THESIS

EFFECT OF MATRIX CONSTITUENTS ON THE DETERMINATION OF PLUTONIUM AND AMERICIUM IN BONE

Submitted by

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ABSTRACT

EFFECT OF MATRIX CONSTITUENTS ON THE DETERMINATION
OF PLUTONIUM AND AMERICIUM IN BONE

There are numerous methods available in the literature for separating and analyzing radionuclides of interest from an array of environmental matrices. The quality of these methods can be affected by the stable elements that are commonly found in many of these samples. The presence of such interfering constituents can result in incomplete separation of the radioisotopes of interest as well as a reduced rate of recovery. This is especially the case when complex matrices such as samples of bone and bone ash are analyzed. Plutonium and americium tend to concentrate in bone, they are therefore often referred to as bone seekers. They accumulate in actively metabolizing portions of bones of mammals including humans. It is therefore extremely important to study and evaluate the accumulation of these radionuclides in human bone by analyzing bone samples. However, calcium, which is present in high concentrations in the hydroxyapatite that constitutes the bone, as well as sodium and potassium, have the potential to strongly affect the efficacy of radiochemical separation methods. The objective of this research is to investigate the influence of the major and minor elemental constituents present in bone on the affinity of plutonium and americium for a variety of commercial extraction chromatographic resins.

ACKNOWLEDGMENTS

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CHAPTER 1: INTRODUCTION

1.1 Background

In the unforeseen event of actinides being released into the environment, radiological and chemical toxicity have been shown to be a result of acute or chronic exposure to the contamination. Radioactive isotopes of some elements have homologs that play a role in the human body. The metabolic behavior of radiostrontium, e.g., mimics the behavior of its natural analog calcium. The behavior of radiocesium in the human body can be inferred from the of metabolic distribution of sodium. In the case of the actinide elements, there are however no natural analogs in living organisms. It is therefore extremely important to understand the interaction between actinides and constituents of cells and tissues. As a result, these interactions could affect normal biochemical reactions, and findings from such research could help scientist to develop more effective treatment for internal contaminations, such as chelation therapy.

While there are many methods in literature for separating and analyzing radionuclides, the rate of recovery for the actinide of interest can be low due to the elemental constituents that are commonly found in many of these samples. This is especially the case when analyzing complex matrices such as samples of bone and bone ash. [4]

A large part of the work focused on studying the distribution of actinides in the human body and in particular in bones is carried out by the United States Transuranium and Uranium Registries (USTUR). The mission of the USTUR is to evaluate health outcomes, cause of death, and the life expectancy of former nuclear workers. Volunteers, who have worked with and been subject to internal contamination from actinide elements, can make either whole or partial body donations

to science post-mortem, thereby allowing the USTUR to preserve tissue samples and make them available for future research. [21]

Some of the unidentified donors have worked at government sites, where plutonium and americium were processed during the development, manufacture or testing of nuclear weapons. Some of these sites include Los Alamos, Savannah River, and Rocky Flats. The more recent donors include uranium mining workers as well as workers from privately owned facilities that handled actinides for industrial use.[21] The study of the biodistribution of plutonium and americium requires the precise determination of very small amounts of these two elements in various tissues and organs. It is therefore of great importance to select radioanalytical separation procedures with a very high chemical yield. Unfortunately, very little is known about how the other elements present in bone affect the uptake of plutonium and americium by many of the commercial extraction chromatographic resins that form the basis of many of the currently utilized separation procedures. This work therefore focuses on studying the impact that the stable constituents of bone have on the adsorption of plutonium and americium on a variety of commercially available extraction chromatographic resins.

The isotopes of greatest concerns have typically been alpha-emitting radionuclides such as ^{239}Pu and ^{241}Am. Plutonium-239 has a half-life of 24,100 years and is commonly found in nuclear power plant and nuclear weapons. According to the Agency for Toxic Substances and Disease Registry, plutonium can enter your body when it is inhaled, swallowed or through wound depositions. [1] It was originally assumed in ICRP 2 that a maximum of 90% of plutonium would be retained in the skeleton, while 10% would be deposited in the liver. [12] In ICRP 48 it assumed that a total of 90% of plutonium would be deposited in the skeleton and liver, while

10% would be deposited in other soft tissues or excreted [12] This assumed biodistribution was further supported by additional data from human autopsies. It was found that 50% is more likely distributed in the skeleton and 30% in the liver. [12]

As for Americium-241, which has a half-life of 432.2 years, it is found in spent nuclear fuel, weapons production waste and smoke detectors. There are limited case reports of internal dose from americium via external wounds. Following an accident when an ion-exchange column containing 100 g of ^{241}Am exploded in the face of a 64-year-old man, lymphopenia, thrombocytopenia, and histological signs of bone marrow peritrabecular fibrosis had occurred. [8] Researchers were able to support these observations by exposing animals to americium via inhalation. [20] There are very few biokinetic studies on americium compared to plutonium. However, alpha-emitting isotopes of plutonium and americium are bone seekers. [12] This means they have accumulated in actively metabolizing portions of bones, where the alpha particles can cause localized damage to blood producing cells in the bone. It is therefore important to study and evaluate the quantity of plutonium and americium in human bone samples.

1.1.1 Plutonium

Plutonium, element 94 was first isolated and produced at the University of California, Berkeley in 1940 by a bombardment of uranium target with 16-MeV deuterons shown below: [18]

$$^{238}_{92}\mathrm{U} + ^{1}_{0}\mathrm{n} \longrightarrow ^{239}_{92}\mathrm{U} \xrightarrow{\beta^-} ^{239}_{93}\mathrm{Np} \xrightarrow{\beta^-} ^{239}_{94}\mathrm{Pu}$$

Neutrons from the fission of ^{235}U are captured by ^{238}U nuclei to form ^{239}U. From there, the beta decay converts a neutron into a proton to form ^{239}Np and another beta decay forming ^{239}Pu. [9]

Plutonium is the first member of the actinide series with a tripositive state that has enough stability in aqueous solution to be useful in separation chemistry. [19] The fact that plutonium can selectively produce either Pu(III) or Pu(IV) in solution is a huge advantage for radiochemistry. In aqueous solution, plutonium may exhibit all oxidation states as positive ions with varying charge and radius. Consequently, plutonium has the tendency to undergo hydrolysis with the lack of complexing anions. This effect is most common with Pu(IV), decreasing for Pu(VI), and even less for Pu(III). [19]

1.1.2 Americium

In 1944, Glenn T. Seaborg and many other American scientist at Metallurgical Laboratory discovered element 95, americium, as a product of the irradiation of plutonium with neutrons shown below: [17]

$$^{239}\mathrm{Pu}(\mathrm{n},\gamma)^{240}\mathrm{Pu}(\mathrm{n},\gamma)^{241}\mathrm{Pu} \xrightarrow{-\beta^-} {}^{241}\mathrm{Am}$$

Americium can also display four oxidation states, III, IV, V, and VI in aqueous solutions and under certain conditions in carbonate media, all four oxidation states can coexist. [2] In aqueous solution, Am(III) is the most common and most stable oxidation state. Solid compounds of Am(III) have been characterized and the preparation steps are well known. The metal can be dissolved in acid, AmO_2 can be dissolved in hot HCl, or using reducing agents such as NH_2OH, SO_2, or KI to reduce higher valent americium compounds. [3]

1.2 Extraction Chromatography

Extraction Chromatography (EXC) is a common technique used to separate a variety of radionuclides from a wide range of samples. [10] An EXC system typically consists of three components: a stationary phase, an inert support, and a mobile phase, as shown in figure 1. The

stationary phase is the active part of the resin. It is comprised of liquid extractants that adhere through physisorption to the inert support. The inert support itself is made of porous silica or organic polymer spheres that range from 50 to 150 µm diameter. The mobile phase is usually an acidic solution, such as nitric acid or hydrochloric acid, which aids in the adsorption (or extraction) of radionuclides by the stationary phase. In addition, a complexant, such as oxalic acid, may be used to enhance resin selectivity of metal ions strongly retained in solution.

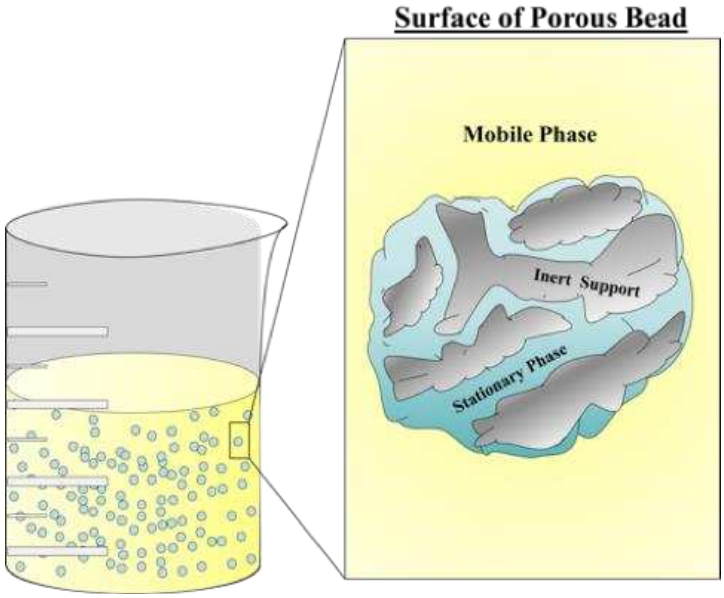
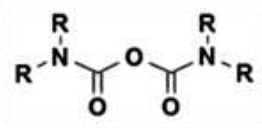
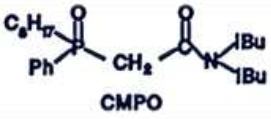
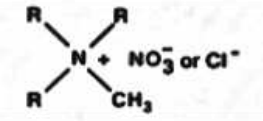
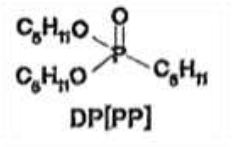


Figure 1. Surface of porous Eichrom resin bead.

1.3 Eichrom Resins

In 1990, Eichrom Technologies, LLC was founded at Argonne National Laboratory to offer commercialize chemical separation technology. Eichrom's line of extraction chromatographic resins have been successfully used for the separation of a variety of samples containing radioisotopes. [10] The four type of resins investigated for the complete separation of plutonium and americium in this study are as follows: DGA, TRU, TEVA, and UTEVA. Table 1 shows the molecular structure for all of these four resins.

Table 1. Molecular Figures of Resins

DGA	TRU	TEVA	UTEVA
			

Depending on the radionuclides present in sample of interest, it can be helpful to use an acid dependency curve when determining a separation plan to ensure that the element of interest is eluted while other elements are retained or vice versa. Figures 3, 5, 7, and 9 show the k'-values for several ions in a system consisting of each of four resins and either nitric acid or hydrochloric acid. These acid dependency curves were also consulted to determine the best experimental conditions for this work. An increase in k' for DGA resin was observed by Horwitz, et al. at a lower nitric acid/hydrochloric acid matrix so 1 M was chosen, while TRU, TEVA, and UTEVA are more easily separated in higher concentrations; therefore, these studies were conducted in a 3 M acid matrix.

1.3.1 DGA Resin

Normal DGA resin extractant system is N, N, N', N' tetraoctyldiglycolamide (TODGA) and has a bed density of 0.38 g/mL shown in figure 2. [6] DGA resin has been performed in nitric acid and hydrochloric acid to observed for uptake of various ions shown in figure 3.

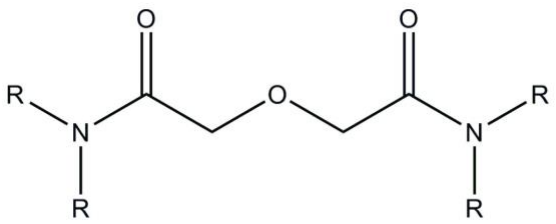


Figure 2. Extractant system of DGA Resin: N,N,N'N' tetraoctyldiglycolamide (TODGA). R-groups are straight chains. [6]

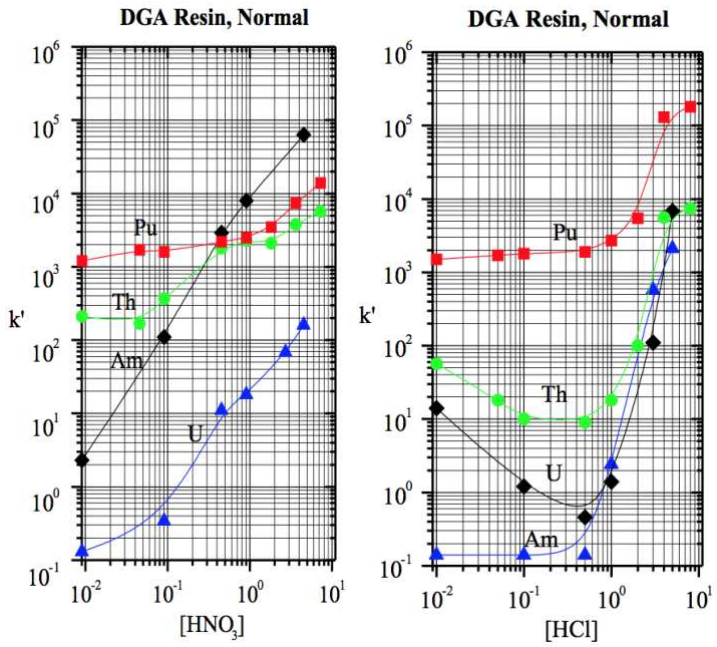


Figure 3. Acid dependency for uptake of various ions by DGA Resins at 23-25℃. [6]

One of DGA resin main applications is separation of Am(III) because of their high affinity for trivalent rare earths and actinides shown in the mechanism below:

$$M^{3+} + 3X^{-} + 3\bar{E} \leftrightarrow \overline{ME_3 \cdot X_3}$$

The DGA resin is an electrically neutral complex and for every one trivalent metal ion, three molecules of nitrate or chloride and three molecules of the organic extractant are required. The DGA resin was demonstrated to have a good affinity for Am(III) with a k' -value of >100 between 0.5 to 5 M HNO_3. Normal DGA resin also has an affinity for tetravalent metals and the mechanism is shown below:

$$M^{4+} + 4X^{-} + 2\bar{E} \leftrightarrow \overline{ME_2 \cdot X_4}$$

In this case, for every one tetravalent metal ion, four molecules of nitrate or chloride and two molecules of the organic extractant are required. Am(III) is can easily be fixed to DGA resin in 5 M HCl or HNO_3 and it can also be eluted with 0.5 M HCl or 0.01 M HNO_3. [6] Characterization

of Pu(IV) also show strong affinity for the DGA resin with a retention factor k'> 3000 over the entire acid range in the study. [6]

1.3.2 TRU Resin

TRU resin extractant system is octylphenyl-N,N-di-isobutylcarbamoylmethylphosphine oxide (abbreviated CMPO) dissolved in tri-n-butyl phosphate (TBP) and has a bed density of 0.37 g/mL shown in figure 4. [6] TRU resin has been performed in nitric acid and hydrochloric acid to observed for uptake of various ions shown in figure 5.

