	1251	16	0	0.229
F-6^	0	0	0	0	0	1564	1349	68	0	0.205

RB012494 D,E,F
count time: 200 ± 1 min.
(sulfate fume)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 5703 ± 229 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 193 counts
Am-241 (Am-06D) 1.05E2 ± 0.10 dpm/ml
100% yield: 6300 ± 378 counts

#D-E Th-229 (Th-09F) 3.96E1 ± 0.10 dpm/ml, 100% yield: 3960 ± 198 counts
#F Th-229 (Th-08B) 4.15E1 ± 0.10 dpm/ml, 100% yield: 4152 ± 250 counts

Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 2230 ± 134 counts
U-232 (U-05AA3) 4.86E1 ± 0.10 dpm/ml
100% yield(decayed): 4505 ± 271 counts
Am-243 (Am-08A) 4.97E1 ± 0.10 dpm/ml
100% yield: 4966 ± 298 counts

TABLE LXXX

THE REDOX EFFECT OF HYDROGEN PEROXIDE ON
THE ELUTION PATTERN OF THE ACTINIDES
EXPERIMENT RB012494 D,E,F
PERCENT RECOVERY

Column-Exp step	Am 241 (±9%)	Am 243 (±9%)	Np 237 (±25%)	Pu 239 (±10%)	Pu 236 (±25%)	U 238 (±9%)	U 232 (±10%)	Th 232 (±18%)	Th 229 (±25%)
				% Recovery					
D-1	89	97	0	0	0	0	0	0	0
E-1	85	85	0	0	0	0	0	0	0
F-1	100	111	0	0	0	0	0	0	0
D-2	0	0	14*	9	**	0	0	63	63*
E-2	0	0	16*	13	**	0	0	78	78*
F-2	0	0	10*	7	**	0	0	80	80*
D-3	0	0	19*	9	**	0	0	5	5*
E-3	0	0	12*	8	7**	0	0	11	11*
F-3	0	0	11*	7	**	0	0	8	8*
D-4	0	0	54*	61	76**	0	0	13	13*
E-4	0	0	65*	63	59**	0	0	12	12*
F-4	0	0	75*	77	75**	0	0	8	8*
D-5	0	0	1*	6	6**	0	0	7	7*
E-5	0	0	0*	2	0**	0	0	3	2*
F-5	0	0	2*	4	**	0	0	2	2*
D-6	0	0	0	0	0	77	69	0	0
E-6	0	0	0	0	0	67	61	1	0
F-6	0	0	0	0	0	80	73	3	0

* estimated percentages, estimated Th-230 to equal 17% of Th-232 counts and estimated Th-229 percent recovery to equal Th-232 percent recovery

** unable to determine due to Ra-224 and Pu-236 energy range overlap, or subtracted estimated Ra-224 counts (equal to Rn-220 counts)

Procedure: Column D-F
CHLORIDE Column:
Step 1. loaded sample in 5.0-M HCl plus 1.0 ml 30% H_2O_2 (15 ml),
5.0 ml 0.15-M H_2O_2 in 5.0-M HCl
Step 2. 6.0 ml 0.15-M H_2O_2 in 1.30-M HCl
Step 3. 2.0 ml 0.15-M H_2O_2 in 1.30-M HCl
Step 4. Column D-F 5.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Column G-I 2.0 ml of 0.10-M NH_4I
3.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 5. 15.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 6. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$

TABLE LXXXI

INDEPENDENT SEPARATION PERFORMED BY MR.TIAM XIE
EXPERIMENT TX111093
RAW DATA

EXP Step	Am 241 (±2%)	Am 243	Np237* Pu242	Pu 239	Pu 236 Counts (total) (±1.65√c)	U 238	U 232	Th 232	Th 229	Cm 244	Detector PIPS/ls #/Eff
TX111093-1											
2	0	0	0	0	0	0	0	447	613	0	24/0.201
4	0	0	846	337	235	0	0	19	0	0	27/0.235
6	0	0	0	0	0	648	669	0	0	0	21/0.221
13	2124	862**	0	0	0	0	0	0	0	607***	17/0.200
TX111093-2											
2	0	0	0	0	0	0	0	549	878	0	25/0.201
4	0	0	1167	527	371	0	0	4	0	0	32/0.227
6	0	0	0	0	0	594	506	0	0	0	22/0.172
13	2051	989**	0	0	0	0	0	0	0	577***	19/0.215
TX111093-3											
2	0	0	0	0	0	0	0	619	914	0	27/0.235
4	0	0	1272	560	353	0	0	4	0	0	17/0.200
6	0	0	0	0	0	672	592	0	0	0	24/0.201
13	(2386**)		0	0	0	0	0	0	0	542***	21/0.221
TX111093-4											
2	0	0	0	0	0	0	0	632	913	0	32/0.227
4	0	0	1101	550	423	0	0	4	0	0	19/0.215
6	0	0	0	0	0	609	535	0	0	0	25/0.201
13	1908	813**	0	0	0	0	0	0	0	670***	22/0.171
TX111093-5											
2	0	0	0	0	0	0	0	543	809	0	17/0.200
4	0	0	667	284	202	0	0	15	0	0	21/0.221
6	0	0	0	0	0	709	766	0	0	0	27/0.235
13	2019	728	0	0	0	0	0	0	0	670***	24/0.201
TX111093-Blank											
2	-	0	0	-	0	-	0	-	117	-	19/0.215
4	-	0	^, ^^	-	368	-	0	-	^^	-	22/0.172
6	-	0	0	-	0	-	437	-	150^^	-	32/0.227
13	-	707	0	-	0	-	0	-	0	163^^^	25/0.201

*counted together due to alpha energy range overlap
**poor resolution, some overlap of Am-241 counts into the Am-243 energy range
*** Ac-225 (daughter of Th-229) counts in Cm energy range
^ no Np-237 added to blank
^^ count contains Th-229 and Pu-242 (unable to determine percentage)
^^^ No Cm-244 added to blank, Ac-225 (daughter of Th-229) counts in Cm-244 energy range

TABLE LXXXI (CONTINUED)

TX111093
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in #6 only)

Th-232 (Th-07F) 2.99E1 ± 0.10 dpm/ml
100% yield: 2992 ± 150 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 3328 ± 134 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 193 counts
Am-241 (Am-06D) 1.05E2 ± 0.10 dpm/ml
100% yield: 10500 ± 525 counts
Cm-244 (Cm-02) 3.20E2 ± 0.10 dpm/ml
100% yield(decayed): 2681 ± 161 counts
Th-229 (Th-09D) 3.96E1 ± 0.10 dpm/ml
100% yield: 3960 ± 198 counts
Am-243 (Am-09) 3.8E1 ± 0.10 dpm/ml
100% yield: 3796 ± 190 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 2299 ± 138 counts
U-232 (U-05AA3) 4.86E1 ± 0.10 dpm/ml
100% yield(decayed): 4510 ± 226 counts
Pu-242 (Pu-22) 4.5E1 ± 0.10 dpm/ml
100% yield: 4540 ± 227 counts

TABLE LXXXII

INDEPENDENT SEPARATION PERFORMED BY MR.TIAM XIE
EXPERIMENT TX111093
PERCENT RECOVERY

EXP Step	Am 241	Am 243	Np237* Pu242	Pu 239	Pu 236	U 238	U 232	Th 232	Th 229	Cm 244
	% Recovery									
	(±12%)	(±12%)	(±18%)	(±10%)	(±13%)	(±10%)	(±11%)	(±10%)	(±10%)	(±18%)
TX111093-1										
CHLORIDE Column										
2	0	0	0	0	0	0	0	74	77	0
4	0	0	43	43	43	0	0	0	0	0
6	0	0	0	0	0	61	68	0	0	0
NITRATE Column (after calcium oxalate coprecipitation)										
13	101	114**	0	0	0	0	0	0	0	113***
TX111093-2										
CHLORIDE Column										
2	0	0	0	0	0	0	0	92	111	0
4	0	0	62	70	71	0	0	0	0	0
6	0	0	0	0	0	73	66	0	0	0
NITRATE Column (after calcium oxalate coprecipitation)										
13	91	121**	0	0	0	0	0	0	0	100***
TX111093-3										
CHLORIDE Column										
2	0	0	0	0	0	0	0	90	100	0
4	0	0	76	84	76	0	0	0	0	0
6	0	0	0	0	0	70	65	0	0	0
NITRATE Column (after calcium oxalate coprecipitation)										
13	(76**)		0	0	0	0	0	0	0	92***
TX111093-4										
CHLORIDE Column										
2	0	0	0	0	0	0	0	92	101	0
4	0	0	61	77	86	0	0	0	0	0
6	0	0	0	0	0	63	59	0	0	0
NITRATE Column (after calcium oxalate coprecipitation)										
13	107	126**	0	0	0	0	0	0	0	147***
TX111093-5										
CHLORIDE Column										
2	0	0	0	0	0	0	0	91	102	0
4	0	0	36	39	40	0	0	0	0	0
6	0	0	0	0	0	63	59	0	0	0

TABLE LXXXII (CONTINUED)

EXP Step	Am 241 (±12%)	Am 243 (±12%)	Np237* Pu242 (±18%)	Pu 239 (±10%)	Pu 236 (±13%)	U 238 (±10%)	U 232 (±11%)	Th 232 (±10%)	Th 229 (±10%)	Cm 244 (±18%)
				% Recovery						
TX111093-5										
NITRATE Column (after calcium oxalate coprecipitation)										
13	96	96	0	0	0	0	0	0	0	92***
TX111093-Blank										
CHLORIDE Column										
2	-	0	0	-	0	-	0	-	14	-
4	-	0	^^	-	93	-	0	-	^^	-
6	-	0	0	-	0	-	59	-	17	-
NITRATE Column (after calcium oxalate coprecipitation)										
13	-	93	0	-	0	-	0	-	0	30^^^

*counted together due to alpha energy range overlap
**poor resolution, some overlap of Am-241 counts into the Am-243 energy range
*** Ac-225 (daughter of Th-229) counts in Cm energy range
^ no Np-237 added to blank
^^ count contains Th-229 and Pu-242 (unable to determine percentage)
^^^ No Cm-244 added to blank, Ac-225 (daughter of Th-229) counts in Cm-244 energy range

Procedure:
CHLORIDE Column:
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2 (15 ml),
4.0 ml 3.0-M HCl,
Step 2. 4.0 ml 1.30-M HCl,(Th fraction)
Step 3. 3.0 ml 1.30-M HCl,
Step 4. 20.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl (Np/Pu fraction)
Step 5. 10.0 ml 0.50-M HCl,
Step 6. 40.0 ml 0.10-M $(NH_4)_2C_2O_4$ (U fraction)
NITRATE Column:
Step 10. coprecipitation using calcium oxalate, redissolved in nitric acid
Step 11. load in 3.0-M HNO_3 plus solid ascorbic acid
rinse 10 ml 3.0-M HNO_3
Step 12. 2.0 ml 8.0-M HCl
Step 13 20 ml 3.0-M HCl (Am/Cm fraction)

TABLE LXXXIII

INDEPENDENT SEPARATION PERFORMED BY MR.TIAM XIE
EXPERIMENT TX012594
RAW DATA

EXP Step	Am 241	Am 243	Np 237	Pu 239 Counts ($\pm 1.65\sqrt{c}$)	Pu 236 (total)	U 238	U 232	Th 232	Th 229	Detector PIPS/ls #/Eff ($\pm$2%)
TX012594-1										
2	0	0	0	0	0	0	0	905	1276	15/0.181
4	0	0	1422	111	105	0	0	19	0	8/0.169
5	0	0	0	0	0	1637	1009	0	0	21/0.223
13	1052	1004	0	0	0	0	0	0	0	1/0.161
TX012594-2										
2	0	0	0	0	0	0	0	878	1264	16/0.175
4	0	0	1348	62	42	0	0	22	0	9/0.170
5	0	0	0	0	0	1911	1096	0	0	22/0.176
13	738	758	0	0	0	0	0	0	0	2/0.166
TX012594-3										
2	0	0	0	0	0	0	0	1219	1763	17/0.200
4	0	0	1335	68	58	0	0	17	0	10/0.172
5	0	0	0	0	0	2014	1110	0	0	23/0.230
13	798	823	0	0	0	0	0	0	0	3/0.170
TX012594-4										
2	0	0	0	0	0	0	0	1273	1698	20/0.215
4	0	0	1186	98	77	0	0	29	0	13/0.162
5	0	0	0	0	0	1959	1075	0	0	26/0.218
13	1041	1052	0	0	0	0	0	0	0	7/0.188
TX012594-Blank-1										
2	-	0	-	-	0	-	0	-	1079	18/0.211
4	-	0	-	-	62	-	0	-	44	11/0.173
5	-	0	-	-	0	-	1155	-	0	26/0.218
13	-	431	-	-	0	-	0	-	0	4/0.187
TX012594-Blank-2										
2	-	0	-	-	0	-	0	-	1311	18/0.217
4	-	0	-	-	67	-	0	-	73	12/0.182
5	-	0	-	-	0	-	1025	-	0	25/0.205
13	-	850	-	-	0	-	0	-	0	5/0.180

count time: 399 ± 1 min., sulfate fume, 22°C, $TiCl_3$ in #5 only
Th-232 (Th-07F) 2.99E1 ± 0.10 dpm/ml, 100% yield: 5971 ± 299 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml, 100% yield: 7585 ± 304 counts
Pu-239 (Pu-236A1) 2.93E1 ± 0.10 dpm/ml, 100% yield: 5845 ± 292 counts
U-238 (U-12) 4.83E1 ± 0.10 dpm/ml, 100% yield: 9636 ± 386 counts
Am-241 (Am-06D) 1.05E2 ± 0.10 dpm/ml, 100% yield: 8379 ± 419 counts
Th-229 (Th-08B) 4.15E1 ± 0.10 dpm/ml, 100% yield: 8283 ± 415 counts
Am-243 (Am-09) 3.8E1 ± 0.10 dpm/ml, 100% yield: 7573 ± 379 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml, 100% yield(decayed): 4449 ± 267 counts
U-232 (U-2-088A1) 2.93E1 ± 0.10 dpm/ml, 100% yield(decayed): 5311 ± 319 counts