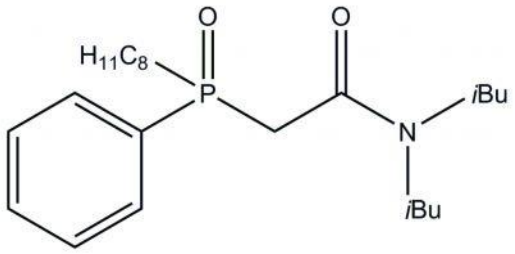


Figure 4. Extractant system of TRU Resin: octylphenyl-N-N-di-isobutyl carbamoylphosphine oxide (CMPO). [6]

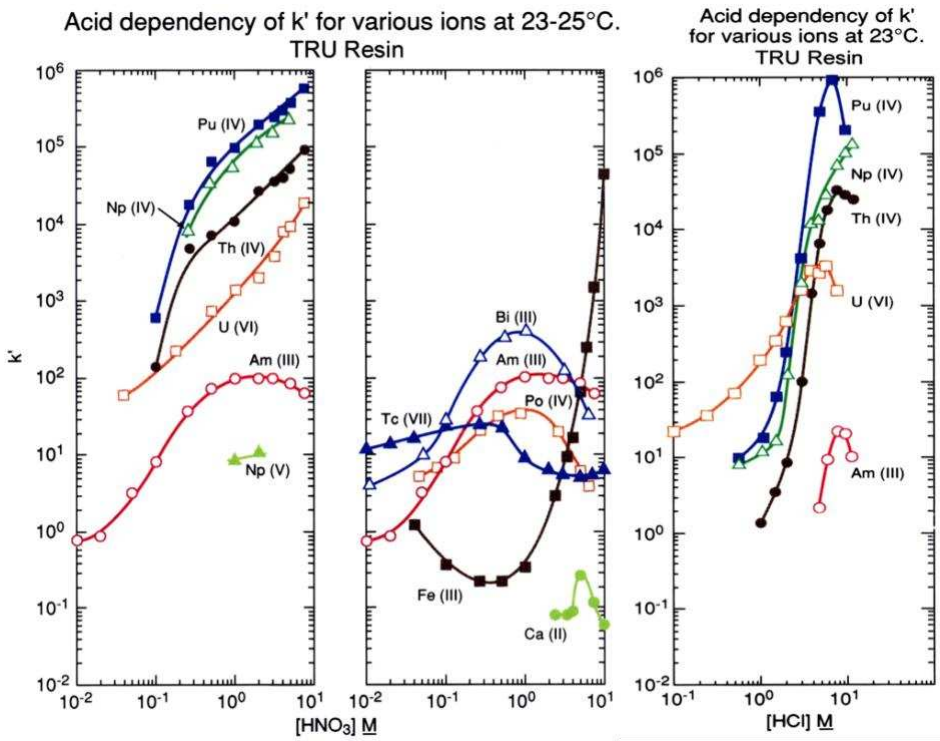


Figure 5. Acid dependency for uptake of various ions by TRU Resins at 23-25℃. [6]

One of TRU resin main applications is separation of actinides and has an affinity for trivalent metal ions shown in the mechanism below:

$$M^{3+} + 3X^- + 2\bar{E} \leftrightarrow \overline{MX_3 \cdot E_2}$$

TRU Resin is an electrically neutral complex and for every one trivalent metal ion, three molecules or nitrate or chloride and 2 molecules of organic extractant are required. The TRU resin was demonstrated to have a good affinity for Am(III) with a k' -value of 100 between 0.5 to 5 M HNO_3. Similarly, TRU resin also has an affinity for tetravalent metals and uranium (VI) shown in the mechanism below:

$$M^{4+} + 4X^- + 2\bar{E} \leftrightarrow \overline{MX_4 \cdot E_2}$$

$$MO_2^{2+} + 2X^- + 2\bar{E} \leftrightarrow \overline{MO_2X_2 \cdot E_2}$$

At >2 M HNO_3, tetravalent actinides show a large retention on the column, with a k'-value between 10^4-10^6. [6]

1.3.3 TEVA Resin

TEVA resin extractant system is trialkyl,methylammonium nitrate or chloride (Aliquat-336) and has a bed density of 0.35 g/mL shown in figure 6. [6] TEVA resin has been performed in nitric acid and hydrochloric acid to observed for uptake of various ions shown in figure 7.

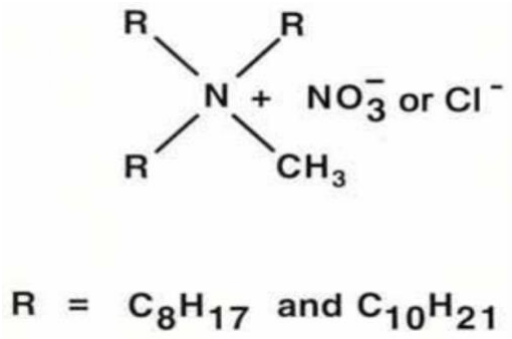


Figure 6. Extractant system of TEVA Resin:trialkyl,methylammonium nitrate or chloride (Aliquat-336). The R groups are C8 or C10 chains. [6]

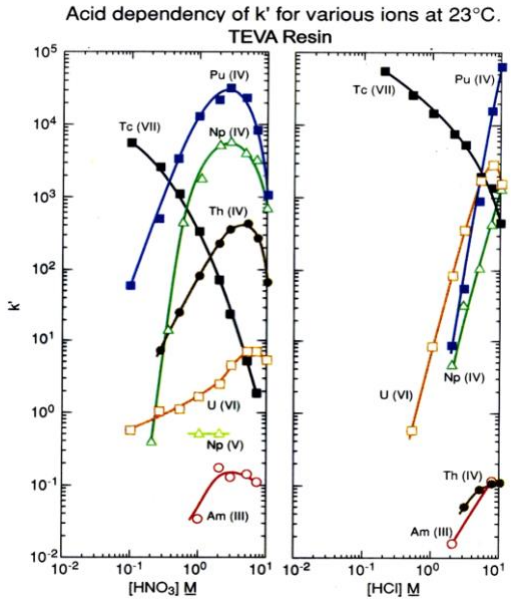


Figure 7. Acid dependency for uptake of various ions by TEVA Resins at 23-25℃. [6]

One of TEVA resin main applications is separation of actinides and has an affinity for tetravalent metal ions shown in the mechanism below:

$$M^{4+} + 4X^{-} + 2\overline{E \cdot X} \leftrightarrow \overline{E_2^{+} \cdot MX_6^{-2}}$$

TEVA resin is a negatively charged complex and for every one tetravalent metal ion, six molecules of nitrate or chloride and 2 molecules of organic extractant are required. In figure 7, Pu(IV) shows a maximum uptake between 2-4 M HNO_3 while the retention is low for Am(III). [6]

1.3.4 UTEVA Resin

UTEVA resin extractant system is diamyl, amylphosphonate (DAAP) and has a bed density of 0.386 g/mL shown in figure 8. [6] UTEVA resin has been performed in nitric acid and hydrochloric acid to observed for uptake of various ions shown in figure 9.

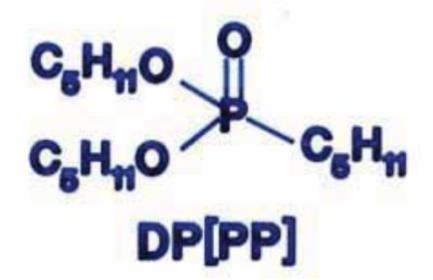


Figure 8. Extractant system of UTEVA Resin: diamyl, amylphosphonate (DAAP) [6]

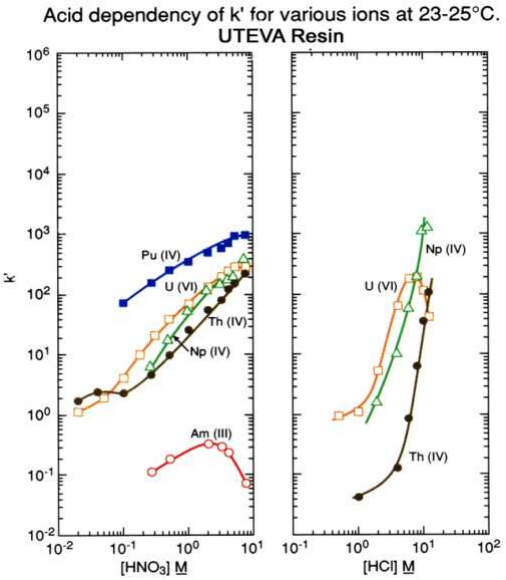


Figure 9. Acid dependency for uptake of various ions by UTEVA Resins at 23-25℃. [6]

One of UTEVA resin main applications is the extraction of tetravalent actinides shown in the mechanism below:

$$M^{4+} + 4X^{-} + 2\bar{E} \leftrightarrow \overline{MX_4 \cdot E_2}$$

UTEVA resin is an electrically neutral complex and for every one tetravalent metal ion, four molecules of nitrate or chloride and 2 molecules of organic are required. From figure 9, all of the tetravalent actinides have a strong k'>100 with HNO_3 acid >5M while Am(III) was not retained at any HNO_3 concentration. [6] Similarly, UTEVA resin also has an affinity for uranium (VI) shown in the mechanism below:

$$MO_2^{2+} + 2X^{-} + 2\bar{E} \leftrightarrow \overline{MO_2X_2 \cdot E_2}$$

1.4 Distribution Ratios (Dw)

In solvent extraction, the distribution ratio describes how much of the total amount of a solute is extracted into the organic phase, regardless of its chemical form. The ratio is defined as the total concentration of the solute in the organic phase relative to the total concentration in the aqueous phase. A similar quantity can be defined in extraction chromatography; however the distribution ratio cannot be obtained directly because the two phases are in different physical states. Instead, it is necessary to calculate the weight distribution ratio D_w. [7] This ratio compares the amount of the analyte that is adsorbed on the resin to the amount remaining in solution. The weight distribution ratio can be calculated using the following equation:

$$D_W = \frac{A_0 - A_S}{A_S} \cdot \frac{\text{mL}}{\text{g}}$$

where:

$A_0 - A_S$ = *activity sorbed on a known weight of resin,* g

A_S = *the activity in a known volume,* mL*, of solution*

The weight distribution ratio can then be used to find the retention factor, k'. The factor k' is defined as the number of free column volumes of eluent that have to pass through the column before the maximum of the elution peak appears and is typically used to describe the retention capabilities of the extraction chromatographic resins. [7] The retention factor, k' can be obtained by taking the distribution ratio, D_w, and dividing it by a constant. In table 1 these conversion values are listed for each of the four resins manufactured by Eichrom Technologies, LLC that are investigated in this work.

Table 1. Extraction chromatography resins manufactured by Eichrom Technology, LLC

Resin	**Extractant System**	**Conversion from D_w to k'**
DGA	N,N,N',N'tetraoctyldiglycolamide (TODGA)	1.75
TRU	Octylphenyl-N–N-di-isobutyl carbamoylphoshine oxide (CMPO)	1.80
TEVA	Aliquat® 336	1.90
UTEVA	Diamyl amylphophonate (DAAP)	1.67

1.5 Batch Distribution Studies

The determination of the uptake of different elements on a specific resin in batch extraction studies plays a key role in evaluation the usefulness of extraction chromatographic separations. To properly evaluate the competition between the element of interest and other major and minor constituents present within the sample, it is important to carry out uptake studies in the presence of some of these elements. For example, the main elements in bone ash that could affect the uptake of plutonium and americium on a variety of resins are calcium and phosphate, along with many other impurities. [12]

The batch technique is commonly used to determine k' trends by observing the retention strengths for individual ions onto the resin. It is a useful way to observe the effects of individual elements and their contributions to the resins. The k' -value typically expresses the affinity of the element to the resin and this value can determine the successfulness of the extraction mechanism. [11] The competition with other elements can be determined by comparing the k' obtained in the absence of an interfering element with the k' -values measured in the presence of varying amounts of individual ions. The k' -value can be obtained by allowing the aqueous solution consisting of the radioisotope in a suitable acid matrix to interact with a known mass of resin for a suitable amount of time to reach a point of equilibrium.

1.6 Liquid Scintillation

Liquid Scintillation Counting (LSC) is a widely used method to determine and quantify the amount of radioactivity contained within samples, mostly beta-emitting and alpha-emitting isotopes. [15] Samples are typically dissolved in a known amount of scintillation cocktail that contains an aromatic organic solvent, such as xylene or toluene, and small amounts of other additives known as 'fluors' or scintillators. A waveshifter, which is a second organic compound, is often required in order to re-emit the photons adsorbed from the primary scintillator. The waveshifter absorbs photons from the primary scintillator and re-emits photons at a longer wavelength (~425 nm), that is then detected by the LSC photomultiplier tubes (PMT). [15] Alpha or beta particles emitted from the sample transfer energy to the solvent molecule in the solution. The energized solvent molecules subsequently transfer their energy to the scintillants, which excite and emit light. The PMT detects the light that is emitted, and the result is measured in cpm (counts per minute). An overview of the scintillation process is shown in figure 6.

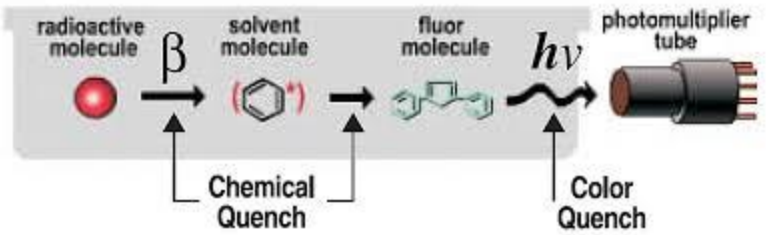


Figure 10. Schematic overview of the scintillation process. [15]

In liquid scintillation counting, the level of quenching that takes place in a given sample is a very important factor. Quenching is defined as the loss of counts due to sample or cocktail characteristics. It may result from a variety of components in a sample and occurs when there is an incomplete transfer of particle energy to solvent molecules, thus reducing the light output for the sample. [15] Quenching can be separated into two broad categories: chemical and optical. Chemical quench occurs when another chemical competes with the primary scintillator for the excitation energy in the solvent. In beta counting, quenching is often encountered when the energy emitted by the beta particle is absorbed by compounds that will not re-emit its energy, thus inducing an incomplete transfer of particle energy to solvent molecules. As follows, no light will reach the detector. Optical or color quench occurs when the light output of the scintillator is absorbed to some degree by the coloring in the sample. A result, the signal detected by the PMT will not represent the total light emitted from the sample. [15] The Special Index of the Transformed External Standard Spectrum (tSIE) value gives an estimate of quench on a scale of 0 (most quenched) to 1,000 (unquenched). A standard of known activity in disintegration per minute (dpm) is used to determine a system's counting efficiency. [15] The standard is analyzed

and the output in counts per minute (cpm) measured to determine the counting efficiency using the equation below:

$$\text{Counting Efficiency (\%)} = \frac{\text{CPM} \cdot 100}{\text{DPM}}$$

Where:

CPM = counts per minute

DPM = disintegration per minute

Vial selection is important due to its effect on background and efficiency. Glass scintillation vials contain ^{40}K, which can create Cerenkov radiation in the scintillation fluid, adding to the background signal. Polyethylene (plastic) vials are therefore preferred in the assay of samples containing low radioactivity. [15]

1.7 Literature Background

There have been a number of recent studies that investigated the use of extraction chromatographic techniques for the separation of actinide elements with different acidic matrices. However, none of these investigated the effect of all of the interfering elements present in bone ash.

1.7.1 Mietelski et al, 2011

While the main technique in this particular investigation is alpha spectroscopy, TEVA resin was mentioned as one of the preparation steps. In this study, the authors present a new method for sampling in-body bone-seeking actinides such as ^{239}Pu, ^{241}Am, and a few other alpha-emitters. Human bones obtained from routine hip or knee joints replacement surgery were analyzed for actinides. This provides a simple and ethical way to obtain bone samples for plutonium and

americium analysis and allow the assessment of actinides background in the general public. Typically, when a person has been exposed to high levels of alpha emitters, an extensive amount of bone tissue (generally taken post mortem) is required for alpha spectrometry. Since most human tissues post-surgery are incinerated and disposed, these tissues are ideal for analysis. This is extremely beneficial for studies, since bone samples from routine surgery are both abundant in number and easily accessible. In addition, patients receiving this routine surgery are typically over 50 years old and have been subjected to global fallout.

A detailed explanation of the procedure can be found in the literature. [16] A total of 23 samples were investigated. The analysis of bone from operations was compared to bone from autopsies and found to be comparable in plutonium concentration. High plutonium activity due to global fallout was confirmed in humans from southern Poland by using the activity ratio (^{238}Pu/ $^{239+240}$Pu) to identify the origin of the Plutonium in bones. Also, since the patients were still living, the opportunities to interview patients to obtain information regarding their lifestyle could help identify their level of internal contamination.

1.7.2 Gharibyan et al. 2014

Gharibyan et al. performed a characterization of seven chromatography resins (TEVA, TRU, DGA(N), Actinide, Ln, Ln2, and Ln3) for Am(III) and Cm(III) from acidic matrices (HNO_3, HCl, and HBr). The uptake of americium and curium by the seven different resins was measured by equilibrating a known amount of each actinide with a known amount of resin of interest. Approximately 50 mg of resin (exact amount measured before preconditioning), in 1.5 mL centrifuge tubes, was preconditioned with 0.800 mL acid of known concentration by mixing with a Labquake Rotisserie shaker for at least one hour. To the preconditioned resin, 0.500 mL of

either ^{241}Am or ^{244}Cm tracer solution (~100 Bq/mL) in the same acid matrix (at 0.01 M) was added and mixed with the shaker for another hour to reach equilibrium. Kinetic studies by Horwitz 2006, have shown that equilibrium is reached for all seven resins within 60 minutes. Samples were allowed to sit overnight, and the aqueous phase was filtered from the resin using a syringe attached to a 0.45 µm Whatman PTFE filter. From the filtered solution, an aliquot of 1.000 mL was mixed with 15 mL of LSC cocktail and analyzed with LSC to determine the total amount of ^{241}Am or ^{244}Cm left in solution after contact with the resin. At high acid concentrations, quenching effects were considered by reanalyzing each data set to include the full ^{241}Am or ^{244}Cm alpha peaks. The final solution conditions were defined by the combination of 0.800 mL of the known concentration of acid from preconditioning and 0.500 mL of 0.01 M acid from the tracer solution. For Actinide resin where k' -values of greater than 105 were encountered, batch studies were repeated with 5 mg of resin instead of 50 mg and the activity of the tracer solutions was increased from ~100 Bq/mL to ~2000 Bq/mL. Under most circumstances, Am(III) and Cm(III) have similar extraction attributes. The result of their experiment provided evidence that separation is possible with DGA(N) and TRU resins with emphasis on the dependence of specific anion in solution in this order NO_3^->Br^->Cl^-. [22]

1.7.3 Daum et al. 2015

Daum et al. focused on how the salinity of an ocean water matrix can impact the isolation, characterization, and determination of ^{239}Pu and ^{241}Am using a variety of resins, in particular the resins Actinide and Diphonix. Ocean water has an 85% salinity concentration of sodium and chloride ions (0.459 and 0.536 M respectively), so the batch contact studies focused on the retention effect on ^{239}Pu in the presence of sodium and chloride ions. None of the sodium chloride matrix concentrations with the acid (nitric acid or hydrochloric acid), significantly

affected the retention of ^{239}Pu on the six extraction chromatographic resins investigated. The retention of ^{241}Am on the six extraction chromatographic resins in the presence of varying concentrations of a synthetic sodium chloride salt solution will be examined in future work. Artificial ocean water was also used with a similar procedure and was prepared by using the American Society for Testing and Materials (ASTM) procedure ASTM D1141-98. The retention of ^{241}Am on DGA resin with nitric acid was found to be reduced by over two orders of magnitude within artificial ocean water. This work is important as ocean water is the largest recipient of environmental contamination by anthropogenic radionuclides. [5]

1.8 Objective of Research

There are numerous methods for separating and analyzing radionuclides of interest from an array of environmental matrices. The efficiency of each methods can be impacted by the presence of elemental constituents that are commonly found in the sample matrix. The objective of this research is to investigate the influence of the major and minor constituents present in bone on the radioanalytical determination of plutonium and americium using extraction chromatographic resins. If the recovery of Pu and Am is reduced due to the presence of Ca or one of the other stable elements present in bone, then the accuracy with which low-levels of Pu or Am can be determined in bones could be affected. The purpose of this work is therefore to ascertain the impact of the presence of Ca^{+}, K^{+} and Mn^{+}, on the efficiency of ^{239}Pu and ^{241}Am uptake on DGA, TRU, TEVA, and UTEVA resins.

CHAPTER 2: MATERIALS AND METHODS

2.1 Batch Distribution Study

In order to observe the contribution of specific metal ions toward the sorption of isotopes of plutonium and americium onto DGA, TRU, TEVA, and UTEVA resins, a batch distribution technique, as described in chapter 1.5 was utilized. The results obtained for the adsorption of plutonium and americium on the different resins at varying concentrations of the stable elements of interest were then converted into k' -values. The k' -values were graphed logarithmically to visualize trends and to identify discrepancies.

A known activity of ^{239}Pu and ^{241}Am together with a given concentration of a stable element of interest were allowed to reach equilibrium for the experiment. The stable elements that were considered as potential interferents included calcium, potassium, and magnesium. Solutions with varying concentrations of 0, 0.001, 0.005, 0.01, 0.05, 0.1, 0.25, 0.5, and 1 M of each of these elements were used for the uptake experiments and analyzed in replicates of four for each concentration observed.

Two different mineral acids (nitric acid and hydrochloric acid) were used to determine the retention factors for ^{239}Pu and ^{241}Am on the four different resins. Nitrates or chlorides were already present in the plutonium or americium standards. Therefore, the overall concentration of nitrate or chloride ions were not altered in each sample. An aliquot of the supernatant was collected from each resin batch sample and placed into individual plastic vials containing liquid scintillation cocktail and counted on a liquid scintillation counter. The average of the four samples analyzed from each concentration was plotted and observed.

2.2 Batch Study Procedure

2.2.1 Resin preparation

To find the ideal conditions for the batch experiments, acid dependency, kinetics and mass independence studies from the literature for each of the extraction chromatographic resins were investigated. Polypropylene microcentrifuge tubes were filled with 50 ± 0.05 mg of the resins (DGA, TRU, TEVA, and UTEVA), as show in figure 11.

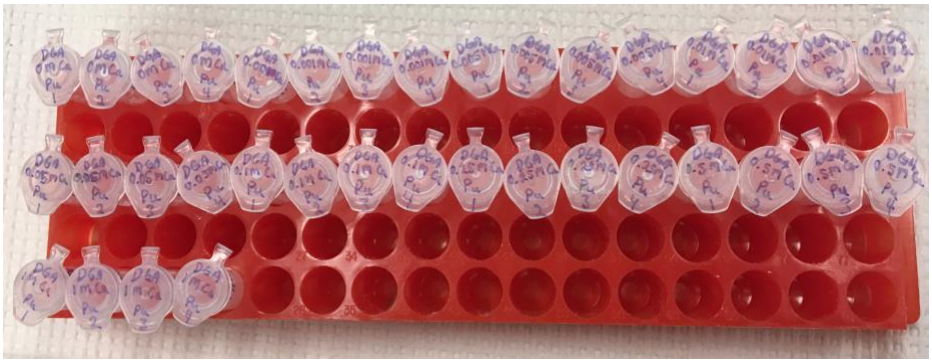


Figure 11. Polypropylene microcentrifuge tubes were filled with 50 ± 0.05 mg of the desired resin.

The resin in each tube was then preconditioned with 0.45 mL of the preferred acid concentration. A nitric acid and hydrochloric acid at a concentration of 1 M was used for experiments with DGA, while the studies with TRU, TEVA, and UTEVA were carried out either in 3 M nitric acid or 3 M hydrochloric acid. For each batch, exactly 36 samples were laid on their sides and agitated for 1 hour using a Thermo Scientific Labquake shaker with a fixed speed. The samples were then allowed to mix and fully swell for >12 h in preparation for the next step, as shown in figure 12.

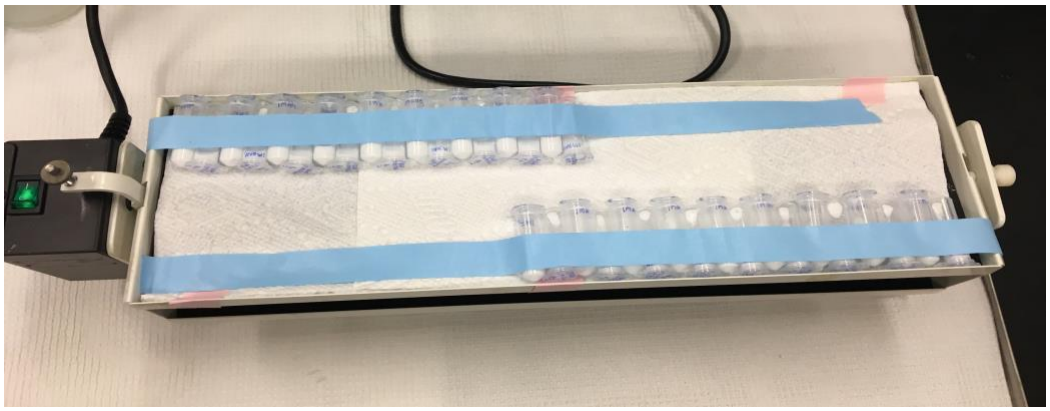


Figure 12. Samples agitated using using a Thermo Scientific Labquake shaker with a fixed speed.