TABLE LXXXIV

INDEPENDENT SEPARATION PERFORMED BY MR.TIAM XIE
EXPERIMENT TX012594
PERCENT RECOVERY

EXP Step	Am 241 (±9%)	Am 243 (±9%)	Np 237 (±8%)	Pu 239 (±19%)	Pu 236 (±19%)	U 238 (±8%)	U 232 (±10%)	Th 232 (±9%)	Th 229 (±9%)
				% Recovery					
TX012594-1									
CHLORIDE Column									
2	0	0	0	0	0	0	0	84	85
4	0	0	110	11	11	0	0	2	0
5	0	0	0	0	0	76	85	0	0
NITRATE Column (after calcium oxalate coprecipitation)									
13	78	82	0	0	0	0	0	0	0
TX012594-2									
CHLORIDE Column									
2	0	0	0	0	0	0	0	84	87
4	0	0	104	6	3	0	0	2	0
5	0	0	0	0	0	112	117	0	0
NITRATE Column (after calcium oxalate coprecipitation)									
13	53	60	0	0	0	0	0	0	0
TX012594-3									
CHLORIDE Column									
2	0	0	0	0	0	0	0	102	106
4	0	0	102	7	4	0	0	2	0
5	0	0	0	0	0	91	91	0	0
NITRATE Column (after calcium oxalate coprecipitation)									
13	56	64	0	0	0	0	0	0	0
TX012594-4									
CHLORIDE Column									
2	0	0	0	0	0	0	0	99	95
4	0	0	96	10	10	0	0	3	0
5	0	0	0	0	0	93	92	0	0
NITRATE Column (after calcium oxalate coprecipitation)									
13	66	74	0	0	0	0	0	0	0
TX012594-Blank-1									
CHLORIDE Column									
2	-	0	-	-	0	-	0	-	62
4	-	0	-	-	8	-	0	-	3
5	-	0	-	-	0	-	108	-	0

TABLE LXXXIV (CONTINUED)

EXP Step	Am 241 (±9%)	Am 243 (±9%)	Np 237 (±8%)	Pu 239 (±19%)	Pu 236 (±19%)	U 238 (±8%)	U 232 (±10%)	Th 232 (±9%)	Th 229 (±9%)
				% Recovery					
NITRATE Column (after calcium oxalate coprecipitation)									
13	-	30	-	-	0	-	0	-	0
TX012594-Blank-2									
CHLORIDE Column									
2	-	0	-	-	0	-	0	-	73
4	-	0	-	-	8	-	0	-	5
5	-	0	-	-	0	-	94	-	0
NITRATE Column (after calcium oxalate coprecipitation)									
13	-	62	-	-	0	-	0	-	0

Procedure:
CHLORIDE Column:
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2 (cooked off)(15 ml), 10.0 ml 5.0-M HCl,
Step 2. 6.0 ml 1.30-M HCl,(Th fraction)
Step 3. 2.0 ml 1.30-M HCl,
Step 4. 15.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl (Np/Pu fraction)
Step 5. 2.0 ml 0.10-M $(NH_4)_2C_2O_4$ (U fraction)
Step 6. wash column with 10 ml 0.10-M HCL, store
NITRATE Column:
Step 10. coppt eluate from step 1, using calcium oxalate, redissolved in 3.0-M nitric acid
Step 11. load solution form step 10 in 3.0-M HNO_3 plus solid ascorbic acid rinse 5 ml 3.0-M HNO_3
Step 12. 1.0 ml 8.0-M HCl
Step 13 15.0 ml 3.0-M HCl (Am fraction)
Step 14 10.0 ml 1.0-M HCl,

TABLE LXXXV

INDEPENDENT SEPARATION PERFORMED BY MR.TIAM XIE
EXPERIMENT TX020994
RAW DATA

EXP Step	Am 241	Am 243	Np 237	Pu 239 Counts (±1.65√c)	Pu 236 (total)	U 238	U 232	Th 232	Th 229	Detector PIPS/ls #/Eff (±2%)
TX020994-1										
2	0	0	0	0	0	0	0	1104	1692	21/0.210
4	0	0	1095	870	722	0	0	57	0	15/0.178
5	0	0	0	0	0	1696	1496	5	0	27/0.192
13	1068	1061	0	0	0	0	0	0	0	8/0.160
TX020994-2										
2	0	0	0	0	0	0	0	1016	1380	22/0.180
4	0	0	1087	875	738	0	0	65	0	16/0.177
5	0	0	0	0	0	1587	1468	0	0	28/0.222
13	1123	1063	0	0	0	0	0	0	0	9/0.173
TX020994-3										
2	0	0	0	0	0	0	0	1097	1610	23/0.227
4	0	0	1479	1160	952	0	0	100	0	17/0.196
5	0	0	0	0	0	1896	1586	0	0	29/0.219
13	899	833	0	0	0	0	0	0	0	10/0.181
TX020994-4										
2	0	0	0	0	0	0	0	1070	1488	24/0.195
4	0	0	1306	1166	868	0	0	92	0	18/0.210
5	0	0	0	0	0	1765	1631	0	0	30/0.211
13	1245	1101	0	0	0	0	0	0	0	11/0.177
TX020994-Blank-1										
2	-	0	-	-	0	-	0	-	1005	25/0.200
4	-	0	-	-	815	-	0	-	127	19/0.221
5	-	0	-	-	0	-	1444	-	0	31/0.243
13	-	915	-	-	0	-	0	-	0	12/0.182
TX020994-Blank-2										
2	-	0	-	-	0	-	0	-	406	26/0.225
4	-	0	-	-	717	-	0	-	93	20/0.207
5	-	0	-	-	0	-	1518	-	156	32/0.213
13	-	926	-	-	0	-	0	-	0	13/0.164

NOTE: Samples counted about 5 days after separation therefore some Ac-225 or Ra-224 may be in Pu-236 peak, no adjustments made.
count time: 399 ± 1 min., sulfate fume, $TiCl_3$ in #5 only

Th-232 (Th-07F) 2.99E1 ± 0.10 dpm/ml, 100% yield: 5971 ± 299 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml,100% yield: 7585 ± 304 counts
Pu-239 (Pu-236A1) 2.93E1 ± 0.10 dpm/ml, 100% yield: 5845 ± 292 counts
U-238 (U-12) 4.83E1 ± 0.10 dpm/ml, 100% yield: 9636 ± 386 counts
Am-241 (Am-06D) 1.05E2 ± 0.10 dpm/ml, 100% yield: 8353 ± 419 counts
Th-229 (Th-08B) 4.15E1 ± 0.10 dpm/ml, 100% yield: 8275 ± 415 counts
Am-243 (Am-09) 3.8E1 ± 0.10 dpm/ml, 100% yield: 7571 ± 379 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml, 100% yield(decayed): 4449 ± 267 counts
U-232 (U-05AA3) 4.86E1 ± 0.10 dpm/ml, 100% yield(decayed): 8997 ± 540 counts

TABLE LXXXVI

INDEPENDENT SEPARATION PERFORMED BY MR.TIAM XIE
EXPERIMENT TX020994
PERCENT RECOVERY

EXP Step	Am 241 (±9%)	Am 243 (±9%)	Np 237 (±9%)	Pu 239 (±9%)	Pu 236 (±15%)	U 238 (±9%)	U 232 (±10%)	Th 232 (±9%)	Th 229 (±9%)
				% Recovery					
TX020994-1									
CHLORIDE Column									
2	0	0	0	0	0	0	0	88	97
4	0	0	81	84	91	0	0	5	0
5	0	0	0	0	0	91	86	0	0
NITRATE Column (after calcium oxalate coprecipitation)									
13	80	87	0	0	0	0	0	0	0
TX020994-2									
CHLORIDE Column									
2	0	0	0	0	0	0	0	94	92
4	0	0	81	84	93	0	0	6	0
5	0	0	0	0	0	74	73	0	0
NITRATE Column (after calcium oxalate coprecipitation)									
13	86	81	0	0	0	0	0	0	0
TX020994-3									
CHLORIDE Column									
2	0	0	0	0	0	0	0	81	86
4	0	0	99	101	107	0	0	8	0
5	0	0	0	0	0	90	80	0	0
NITRATE Column (after calcium oxalate coprecipitation)									
13	59	60	0	0	0	0	0	0	0
TX020994-4									
CHLORIDE Column									
2	0	0	0	0	0	0	0	92	92
4	0	0	82	95	92	0	0	7	0
5	0	0	0	0	0	87	86	0	0
NITRATE Column (after calcium oxalate coprecipitation)									
13	84	82	0	0	0	0	0	0	0
TX020994-Blank-1									
CHLORIDE Column									
2	-	0	-	-	0	-	0	-	60
4	-	0	-	-	82	-	0	-	7
5	-	0	-	-	0	-	66	-	0

TABLE LXXXVI (CONTINUED)

EXP Step	Am 241 (±9%)	Am 243 (±9%)	Np 237 (±9%)	Pu 239 (±9%)	Pu 236 (±15%)	U 238 (±9%)	U 232 (±10%)	Th 232 (±9%)	Th 229 (±9%)
				% Recovery					
NITRATE Column (after calcium oxalate coprecipitation)									
13	-	66	-	-	0	-	0	-	0
TX020994-Blank-2									
CHLORIDE Column									
2	-	0	-	-	0	-	0	-	22
4	-	0	-	-	77	-	0	-	4
5	-	0	-	-	0	-	79	-	9
NITRATE Column (after calcium oxalate coprecipitation)									
13	-	74	-	-	0	-	0	-	0

Procedure:
CHLORIDE Column:
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2(15 ml),
10.0 ml 5.0-M HCl,
Step 2. 6.0 ml 1.30-M HCl,(Th fraction)
Step 3. 3.0 ml 1.30-M HCl,
Step 4. 15.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl (Np/Pu fraction)
Step 5. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$ (U fraction)
10.0 ml 0.10-M HCl
Step 6. wash column with 10 ml 0.10-M HCl, store
NITRATE Column:
Step 10. coppt eluate form step 1, using calcium oxalate, redissolved in 3.0-M nitric acid
Step 11. load solution form step 10 in 3.0-M HNO_3 plus solid ascorbic acid
rinse 5 ml 3.0-M HNO_3
Step 12. 1.0 ml 8.0-M HCl
Step 13 15.0 ml 3.0-M HCl (Am fraction)
Step 14 10.0 ml 1.0-M HCl,

NOTE:
- counted about 5 days after separation
- some Ac-225 or Ra-224 may be in Pu-236 peak, no adjustments made

TABLE LXXXVII

SEPARATION OF ACTINIDE IN THE PRESENCE
OF VARIOUS SOIL MATRIX IONS
EXPERIMENT RB051693

EXP Step	Am-241 Counts (total) (±1.65√c)	U-238 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Am 241 dpm	U 238 dpm	Am 241 %Recovery	U 238 %Recovery
3	0	1398	9/0.167	0	42±4	0	83±10

Soil Sample Matrix:
Weston (1.0g soil): U-238 (2.5 dpm), U-234 (2.3 dpm) [IT]

RB051693
count time: 200 ± 1 min.
(sulfate fume)
(22°C)
($TiCl_3$)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 2982 ± 149 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 9660 ± 387 counts
+ 500 ± 50 dpm (soil)

Procedure:
Step 1. loaded mixture in 3.0-M HCl(13ml)
5.0 ml 3.0-M HCl
Step 2. 20 ml 1.0-M HCl
Step 3. 15 ml 0.1 M $NH_4HC_2O_4$

TABLE LXXXVIII

SEPARATION OF ACTINIDE IN THE PRESENCE
OF VARIOUS SOIL MATRIX IONS
EXPERIMENT RB052793

EXP Step	Am-241 Counts (total) (±1.65√c)	Th-232 Counts (total) (±1.65√c)	U-238 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Am 241	Th 232 %Recovery	U 238
2	0	1243	0	19/0.233	0	38±4	0
3	0	33	0	20/0.223	0	1±1	0
4	0	30	4571	21/0.242	0	1±1	58±6
5	0	7	124	22/0.196	0	0	2±1

Soil Sample Matrix: Fernald, OH (2.0 g soil)
U-238 (28.8 dpm/g), U-234 (27.5 dpm/g)[IT]
Th-232 (1.7 dpm/g), Th-230 (5.1 dpm/g)[IT]

RB052793
count time: 399 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 6107 ± 308 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 5945 ± 298 counts
Th-232 (Th-07E) 2.99E1 ± 0.10 dpm/ml
100% yield: 5965 ± 300 counts
+ 2034 ± 204 (soil)
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 9636 ± 388 counts
+ 23040 ± 2310? (soil)

Procedure:
Step 1. loaded sample mixture in 5.0-M HCl and H2O2(15ml),
Slowed flow rate, 10 ml 5.0-M HCl
Step 2. 5.0 ml 1.0-M HCl, Slowed flow rate
Step 3. 17.0 ml 1.0-M HCl,
Step 4. 5.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 5. 15.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl, 10 ml 3.0-M HCl,
10 ml 1.0-M HCl

TABLE LXXXIX

SEPARATION OF ACTINIDE IN THE PRESENCE
OF VARIOUS SOIL MATRIX IONS
EXPERIMENT RB061893

EXP Step	Am-241 Counts (total) ($\pm 1.65\sqrt{c}$)	Th-243 Counts (total) ($\pm 1.65\sqrt{c}$)	U-238 Counts (total) ($\pm 1.65\sqrt{c}$)	Detector PIPS/ls #/Eff ($\pm 2\%$)	Am 241 %Recovery	Am 243 %Recovery	U 238 %Recovery
2	130	137	0	18/0.218	10±2	11±2	0
0	1476	1351	0	17/0.207	116±10	110±9	0

Soil Sample Matrix
Weston(2.0 g) U-238 (2.5 dpm/g), U-234 (2.3 dpm/g) [IT]
Mount, Ohio

RB061893
count time: 399 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 6107 ± 308 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 5945 ± 298 counts
U-238 1995 ± 200 (soil)

Procedure:
Step 1. loaded sample mixture in 3.0-M HNO_3 (8ml)- column yellow;
rinse 3.0-M HNO_3, 4ml, yellow elute off
switch 8.0-M HCl, 2 ml
Step 2. 12.0 ml 4.0-M HCl, Slowed flow rate
Step 3. 10.0 ml 1.0-M HCl
Step 0. Standard check: Standards wet ashed with H2O2, no column, NdF_3 ppt, alpha counted

TABLE XC

SEPARATION OF ACTINIDE IN THE PRESENCE
OF VARIOUS SOIL MATRIX IONS
EXPERIMENT RB060893

EXP Step	Am-241 Counts (total) (±1.65√c)	Th-232 Counts (total) (±1.65√c)	U-238 Counts (total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Am 241 %Recovery	Th 232 %Recovery	U 238 %Recovery
RB060893 soil#2							
2	0	1026*	0	17/0.207	0	83±7	0
3	0	3	2166*	25/0.206	0	0	80±9
8	259	0	0	21/0.242	35±5	0	0
RB060893 soil#7							
2	0	900***	737***	19/0.233	0	89±7	4±1
3	0	2	1527**	26/0.215	0	0	46±6
8	344	0	0	22/0.196	59±7	0	0
RB060893 soil#12							
2	0	1309**	3	20/0.224	0	70±6	0
3	0	20	1222*	27/0.236	0	0	78±9
8	363	0	0	23/0.214	57±7	0	0

* 1/3 of sample volume counted
** 1/20 of sample volume counted
***1/4 of sample volume counted

Separating Am from high levels of Th and/or U

Nuclide Ratio:
2 15 dpm Am-241 : 89 dpm Th-232 : 198 dpm U-232
7 15 dpm Am-241 : 86 dpm Th-232 : 1550 dpm U-232
#12 15 dpm Am-241 : 831 dpm Th-232 : 100 dpm U-232

Soil Sample Matrix: (2.0 g)
2 Weston U-238 (2.5 dpm/g), U-234 (2.3 dpm/g)
7 Fernald U-238 (28.8 dpm/g), U-234 (27.5 dpm/g)
Th-232 (1.7 dpm/g), Th-230 (5.1 dpm/g)
#12 Denver U-238 (1.5 dpm/g), Am-241 (0.030 dpm/g)

RB060893 soil#2
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in 2,3)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 2980 ± 149 counts
Th-232 (Th-07E) 2.99E1 ± 0.10 dpm/ml
100% yield: 17840 ± 879 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 38640 ± 1546 counts + 1000 ± 100? (soil)

TABLE XC (CONTINUED)

RB060893 soil#7
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in 2,3)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 2980 ± 149 counts
Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 16620 ± 499 counts
+ 680 ± 68? counts (soil)
U-238 made from solid $UO_2(NO_3)_2 \cdot 6H_2O$
0.00285 grams, 195164 ± 9759 counts*
+ 115200 ± 11520? counts (soil)

* 1/30 of sample measured PIPS/ls, 1501 total count, efficiency 0.223, count time 200 min., 976 dpm.