2.2.2 Spiking the Resins with Ions and ^{239}Pu, ^{241}Am

Next, the resin tubes were spiked with 1 mL of the desired salt solution at each given concentration (0, 0.001, 0.005, 0.01, 0.05, 0.1, 0.25, 0.5, and 1 M) and 50 µL of 1000 Bq/mL (~50 Bq) ^{239}Pu or ^{241}Am in the respective acid concentration. The microcentrifuge tubes were agitated for one hour using the Thermo Scientific Labquake shaking table, thoroughly mixing the contents and ensuring all extraction reactions would reach a point of equilibrium.

2.2.3 Filtration

Following agitations, the samples were subsequently filtered through a 0.45 µm PTFE Whatman syringe filter. The filters were attached, screwed onto Luer-Lok syringes and used to separate the supernatant from the resins into new labeled microcentrifuge tubes shown in figure 13.

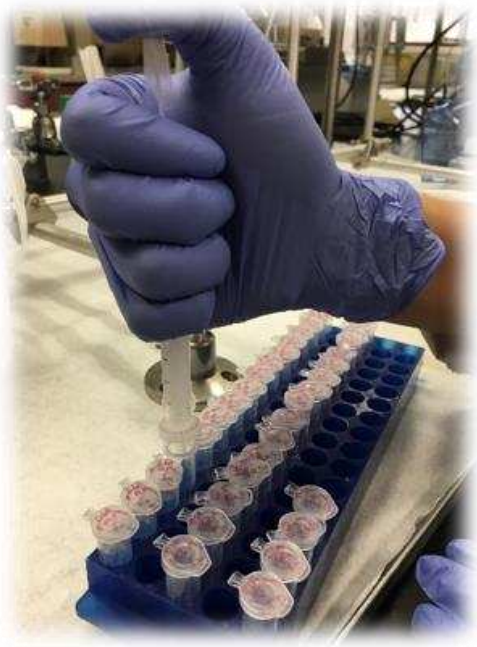


Figure 13. Separating the supernatant from the resins into new labeled microcentrifuge tubes.

2.4 Liquid Scintillation Counting Procedure

All plastic scintillation vials were prepared by addition of 15 mL of liquid scintillation cocktail and were measured using a Perkin Elmer Model Tri-Carb 5110TR liquid scintillation counter and analyzed using QuantaSmartTM Software. The ^{239}Pu and ^{241}Am standards were assessed by placing 50 µL (1000 Bq/mL) of a plutonium or americium standard solution in a plastic vial containing LSC cocktail, and counting efficiency was assessed after subsequent measurement. A 0.9 mL aliquot of each filtered solution from each resin tube sample was added to a plastic vial containing LSC cocktail. Blank samples were prepared by replacing the aliquot of filtered radionuclide solution with 0.9 mL of diluted nitric acid or diluted hydrochloric acid to imitate the sample matrix. The blank samples and standards were counted at the same time as the samples and used to background subtract shown in figure 14.

Figure 14. Samples counted with Perkin Elmer Model Tri-Carb 5110TR liquid scintillation counter and analyzed using QuantaSmartTM Software.

Each vial was counted for 60 minutes and the average of the four replicates resin samples at each concentration was calculated (the standard deviation was used to calculate for the uncertainty of the replicate measurements). The count mode was set to normal and the pre-count delay was set to 0 minutes. The average CPM for the blank samples were subtracted from each resin sample to remove the background counts using Microsoft Excel. A channel region from channel 200 through channel 2000 was used to collect counts. The average count rate per set of replicates was used to calculate the percent yield of ^{241}Am or ^{239}Pu from the separation experiment.

2.5 Materials

A list of materials used for all experiments can be found in appendix II.

CHAPTER 3: RESULTS

3.1 Data Analysis

3.1.2 Batch Distribution Studies

The effect of each potentially interfering stable element was investigated at nine different concentrations, 0, 0.001, 0.005, 0.01, 0.05, 0.1, 0.25, 0.5, and 1 M. Experiments at each concentration were performed and analyzed in replicates of four. The data points shown are the averages of the four replicates. The standard deviation was used to evaluate uncertainty and forms the basis for the error bars plotted in the graphs in this chapter. In some cases, the size of the error bars is smaller than the size of the data points displayed.

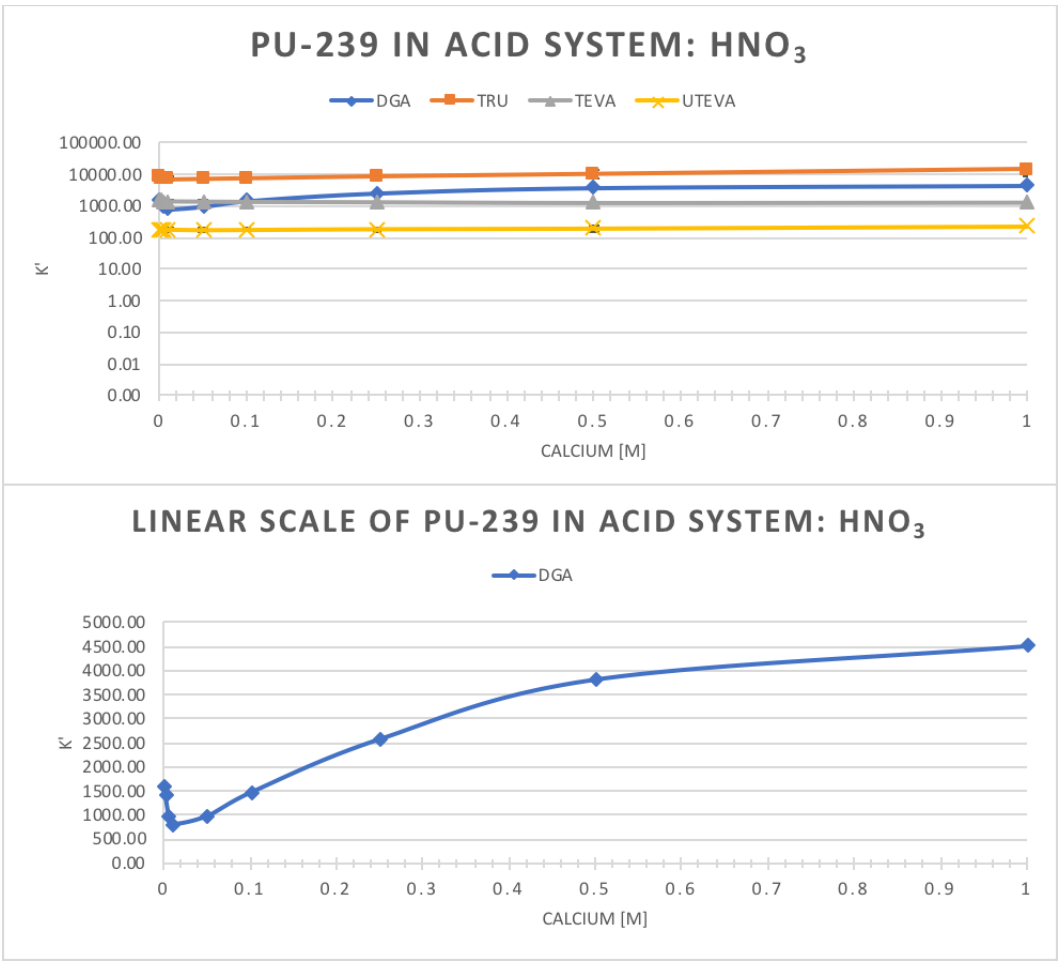


Figure 15. Contribution of calcium in nitric acid on the uptake of ^{239}Pu from a calcium nitrate solution. See Appendix III, table 3.

To observe the elemental effects of calcium, calcium nitrate dissolved in 1 M nitric acid for DGA and in 3 M nitric acid for the other three resins (TRU, TEVA, UTEVA) was used. The results shown in figure 15 suggest that calcium inhibits the sorption of ^{239}Pu to DGA resin slightly at concentrations between 0-0.05 M. The affinity for ^{239}Pu on to the DGA resin slowly increases after the calcium concentration exceeds 0.05 M. Figure 15 also shows that the retention of ^{239}Pu on the remaining three resins with nitric acid was not affected by the calcium solution.

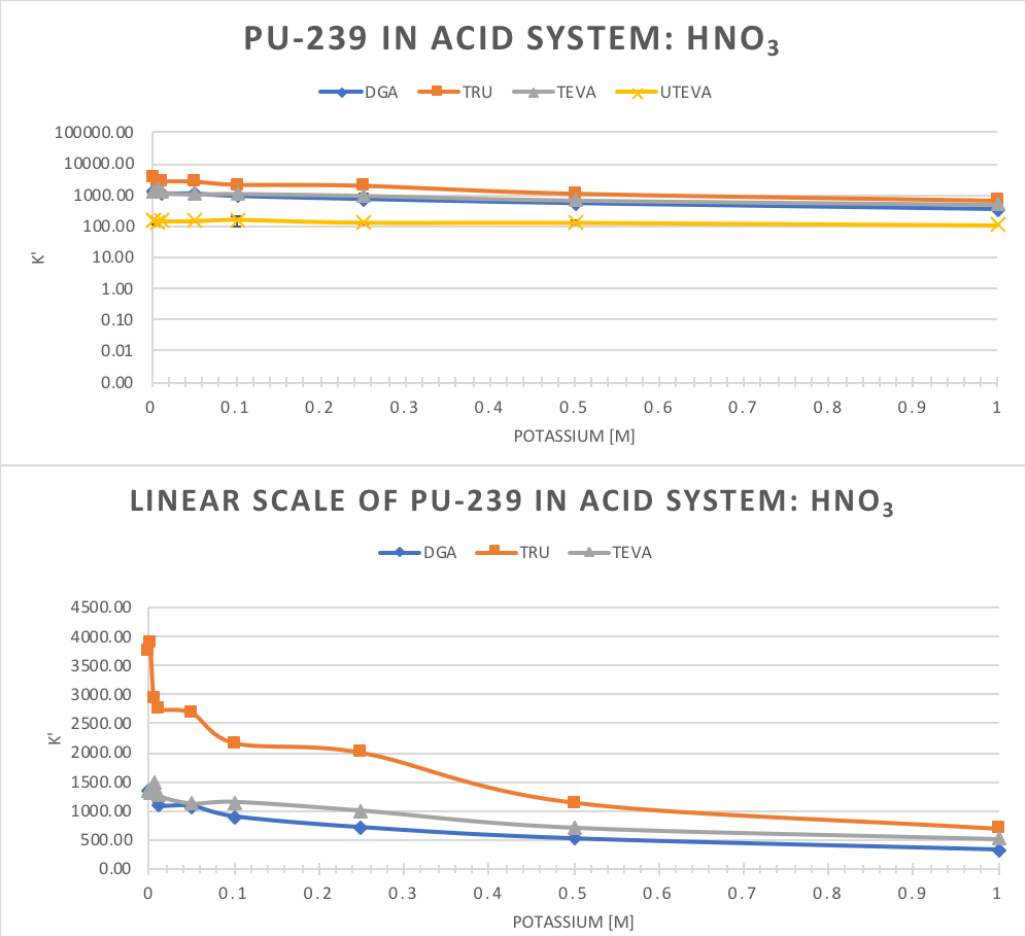


Figure 16. Contribution of potassium in nitric acid on the uptake of ^{239}Pu from a potassium nitrate solution. See Appendix III, table 4.

Potassium nitrate dissolved in 1 M nitric acid for DGA and in 3 M nitric acid for the other three resins (TRU, TEVA, UTEVA) was used to observe the elemental effects of potassium. The results shown in figure 16 suggests that potassium slightly inhibits the sorption of ^{239}Pu to DGA, TRU, and TEVA resin slightly over the potassium concentration rage from 0-1 M. The retention of ^{239}Pu on the remaining UTEVA resin with nitric acid was not affected by the potassium nitrate solution.