RB060893 soil#12
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in 2,3)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 3061 ± 154 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 2980 ± 149 counts
+ 12 ± 6? counts(soil)
Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 166200 ± 4986 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 19320 ± 773 counts
+ 600 ± 60? counts(soil)

Procedure:
Step 1. loaded sample mixture in 5.0-M HCl and H2O2 (10ml),
5 ml 5.0-M HCl (eluted solution is wet ashed and converted to 3.0-M HNO_3)
Step 2. Elute Th 20.0 ml 1.0-M HCl,
Step 3. Elute U 20.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 4. Convert column to HNO_3, eluted with 8.0-M HNO_3 and 3.0-M HNO_3, eluate tested negative for Cl^- using $AgNO_3$
Step 5. reloaded step 1 eluate in HNO_3 after addition of ascorbic acid (tested sample for iron(III) ions with ammonium thiocyanate)
Step 6 10 ml 3.0-M HNO_3, 10 ml 2.0-M HNO_3/0.05 oxalic acid
Step 7. Switch to HCl, 2.0 ml 9.0-M HCl
Step 8 Elute Am 20.0 ml 4.0-M HCl

TABLE XCI

SEPARATION OF ACTINIDE IN THE PRESENCE
OF VARIOUS SOIL MATRIX IONS
EXPERIMENT RB070193

EXP Step	Am 241	Np 237 Counts (total) (±1.65√c)	Pu 239	U 238	Th 232 #/Eff	Detector PIPS/ts (±2%)	Am 241 (±9%)	Np 237 % Recovery (±9%)	Pu 239 (±9%)	U 238 (±9%)	Th 232 (±10%)
Soil # 13											
2	0	34*	0	0	555	22/0.196	0	5	0	0	73
3	0	376	281	0	10	23/0.214	0	46	32	0	1
4	0	26	405	0	0	25/0.206	0	3	47	0	0
5	0	17	9	0	0	26/0.215	0	2	1	0	0
6	0	0	0	29	0	28/0.223	0	0	0	3	0
7	0	0	0	0	0	29/0.227	0	0	0	0	0
10	(nitrate column not performed due to technical error)										
Soil # 14											
2	0	20*	0	0	695	30/0.219	0	2	0	0	82
3	0	699	288	0	0	32/0.235	0	79	30	0	0
4	0	11	460	0	0	18/0.218	0	1	51	0	0
5	0	31	14	0	0	21/0.242	0	1	1	0	0
6	0	0	0	566	0	22/0.196	0	0	0	70	0
7	0	0	0	12	0	23/0.214	0	0	0	1	0
10	817	0	0	0	0	26/0.215	93	0	0	0	0
11	0	0	0	0	0	28/0.223	0	0	0	0	0

*estimated due to energy region overlap of Np-237 and Th-230

Soil Samples: #13,14 Denver(2.0g): U-238 (1.5 dpm), Am-241 (0.030 dpm) [IT]

RB070193
count time: 200 ± 1 min.
(sulfate fume)
(22°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4082 ± 205 counts
+ 6 ± 3 counts (soil)
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 3864 ± 155 counts
+ 300 ± 30 counts (soil)
Th-232 (Th-07E) 3.0E1 ± 0.10 dpm/ml
100% yield: 3888 ± 195 counts

Calcium Oxalate Procedure
1. Diluted soil sample to 100 ml with deionized HOH
2. Added 80 ml saturated oxalic acid solution (10g/100 ml)
3. Added 2.0 ml Ca carrier (50 mg/ml Ca in 0.40-M HNO_3)
4. Adjust pH to 1.6-1.7 with NH_4OH
5. Sat 2.0 hrs., transferred sample to 200 ml centrifuge tube, centrifuge, decant, dissolve ppt in concentrated HNO_3
6. Transfer to glass beaker, wet ash, muffle 475°C, 30 min., cool
7. Dissolve sample in HNO_3 and H2O2, wet ash, repeat twice more
8. Sample dissolved in 5.0-M HCl and 15 d H2O2

TABLE XCI (CONTINUED)

Column Procedure: For both soil samples
CHLORIDE Column:
Step 1. loaded sample mixture in 5.0-M HCl + 15 d H2O2(11 ml), 5.0 ml 5.0-M HCl
Step 2. 5.0 ml 1.0-M HCl,
Step 3. 15.0 ml 1.0-M HCl,
Step 4. 15.0 ml 0.10-M NH_4I in 1.0-M HCl
Step 5. 15.0 ml 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 6. 15.0 ml 0.1 M $NH_4HC_2O_4$ in 0.10-M HCl
Step 7. 15.0 ml 0.1 M $(NH_4)_2C_2O_4$ in deionized HOH
NITRATE Column:
Step 8. Wet ash eluate from step 1 with HNO_3 and H_2O_2, dissolve in 3.0-M HNO_3, add 2.0 ml 0.30-M ascorbic acid, tested for iron(III) with NH_4SCN, sat 2 min. load sample onto nitrate prepared column rinse 3.0 ml 3.0-M HNO_3
Step 9. 2.0 ml 8.0-M HCl
Step 10. 15.0 ml 3.0-M HCl
Step 11. 10.0 ml 0.1-M HCl

TABLE XCII

SEPARATION OF ACTINIDE IN THE PRESENCE
OF VARIOUS SOIL MATRIX IONS
EXPERIMENT RB082793
RAW DATA

EXP Step	Am 241	Am 243	Np 237	Pu 239	Pu 236	U 238	U 232	Th 232	Th 229	Cm 244	Detector PIPS/ls #/Eff (±2%)
					Counts (total) (±1.65√c)						
RB082793A Soil #1											
2	0	0	0	0	0	0	0	196	148	-	3/0.179
3	0	0	0	0	0	0	0	109	78	-	15/0.185
4	0	0	699	703	520	0	0	0	0	-	9/0.178
5	0	0	0	0	0	0	0	0	0	-	24/0.209
6	0	0	0	0	0	821	379	0	0	-	1/0.171
12	30	25	0	0	0	0	0	0	0	-	13/0.166
13	60	80	0	0	0	0	0	0	0	-	1/0.171
22	55	52	0	0	0	0	0	0	0	-	21/0.228
23	276	313	0	0	0	0	0	0	0	-	7/0.184
24	0	0	0	0	0	0	0	181	118	-	17/0.205
RB082793B Soil #2											
2	0	0	0	0	0	0	0	103	93	0	4/0.194
3	0	0	0	0	0	0	0	6	7	0	16/0.185
4	0	0	647	618	495	0	0	0	0	0	10/0.173
5	0	0	0	0	0	0	0	0	0	0	25/0.206
6	0	0	0	0	0	693	313	0	0	0	2/0.172
12	42	49	0	0	0	0	0	0	0	37	14/0.165
13	161	202	0	0	0	0	0	0	0	87	2/0.172
22	60	56	0	0	0	0	0	0	0	111	22/0.192
23	303	339	0	0	0	0	0	0	0	166	8/0.186
24	0	0	0	0	0	0	0	269	194	0	19/0.220
RB082793C Soil #5											
2	0	0	0	0	0	0	0	181	150	-	5/0.176
3	0	0	0	0	0	0	0	121	100	-	17/0.205
4	0	0	488	593	472	0	0	0	0	-	11/0.194
5	0	0	0	0	0	0	0	0	0	-	26/0.205
6	0	0	0	0	0	960	289	0	0	-	3/0.179
12	5	6	0	0	0	0	0	0	0	-	15/0.185
13	19	19	0	0	0	0	0	0	0	-	3/0.179
22	64	52	0	0	0	0	0	0	0	-	24/0.209
23	211	257	0	0	0	0	0	0	0	-	9/0.178
24	0	0	0	0	0	0	0	153	93	-	21/0.228
RB082793D Soil #6											
2	0	0	0	0	0	0	0	447	319	0	6/0.157
3	0	0	0	0	0	0	0	445	309	0	19/0.220
4	0	0	659	643	528	0	0	0	0	0	12/0.182
5	0	0	0	0	0	0	0	0	0	0	30/0.209
6	0	0	0	0	0	986	416	0	0	0	4/0.194
12	13	14	0	0	0	0	0	0	0	11	16/0.185
13	30	32	0	0	0	0	0	0	0	17	4/0.194
22	69	57	0	0	0	0	0	0	0	104	25/0.206
23		no results - precipitation error									10/0.194
24	0	0	0	0	0	0	0	13	7	0	22/0.192

TABLE XCII (CONTINUED)

EXP Step	Am 241	Am 243	Np 237	Pu 239	Pu 236	U 238	U 232	Th 232	Th 229	Cm 244	Detector PIPS/ls #/Eff (±2%)
					Counts (total) (±1.65√c)						
RB082793S No soil											
2	0	0	0	0	0	0	0	574	523	0	7/0.184
3	0	0	0	0	0	0	0	78	98	0	21/0.228
4	0	0	533	541	434	0	0	18	0	0	13/0.166
5	0	0	0	0	0	0	0	0	0	0	31/0.239
6	0	0	0	0	0	432	402	0	0	0	5/0.176
12	15	15	0	0	0	0	0	0	0	54	17/0.205
13	288	342	0	0	0	0	0	0	0	214	5/0.176
22	15	13	0	0	0	0	0	0	0	53	26/0.205
23	357	364	0	0	0	0	0	0	0	256	11/0.194
24	0	0	0	0	0	0	0	0	0	0	24/0.209
RB082793T No soil, Tracers only											
2	-	0	-	-	-	-	0	-	324	-	8/0.186
3	-	0	-	-	-	-	0	-	146	-	22/0.192
4	-	0	-	-	-	-	0	-	0	-	14/0.165
5	-	0	-	-	-	-	0	-	0	-	32/0.220
6	-	0	-	-	-	-	310	-	61	-	6/0.157
12	-	7	-	-	-	-	0	-	0	-	19/0.220
13	-	313	-	-	-	-	0	-	0	-	6/0.157
22	-	24	-	-	-	-	0	-	0	-	28/0.224
23	-	301	-	-	-	-	0	-	0	-	12/0.182
24	-	0	-	-	-	-	0	-	0	-	25/0.206

A: 2 g soil 81093-1 Tracers & spikes added during digestion
B: 2 g soil 81093-2 Tracers& spikes added after digestion
C: 2 g soil 81093-5 Tracers &spikes added during digestion
D: 2 g soil 81093-6 Tracers & spikes added after digestion
S: no soil, Tracers and spikes added
T: no soil, Tracers only (Cm in samples B, D, and S only)

Soil Sample Matrix:

1,2: Np/Pu < 2.7 dpm/2g Th-232 4.7 ± 1.1 dpm/2g (23%),
U-238 38.35 ± 8.44 dpm/2g (22%)

5,6: Np/Pu < 2.7 dpm/2g Th-232 7.77 ± 2.18 dpm/2g(28%),
U-238 55.5 ± 12.2 dpm/2g (22%), Ra-226 5.15 dpm/2g, Ra-228 9.5 dpm/2g

RB082793
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in #6 only)

Additional from Soil Samples:
1,2: Th-232 940 ± 217 counts
U-238 7670 ± 1688 counts
5,6: Th-232 1554 ± 436 counts
U-238 11100 ± 2442 counts

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 3864 ± 155 counts
Am-241 (Am-06A) 2.55E1 ± 0.10 dpm/ml
100% yield: 4072 ± 205 counts
Cm-244 (Cm-02) 3.20E1 ± 0.10 dpm/ml
100% yield(decayed): 3240 ± 205 counts
Th-229 (Th-09D) 4.01E1 ± 0.10 dpm/ml
100% yield: 4010 ± 201 counts
Am-243 (Am-08A) 4.97E2 ± 0.10 dpm/ml
100% yield: 3972 ± 199 counts
Pu-236 (Pu-19A) 3.44E1 ± 0.10 dpm/ml
100% yield(decayed): 3078 ± 154 counts
U-232 (U-05AA2) 4.86E1 ± 0.10 dpm/ml
100% yield(decayed): 3600 ± 180 counts

TABLE XCIII

SEPARATION OF ACTINIDE IN THE PRESENCE
OF VARIOUS SOIL MATRIX IONS
EXPERIMENT RB082793
PERCENT RECOVERY

EXP Step	Am 241 (±18%)	Am 243 (±18%)	Np 237 (±10%)	Pu 239 (±10%)	Pu 236 (±10%)	U 238 (±12%)	U 232 (±11%)	Th 232 (±18%)	Th 229 (±18%)	Cm 244 (±18%)
					% Recovery					
RB082793A Soil #1										
CHLORIDE Column										
2	0	0	0	0	0	0	0	21	21	-
3	0	0	0	0	0	0	0	12	10	-
4	0	0	102	94	95	0	0	0	0	-
5	0	0	0	0	0	0	0	0	0	-
6	0	0	0	0	0	41	61	0	0	-
NITRATE Column										
12	9	8	0	0	0	0	0	0	0	-
13	17	24	0	0	0	0	0	0	0	-
NITRATE Column (after calcium oxalate coprecipitation)										
22	12	12	0	0	0	0	0	0	0	-
23	73	86	0	0	0	0	0	0	0	-
24	0	0	0	0	0	0	0	35	29	-
RB082793B Soil #2										
CHLORIDE Column										
2	0	0	0	0	0	0	0	10	12	0
3	0	0	0	0	0	0	0	6	7	0
4	0	0	100	88	92	0	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	35	51	0	0	0
NITRATE Column										
12	12	14	0	0	0	0	0	0	0	13
13	45	59	0	0	0	0	0	0	0	30
NITRATE Column (after calcium oxalate coprecipitation)										
22	15	15	0	0	0	0	0	0	0	36
23	79	91	0	0	0	0	0	0	0	30
24	0	0	0	0	0	0	0	48	44	0
RB082793C Soil #5										
CHLORIDE Column										
2	0	0	0	0	0	0	0	18	21	-
3	0	0	0	0	0	0	0	10	12	-
4	0	0	68	75	79	0	0	0	0	-
5	0	0	0	0	0	0	0	0	0	-
6	0	0	0	0	0	36	45	0	0	-
NITRATE Column										
12	1	1	0	0	0	0	0	0	0	-
13	5	5	0	0	0	0	0	0	0	-
NITRATE Column (after calcium oxalate coprecipitation)										
22	15	13	0	0	0	0	0	0	0	-
23	58	73	0	0	0	0	0	0	0	-
24	0	0	0	0	0	0	0	24	20	-

TABLE XCIII (CONTINUED)

EXP Step	Am 241 (±18%)	Am 243 (±18%)	Np 237 (±10%)	Pu 239 (±10%)	Pu 236 (±10%)	U 238 (±12%)	U 232 (±11%)	Th 232 (±18%)	Th 229 (±18%)	Cm 244 (±18%)
					% Recovery					
RB082793D Soil #6										
CHLORIDE Column										
2	0	0	0	0	0	0	0	50	51	0
3	0	0	0	0	0	0	0	36	35	0
4	0	0	95	85	94	0	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	34	59	0	0	0
NITRATE Column										
12	3	4	0	0	0	0	0	0	0	4
13	30	32	0	0	0	0	0	0	0	4
NITRATE Column (after calcium oxalate coprecipitation)										
22	16	14	0	0	0	0	0	0	0	31
23	no results									
24	0	0	0	0	0	0	0	1	1	0
RB082793S No soil										
CHLORIDE Column										
2	0	0	0	0	0	0	0	77	72	0
3	0	0	0	0	0	0	0	8	11	0
4	0	0	84	78	84	0	0	3	0	0
5	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	63	63	0	0	0
NITRATE Column										
12	4	4	0	0	0	0	0	0	0	16
13	80	98	0	0	0	0	0	0	0	74
NITRATE Column (after calcium oxalate coprecipitation)										
22	4	3	0	0	0	0	0	0	0	16
23	90	95	0	0	0	0	0	0	0	80
24	0	0	0	0	0	0	0	0	0	0
RB082793T No soil, Tracers only										
CHLORIDE Column										
2	-	0	-	-	-	-	0	-	44	-
3	-	0	-	-	-	-	0	-	19	-
4	-	0	-	-	-	-	0	-	0	-
5	-	0	-	-	-	-	0	-	0	-
6	-	0	-	-	-	-	55	-	10	-
NITRATE Column										
12	-	2	-	-	-	-	0	-	0	-
13	-	100	-	-	-	-	0	-	0	-
NITRATE Column (after calcium oxalate coprecipitation)										
22	-	5	-	-	-	-	0	-	0	-
23	-	83	-	-	-	-	0	-	0	-
24	-	0	-	-	-	-	0	-	0	-

A: 2 g soil 81093-1 Tracers and spikes added during digestion
B: 2 g soil 81093-2 Tracers and spikes added after digestion
C: 2 g soil 81093-5 Tracers and spikes added during digestion
D: 2 g soil 81093-6 Tracers and spikes added after digestion
S: no soil, Tracers and spikes added
T: no soil, Tracers only
(Cm in samples B, D, and S only)

TABLE XCIII (CONTINUED)

Procedure:
CHLORIDE Column:
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2 (50-60 ml),
4.0 ml 3.0-M HCl,
Step 2. 4.0 ml 1.30-M HCl,
Step 3. 3.0 ml 1.30-M HCl,
Step 4. 20.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 5. 10.0 ml 0.50-M HCl,
Step 6. 40.0 ml 0.10-M $(NH_4)_2C_2O_4$
NITRATE Column:
Step 10. The load elution from step#1 was divided into two equal samples.
The first half was converted to nitric acid and loaded onto a column.
The second half was coppt using calcium oxalate, redissolved in nitric acid and loaded onto a column.
Step 11. load first half in 3.0-M HNO_3 plus solid ascorbic acid rinse 10 ml 3.0-M HNO_3
Step 12. 7 ml 2.0-M HNO_3 plus 0.05 $H_2C_2O_4$
2.0 ml 8.0-M HCl
Step 13 20 ml 3.0-M HCl

Step 21. load second half in 3.0-M HNO_3 plus solid ascorbic acid
rinse 10 ml 3.0-M HNO_3
Step 22. 7 ml 2.0-M HNO_3 plus 0.05-M $H_2C_2O_4$
2.0 ml 8.0-M HCl
Step 23 25 ml 1.0-M HCl,
(NOTE) 1-2 ml of 1.0-M HCl was discarded prior to Step 24
Step 24 10 ml 0.5-M HCl

TABLE XCIV

SEPARATION OF ACTINIDE IN THE PRESENCE
OF VARIOUS SOIL MATRIX IONS
EXPERIMENT RB071993 A,B

EXP Step	Am 241	Np 237 Counts (total) (±1.65√c)	Pu 239	U 238	Th 232	Detector PIPS/ls #/Eff (±2%)	Am 241 (±12%)	Np 237 % Recovery (±10%)	Pu 239 (±10%)	U 238 (±14%)(	Th 232 ±10%)
RB071993A											
2	0	30*	29	0	625	1/0.171	0	5	4	0	81
3	0	467*	158	0	110	2/0.172	0	72	22	0	14
4	0	178	481	0	0	3/0.179	0	26	65	0	0
5	0	2	3	0	2	4/0.194	0	0	0	0	0
6	0	0	0	1209	0	5/0.176	0	0	0	71	0
7	0	0	0	41	0	6/0.157	0	0	0	3	0
4n	617	0	0	0	0	19/0.220	69	0	0	0	0
RB071993B											
2	0	0	0	0	0	7/0.184	0	0	0	0	0
3	0	3	8 0	5	8/0.186	0	0	1	0	0	
4	0	3	21	0	35	9/0.178	0	0	3	0	4
5	0	5*	27	0	121	10/0.173	0	1	4	0	16
6	0	93*	62	0	467	11/0.194	0	13	8	0	54
7	0	233	199	0	0	12/0.182	0	34	26	0	0
8	0	110	212	0	0	13/0.166	0	17	31	0	0
9	0	0	0	0	0	15/0.186	0	0	0	0	0
10	0	0	0	1268	0	16/0.185	0	0	0	71	0
11	0	0	0	87	0	17/0.205	0	0	0	4	0
4n	505	0	0	0	0	20/0.212	59	0	0	0	0

* estimated due to energy region overlap of Np-237 and Th-230.

Soil Sample Matrix
#8,9 Fernald U-238 (28.8 dpm), U-234 (27.5 dpm),
Th-232 (1.7 dpm), Th-230 (5.1 dpm)

RB071993A,B Soil #8,9
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4082 ± 205 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 3864 ± 155 counts
+ 5760 ± 576 (soil)
Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
+ 340 ± 34 (soil)

Procedure: Sample A - Soil # 8
CHLORIDE column
Step 1. loaded sample mixture in 5.0-M HCl, 15d H2O2 (20 ml),
Step 2. 5.0 ml 1.0-M HCl,
Step 3. 6.0 ml 1.0-M HCl,

TABLE XCIV (CONTINUED)

Step 4. 20.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 5. 10.0 ml 1.0-M HCl
Step 6. 20.0 ml 0.1 M $(NH_4)_2C_2O_4$, 3.0 ml 1.0-M HCl
Step 7. Stored column overnight, drained column, eluted with 15.0 ml 0.1 M $(NH_4)_2C_2O_4$, 5.0 ml 1.0-M HCl
Procedure: Sample B - Soil # 9 (Same except use 2.0-M HCl)
CHLORIDE column
Step 1. loaded sample mixture in 5.0-M HCl, 15d H2O2 (20 ml),
Step 2. 5.0 ml 2.0-M HCl,
Step 3. 5.0 ml 2.0-M HCl,
Step 4. 5.0 ml 2.0-M HCl,
Step 5. 5.0 ml 2.0-M HCl,
Step 6. 5.0 ml 1.0-M HCl,
Step 7. 6.0 ml 1.0-M HCl,
Step 8. 20.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 9. 10.0 ml 1.0-M HCl
Step 10. 15.0 ml 0.1 M $(NH_4)_2C_2O_4$, 3.0 ml 1.0-M HCl
Step 11. Stored column overnight, drained column, eluted with 15.0 ml 0.1 M $(NH_4)_2C_2O_4$, 5.0 ml 1.0-M HCl
Procedure: Samples A and B the same
NITRATE column
Step 1n. Calcium Oxalate ppt of eluate from step #1, sat overnight, muffle 30 min. 475°C
Step 2n. Load 3.0-M HNO_3 (no ascorbic acid added) (50 ml) rinse 5.0 ml 3.0-M HNO_3
Step 3n. 2.0 ml 8.0-M HCl
Step 4n. 15 ml 3.0-M HCl

TABLE XCV

SEPARATION OF ACTINIDE IN THE PRESENCE
OF VARIOUS SOIL MATRIX IONS
EXPERIMENT RB072093

EXP Step	Am 241	Np 237	Pu 239	U 238	Th 232	Detector PIPS/ls #/Eff (±2%)	Am 241 (±12%)	Np 237	Pu 239	U 238	Th 232
		Counts (total) (±1.65√c)						% Recovery (±10%)	(±10%)	(±14%)	(±10%)
2	0	2	12	0	4	18/0.225	0	0	1	0	0
3	0	3	23	0	7	19/0.220	0	0	3	0	0
4	0	157*	109	0	807	20/0.212	0	20	12	0	85
5	0	352	270	0	2	21/0.228	0	41	27	0	0
6	0	143	324	0	0	22/0.192	0	20	41	0	0
7	0	0	0	0	0	23/0.212	0	0	0	0	0
8	0	0	0	1219	0	24/0.209	0	0	0	61	0
4n	790	0	0	0	0	21/0.228	85	0	0	0	0
5n	0	0	0	0	0	26/0.205	0	0	0	0	0
6n	0	0	0	0	0	30/0.209	0	0	0	0	0

* estimated due to energy region overlap of Np-237 and Th-230.

Soil Sample: #10 Fernald U-238 (28.8 dpm), U-234 (27.5 dpm)
Th-232 (1.7 dpm), Th-230 (5.1 dpm)

RB072093 Soil #10
count time: 200 ± 1 min.
(sulfate fume)(22°C)
($TiCl_3$ in all)

Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4082 ± 205 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4160 ± 167 counts
U-238 (U-11) 4.83E1 ± 0.10 dpm/ml
100% yield: 3864 ± 155 counts
+ 5760 ± 576 (soil)
Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4155 ± 208 counts
+ 340 ± 34 (soil)

Procedure: Sample A - Soil # 10
CHLORIDE column
Step 1. loaded sample mixture in 5.0-M HCl plus 10 drops H2O2 (15 ml), 5.0 ml 5.0-M HCl,
Step 2,3. 5.0 ml 2.0-M HCl,
Step 4,5. 5.0 ml 1.0-M HCl,
Step 6. 25.0 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 7. 15.0 ml 1.0-M HCl
Step 8. 25.0 ml 0.1 M $(NH_4)_2C_2O_4$, 5.0 ml 1.0-M HCl
NITRATE column
Step 1n. Calcium Oxalate ppt of eluate from step #1, sat overnight, muffle 30 min. 475°C
Step 2n. Load 3.0-M HNO_3 (no ascorbic acid added) (50 ml) rinse 5.0 ml 3.0-M HNO_3
Step 3n. 2.0 ml 8.0-M HCl
Step 4n. 15 ml 3.0-M HCl
Step 5n. 10.0 ml 1.0-M HCl, 10 ml 0.10-M NH_4I and 0.030-M $H_2C_2O_4$ in 1.0-M HCl
Step 6n. 20.0 ml 0.1 M $(NH_4)_2C_2O_4$, 5.0 ml 1.0-M HCl

TABLE XCVI

SEPARATION OF ACTINIDE FROM HARD WATER MATRICES
EXPERIMENT RB111993
RAW DATA

EXP Step	Am 241	Am 243	Np 237	Pu 239	Pu 236	U 238	U 232	Th 232	Th 229	Cm 244	Detector PIPS/ls #/Eff (±2%)
					Counts (total) (±1.65√c)						
RB111993-Al											
2	0	0	927*	1046	623	0	0	499	927*	0	1/0.169
3^	0	0	95	63	32	0	0	3	0	0	24/0.192
4	0	0	77	130	68	0	0	0	0	0	7/0.188
5	0	0	0	25	0	10	15	6	0	0	12/0.216
13^**	100	100	0	0	0	0	0	188	163	107	1/0.168
14^	0	0	0	0	0	0	0	6	3	0	7/0.181
15^	0	0	0	2	6	0	0	5	5	0	12/0.184
16^	0	0	0	0	0	4	1	0	0	0	17/0.206
RB111993-Blank											
2	0	0	0	0	0	0	0	1290	1002	0	2/0.174
3^	0	0	65*	2	3	0	0	62	65*	0	25/0.197
4	0	0	1107	1452	816	0	0	166	0	0	8/0.180
5	0	0	0	113	65	711	631	5	0	0	13/0.167
13^**	553	729	0	0	0	0	0	0	0	457	2/0.180
14^	0	0	0	0	0	0	0	2	1	0	8/0.178
15^	0	0	6	12	5	0	0	0	0	0	13/0.165
16^	0	0	0	0	0	67	47	0	0	0	19/0.217
RB111993-Ca											
2	0	0	1388*	77	0	0	0	1271	1388*	0	3/0.170
3^	0	0	67	19	16	0	0	18	0	0	27/0.213
4	0	0	61*	1317	673	0	0	56	61*	0	9/0.178
5	0	0	0	100	61	706	573	33	0	0	14/0.160
13^**	80	80	77	0	0	0	0	0	0	25	3/0.169
14^	0	0	0	0	0	0	0	9	8	0	9/0.183
15^	0	0	0	29	18	13	0	4	0	0	14/0.167
16^	0	0	0	0	0	61	71	0	0	0	20/0.216
RB111993-Fe											
2	0	0	0	182	108	0	0	104	96	0	4/0.186
3^	0	0	3	45	35	0	0	3	0	0	31/0.234
4	0	0	30	909	430	0	0	0	0	0	10/0.170
5	0	0	0	8	2	108	76	2	0	0	15/0.190
13^**	513	799	0	0	0	0	0	0	0	435	4/0.184
14^	0	0	0	0	0	0	0	549	401	0	10/0.177
15^	0	0	0	225	74	68	0	41	0	0	15/0.185
16^	0	0	0	0	0	17	11	5	0	0	21/0.216
RB111993-Mg											
2	0	0	1586*	0	0	0	0	1244	1586*	0	5/0.180
3^	0	0	68*	117	59	0	0	36	68*	0	32/0.211
4	0	0	55	1134	516	0	0	30	0	0	11/0.193
5	0	0	0	76	22	674	535	33	0	0	16/0.200

TABLE XCVI (CONTINUED)

EXP Step	Am 241	Am 243	Np 237	Pu 239	Pu 236 Counts (±1.65√c)	U 238 (total)	U 232	Th 232	Th 229	Cm 244	Detector PIPS/s #/Eff (±2%)
13^	9	11	0	0	0	0	0	5	0	5	5/0.182
14^	0	0	0	0	0	0	0	0	9	0	11/0.177
15^	0	0	1	5	9	0	0	0	0	0	16/0.179
16^	0	0	0	0	0	42	44	3	0	0	22/0.175

* Th-229 and Np-237 combined, estimated percentages
** poor resolution
^ 200 min count time

10.0g Simulated Hard Water Samples:
Solution made by dissolving the following into 200 ± 50 ml of 5.0-M HCl. The amounts used were calculated to yield 10 grams of the metal ion in solution.