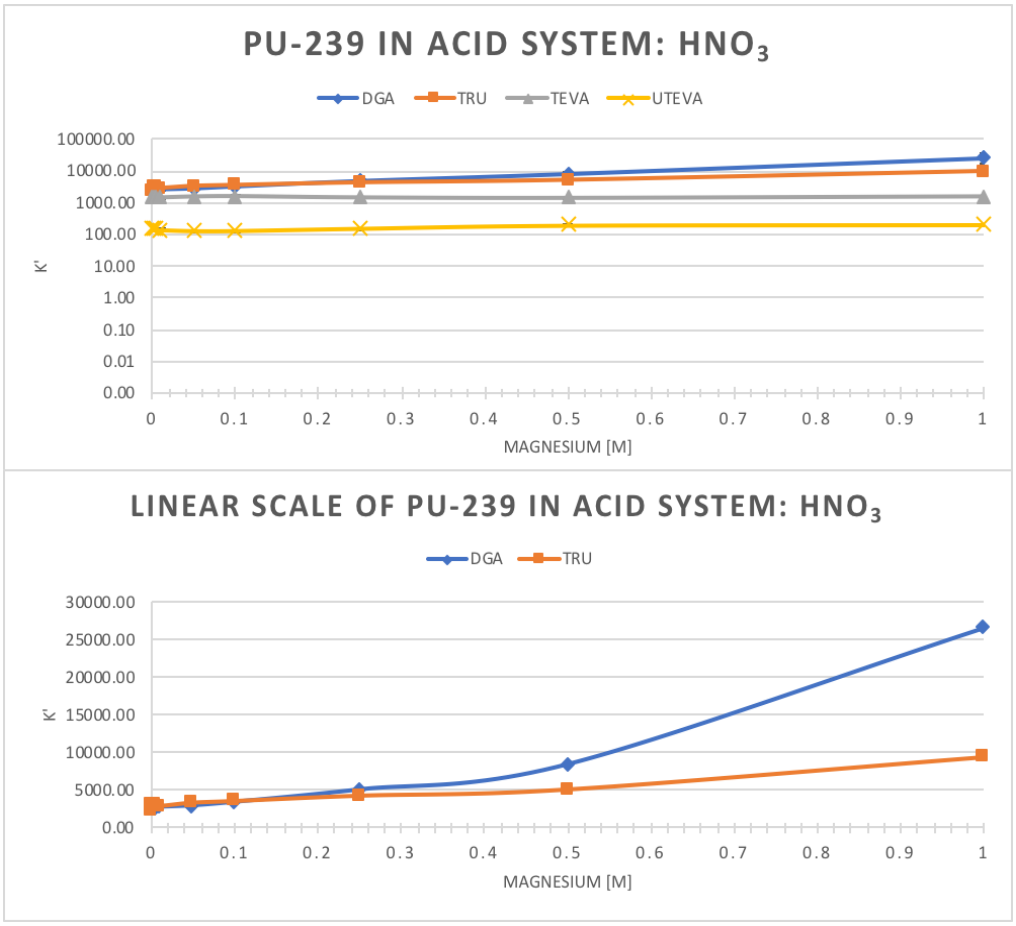


Figure 17. Contribution of magnesium in nitric acid on the separation of ^{239}Pu from a magnesium nitrate solution. See Appendix III, table 5.

To observe the elemental effects of magnesium, magnesium nitrate dissolved in 1 M nitric acid for DGA and in 3 M nitric acid for the other three resins (TRU, TEVA, UTEVA) was used. The results shown in figure 17 suggest a synergism effect of the magnesium nitrate that leads to an increase of the k' -value for ^{239}Pu on two of the resins (DGA and TRU). The retention of ^{239}Pu on the remaining two resins from nitric acid was not affected by the magnesium nitrate solution.

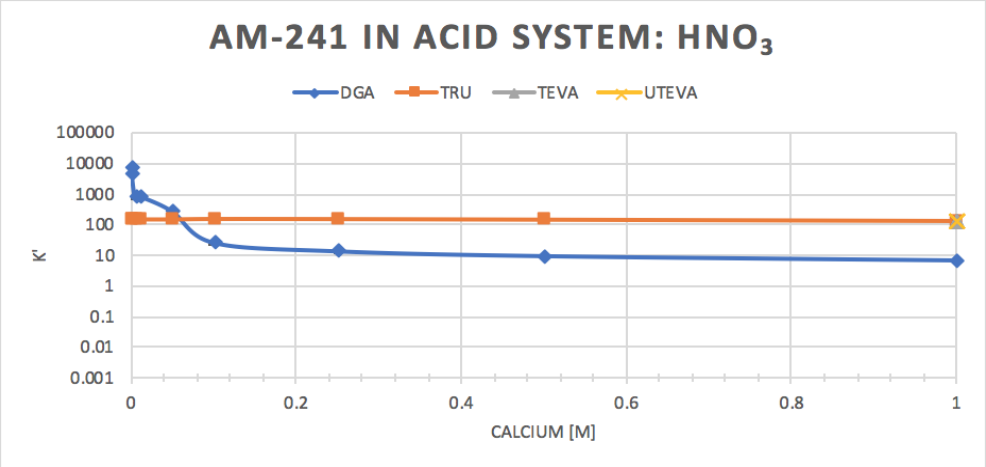


Figure 18. Contribution of calcium in nitric acid on the uptake of ^{241}Am from a calcium nitrate solution. See Appendix III, table 6.

The results for the uptake of ^{241}Am on the four resins from nitric acid in the presence of calcium are shown in figure 18. The data suggests that calcium inhibits the sorption of ^{241}Am to DGA resin even at low concentrations. This is evident from the rapid decrease in k' -value between 0-0.1 M, which then slowly tapers off at calcium concentrations greater than 0.1 M. The increase at a calcium concentration of approximately 0.01 M was due to solution loss during the shaking process for one of the replicates. The retention of ^{241}Am on TRU resin with nitric acid was not impacted by calcium. The two remaining resins exhibit negative k'-values and anything below zero on a logarithmic scale will not show up on the graph. The negative k' is due to a volume reduction in the sample. When water is adsorbed by the resin without a simultaneous uptake of the radionuclide, then the activity concentration of the sample appears to increase, resulting in a greater count rate than for the standard solutions.

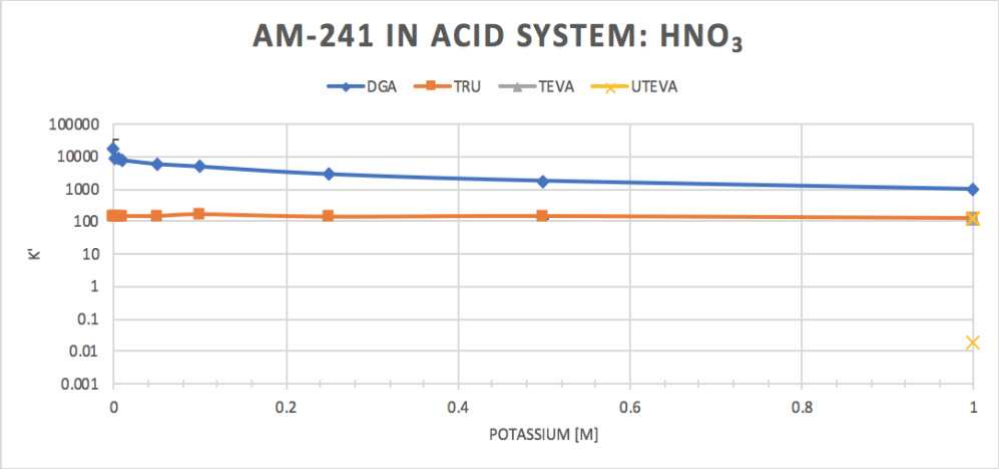


Figure 19. Contribution of potassium in nitric acid on the uptake of ^{241}Am from a potassium nitrate solution. See Appendix III, table 7.

The results for the uptake of ^{241}Am on the four resins from nitric acid in the presence of potassium are shown in figure 19. It can be seen that potassium inhibits the adsorption of ^{241}Am on DGA resin. The Am-241 absorption rapidly decreases as the potassium concentration increases from 0-1 M, resulting in a reduction of the k'-value. The retention of ^{241}Am on TRU resin with nitric acid was not affected by the calcium solution. The two remaining resins showed no uptake on the resin.

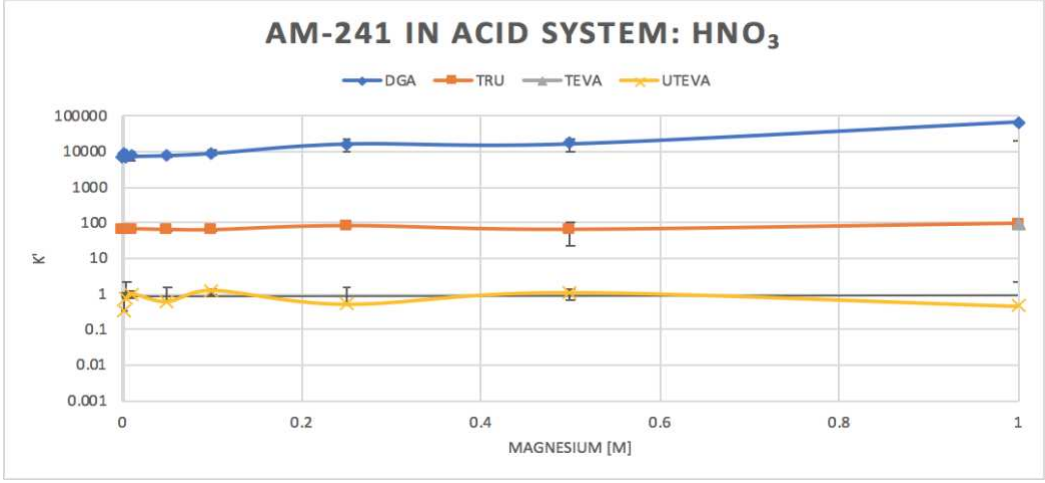


Figure 20. Contribution of magnesium in nitric acid on the uptake of ^{241}Am from a magnesium nitrate solution. See Appendix III, table 8.

The results for the uptake of ^{241}Am on the four resins from nitric acid in the presence of magnesium are shown in in figure 20. The data suggests a synergism effect due to the presence of the magnesium nitrate. The retention of ^{241}Am on TRU resin with nitric acid was not affected by the magnesium in solution. There is some minor fluctuation for UTEVA with only 0.42 standard deviation from the mean. TEVA resin showed no uptake for americium.

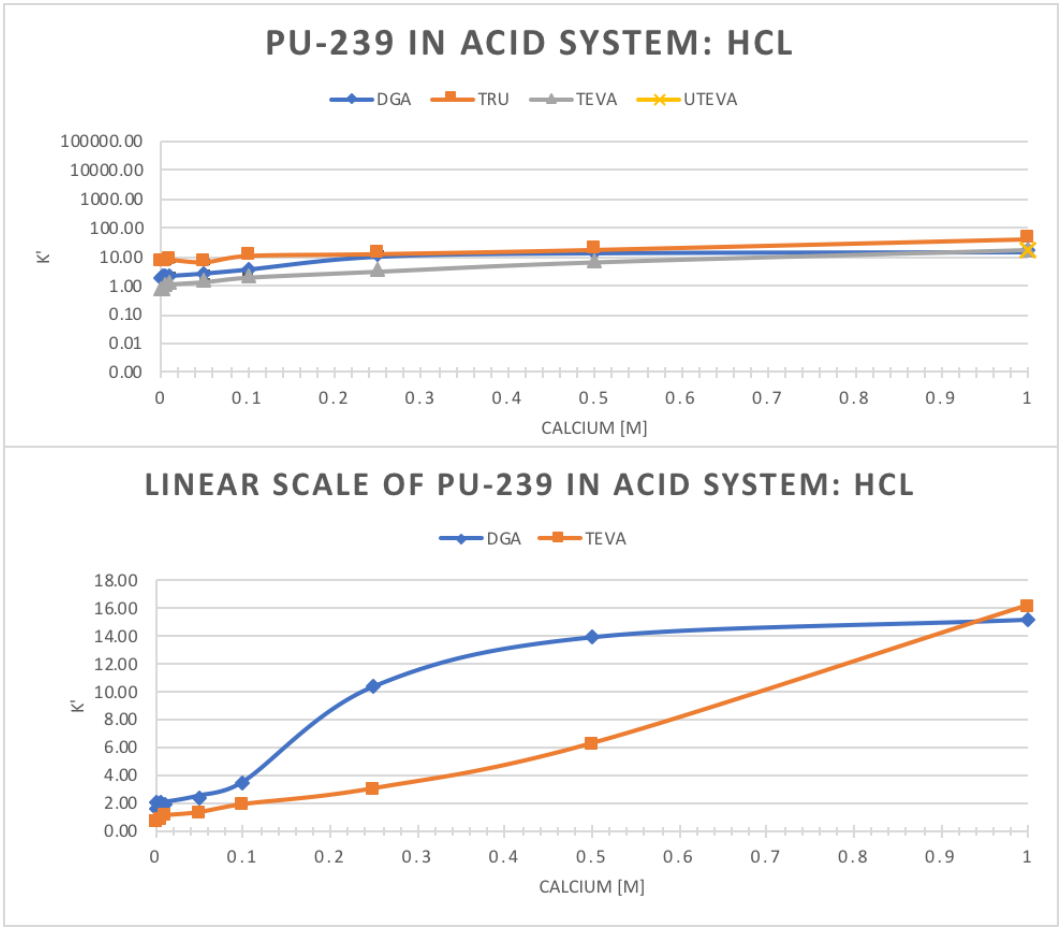


Figure 21. Contribution of calcium in hydrochloric acid on the uptake of ^{239}Pu from a calcium chloride solution. See Appendix III, table 9.

Calcium chloride dissolved in 1 M nitric acid for DGA and in 3 M nitric acid for the other three resins (TRU, TEVA, UTEVA) was used to observe the elemental effects of calcium. The results for the uptake of ^{239}Pu on the four resins from nitric acid in the presence of calcium are shown in

figure 21. The data suggest synergism between calcium chloride and ^{239}Pu onto two of the resins (DGA and TEVA). There is an increase in k'-value for DGA from 0-0.2 M then it slowly tapers off. There is a gradual increase in k'-value for TEVA up until 0.5 M where it tapers off. The retention of ^{239}Pu on the remaining TRU resins appears not to be affected by the calcium solution. The remaining UTEVA resin showed no uptake for plutonium.

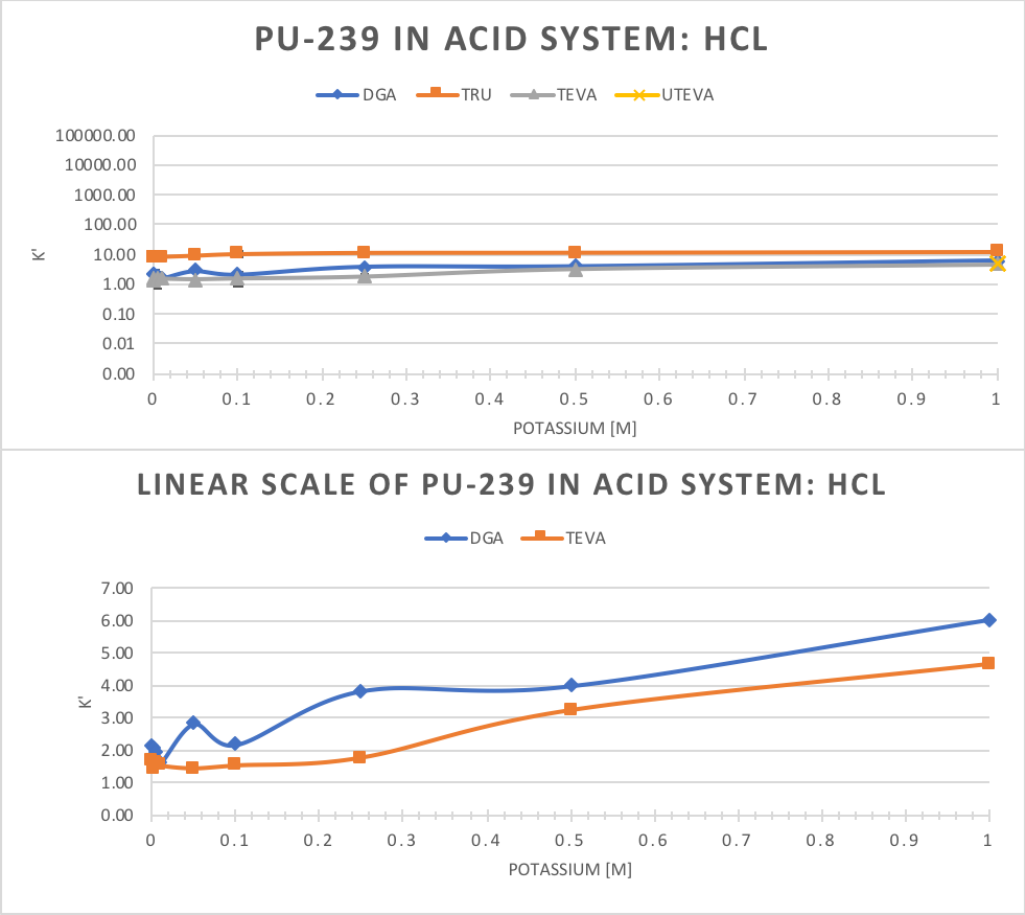


Figure 22. Contribution of potassium in hydrochloric acid on the uptake of ^{239}Pu from a potassium chloride solution. See Appendix III, table 10.

Potassium chloride dissolved in 1 M nitric acid for DGA and in 3 M nitric acid for the other three resins (TRU, TEVA, UTEVA) was used to observe the elemental effects of potassium. The results for the uptake of ^{239}Pu on the four resins from nitric acid in the presence of calcium are

shown in figure 22. There appears to be a slightly synergistic relationship between potassium chloride and ^{239}Pu on two of the resins (DGA and TEVA). The retention of ^{239}Pu on TRU resin with was not significantly affected by the potassium solution. There was no uptake of plutonium on the UTEVA resin.

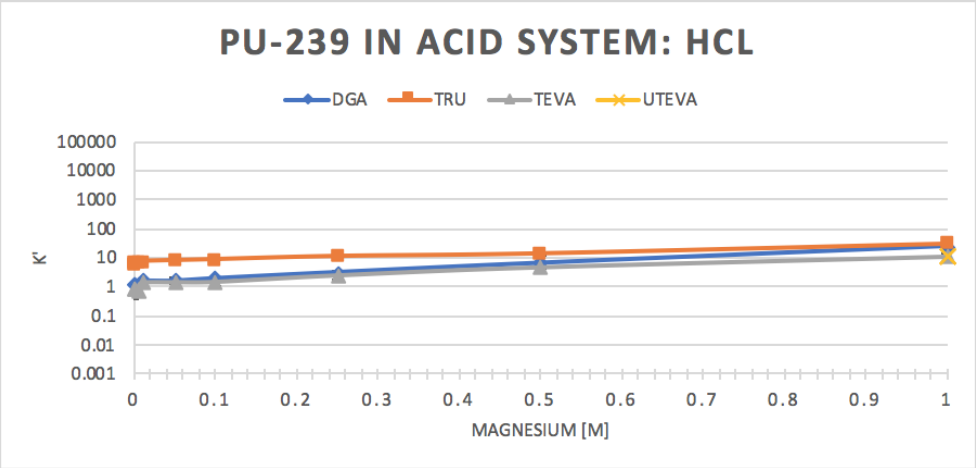


Figure 23. Contribution of magnesium in hydrochloric acid on the uptake of ^{239}Pu from a magnesium chloride solution. See Appendix III, table 11.

Magnesium chloride dissolved in 1 M nitric acid for DGA and in 3 M nitric acid for the other three resins (TRU, TEVA, UTEVA) was used to observe the elemental effects of magnesium. The results for the uptake of ^{239}Pu on the four resins from nitric acid in the presence of calcium are shown in figure 23. Again, there appears to be a slight synergistic effect caused by the magnesium chloride on two of the resins (DGA and TEVA). The retention of ^{239}Pu on TRU resin with was not affected by the magnesium solution. There was no uptake of plutonium on the UTEVA resin.

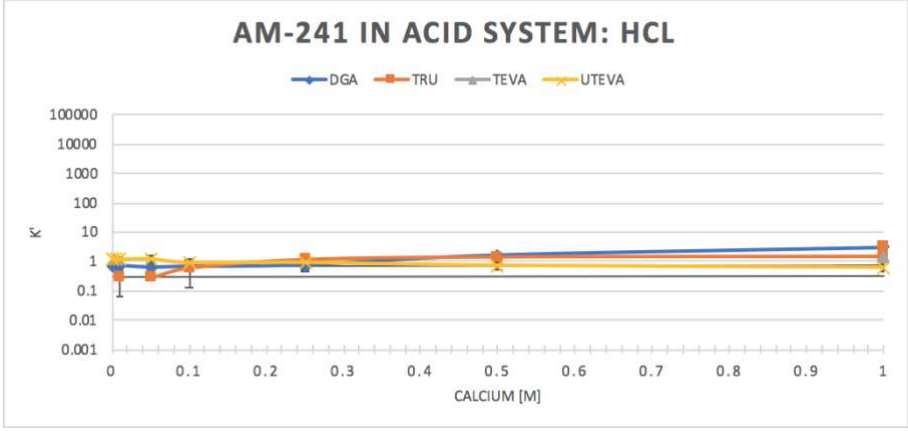


Figure 24. Contribution of calcium in hydrochloric acid on the uptake of ^{241}Am from a calcium chloride solution. See Appendix III, table 12.

The results for the uptake of ^{241}Am on the four resins from nitric acid in the presence of calcium are shown in figure 24. The retention of ^{241}Am from hydrochloric acid does not appear to be affected by the presence of the calcium chloride solution on any of the resins investigated.

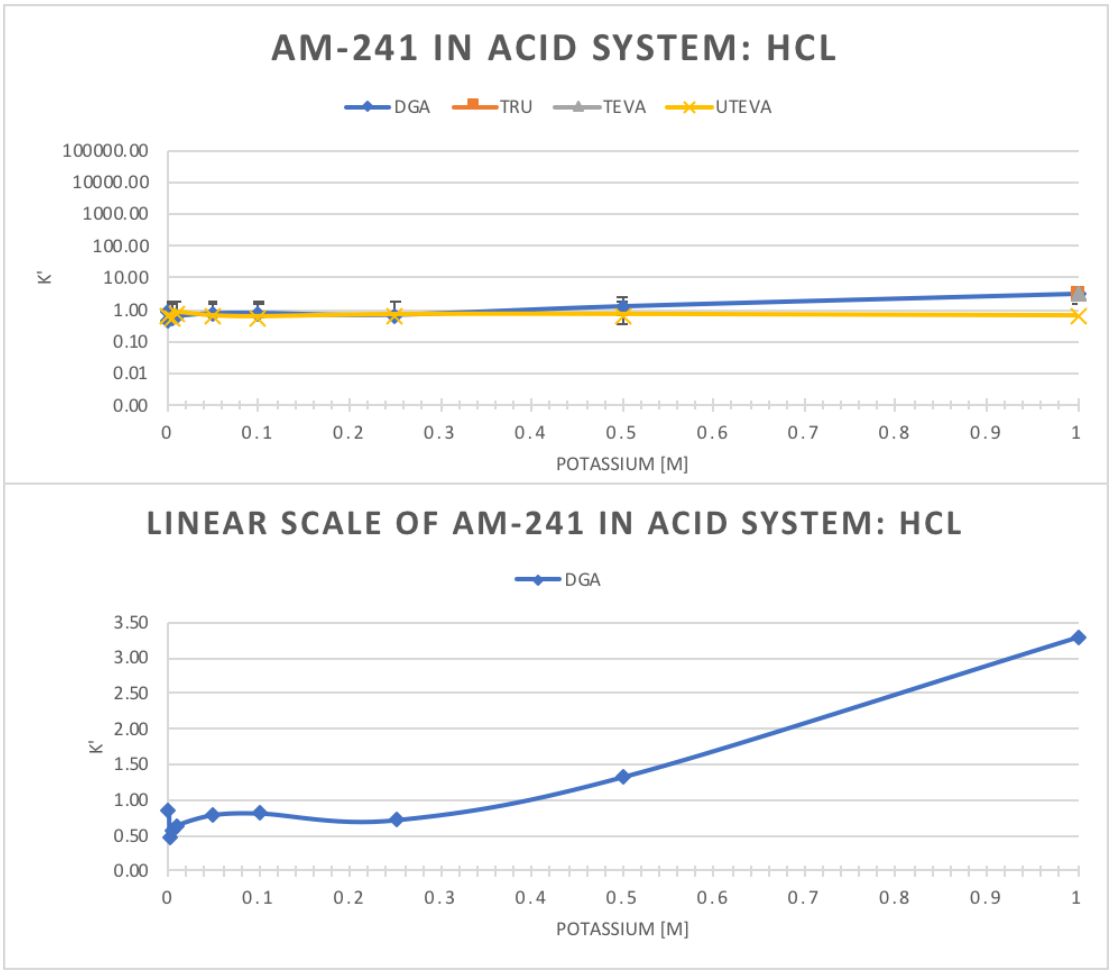


Figure 25. Contribution of potassium in hydrochloric acid on the uptake of ^{241}Am from a potassium chloride solution. See Appendix III, table 13.

The results for the uptake of ^{241}Am on the four resins from nitric acid in the presence of potassium are shown in figure 25. For DGA, an increase in k'-value suggest a synergistic effect due to the presence of the potassium chloride solution. There is a gradual increase in k'-value for DGA in the magnesium concentration range from 0.25-1 M. The retention of ^{241}Am on UTEVA resin from hydrochloric acid was not significantly affected by the potassium solution. No uptake of americium was seen for the TRU and TEVA resins.

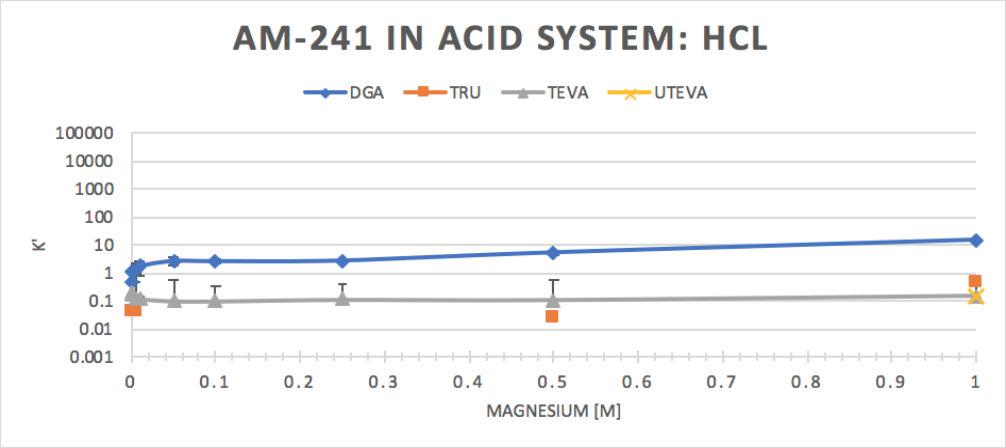


Figure 26. Contribution of magnesium in hydrochloric acid on the uptake of ^{241}Am from a magnesium chloride solution. See Appendix III, table 14.

The results for the uptake of ^{241}Am on the four resins from nitric acid in the presence of magnesium are shown in figure 26. Again, there appears to be a slightly synergistic effect on the uptake of americium on DGA resin due to the presence of the magnesium chloride solution. There is a gradual increase in k'-value for DGA in the magnesium concentration range from 0.25-1 M. The retention of ^{241}Am on TEVA resin from hydrochloric acid was not significantly affected by the magnesium solution. No uptake of americium was seen for the TRU and TEVA resins.