83.61 g of MgCl2.6 HOH
27.69 g of CaCl2
89.44 g AlCl3.6HOH (warmed to dissolve)
48.45 g FeCl3.6HOH
blank 5.0-M HCl about 200 ml

All samples spiked and traced after they were dissolved.
Initial flow rates 1-2 ml /min (flow rates varies over time)

Blank 2-3 ml/min flow rates, total HCl-load time 5.5 hrs
Al 3-0.5 ml/min flow rates, total HCl-load time 10 hrs
Ca 3-1 ml/min flow rates, total HCl-load time 10.5 hrs
Fe 3-0.5 ml/min flow rates, total HCl-load time 10 hrs
Mg 3-0.5 ml/min flow rates, total HCl-load time 7.5 hrs

Columns capped and finished elutions the next day

RB111993
count time: 399 ± 1 min.
(sulfate fume)
($TiCl_3$ in U fraction only)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 9947 ± 498 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 7585 ± 304 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 9959 ± 399 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 5758 ± 231 counts
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 5089 ± 255 counts
Cm-244 (Cm-02) 3.20E2 ± 0.10 dpm/ml
100% yield(decayed): 5351 ± 322 counts
Th-229 (Th-09F) 3.96E1 ± 0.10 dpm/ml
100% yield: 7900 ± 396 counts
Am-243 (Am-08A) 4.965E2 ± 0.10 dpm/ml
100% yield: 7924 ± 397 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 4588 ± 275 counts
U-232 (U-05AA3) 4.86E1 ± 0.10 dpm/ml
100% yield(decayed): 5398 ± 324 counts

TABLE XCVII

SEPARATION OF ACTINIDE FROM HARD WATER MATRICES
EXPERIMENT RB111993
PERCENT RECOVERY

EXP Step	Am 241 (±20%)	Am 243 (±20%)	Np 237 (±20%)	Pu 239 (±10%)	Pu 236 (±11%)	U 238 (±20%)	U 232 (±20%)	Th 232 (±12%)	Th 229 (±20%)	Cm 244 (±20%)
					% Recovery					
RB111993-Al										
CHLORIDE Column										
2	0	0	40*	62	80	0	0	29	30*	0
3	0	0	13	7	7	0	0	0	0	0
4	0	0	5	7	7	0	0	0	0	0
5	0	0	0	1	0	1	1	0	0	0
NITRATE Column (after calcium oxalate coprecipitation)										
13	22	14	0	0	0	0	0	22	24	24
14	0	0	0	0	0	0	0	0	0	0
15	0	0	0	0	0	0	0	0	0	0
16	0	0	0	0	0	0	0	0	0	0
Total %	22	14	58	77	94	1	1	51	54	24
RB111993-Blank										
CHLORIDE Column										
2	0	0	0	0	0	0	0	76	75	0
3	0	0	4*	0	0	0	0	6	5*	0
4	0	0	81	81	98	0	0	9	0	0
5	0	0	0	7	8	72	69	0	0	0
NITRATE column (after calcium oxalate coprecipitation)										
13	121	102	0	0	0	0	0	0	0	95
14	0	0	0	0	0	0	0	0	0	0
15	0	0	0	1	1	0	0	0	0	0
16	0	0	0	0	0	11	8	0	0	0
Total %	121	102	85	89	117	83	77	91	80	95
RB111993-Ca										
CHLORIDE Column										
2	0	0	28*	5	0	0	0	75	75*	0
3	0	0	9	2	3	0	0	2	0	0
4	0	0	2*	73	80	0	0	3	2*	0
5	0	0	0	6	8	77	66	2	0	0
NITRATE Column (after calcium oxalate coprecipitation)										
13	18	12	12	0	0	0	0	0	0	5
14	0	0	0	0	0	0	0	0	0	0
15	0	0	4	2	2	0	0	0	0	0
16	0	0	0	0	0	10	12	0	0	0
Total %	18	12	46	87	92	87	78	81	77	5
RB111993-Fe										
CHLORIDE Column										
2	0	0	0	10	12	0	0	5	6	0
3	0	0	0	4	7	0	0	0	0	0
4	0	0	2	54	55	0	0	0	0	0
5	0	0	0	0	0	10	7	0	0	0

TABLE XCVII (CONTINUED)

EXP Step	Am 241 (±20%)	Am 243 (±20%)	Np 237 (±20%)	Pu 239 (±10%)	Pu 236 (±11%)	U 238 (±20%)	U 232 (±20%)	Th 232 (±12%)	Th 229 (±20%)	Cm 244 (±20%)
					% Recovery					
NITRATE Column (after calcium oxalate coprecipitation)										
13	112	112	0	0	0	0	0	0	0	90
14	0	0	0	0	0	0	0	61	56	0
15	0	0	32	8	16	0	0	5	0	0
16	0	0	0	0	0	3	2	0	0	0
Total %	112	112	34	76	90	13	9	71	62	90
RB111993-Mg										
CHLORIDE Column										
2	0	0	40	0	0	0	0	69	70*	0
3	0	0	2	11	12	0	0	3	2	0
4	0	0	4	60	59	0	0	2	0	0
5	0	0	0	5	2	73	62	2	0	0
NITRATE Column (after calcium oxalate coprecipitation)										
13	2	2	0	0	0	0	0	1	0	2
14	0	0	0	0	0	0	0	0	1	0
15	0	0	0	1	2	0	0	5	0	0
16	0	0	0	0	0	8	9	0	0	0
Total %	2	2	46	77	77	73	71	82	73	2

* Th-229 and Np-237 combined, estimated percentages
** poor resolution
^ 200 min count time

10.0g Hard Water Samples

Procedure:
CHLORIDE Column:
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2 (200±50 ml),
5.0 ml 5.0-M HCl,
Step 2. 6.0 ml 1.30-M HCl,(Th fraction)
Step 3. 2.0 ml 1.30-M HCl,
Step 4. 15.0 ml 0.10-M NH_4I and 0.03-M $H_2C_2O_4$ in 0.50-M HCl (Np/Pu fraction)
10.0 ml 0.50-M HCl,
Step 5. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$ (U fraction)
10 ml 0.10-M HCl
NITRATE Column:
Step 10. coppt using calcium oxalate, redissolved in nitric acid
Step 11. loaded solution from step 10 onto column in 3.0-M HNO_3 plus solid
ascorbic acid (load vol: Al 13ml, Blank 13ml, Ca 62ml, Fe 12ml, Mg 44ml), rinse 10 ml 3.0-M HNO_3
Step 12. 1.0 ml 8.0-M HCl
Step 13 15 ml 3.0-M HCl (Am/Cm fraction)
Step 14 6.0 ml 1.30-M HCl
Step 15 15 ml 0.03-M H2C2O4
Step 16 30.0 ml 0.10-M $(NH_4)_2C_2O_4$

TABLE XCVIII

SEPARATION OF ACTINIDE FROM HARD WATER MATRICES
EXPERIMENT RB021594
RAW DATA

Column Exp step	Am 241	Am 243	Np 237	Pu 239	Pu 236	U 238	U 232	Th 232	Th 229	Detector PIPS/s #/Eff (±2%)
				Counts (total) (±1.65√c)						
RB021594 A	Blank, deionized HOH									
2	0	0	0	0	0	0	0	668	550	1/0.164
3	0	0	61*	9	0	0	0	15	61*	2/0.171
4	0	0	860*	749	349	0	0	10	860*	10/0.181
5	0	0	0	0	0	466	375	6	0	15/0.178
8^	2613	2497	0	0	0	0	0	0	0	25/0.200
9^	0	0	25**	11	0	15	^^	9	25**	29/0.219
RB021594 B	1.0 g Al ions									
2	0	0	0	0	0	0	0	750	567	4/0.188
3	0	0	63*	18	^^	0	0	16	63*	5/0.174
4	0	0	816*	860	355	0	0	4	0	11/0.177
5	0	0	0	0	10	473	436	20	^^	16/0.177
8^	2476	2188	0	0	0	0	0	0	0	26/0.225
9^	0	0	41**	7	^^	25	^^	6	41**	30/0.211
RB021594 C	1.0 g Ca ions									
2	0	0	0	0	0	0	0	678	534	6/0.153
3	0	0	50*	9	^^	0	0	22	50*	7/0.205
4	0	0	942*	847	405	0	0	11	0	12/0.182
5	0	0	0	0	0	506	440	5	0	1/0.164
8^	1318	1261	0	0	0	0	0	0	0	27/0.192
9^	0	0	36**	8	^^	34	^^	3	36**	31/0.243
RB021594 D	1.0 g Fe ions									
2	0	0	146*	10	^^	0	0	162	146*	8/0.160
3	0	0	45*	30	^^	0	0	10	45*	9/0.173
4	0	0	717	214	125	0	0	0	0	13/0.164
5	0	0	0	0	0	297	294	4	0	2/0.171
8^	2287	2241	0	0	0	0	0	0	0	28/0.222
9^	0	0	1260*	1155	387	0	0	1442	1260*	32/0.213

* contains counts from alpha decay of Th-229, Th-230, and Np-237. Estimated the amount of Th-229(equal to the percent recovery of Th-232), Th-230 (equal to 0.17 of Th-232 counts), and assigned the remaining counts to Np-237.

** contains counts from alpha decay of Th-229, Th-230, U-234, and Np-237. Estimated the amount of Th-229(equal to the percent recovery of Th-232), Th-230 (equal to 0.17 of Th-232 counts), U-234 (equal to U-238), and assigned the remaining counts to Np-237.

^ count time 399 ±1 min.

^^ unable to determine due to energy overlap with other nuclides

TABLE XCVIII (CONTINUED)

Simulated Hard Water (1.0g)

Four columns		LOAD TIME
A	blank, 5.0-M HCl	22
B	Al, 1.0 g metal, 8.94 g $AlCl_3 \cdot 6HOH$	152
C	Ca, 1.1 g metal, 3.083 g $CaCl_2$	35
D	Fe, 1.0 g metal, 4.845 g $FeCl_3 \cdot 6HOH$	55

RB021594
count time: 200 ± 1 min.
(sulfate fume)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 5703 ± 229 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 193 counts
Am-241 (Am-06D) 1.05E2 ± 0.10 dpm/ml
100% yield: 6300 ± 378 counts
Th-229 (Th-08B) 4.15E1 ± 0.10 dpm/ml
100% yield: 4152 ± 250 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 2230 ± 134 counts
U-232 (U-05AA3) 4.86E1 ± 0.10 dpm/ml
100% yield(decayed): 4505 ± 271 counts
Am-243 (Am-08A) 4.97E1 ± 0.10 dpm/ml
100% yield: 4966 ± 298 counts

Procedure:
CHLORIDE Column:
Step 1. loaded sample in 5.0-M HCl plus H_2O_2 (20-25 ml),
5.0 ml 5.0-M HCl
20.0 ml 3.0-M HCl
Step 2. 6.0 ml 1.30-M HCl
Step 3. 3.0 ml 1.30-M HCl
Step 4. 20.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 5. 40.0 ml 0.10-M $(NH_4)_2C_2O_4$
Step 6. 10 ml 0.10-M HCl
Calcium Oxalate Coprecipitation of eluate from step 1.
NITRATE column
Step 7. load 3.0-M HNO_3 plus solid ascorbic acid
5.0 ml 3.0-M HNO_3
1.0 ml 8.0-M HCl
Step 8. 15.0 ml 3.0-M HCl
Step 9. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$
10.0 ml 0.10-M HCl

TABLE XCIX

SEPARATION OF ACTINIDE FROM HARD WATER MATRICES
EXPERIMENT RB021594
PERCENT RECOVERY

Column Exp step	Am 241 (±9%)	Am 243 (±10%)	Np 237 (±15%)	Pu 239 (±9%)	Pu 236 (±12%)	U 238 (±11%)	U 232 (±12%)	Th 232 (±9%)	Th 229 (±15%)
					% Recovery				
RB021594 A	Blank, deionized HOH								
2	0	0	0	0	0	0	0	82	81
3	0	0	5*	1	0	0	0	2	2*
4	0	0	84*	83	86	0	0	1	1*
5	0	0	0	0	0	54	47	1	0
8^	104	125	0	0	0	0	0	0	0
9^	0	0	0	0	0	1	0	0	0
RB021594 B	1.0 g Al ions								
2	0	0	0	0	0	0	0	80	73
3	0	0	5*	2	0	0	0	2	2*
4	0	0	81*	97	90	0	0	0	0*
5	0	0	0	0	1	56	55	3	^^
8^	88	98	0	0	0	0	0	0	0
9^	0	0	0	0	0	1	0	0	0
RB021594 C	1.0 g Ca ions								
2	0	0	0	0	0	0	0	89	84
3	0	0	2*	1	0	0	0	2	2*
4	0	0	91*	93	100	0	0	1	1*
5	0	0	0	0	0	64	59	0	0
8^	55	66	0	0	0	0	0	0	0
9^	0	0	0	0	0	1	0	0	0
RB021594 D	1.0 g Fe ions								
2	0	0	1*	1	^^	0	0	20	20
3	0	0	4*	3	^^	0	0	1	1*
4	0	0	77	26	35	0	0	0	0
5	0	0	0	0	0	36	38	0	0
8^	82	102	0	0	0	0	0	0	0
9^	0	0	3*	55	40	0	0	68	68*

* contains counts from alpha decay of Th-229, Th-230, and Np-237. Estimated the amount of Th-229(equal to the percent recovery of Th-232), Th-230 (equal to 0.17 of Th-232 counts), and assigned the remaining counts to Np-237.
** contains counts from alpha decay of Th-229, Th-230, U-234, and Np-237. Estimated the amount of Th-229(equal to the percent recovery of Th-232), Th-230 (equal to 0.17 of Th-232 counts), U-234 (equal to U-238), and assigned the remaining counts to Np-237.
^ count time 399 ±1 min.
^^ unable to determine due to energy overlap with other nuclides
PROCEDURE: (Simulated Hard Water (1.0g)) CHLORIDE Column:
Step 1. loaded sample in 5.0-M HCl plus H_2O_2 (20-25 ml), 5.0 ml 5.0-M HCl, 20.0 ml 3.0-M HCl
Step 2. 6.0 ml 1.30-M HCl
Step 3. 3.0 ml 1.30-M HCl
Step 4. 20.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 5. 40.0 ml 0.10-M $(NH_4)_2C_2O_4$
Step 6. 10 ml 0.10-M HCl
Calcium Oxalate Coprecipitation of eluate from step 1., NITRATE column
Step 7. load 3.0-M HNO_3 plus solid ascorbic acid, 5.0 ml 3.0-M HNO_3, 1.0 ml 8.0-M HCl
Step 8. 15.0 ml 3.0-M HCl
Step 9. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$, 10.0 ml 0.10-M HCl

TABLE C

SEPARATION OF ACTINIDE FROM HARD WATER MATRICES
EXPERIMENT RB012994
RAW DATA

Column Exp step	Am 241	Am 243	Np 237	Pu 239	Pu 236 Counts (total) (±1.65√c)	U 238	U 232	Th 232	Th 229	Detector PIPS/ls #/Eff (±2%)
RB012994 A	Blank, deionized HOH									
2	0	0	0	0	0	0	0	746	615	1/0.161
3	0	0	30*	6	0	0	0	16	30*	9/0.170
4	0	0	1245*	1043	467	0	0	65	1245*	17/0.200
5	0	0	0	0	0	634	541	15	12	1/0.161
7	959	937	0	0	0	0	0	0	0	9/0.170
8	6	4	0	14	0	15	14	0	0	17/0.200
RB012994 B	0.10 g Al ions									
2	0	0	0	0	0	0	0	706	535	2/0.166
3	0	0	0	2	0	0	0	35	23	10/0.172
4	0	0	1125*	1025	486	0	0	93	1125*	18/0.211
5	0	0	0	16	0	543	430	21	14	2/0.166
7	842	945	0	0	0	0	0	0	0	10/0.172
8	4	2	0	5	0	33	18	0	0	18/0.211
RB012994 C	0.010 g Al ions									
2	0	0	0	0	0	0	0	685	488	3/0.170
3	0	0	19*	1	0	0	0	15	19*	11/0.173
4	0	0	1154*	1035	503	0	0	61	1154*	19/0.217
5	0	0	0	10	0	540	507	18	10	3/0.170
7	958	844	0	0	0	0	0	0	0	11/0.173
8	5	3	0	0	0	31	25	0	0	19/0.217
RB012994 D	0.10 g Ca ions									
2	0	0	0	0	0	0	0	712	552	4/0.187
3	0	0	20*	2	0	0	0	32	20*	12/0.182
4	0	0	1191*	982	489	0	0	106	1191*	20/0.215
5	0	0	0	0	0	585	471	20	15	4/0.187
7	1017	957	0	0	0	0	0	0	0	12/0.182
8	0	0	0	0	0	34	23	4	0	20/0.215
RB012994 E	0.50 g Ca ions									
2	0	0	0	0	0	0	0	672	509	5/0.180
3	0	0	29*	4	0	0	0	32	29*	13/0.162
4	0	0	1292*	1084	499	0	0	100	1292*	21/0.223
5	0	0	0	20	0	511	481	20	13	5/0.180
7	840	777	0	0	0	0	0	0	0	13/0.162
8	0	0	0	18	6	38	21	2	0	21/0.223
RB012994 F	0.10 g Fe ions									
2	0	0	0	0	0	0	0	697	561	7/0.188
3	0	0	48*	53	18	0	0	6	48*	15/0.181
4	0	0	1106**	705	329	21	11	0	0	22/0.177
5	0	0	0	0	0	569	495	3	0	7/0.188
7	1037	863	0	0	0	0	0	0	0	15/0.181
8	0	0	60*	151	54	6	0	10	60*	22/0.177

TABLE C (CONTINUED)

Column Exp step	Am 241	Am 243	Np 237	Pu 239	Pu 236	U 238	U 232	Th 232	Th 229	Detector PIPS/ls #/Eff (±2%)
					Counts (total) (±1.65√c)					
RB012994 G 0.010 g Fe ions										
2	0	0	0	0	0	0	0	746	593	8/0.169
3	0	0	38*	19	14	0	0	6	38*	16/0.175
4	0	0	1178	944	464	12	8	6	0	23/0.229
5	0	0	0	0	0	661	549	10	1	8/0.169
7	1036	919	0	0	0	0	0	0	0	16/0.175
8	0	0	32**	21	10	8	10	0	32*	23/0.229

* contains counts from alpha decay of Th-229, Th-230, and Np-237. Estimated the amount of Th-229 (equal to the percent recovery of Th-232), Th-230 (equal to 0.17 of Th-232 counts), and assigned the remaining counts to Np-237.

** contains U-234 and Np-237, used the amount of U-238 to estimate the amount of U-234 counts.

Simulated Hard Water (0.010 - 0.50 g)

7 columns		LOAD TIME
A	blank, 5.0-M HCl only plus tracers and spikes	81 min
B	Al, 0.10 g metal, 0.894 g $AlCl_3 \cdot 6HOH$	100
C	Al, 0.010 g metal, 0.0894 g $AlCl_3 \cdot 6HOH$	90
D	Ca, 0.10 g metal, 0.2769 g $CaCl_2$	91
E	Ca, 0.50 g metal, 1.385 g $CaCl_2$	80
F	Fe, 0.10 g metal, 0.485 g $FeCl_3 \cdot 6HOH$	82
G	Fe, 0.010 g metal, 0.0485 g $FeCl_3 \cdot 6HOH$	82

RB012994
count time: 200 ± 1 min.
(sulfate fume)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 5703 ± 229 counts
Pu-239 (Pu-13D) 8.32E1 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 193 counts
Am-241 (Am-06D) 1.05E2 ± 0.10 dpm/ml
100% yield: 6300 ± 378 counts
Th-229 (Th-08B) 4.15E1 ± 0.10 dpm/ml
100% yield: 4152 ± 250 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 2230 ± 134 counts
U-232 (U-05AA3) 4.86E1 ± 0.10 dpm/ml
100% yield(decayed): 4505 ± 271 counts
Am-243 (Am-08A) 4.97E1 ± 0.10 dpm/ml
100% yield: 4966 ± 298 counts

TABLE CI

SEPARATION OF ACTINIDE FROM HARD WATER MATRICES
EXPERIMENT RB012994
PERCENT RECOVERY

Column-Exp step	Am 241 (±9%)	Am 243 (±10%)	Np 237 (±15%)	Pu 239 (±9%)	Pu 236 (±11%)	U 238 (±10%)	U 232 (±10%)	Th 232 (±9%)	Th 229 (±15%)
				% Recovery					
RB012994 A Blank, deionized HOH									
2	0	0	0	0	0	0	0	93	92
3	0	0	2*	1	0	0	0	2	2*
4	0	0	104*	105	104	0	0	7	7*
5	0	0	0	0	0	82	75	2	2
7	88	109	0	0	0	0	0	0	0
8	0	0	0	1	0	2	2	0	0
RB012994 B 0.10 g Al ions									
2	0	0	0	0	0	0	0	85	78
3	0	0	0	0	0	0	0	4	2
4	0	0	86*	97	103	0	0	9	9*
5	0	0	0	0	0	65	58	3	2
7	88	101	0	0	0	0	0	0	0
8	0	0	0	0	0	3	2	0	0
RB012994 C 0.010 g Al ions									
2	0	0	0	0	0	0	0	81	69
3	0	0	0	0	0	0	0	2	2
4	0	0	88*	96	104	0	0	6	6*
5	0	0	0	0	0	66	66	2	1
7	89	99	0	0	0	0	0	0	0
8	0	0	0	0	0	3	3	0	0
RB012994 D 0.10 g Ca ions									
2	0	0	0	0	0	0	0	76	71
3	0	0	0	0	0	0	0	4	3
4	0	0	89*	92	102	0	0	10	10*
5	0	0	0	0	0	63	60	2	2
7	89	102	0	0	0	0	0	0	0
8	0	0	0	0	0	3	2	0	0
RB012994 E 0.50 g Ca ions									
2	0	0	0	0	0	0	0	75	68
3	0	0	0	0	0	0	0	4	4
4	0	0	94*	98	100	0	0	9	9*
5	0	0	0	2	0	59	60	2	2
7	87	91	0	0	0	0	0	0	0
8	0	0	0	2	1	4	2	0	0
RB012994 F 0.10 g Fe ions									
2	0	0	0	0	0	0	0	75	72
3	0	0	4*	6	4	0	0	1	1*
4	0	0	107**	80	80	2	1	0	0
5	0	0	0	0	0	63	59	0	0
7	91	96	0	0	0	0	0	0	0
8	0	0	5*	17	14	1	0	1	1

TABLE CI (CONTINUED)

Column-Exp step	Am 241 (±9%)	Am 243 (±10%)	Np 237 (±15%)	Pu 239 (±9%)	Pu 236 (±11%)	U 238 (±10%)	U 232 (±10%)	Th 232 (±9%)	Th 229 (±15%)
				% Recovery					
RB012994 G 0.010 g Fe ions									
2	0	0	0	0	0	0	0	88	84
3	0	0	4*	2	4	0	0	1	1*
4	0	0	90	83	90	1	1	1	0
5	0	0	0	0	0	81	72	1	0
7	94	106	0	0	0	0	0	0	0
8	0	0	2**	2	2	1	1	0	0

* contains counts from alpha decay of Th-229, Th-230, and Np-237. Estimated the amount of Th-229(equal to the percent recovery of Th-232), Th-230 (equal to 0.17 of Th-232 counts), and assigned the remaining counts to Np-237.

** contains U-234 and Np-237, used the amount of U-238 to estimate the amount of U-234 counts.

Simulated Hard Water (0.010 - 0.50 g)

7 columns		LOAD TIME
A	blank, 5.0-M HCl only plus tracers and spikes	81 min
B	Al, 0.10 g metal, 0.894 g $AlCl_3$·6HOH	100
C	Al, 0.010 g metal, 0.0894 g $AlCl_3$·6HOH	90
D	Ca, 0.10 g metal, 0.2769 g $CaCl_2$	91
E	Ca, 0.50 g metal, 1.385 g $CaCl_2$	80
F	Fe, 0.10 g metal, 0.485 g $FeCl_3$·6HOH	82
G	Fe, 0.010 g metal, 0.0485 g $FeCl_3$·6HOH	82

Procedure:

CHLORIDE Column:

Step 1. loaded sample in 5.0-M HCl plus H_2O_2 (25 ml),
5.0 ml 5.0-M HCl
20.0 ml 3.0-M HCl

Step 2. 6.0 ml 1.30-M HCl

Step 3. 2.0 ml 1.30-M HCl

Step 4. 15.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl

Step 5. 20.0 ml 0.10-M $(NH_4)_2C_2O_4$

CALCIUM OXALATE coprecipitation of eluate from step 1.

NITRATE column

Step 6. load 3.0-M HNO_3 plus solid ascorbic acid
5.0 ml 3.0-M HNO_3
1.0 ml 8.0-M HCl

Step 7. 15.0 ml 3.0-M HCl

Step 8. 20.0 ml 0.10-M $(NH_4)_2C_2O_4$
5.0 ml 0.10-M HCl

TABLE CII

SEPARATION OF ACTINIDE IONS FROM SOIL MATRICES
USING BARIUM SULFATE COPRECIPITATION
EXPERIMENT RB012594
INITIAL COUNTS

EXP Step	Am 243	Np 237	Pu 236 Counts (total) (±1.65√c)	U 238	Th 232	Det. Eff. (±2%)	Am 243 (±15%)	Np 237 % Recovery (±25%)	Pu 236 (±10%)	U 238 (±16%)	Th 232 (±10%)
$BaSO_4$-1 resolution poor, total counts=6324*						0.166	95±9 dpm /approx.200 dpm added**				
$BaSO_4$-2 resolution poor, total counts=6742*						0.170	99±9 dpm /approx. 200 dpm added**				
$BaSO_4$-3 resolution poor, total counts=6903*						0.187	93±9 dpm /approx. 200 dpm added**				
Filtrate-1	162	160^	39	126	226	0.180	9	8	8	7	13
Filtrate-2	211	175^	64	149	290	0.188	11	8	13	8	16
Filtrate-3	174	137^	76	137	215	0.169	10	7	17	8	13

* significant amounts of self-adsorption, no background correction

**includes the five parent nuclides listed above and an estimated value of the following daughters Th-230, Th-228, U-234, Ra-224, Bi-212, Rn-220

^ Np-237 count obtained by subtracting U-234 (equal to U-238 counts) and Th-230 (equal to 0.17 time Th-232 counts) from total counts in energy range

SEE TABLE CIII for Procedure.

RB012594 $BaSO_4$ coppt
count time: 399 ± 1 min.
Detector: PIPS/ls
($TiCl_3$)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 9939 ± 500 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 11377 ± 458 counts8
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 2669 ± 162 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 9596 ± 482 counts
Am-243 (Am-08A) 4.97E1 ± 0.10 dpm/ml
100% yield: 9907 ± 596 counts

TABLE CIII

SEPARATION OF ACTINIDE IONS FROM SOIL MATRICES
USING BARIUM SULFATE COPRECIPITATION
EXPERIMENT RB012594
TRU-Resin™ SEPARATION

EXP Step	Am 243	Np 237	Pu 236	U 238	Th 232	Det. Eff. (±2%)	Am 243 (±10%)	Np 237 (±10%)	Pu 236 (±10%)	U 238 (±16%)	Th 232 (±10%)
		Counts (total) (±1.65√c)					% Recovery				
A-2	134	0	0	0	0	0.170	16	0	0	0	0
B-2	171	0	0	0	0	0.162	21	0	0	0	0
C-2	198	0	0	0	0	0.211	19	0	0	0	0
A-3	0	16	8	0	144	0.172	0	2	3	0	13
B-3	0	25	1	0	126	0.181	0	2	0	0	14
C-3	0	28	1	0	120	0.217	0	3	0	0	11
A-5	0	146	22	0	3	0.173	0	15	10	0	0
B-5	0	141	30	0	12	0.175	0	14	13	0	1
C-5	0	160	37	0	17	0.215	0	13	13	0	2
A-6	0	0	0	2	0	0.182	0	0	0	0	0
B-6	0	0	0	3	0	0.200	0	0	0	0	0
C-6	198	0	0	4	0	0.223	0	0	0	0	0

Procedure:
-prepared 3 samples: pipet tracers, added 4 ml of conc. H_2SO_4, warmed, added 2.0 ml 0.45% $BaCl_2 \cdot 2HOH$, heated on hot plate till fumed (barium sulfate dissolved), cooled in cold tap water bath about 3 min., added 20 ml of DI HOH, observed fine ppt., returned to HOH bath for 10 min., swirled periodically
-poured samples onto filters, saved filtrate, filter wash (1-M H_2SO_4 then HOH) dried, mounted, counted filters
-poured filtrate back into beakers, fumed, diluted with 3-M HCl, ppt with NdF_3 procedure, counted
-AFTER BEING COUNTED, the discs with Barium sulfate ppt were placed into a crucible with NaCl and muffled (850°C for 400 min.), removed from oven: observed white crystalline solid with brown ring in middle, added 5-M HCl, plate, wet ashed 3 times with 8-M HNO_3 and H2O2, clear colorless solution with solid?
-Passed liquid through TRU-Resin™ column. Procedure:
Step 1. loaded samples in 3.0-M HNO_3 plus ascorbic acid (20ml), 3.0 ml 3.0-M HNO_3, 1.0 ml 8-M HCl
Step 2. 15.0 ml 3.0-M HCl,(elute Am)
Step 3. 6.0 ml 1.30-M HCl,
Step 4. 2.0 ml 1.30-M HCl,
Step 5. 20.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl
Step 6. 20.0 ml 0.10-M $(NH_4)_2C_2O_4$, 5.0 ml 0.10-M HCl