CHAPTER 4: DISCUSSION

4.1 Batch Distribution Studies

The batch distribution studies performed provide an important method to assess the influence of common elements found in bone on the adsorption of ^{239}Pu and ^{241}Am on DGA, TRU, TEVA, and UTEVA resins from Eichrom Technologies. The interfering ions of interest investigated were calcium, potassium, and magnesium, all of them common major or minor constituents of bones.

Calcium, potassium, and magnesium all affect the retention of ^{241}Am onto DGA in some way. The contribution of calcium from calcium nitrate with nitric acid reduced adsorption of ^{241}Am on DGA resin in the calcium concentration range from 0-0.1 M. Potassium, from potassium nitrate with nitric acid also gave rise to a rapid decrease in k' for potassium concentrations between 0-1 M. Magnesium from magnesium nitrate caused an increase of k' between 0-1 M due salting out, which is common in aqueous solutions of high ionic strength. In solution, both Mg(II) and Am(III) are competing for the resin binding site. However, as the concentration of the salt increases, some of the molecules of water are attracted to the salt ions resulting in a decrease of water molecules available to interacts with the charged part of Am(III). Thus, more Am(III) is readily available to bind itself to the binding sites on the resin thereby increasing the k'-value. [23] It should be noted with the addition of the salt, this will also lead to an increase in the nitrate or chloride concentration. An example mechanism is shown below:

$$3Mg + 8HNO_3 = 4H_2O + 3Mg(NO_3)_2 + 2NO$$

Since the cation salts are attracted to the acids which are anions, there will be less competition for Am(III) to bind to the binding sites on the resin potentially reducing the activity in the solution giving a higher k'-value.

Table 2 is a summary of ^{239}Pu and ^{241}Am results where resins were not significantly affected. In order for a retention factor to be appreciatively influenced, a change of more than an order of magnitude in value for k' should be demonstrated.

Table 2. Combined uptake results for Pu-239 and Am-241 for all sets of data

	Hydrochloric Acid		Nitric Acid		
	Pu-239	**Am-241**	**Pu-239**	**Am-241**	
DGA	Slight increase of k' from 0-0.2 M, then tapers off.	Slight increase of k' from 0.25-1 M	Slight decrease of k' up to 0.5 M, then increase of k'	Rapid decrease of k' from 0-0.1 M, then tapers off	**Ca**
	Slight increase of k' from 0.25-1 M	Slight increase of k' from 0.25-1M	Slight decrease of k' from 0-1 M	Rapid decrease of k' from 0-1 M	**K**
	Slight increase of k' from 0-1 M	Slight increase of k' from 0-0.25 M then larger increase after 0.25	Slight increase of k' from 0-1 M	Rapid increase of k' from 0-1 M	**Mg**
TRU	No effect	No effect	No effect	No effect	**Ca**
	No effect	No effect	Slight decrease of k' from 0-1 M	No effect	**K**
	No effect	Small or no uptake	Slight increase of k' from 0-1 M	No effect	**Mg**
TEVA	Slight increase of k' from 0-0.5 M, then tapers off	No uptake	No effect	No uptake	**Ca**
	Slight increase of k' from 0.25-1 M	No uptake	Slight decrease of k' from 0-1 M	No uptake	**K**
	Slight increase of k' from 0-1 M	No effect	No effect	No uptake	**Mg**
UTEVA	No uptake	Slight decrease of k' from 0-1 M	No effect	No uptake	**Ca**
	No uptake	No effect	No effect	No uptake	**K**
	No uptake	No uptake	No effect	Minor fluctuation of k'	**Mg**

CHAPTER 5: CALCULATIONS & UNCERTAINTY ANALYSIS

5.1 Data Analysis

5.1.1 Mean Calculations

The mean was calculated for all data sets including blank samples (Knoll, 2010). The equation for the mean, $\bar{x}$, is shown below:

$$\bar{x} = \sum \frac{x_i}{N}$$

Where:

x_i = number of counts obtained for each sample under uniform conditions

N = number of samples

5.1.2 Standard Deviation Calculations

The standard deviation, σ, was calculated to determine the deviation from the mean. The equation is shown below:

$$\sigma = \sqrt{x}$$

Where:

x = number of counts of each sample

Microsoft Excel was used to calculate the mean and standard deviation for all data set.

5.2 Uncertainty during the Sample Preparation Process

Uncertainty introduced during the preparation of samples included static charged causing the resins to stick on the rims of the microcentrifuge tubes. Consequently, the lids were not able to be snapped completely. This resulted in loss of samples during the shaking process, thus, contributed to the final uncertainty of the mean.

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APPENDIX I: CHEMICALS

Nitric Acid, ACS Grade

CAS 7697-37-2

Hydrochloric Acid, ACS Grade

CAS 7647-01-0

Calcium nitrate

CAS 35054-52-5

Potassium nitrate

CAS 7757-79-1

Magnesium nitrate

CAS 13446-18-9

Potassium chloride

CAS 7447-40-7

Calcium chloride

CAS 10043-52-4

Magnesium chloride

CAS 7786-30-3

^{241}Am, Isotope Product

^{239}Pu, Isotope Product

APPENDIX II: MATERIALS AND REAGENTS

^{241}Am in 1 mol L^{-1} nitric acid, 1000 Bq mL^{-1}

^{241}Am in 3 mol L^{-1} nitric acid, 1000 Bq mL^{-1}

^{241}Am in 1 mol L^{-1} hydrochloric acid, 1000 Bq mL^{-1}

^{241}Am in 3 mol L^{-1} hydrochloric acid, 1000 Bq mL^{-1}

^{239}Pu in 1 mol L^{-1} nitric acid, 1000 Bq mL^{-1}

^{239}Pu in 3 mol L^{-1} nitric acid, 1000 Bq mL^{-1}

^{239}Pu in 1 mol L^{-1} hydrochloric acid, 1000 Bq mL^{-1}

^{239}Pu in 3 mol L^{-1} hydrochloric acid, 1000 Bq mL^{-1}

DGA resin, loose, Eichrom Technologies

TRU resin, loose, Eichrom Technologies

TEVA resin, loose, Eichrom Technologies

UTEVA resin, loose, Eichrom Technologies

Nitric Acid, 1 mol L^{-1}

Nitric Acid, 3 mol L^{-1}

Hydrochloric acid, 1 mol L^{-1}

Hydrochloric acid, 3 mol L^{-1}

Thermo Scientific Labquake Shaker (fixed speed)

Whatman PTFE membrane Syringe Filters, 0.45 µm pore size

APPENDIX III

Table 3. Calcium contribution from calcium nitrate on the separation of ^{239}Pu

Ionic Species: Calcium	^{239}Pu in Acid System: Nitric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	1579.50	160.48
	0.001	1414.63	22.38
	0.005	990.79	42.67
	0.01	812.68	29.63
	0.05	980.50	28.46
	0.1	1465.88	65.60
	0.25	2578.85	117.20
	0.5	3818.36	421.48
	1	4514.41	571.51
TRU	Concentration [M]	k' value	Standard Deviation
	0	8073.78	1039.44
	0.001	7405.57	599.20
	0.005	7155.82	586.85
	0.01	7206.58	951.96
	0.05	7456.88	592.91
	0.1	7799.04	1248.59
	0.25	8850.46	1285.46
	0.5	10446.47	3199.35
	1	14542.50	5414.44
TEVA	Concentration [M]	k' value	Standard Deviation
	0	1500.41	249.62
	0.001	1463.08	68.96
	0.005	1411.84	60.57
	0.01	1438.92	46.40
	0.05	1415.09	42.87
	0.1	1388.31	111.41
	0.25	1343.47	49.24
	0.5	1268.29	31.31
	1	1298.30	69.12
UTEVA	Concentration [M]	k’ value	Standard Deviation
	0	179.45	13.98
	0.001	168.71	10.03
	0.005	175.21	3.65
	0.01	182.43	12.99
	0.05	177.70	23.67
	0.1	179.91	16.25
	0.25	189.69	32.15
	0.5	198.03	42.24
	1	229.20	19.25

Table 4. Potassium contribution from potassium nitrate on the separation of ^{239}Pu

Ionic Species: Potassium	^{239}Pu in Acid System: Nitric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	1347.91	97.85
	0.001	1324.10	36.66
	0.005	1303.33	194.39
	0.01	1087.99	81.62
	0.05	1076.65	120.82
	0.1	898.10	24.02
	0.25	712.87	34.34
	0.5	523.11	13.41
	1	333.30	5.30
TRU	Concentration [M]	k' value	Standard Deviation
	0	3767.73	493.80
	0.001	3885.37	314.00
	0.005	2926.02	301.41
	0.01	2750.38	153.92
	0.05	2684.13	211.63
	0.1	2169.38	51.78
	0.25	2003.20	91.31
	0.5	1149.68	31.05
	1	701.91	24.03
TEVA	Concentration [M]	k' value	Standard Deviation
	0	1362.70	57.26
	0.001	1306.55	49.65
	0.005	1483.07	415.70
	0.01	1261.64	107.11
	0.05	1118.31	221.58
	0.1	1147.53	11.56
	0.25	999.99	29.73
	0.5	710.45	23.63
	1	517.84	20.03
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	161.43	16.9
	0.001	147.83	13.21
	0.005	139.90	2.42
	0.01	143.00	4.26
	0.05	143.68	13.97
	0.1	156.01	51.08
	0.25	131.30	4.69
	0.5	129.67	15.08
	1	111.51	8.79

Table 5. Magnesium contribution from magnesium nitrate on the separation of ^{239}Pu

Ionic Species: Magnesium	^{239}Pu in Acid System: Nitric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	3166.39	184.02
	0.001	2803.96	292.63
	0.005	2694.30	441.80
	0.01	2770.31	429.34
	0.05	2960.21	119.92
	0.1	3424.27	89.14
	0.25	5077.90	499.68
	0.5	8448.10	1023.26
	1	26664.48	10120.27
TRU	Concentration [M]	k' value	Standard Deviation
	0	2253.42	661.70
	0.001	3056.88	248.15
	0.005	3257.92	526.94
	0.01	2966.82	293.31
	0.05	3408.87	426.26
	0.1	3623.75	250.34
	0.25	4329.69	461.15
	0.5	5155.08	817.82
	1	9428.25	1944.31
TEVA	Concentration [M]	k' value	Standard Deviation
	0	1508.80	40.88
	0.001	1506.59	102.02
	0.005	1464.62	71.93
	0.01	1481.88	108.52
	0.05	1526.94	42.92
	0.1	1541.78	101.46
	0.25	1482.60	33.90
	0.5	1461.35	117.12
	1	1528.74	70.29
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	149.12	6.80
	0.001	145.22	8.59
	0.005	146.71	9.46
	0.01	144.33	3.16
	0.05	135.01	8.55
	0.1	136.00	4.15
	0.25	160.17	1.42
	0.5	199.46	9.96
	1	209.27	18.62

Table 6. Calcium contribution from calcium nitrate on the separation of ^{241}Am

Ionic Species: Calcium	^{241}Am in Acid System: Nitric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	6728.86	434.85
	0.001	4550.66	170.46
	0.005	795.96	18.04
	0.01	859.02	144.09
	0.05	275.78	4.50
	0.1	25.67	0.62
	0.25	13.37	0.25
	0.5	9.32	0.44
	1	6.75	0.45
TRU	Concentration [M]	k' value	Standard Deviation
	0	145.61	1.59
	0.001	144.08	1.71
	0.005	145.16	3.68
	0.01	142.16	6.96
	0.05	141.76	4.36
	0.1	145.55	2.45
	0.25	144.51	2.89
	0.5	139.49	3.52
	1	128.51	1.69
TEVA	Concentration [M]	k’ value	Standard Deviation
	0	-0.35	0.39
	0.001	-0.53	0.63
	0.005	-0.65	0.40
	0.01	-0.80	0.34
	0.05	-0.72	0.93
	0.1	-0.67	0.09
	0.25	-1.28	0.87
	0.5	-1.12	0.77
	1	-1.05	0.71
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.46	0.67
	0.001	-0.80	0.49
	0.005	-0.32	1.34
	0.01	-0.52	0.71
	0.05	-0.45	1.16
	0.1	-0.85	0.42
	0.25	-0.53	0.76
	0.5	-0.39	0.65
	1	-0.44	0.96

Table 7. Potassium contribution from potassium nitrate on the separation of ^{241}Am

Ionic Species: Potassium	^{241}Am in Acid System: Nitric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	16762.67	17276.65
	0.001	8745.56	729.46
	0.005	8157.38	493.09
	0.01	7937.60	931.50
	0.05	6027.47	316.44
	0.1	4998.07	220.59
	0.25	2867.38	148.18
	0.5	1759.14	27.10
	1	954.84	21.08
TRU	Concentration [M]	k' value	Standard Deviation
	0	144.08	3.80
	0.001	147.01	3.01
	0.005	147.24	2.76
	0.01	147.75	3.77
	0.05	147.61	4.35
	0.1	165.28	25.50
	0.25	141.42	4.71
	0.5	146.74	20.65
	1	129.41	7.23
TEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.80	0.35
	0.001	-0.83	0.36
	0.005	-0.97	0.23
	0.01	-0.47	0.42
	0.05	-0.54	0.16
	0.1	-0.86	0.27
	0.25	-1.04	0.48
	0.5	-0.55	0.20
	1	-0.84	0.43
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.99	1.06
	0.001	-1.32	0.62
	0.005	-1.13	0.49
	0.01	-1.61	0.82
	0.05	-1.30	0.66
	0.1	-0.58	0.23
	0.25	-1.16	0.28
	0.5	-0.60	0.39
	1	0.02	1.74