RB012594 TRU column
count time: 200 ± 1 min.
Detector: PIPS/ls
($TiCl_3$)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4982 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 5703 ± 229 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 1338 ± 81 counts

U-238 (U-12) 4.81E1 ± 0.10 dpm/ml 100% yield: 4810 ± 241 counts
Am-243(Am-08A) 4.97E1 ± 0.10 dpm/ml 100% yield: 4966 ± 298 counts

TABLE CIV

SEPARATION OF ACTINIDE IONS FROM SOIL MATRICES
USING BARIUM SULFATE COPRECIPITATION
EXPERIMENT RB020894

EXP Step	Am 243	Np 237	Pu 239	U 238	Th 232	Detector PIPS/ls #/Eff (±2%)	Am 243 (±20%)	Np 237 (±20%)	Pu 239 % Recovery (±15%)	U 238 (±15%)	Th 232 (±15%)
		Counts (total) (±1.65√c)									
A-8	24	792*	140	480	35	5/0.180	1	27*	8	49	3
B-8	199	341*	632	104	396	6/0.151	35	20*	84	14	53
C-8	148	327*	248	105	433	7/0.188	21	14*	27	12	47
D-8	110	345*	191	123	388	8/0.169	17	18*	23	15	46
A-12^	16	35*	8	8	11	17/0.196	1	1*	0	0	0
B-12^	5	42*	13	6	8	18/0.210	0	1*	1	0	0
C-12^	13	26*	8	4	21	19/0.221	1	0*	0	0	1
D-12^	24	79*	18	20	16	20/0.207	1	2*	1	1	1
A-13^	14	114*	18	10	19	21/0.210	1	4*	1	0	1
B-13^	7	36*	6	4	4	22/0.180	0	1*	0	0	0
C-13^	22	28*	26	8	17	23/0.227	1	0*	1	0	1
D-13^	8	29*	8	5	3	24/0.195	0	1*	0	0	0
					Total % recovered						
						A	3	32	9	49	4
						B	35	22	85	14	53
						C	23	14	28	12	49
						D	18	21	24	16	47

* estimated because U-234, Th-230, and Np-237 energy range overlaps. Estimated U-234 (to equal U-238 counts), Th-230 (0.17 of Th-232 counts, and the rest was assigned to Np-237.
^ count time 399 ± 1 min, significant scatter observed

Barium Sulfate, redox,
Procedure:
prepared 4 samples:

1. pipet tracers, wet ashed
2. added 4 ml of conc. H_2SO_4, fumed about 3 min., cooled in cold tap HOH bath
3. added 20 ml DI HOH
4. added redox reagents: A: 0.30 g $K_2S_2O_4$ plus 4 drops 1% $AgNO_3$
   B: 7 d 30% H2O2
   C: 3 d 20% $TiCl_3$
   D: 0.15g ascorbic acid + 4 drops 0.004g/ml Fe ions
   swirled, sat 10 min.
5. added 2.0 ml 0.45% $BaCl_2 \cdot 2HOH$ (mg Ba), sat 1 min.
6. heated on hot plate, boiled one min., cooled in cold tap water bath 10 min.
7. filtered solid $BaSO_4$ coppt from solution (using metricel filters)
8. wet ashed solution with 3-M HNO_3 and H_2O_2, dissolved in 3-M HCl, precipitated using NdF_3 procedure with $TiCl_3$ added, alpha counted
9. the discs with Barium sulfate ppt were placed into a crucible with NaCl and fine carbon and the sample was then muffled with crucible lid tilted, 850°C for 480 min.
10. removed from oven: white crystalline solid with very little black remaining
11. added 5-M HCl, wet ashed, repeated with 8-M HCl, then conc. HNO_3 plus H_2O_2, then conc HCl plus H2O2, then dissolved in 15 ml 3.0-M HCl, warmed
12. filtered out any remaining undissolved solid, (metricel filters), counted 13. NdF_3 ppt procedure on remaining solution, used $TiCl_3$, counted

TABLE CIV (CONTINUED)

RB020894
$BaSO_4$ coppt
count time: 200 ± 1 min.
($TiCl_3$)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 5703 ± 229 counts
Pu-239 (Pu-13D) 8.3E1 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 241 counts
Am-243 (Am-09) 3.80E1 ± 0.10 dpm/ml
100% yield: 3794 ± 190 counts

TABLE CV

SEPARATION OF ACTINIDE IONS FROM SOIL MATRICES
USING BARIUM SULFATE COPRECIPITATION
EXPERIMENT RB021494

EXP Step	Am 243	Np 237 Counts (total) (±1.65√c)	Pu 239	U 238	Th 232 #/Eff (±2%)	Det.	Am 243 % Recovery (±15%)	Np 237 (±20%)	Pu 239 (±15%)	U 238 (±15%)	Th 232 (±15%)
A-6	211	840*	500	354	273	0.188	30	41*	53	39	29
B-6	13	32*	4	8	18	0.174	1	2*	0	1	2
C-6	0	1198*	212	573	644	0.205	0	37*	21	58	63
D-6	327	800*	510	441	366	0.153	56	42*	67	50	58

* estimated because U-234, Th-230, and Np-237 energy range overlaps. Estimated U-234 (to equal U-238 counts), Th-230 (0.17 of Th-232 counts), and assigned the rest to Np-237.

Barium Sulfate, matrix
A: blank, DI HOH
B: Al, 1.0 g metal, 8.94 g $AlCl_3 \cdot 6HOH$, 0.00370 mole
C: Ca, 1.0 g metal, 3.083 g $CaCl_2$, 0.00179 mole
D: Fe, 1.0 g metal, 4.845 g $FeCl_3 \cdot 6HOH$, 0.002778 mole

RB021494
$BaSO_4$ coppt
count time: 200 ± 1 min.
($TiCl_3$)
Detectors: PIPS/s

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 5703 ± 229 counts
Pu-239 (Pu-13D) 8.3E1 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 241 counts
Am-243 (Am-09) 3.80E1 ± 0.10 dpm/ml
100% yield: 3794 ± 190 counts

Procedure:
1. mixed tracers and metal ions solutions dissolved in 3-M HCl,
2. added 4 ml of conc. H_2SO_4, warmed,
3. added 2.0 ml 0.45% $BaCl_2 \cdot 2HOH$, fumed about 3 min.,
(lots of ppt in Al, Fe, and Ca; Ca cleared up by end of fume)
cooled in cold tap water bath about 3 min.
4. cooled, added 20 ml DI HOH, lots of ppt in Al, Fe, Ca
5. centrifuged, decanted, saved ppt and supernatant separately
6. fumed supernatant sample,
precipitated using NdF_3 procedure with $TiCl_3$ added, alpha counted

TABLE CVI

SEPARATION OF ACTINIDE IONS AS ONE GROUP USING DIPHONIX RESIN
EXPERIMENT RB012794

EXP Step	Am 243	Np 237 Counts (total) (±1.65√c)	Pu 236	U 238	Th 232	Detector PIPS/ls #/Eff (±2%)	Am 243 (±15%)	Np 237 (±8%)	Pu 236 % Recovery (±10%)	U 238 (±16%)	Th 232 (±10%)
A-1	98	1416	21	0	0	9/0.170	6	74	5	0	0
B-1	0	1366	0	0	0	10/0.172	0	69	0	0	0
C-1	23	590	0	0	0	13/0.162	1	32	0	0	0
A-2	601	361*	19	292	9	11/0.173	35	3*	4	18	0
B-2	541	271*	24	134	1	12/0.182	30	7*	5	8	0
C-2	589	112*	37	103	0	15/0.181	33	0*	8	6	0
A-3	823	405*	29	404	14	19/0.217	38	0*	5	19	1
B-3	958	442*	32	455	9	20/0.215	45	0*	6	22	0
C-3	753	348*	70	379	13	21/0.223	34	0*	13	18	1
					Total % eluted						
						A	79	77	14	37	1
						B	75	76	11	30	0
						C	68	32	21	24	1

* estimated because U-234 and Np-237 energy range overlaps, U-234 estimated to equal U-232 counts

Procedure: column A & B
Step 1: loaded 1.0-M HNO_3 (10ml), 2.5 ml 1.0-M HNO_3
Step 2: 10 ml 5.0-M HCl
Step 3: 50 ml 5.0-M HCl
Procedure: column C
Step 1: loaded 1.0-M HNO_3 plus solid ascorbic acid (10ml), 2.5 ml 1.0-M HNO_3 plus ascorbic acid
Step 2: 10 ml 5.0-M HCl plus ascorbic acid
Step 3: 50 ml 5.0-M HCl plus ascorbic acid

RB012794
count time: 399 ± 1 min.
Detectors: PIPS/ls
($TiCl_3$)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 9939 ± 500 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 11377 ± 458 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 2669 ± 162 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 9596 ± 482 counts
Am-243 (Am-08A) 4.97E1 ± 0.10 dpm/ml
100% yield: 9907 ± 596 counts

TABLE CVII

SEPARATION OF ACTINIDE IONS AS ONE GROUP USING DIPHONIX RESIN
EXPERIMENT RB020794

EXP Step	Am 243	Np 237	Pu 239	U 238	Th 232	Det. Eff (±2%)	Am 243 (±12%)	Np 237 (±20%)	Pu 239 (±12%)	U 238 (±12%)	Th 232 (±12%)
		Counts (total) (±1.65√c)						% Recovery			
A-2	7	77	4	0	0	0.188	1	7	0	0	0
B-2	8	37	9	0	0	0.169	1	4	1	0	0
C-2	370	138*	445	7	7	0.170	58	13*	52	1	1
D-2	5	5*	6	0	2	0.172	1	0*	1	0	0

* estimated because U-234, Th-230, and Np-237 energy range overlaps. Estimated U-234 (to equal U-238 counts), Th-230 (0.17 of Th-232 counts, and the rest was assigned to Np-237.

Procedure:

1. pipet tracers into 10 ml 1.0-M HCl, added redox reagents:
   A: 0.30 g $K_2S_2O_4$ plus 4 drops 1% $AgNO_3$
   B: 7 d 30% H2O2
   C: 3 d 20% $TiCl_3$
   D: 0.15 g ascorbic acid plus 4 drops 0.004 g/ml Fe ions
   swirled, sat 10 min. total volume 10 ml, poured samples onto columns,
   rinse 2.5 ml 1.0-M HCl
2. collected filtrate, fumed, precipitated using NdF_3 procedure with $TiCl_3$ added, alpha counted
3. the diphonix resin samples were placed into crucibles with K2CO3 (mp 891oC),
   The sample was then muffled with crucible lid tilted, 900°C for 480 min.
   Dissolution unsuccessful.

RB020794 diphonix
count time: 200 ± 1 min.
($TiCl_3$)
Detector: PIPS/ls

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 250 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 5703 ± 229 counts
Pu-239 (Pu-13D) 8.3E1 ± 0.10 dpm/ml
100% yield: 4992 ± 200 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 241 counts
Am-243 (Am-09) 3.80E1 ± 0.10 dpm/ml
100% yield: 3794 ± 190 counts

TABLE CVIII

SEPARATION OF ACTINIDE IONS AS ONE GROUP USING DIPHONIX RESIN
EXPERIMENT RB021694

EXP Step	Am 243	Np 237	Pu 239	U 238	Th 232	Det. Eff. (±2%)	Am 243	Np 237	Pu 239	U 238	Th 232
		Counts (total) (±1.65√c)					% Recovery				
							(±20%)	(±20%)	(±15%)	(±12%)	(±12%)
A-3	54	29*	16	11	10	0.153	5	1*	1	1	1
B-3	37	6*	18	10	1	0.205	2	0*	1	0	0
C-3	42	37*	15	8	8	0.160	3	2*	1	0	0
D-3	39	26*	35	11	6	0.173	3	1*	2	1	0
A-12	1826**	1287*	411	309	1588	0.164	19**	38*	25	20	97
B-12	1425**	1211*	537	283	1425	0.171	23**	35*	32	17	84
C-12	2711**	3177*	1621	1160	1827	0.188	62**	80*	87	64	98
D-12	2791**	3441*	1686	1299	1829	0.174	68**	95*	97	78	106

* estimated because U-234, Th-230, and Np-237 energy range overlaps. Estimated U-234 (to equal U-238 counts), Th-230 (0.17 of Th-232 counts, and the rest was assigned to Np-237.
** estimated because Th-228 and Am-243 energy range overlaps. Estimated Th-228 (equal to Th-232 counts) and assigned the rest to Am-243.

Sample A: Ceramic crucible with NaCl
Sample B: Ceramic crucible with NaCl
Sample C: Alumina crucible with K2CO3
Sample D: Alumina crucible with K2CO3

Procedure:
Sep 1: combined tracer samples and resin, swirled periodically, sat 2 hours,
Step 2: poured resins with solutions into columns, rinse 3 ml 1.0-M HCl
Step 3: filtrate was wet ashed, NdF3 ppt procedure, alpha counted
Step 4: Transferred resin from columns to crucibles, dried
Step 5: muffled resins, 700°C for 480 min.
Step 10. removed from oven: diphonix resin appears to be gone, material is crumbly, NaCl samples have a light blue color
Step 11. added 5-M HCl, transfer to beakers, (K2CO3 trn easily, NaCl had a roughness on the bottom of the crucible)
Step 12. wet ashed, fumed, dissolved in 3-M HCl, NdF3 ppt procedure, alpha count (filtered small amt of solid from A and B prior to ppt - counted also)

RB021694
Detectors: PIPS/ls
count time: 399 ± 1 min.
($TiCl_3$)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 9947 ± 498 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 11377 ± 456 counts
Pu-239 (Pu-13D) 8.3E1 ± 0.10 dpm/ml
100% yield: 9959 ± 399 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 9596 ± 480 counts
Am-243 (Am-09) 3.80E1 ± 0.10 dpm/ml
100% yield: 7573 ± 455 counts

TABLE CIX

SEPARATION OF ACTINIDE IONS AS ONE GROUP USING DIPHONIX RESIN
EXPERIMENT RB022194

EXP Step	Am 241\243 Counts(total) (±1.65√c)	Detector PIPS/ls #/Eff (±2%)	Am 241\243 % Recovery (±20%)
A-2	14	25/0.200	0
B-2	40	27/0.192	1
A-14	1251	26/0.225	39
B-14	958	28/0.222	30

-spectral resolution was poor so Am-241 and Am-243 counted together

Procedure: Sample A
step 1:loaded column, load time about 8 hours
step 2:evaporated filtrate, diluted with 3-M HCl, NdF3 ppt, count
Procedure: Sample B
Step 1: Sample stirred for 3 hrs., allowed resin to settle for 30 min., poured off 600 ml of clear supernatant, filtered resin from rest of supernatant using 41 ashless Whatman filter paper, combined solutions.
Step 2: evaporated supernatant, diluted with 3-M HCl, NdF3 ppt, count
****
Sample A and B
Step 3: Transferred resin from column to alumina crucible and from filter paper to alumina crucible, dried, added 1.9 g K2CO3
Step 5: muffled resins at 700°C for 480 min.
Step 10. removed from oven: black remained in both crucibles
Step 11. remuffled at higher temperature: 900°C for 480 min.
Step 12. removed from oven: light pink cloudy material,
Step 13. dissolved easily in 8-M HNO3 then conc HNO3, trn to beaker. wet ashed, added HCl, wet ashed twice,
Step 14 fumed, NdF3 ppt, counted

RB022194
count time: 200 ± 1 min.

Am-241 (Am-06D) 1.05E2 ± 0.10 dpm/ml
100% yield: 10500 ± 525 counts
Am-243 (Am-09) 3.80E1 ± 0.10 dpm/ml
100% yield: 3796 ± 228 counts

TABLE CX

SEPARATION OF ACTINIDE IONS AS ONE GROUP USING DIPHONIX RESIN
EXPERIMENT RB030494

EXP Step	Am 241	U 238 Counts (total) (±1.65√c)	Th 232	Detector PIPS/ls #/Eff (±2%)	Am 241 (±15%)	U 238 % Recovery (±22%)	Th 232 (±30%)
E-Resin-2	0	604	113	31/0.243	0	34	8
E-Resin-5	838	0	0	27/0.192	104	0	0
E-Resin-6	0	0	192	31/0.243	0	0	14
E-Filtrate-2	0	536	28	29/0.219	0	34	2
E-Filtrate-5	65*	0	149	25/0.200	8	0	13
E-Filtrate-6	0	4	178	29/0.219	0	0	14
				tot %	112	68	51
F-Resin-2	0	610	174	32/0.213	0	39	15
F-Resin-5	772*	0	40	28/0.222	83	0	3
F-Resin-6	0	0	232	32/0.213	0	0	19
F-Filtrate-2	0	543	443	30/0.211	0	35	37
F-Filtrate-5	71*	0	20	26/0.225	8	0	2
F-Filtrate-6	0	0	172	30/0.211	0	0	15
				tot %	91	74	91

* estimated because Th-228 and Am-241 energy range overlaps. Estimated Th-228 (equal Th-232 counts) and the rest was assigned to Am-241.

Muffle Method: put 1.00 g K2CO3 (mp 891oC) in crucible, filter containing diphonix on top. put in drying oven (about 100oC) for 1 hr.,
the sample was then muffled with crucible lid tilted, 900°C for 480 min.
removed from muffle, dissolved in 8-M HCl, wet ashed,
repeated wet ash with HNO3 plus H2O2
repeated wet ash with HCl plus H2O2
dissolved muffled resin taken through TRU-Resin™ columns procedures

CHLORIDE COLUMN
Step 1. Load 5-M HCl plus H2O2 + beaker rinse
(35 ml resin solutions, 60 ml filtrate solutions)
10 ml 3-M HCl
Step 2. 5.0 ml 0.10-M HCl, 40.0 ml 0.10-M (NH4)2C2O4, 10.0 ml 0.10-M HCl
Step 3. Calcium oxalate coppt on step 1 filtrate, dissolve in 3.0-M HNO3
NITRATE COLUMN
Step 4. load solution from step 3 plus solid ascorbic acid
rinse 5.0-M 3.0-M HNO3, switch 1.0 ml 8.0-M HCl
Step 5. 20.0 ml 3.0-M HCl
Step 6. 5.0 ml 0.10-M HCl, 30.0 ml 0.10-M (NH4)2C2O4, 10.0 ml 0.10-M HCl

RB030494 DIPHONIX, soil
count time: 200 ± 1 min.
($TiCl_3$)
Activity from soil samples:
U-238: 2442 ± 502 counts
Th-232: 635 ± 178 counts

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 4986 ± 250 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 241 counts
Am-241 (Am-06D) 1.05E2 ± 0.10 dpm/ml
100% yield: 4200 ± 252 counts

TABLE CXI

SEPARATION OF ACTINIDE IONS FROM SOIL MATRICES
USING CALCIUM FLUORIDE COPRECIPITATION
EXPERIMENT RB060294

EXP Step	Am 241	Np 237 Counts (total) (±1.65√c)	Pu 236	U 238	Th 232 #/Eff (±2%)	Detector PIPS/ls	Am 241 (±10%)	Np 237 % Recovery (±15%)	Pu 236 (±12%)	U 238 (±14%)	Th 232 (±10%)
CHLORIDE Column (Dissolved CaF_2)											
A-2	0	371*	0	0	656	1/0.162	0	43*	0	0	90
B-2	0	116*	0	0	596	2/0.157	0	2*	0	0	85
A-4	0	88	464	0	0	7/0.209	0	11	78	0	0
B-4	0	500	443	0	0	8/0.161	0	83	96	0	0
A-5	0	0	0	199	0	13/0.165	0	0	0	25	0
B-5	0	0	0	236	0	15/0.177	0	0	0	28	0
NITRATE Column (Dissolved CaF_2)											
A-7	837	0	0	0	0	27/0.189	109	0	0	0	0
B-7	869	0	0	0	0	28/0.219	97	0	0	0	0
A-10	0	71*	0	0	52	19/0.201	0	8*	0	0	6
B-10	0	32*	0	0	40	21/0.208	0	3*	0	0	4
NITRATE Column (Supernatant)											
A/S-7	11	0	0	0	0	32/0.206	1	0	0	0	0
B/S-7	13	0	0	0	0	33/0.165	2	0	0	0	0
A/S-8	0	0	0	0	6	5/0.174	0	0	0	0	0
B/S-8	0	0	0	0	5	6/0.149	0	0	0	0	0
A/S-9	0	28	7	0	0	11/0.169	0	2	1	0	0
B/S-9	0	35	10	0	0	12/0.180	0	5	1	0	0
A/S-10	0	0	0	402	0	25/0.210	0	0	0	40	0
B/S-10	0	0	0	478	0	26/0.206	0	0	0	48	0
					Total % recovered						
						A-CaF_3	109	62	78	25	96
						B-CaF_3	97	88	96	28	89
						A-Sup.	1	2	1	40	1
						B-Sup.	2	5	1	48	1

* estimated because Th-230 and Np-237 alpha energy range overlaps. Estimated % recovery of Th-230 (equal to 0.17 of Th-232 counts) and assigned the remaining counts to Np-237.
^ count estimated from alpha energy at 5770 keV (68%), alpha emitted at 5722 keV (32%) overlaps with Ra-224 at 5685 keV
** estimated because Np-237 and U-234 alpha energy range overlaps. Estimated U-234 (equal to U-238 counts) and assigned the remaining counts to Np-237.

Procedure: CHLORIDE A-CaF_2, A-CaF_2
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2 (20 ml),
5.0 ml 5.0-M HCl
Step 2. 6.0 ml 1.30-M HCl,
Step 3. 3.0 ml 1.30-M HCl,
Step 4. 15.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl,
Step 5. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$, 10 ml 0.1-M HCl
****

NO CALCIUM OXALATE COPPT
****

TABLE CXI (CONTINUED)

Procedure: NITRATE A-CaF_2,A-Supernatant,B-CaF_2,B-Supernatant,
Step 6. wet ash eluate from step 1 plus A-sup and B-sup,
added HNO3, wet ash twice
dissolve in 3.0-M HNO3 plus solid ascorbic acid, load onto column
7.0 ml 3-M HNO3, 1.0-ml 8.0-M HCl
Step 7. 15.0 ml 3.0-M HCl
Step 8. 6.0-M 1.30-M HCl (supernatants only)
Step 9. 15.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl (supernatants only)
Step 10. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$, 10.0 ml 0.10-M HCl

RB060294 CaF_2
count time: 200 ± 1 min.
(sulfate fume)
(TiCl3 in steps 5 & 10)
Assisted by J. Check

Th-232 (Th2-186-A6) 2.99E1 ± 0.10 dpm/ml
100% yield: 4810 ± 241 counts
Np-237 (Np-020-A3) 9.3E1 ± 0.10 dpm/ml
100% yield: 3822 ± 149 counts
U-238 (U8-282-A) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 241 counts
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 4200 ± 210 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 2860.± 172 counts

TABLE CXII

SEPARATION OF ACTINIDE IONS FROM SOIL MATRICES
USING CALCIUM FLUORIDE COPRECIPITATION
EXPERIMENT RB031094
RAW DATA

Column Exp step	Am 241	Am 243	Np 237 Counts (total) ($\pm 1.65\sqrt{c}$)	Pu 239	Pu 236	U 238	U 232	Th 232	Th 229	Detector PIPS/ls #/Eff (±2%)
A-2	0	0	0	0	0	0	0	514	650	1/0.164
B-2	0	0	0	0	0	0	0	473	639	2/0.171
A-4	0	0	116*	55	45	0	0	14	166*	4/0.188
B-4	0	0	106	59	43	0	0	5	0	5/0.174
A-5	0	0	0	0	0	557	468	0	0	7/0.205
B-5	0	0	0	0	0	494	459	0	0	8/0.160
A-7	503	590	25*	0	0	0	0	1	25*	9/0.173
B-7	441	461	50*	0	0	0	0	0	50*	10/0.181
A-8	0	0	28*	0	0	0	0	10	28*	11/0.177
B-8	0	0	29*	9	8	0	0	2	29*	12/0.182
A-10	0	0	206**	190	146	10	5	5	0	13/0.164
B-10	0	0	127**	136	117	11	3	0	0	15/0.178

* estimated because Th-229 and Np-237 alpha energy range overlaps. Estimated % recovery of Th-229(equal to % recovery of TH-232), Th-230 (equal to 0.17 of Th-232 counts) and assigned the remaining counts to Np-237.
** estimated because Np-237 and U-234 alpha energy range overlaps. Estimated U-234 (equal to U-238 counts) and assigned the remaining counts to Np-237.

RB031094 CaF_2 plus Nd^{+3} and Ti^{+3}
count time: 200 ± 1 min.
(sulfate fume)
(TiCl3 in steps 5 & 10)

Th-232 (Th-10) 8.31E2 ± 0.10 dpm/ml
100% yield: 3324 ± 167 counts
Np-237 (Np-01B) 9.5E1 ± 0.10 dpm/ml
100% yield: 3802 ± 153 counts
Pu-239 (Pu-9-236A2) 2.93E1 ± 0.10 dpm/ml
100% yield: 2930 ± 147 counts
U-238 (U-12) 4.81E1 ± 0.10 dpm/ml
100% yield: 4810 ± 193 counts
Am-241 (Am-06A) 2.55E2 ± 0.10 dpm/ml
100% yield: 2551 ± 130 counts
Th-229 (Th-08B) 4.15E1 ± 0.10 dpm/ml
100% yield: 4152 ± 208 counts
Pu-236 (Pu-19B) 3.31E1 ± 0.10 dpm/ml
100% yield(decayed): 2210 ± 133 counts
U-232 (U-05AA3) 4.86E1 ± 0.10 dpm/ml
100% yield(decayed): 4505 ± 271 counts
Am-243 (Am-08A) 4.97E1 ± 0.10 dpm/ml
100% yield: 2980 ± 150 counts

TABLE CXIII

SEPARATION OF ACTINIDE IONS FROM SOIL MATRICES
USING CALCIUM FLUORIDE COPRECIPITATION
EXPERIMENT RB031094
PERCENT RECOVERY

Column-Exp step	Am 241 (±10%)	Am 243 (±11%)	Np 237 (±20%)	Pu 239 (±18%)	Pu 236 (±18%)	U 238 (±10%)	U 232 (±11%)	Th 232 (±10%)	Th 229 (±11%)
				% Recovery					
A-2	0	0	0	0	0	0	0	95	96
B-2	0	0	0	0	0	0	0	83	90
A-4	0	0	13	10	10	0	0	2	2*
B-4	0	0	16	12	10	0	0	0	0
A-5	0	0	0	0	0	57	51	0	0
B-5	0	0	0	0	0	64	63	0	0
A-7	114	114	4*	0	0	0	0	0	0*
B-7	96	86	7*	0	0	0	0	0	0*
A-8	0	0	2*	0	0	0	0	1	1*
B-8	0	0	4*	2	2	0	0	0	0*
A-10	0	0	30**	40	40	1	0	0	0
B-10	0	0	18**	26	30	1	0	0	0

* estimated because Th-229 and Np-237 alpha energy range overlaps. Estimated % recovery of Th-229(equal to % recovery of TH-232), Th-230 (equal to 0.17 of Th-232 counts) and assigned the remaining counts to Np-237.
** estimated because Np-237 and U-234 alpha energy range overlaps. Estimated U-234 (equal to U-238 counts) and assigned the remaining counts to Np-237.

RB031094 CaF_2 plus Nd^{+3} and Ti^{+3}
Procedure: CHLORIDE
Step 1. loaded sample mixture in 5.0-M HCl plus H2O2(19 ml), 5.0 ml 3.0-M HCl
Step 2. 6.0 ml 1.30-M HCl,
Step 3. 3.0 ml 1.30-M HCl,
Step 4. 15.0 ml 0.03-M $H_2C_2O_4$ in 0.50-M HCl,
Step 5. 30.0 ml 0.10-M $(NH_4)_2C_2O_4$, 10 ml 0.1-M HCl
****
NO CALCIUM OXALATE COPPT
****
Procedure: NITRATE
Step 6. wet ash eluate from step 1, added HNO3, wet ash twice
dissolve in 3.0-M HNO3 plus solid ascorbic acid, load onto column
5.0 ml 3-M HNO3, 1.0-Ml 8.0-M HCl
Step 7. 15.0 ml 3.0-M HCl
Step 8. 6.0-M 1.30-M HCl
Step 9. 3.0-M 1.30-M HCl
Step 10. 50.0 ml 0.10-M $(NH_4)_2C_2O_4$

VITA

Rose Ann Boll was born in Portsmouth, Ohio, on January 18, 1953. She graduated from Wheelersburg High School of Wheelersburg, Ohio, in May 1971. In 1975, she graduated with a B.S. in Biology and Chemistry from Ohio Dominican College, Columbus, Ohio. After a 12 month Medical Technology internship at St. Thomas Hospital in Akron, Ohio, Rose successfully passed the 1976 American Society of Clinical Pathologist registry exam for Medical Technology. From 1977-81, she taught High School chemistry and biology classes at St. Francis DeSales H.S. and Watterson H.S., both in Columbus Ohio. During this time, she complete education courses at Ohio Dominican College and obtained her Ohio Teaching Certificate. From 1981-1989, she worked at St. Francis-St. George Hospital in Cincinnati, Ohio, as a Medical Technologist. In 1983, Rose advanced to the position of Unit Manager. In the fall of 1989, she entered the Graduate School of The University of Tennessee, where she held a teaching assistantship.

The author has presented posters at the 1993 and 1995 ORNL-DOE Analytical Chemistry in Energy Technology Conference in Gatlinburg, TN. She has presented seminars at ORNL and UTK in 1996. She comes from a wonderful family of 11 children. She was a member of the Dominican Sisters of St. Mary of the Springs in Columbus, Ohio, from 1979-1988. She is currently living in Knoxville, TN, with a dog, a bird, a chinchilla, a hamster, a rabbit, and eight fish.