Table 8. Magnesium contribution from Magnesium nitrate on the separation of ^{241}Am

Ionic Species: Magnesium	^{241}Am in Acid System: Nitric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	7007.44	739.01
	0.001	8989.21	1909.20
	0.005	7052.86	785.69
	0.01	7713.38	2170.52
	0.05	8159.21	36.13
	0.1	9377.24	1383.00
	0.25	16781.54	6678.07
	0.5	16933.07	6898.67
	1	66250.44	45914.11
TRU	Concentration [M]	k' value	Standard Deviation
	0	65.55	1.88
	0.001	66.48	3.38
	0.005	61.57	1.43
	0.01	67.37	8.10
	0.05	65.11	1.22
	0.1	64.30	1.70
	0.25	80.88	11.11
	0.5	65.65	43.87
	1	92.47	10.46
TEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.10	0.78
	0.001	-0.26	0.74
	0.005	-0.26	0.25
	0.01	-0.55	0.56
	0.05	-0.27	0.38
	0.1	0.61	1.53
	0.25	-1.01	0.52
	0.5	-0.70	0.17
	1	-0.63	0.07
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.19	1.07
	0.001	0.33	0.74
	0.005	0.73	1.42
	0.01	0.99	0.19
	0.05	0.62	0.97
	0.1	1.19	0.24
	0.25	0.53	1.01
	0.5	1.04	0.39
	1	0.47	1.81

Table 9. Calcium contribution from calcium chloride on the separation of ^{239}Pu

Ionic Species: Calcium	^{239}Pu in Acid System: Hydrochloric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	1.71	0.87
	0.001	2.10	0.39
	0.005	2.10	0.32
	0.01	2.06	0.83
	0.05	2.52	0.75
	0.1	3.51	0.46
	0.25	10.42	0.78
	0.5	13.98	0.59
	1	15.25	1.45
TRU	Concentration [M]	k' value	Standard Deviation
	0	6.33	0.36
	0.001	6.48	0.32
	0.005	6.41	0.06
	0.01	7.82	0.60
	0.05	6.47	1.04
	0.1	10.97	2.16
	0.25	12.71	2.85
	0.5	18.00	0.39
	1	43.39	2.34
TEVA	Concentration [M]	k' value	Standard Deviation
	0	0.75	0.19
	0.001	0.73	0.17
	0.005	0.94	0.23
	0.01	1.17	0.14
	0.05	1.40	0.45
	0.1	1.97	0.24
	0.25	3.11	0.34
	0.5	6.33	0.73
	1	16.20	1.62
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	-3.42	5.65
	0.001	-0.12	0.23
	0.005	-0.71	0.39
	0.01	-0.37	0.13
	0.05	-0.47	0.30
	0.1	-0.95	0.41
	0.25	-0.69	0.69
	0.5	-0.89	0.45
	1	-0.90	0.10

Table 10. Potassium contribution from potassium chloride on the separation of ^{239}Pu

Ionic Species: Potassium	^{239}Pu in Acid System: Hydrochloric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	2.13	0.26
	0.001	2.03	0.16
	0.005	1.95	0.20
	0.01	1.62	0.31
	0.05	2.82	0.28
	0.1	2.17	0.29
	0.25	3.83	0.26
	0.5	3.98	0.43
	1	6.02	0.80
TRU	Concentration [M]	k' value	Standard Deviation
	0	7.44	0.45
	0.001	8.17	0.73
	0.005	8.28	0.31
	0.01	8.34	1.06
	0.05	9.14	1.78
	0.1	10.34	1.89
	0.25	11.42	0.86
	0.5	11.59	0.49
	1	12.26	0.69
TEVA	Concentration [M]	k' value	Standard Deviation
	0	1.66	0.25
	0.001	1.42	1.43
	0.005	1.63	0.94
	0.01	1.51	0.15
	0.05	1.42	0.15
	0.1	1.51	0.70
	0.25	1.76	0.50
	0.5	3.22	1.14
	1	4.64	1.56
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.27	0.11
	0.001	-0.54	0.12
	0.005	-0.35	0.18
	0.01	-0.53	0.06
	0.05	-0.66	0.10
	0.1	-0.66	0.36
	0.25	-0.91	0.66
	0.5	-1.20	0.45
	1	-0.56	0.73

Table 11. Magnesium contribution from magnesium chloride on the separation of ^{239}Pu

Ionic Species: Magnesium	^{239}Pu in Acid System: Hydrochloric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	1.20	0.24
	0.001	1.34	0.20
	0.005	1.35	0.37
	0.01	1.66	0.10
	0.05	1.68	0.33
	0.1	2.04	0.25
	0.25	3.26	0.22
	0.5	6.71	0.67
	1	24.59	0.79
TRU	Concentration [M]	k' value	Standard Deviation
	0	6.29	1.06
	0.001	7.19	0.41
	0.005	7.26	0.66
	0.01	7.76	0.33
	0.05	8.32	0.87
	0.1	8.99	0.58
	0.25	11.70	0.35
	0.5	14.52	0.90
	1	32.45	2.72
TEVA	Concentration [M]	k' value	Standard Deviation
	0	0.93	0.53
	0.001	0.84	0.20
	0.005	0.80	0.11
	0.01	1.46	0.42
	0.05	1.55	0.15
	0.1	1.59	0.48
	0.25	2.67	1.01
	0.5	5.07	0.81
	1	11.84	2.18
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.86	0.23
	0.001	-0.83	0.53
	0.005	-0.53	0.28
	0.01	-0.30	0.03
	0.05	-1.24	0.30
	0.1	-1.04	0.63
	0.25	-0.62	0.36
	0.5	-0.97	0.19
	1	-1.17	0.47

Table 12. Calcium contribution from calcium chloride on the separation of ^{241}Am

Ionic Species: Calcium	^{241}Am in Acid System: Hydrochloric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	0.82	0.10
	0.001	0.71	0.28
	0.005	0.78	0.17
	0.01	0.71	0.27
	0.05	0.61	0.14
	0.1	0.65	0.22
	0.25	0.74	0.27
	0.5	1.62	0.13
	1	3.06	0.55
TRU	Concentration [M]	k' value	Standard Deviation
	0	-0.51	0.18
	0.001	-0.26	0.91
	0.005	-0.11	0.43
	0.01	0.28	0.22
	0.05	0.30	0.31
	0.1	0.62	0.49
	0.25	1.18	0.17
	0.5	1.44	0.19
	1	1.50	0.84
TEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.04	0.09
	0.001	-0.07	0.16
	0.005	-0.65	1.57
	0.01	-0.02	0.25
	0.05	-0.37	0.36
	0.1	-0.20	0.31
	0.25	-0.05	0.23
	0.5	-0.07	0.17
	1	-0.73	0.88
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	1.30	0.42
	0.001	1.30	0.26
	0.005	1.15	0.29
	0.01	1.18	0.23
	0.05	1.24	0.32
	0.1	0.91	0.29
	0.25	0.94	0.42
	0.5	0.74	0.23
	1	0.63	0.17

Table 13. Potassium contribution from potassium chloride on the separation of ^{241}Am

Ionic Species: Potassium	^{241}Am in Acid System: Hydrochloric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	0.87	0.44
	0.001	0.50	0.28
	0.005	0.58	0.17
	0.01	0.66	0.20
	0.05	0.80	0.34
	0.1	0.82	0.56
	0.25	0.73	0.12
	0.5	1.33	0.27
	1	3.29	0.46
TRU	Concentration [M]	k' value	Standard Deviation
	0	-0.13	0.12
	0.001	-0.67	0.62
	0.005	-0.17	0.63
	0.01	-0.50	0.87
	0.05	-0.41	0.13
	0.1	-0.21	0.38
	0.25	-0.02	0.22
	0.5	-0.16	0.62
	1	-0.27	0.89
TEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.29	0.16
	0.001	-0.70	0.24
	0.005	-0.35	0.35
	0.01	-0.48	0.10
	0.05	-0.61	0.43
	0.1	-0.88	0.35
	0.25	-0.45	0.06
	0.5	-0.92	0.06
	1	-0.86	0.38
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	0.63	0.33
	0.001	0.69	0.18
	0.005	0.62	0.33
	0.01	0.84	0.13
	0.05	0.64	0.28
	0.1	0.60	0.22
	0.25	0.71	0.40
	0.5	0.72	0.04
	1	0.64	0.21

Table 14. Magnesium contribution from magnesium chloride on the separation of ^{241}Am

Ionic Species: Magnesium	^{241}Am in Acid System: Hydrochloric Acid		
DGA	Concentration [M]	k' value	Standard Deviation
	0	0.53	0.03
	0.001	1.12	1.05
	0.005	1.28	0.81
	0.01	1.77	0.90
	0.05	2.70	0.83
	0.1	2.66	0.47
	0.25	2.81	0.34
	0.5	5.43	0.58
	1	15.17	0.93
TRU	Concentration [M]	k' value	Standard Deviation
	0	0.04	0.18
	0.001	-0.04	0.11
	0.005	0.04	0.18
	0.01	-0.04	0.11
	0.05	-0.07	0.18
	0.1	-0.38	0.76
	0.25	0.00	0.28
	0.5	0.03	0.41
	1	0.52	0.09
TEVA	Concentration [M]	k' value	Standard Deviation
	0	0.19	0.35
	0.001	0.17	0.36
	0.005	0.13	0.33
	0.01	0.12	0.04
	0.05	0.10	0.45
	0.1	0.10	0.24
	0.25	0.12	0.28
	0.5	0.11	0.44
	1	0.16	0.19
UTEVA	Concentration [M]	k' value	Standard Deviation
	0	-0.11	0.31
	0.001	-0.50	0.57
	0.005	-0.73	0.21
	0.01	-0.16	0.47
	0.05	-0.04	0.53
	0.1	-0.03	0.48
	0.25	-0.06	0.32
	0.5	-0.10	0.30
	1	-0.53	0.43

CURRICULUM VITAE

Nhung Nguyen
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EDUCATION:

2017-Present ***M.S. Health Physics (In Progress)*** - Colorado State University
2015-2016 ***M.S. Toxicology*** - Colorado State University
2010-2014 ***B.S. Biology Major & Sociology Minor*** - University of Colorado Denver

WORK EXPERIENCES:

2017-Present **Graduate Research Assistant –** Colorado State University (*Fort Collins, Colorado)*
- ❖ Perform radioanalytical determination of actinide for Health Physics and Radiochemistry

2017-2018 **Post-Master's Internship** – Los Alamos National Laboratory (*Los Alamos, New Mexico*)
- ❖ Calculated dose and shielding using MCNP programing as a health physics intern
- ❖ Peered review Radiological Engineering Design Analysis for various projects

2014-2015 **Dental Office Manager** – Dr. Michael Milausnic, D.D.S. (*Lakewood, Colorado*)
- ❖ Maintained and managed patient records in compliance with privacy and security regulations
- ❖ Created teeth models for patients and sterilized dental surgical equipment and tools
- ❖ Responsible for over $5000 in transactions per day

2013-2015 **Program Assistant** - Colorado Area Health Education Center (*CU Anschutz Medical Campus*)
- ❖ Editor-in-chief and graphic layouts designer for the newsletter *Health Matters*
- ❖ Coordinated job shadowing for 65 undergraduate pre-health students
- ❖ Performed quality statistical analysis of data and responsible for associated reports
- ❖ Coordinated training for over 500 MD, PA, and nursing students in medical procedures for free health screening at the National Western Stock Show
- ❖ Developed anatomy-in-clay lectures for undergraduates specifically the digestive tract

2012-2013 **Student Assistant II** - Pre-Collegiate Health Career (*CU Anschutz Medical Campus*)
- ❖ Coordinated standardized test workshops and mentor for minority H.S. students to prepare them for college

AWARDS & SCHOLARSHIPS:

2019 Roy G. Post Foundation Graduate Level Scholarship through Waste Management
2018 National Registry of Radiation Protection Technologists Student Scholarship
2018 Health Physics Society Travel Grant
2018 Fukushima Ambassador Program Scholarship
2018 Selected for Colorado State University Scholarship Video (1/3 students)
2017 Colorado State University Radiation Health Scholarship
2017 U.S. Nuclear Regulatory Commission Fellowship
2015 The Colorado Health Symposium University Scholarship (*1/7 awards in Colorado*)
2014 Colorado Collegiate Health Professions Development (*Co-HPD Scholar – 1/35 awardees*)
- ❖ Focused on understanding current issues and events that are shaping the field of healthcare today

2012 Colorado Area Health Education Centers Program (*CREATE Health Scholar – 1/30 awardees*)
- ❖ Focused on hands-on training and exposure to a variety of health careers

PUBLICATION AND RESEARCH:

2019 Oral Presentation at the Colorado Rocky Mountain Chapter Health Physics Society Meeting on *The Determination of Actinides in Human Bones and the Impact of Matrix Constituents*

2019 Poster Presentation at the Waste Management Symposia on *Actinide Determination in Bone Sample - Effect of Matrix Constituents*

2019 Poster Presentation at the 20th Annual CVMBS Research Day on *Effect of Matrix Constituents on the Separation of Plutonium and Americium from Bone Samples*

2018 Oral presentation at the ERHS student seminar on *The Fukushima Ambassador Program*

2018 Oral presentation at the annual Health Physics Society conference on *The Determination of Actinides in Human Bones and the Impact of Matrix Constituents*

2018 Poster presentation at the Methods and Applications of Radioanalytical Chemistry conference on *Actinide Determination in Bone Sample - Effect of Matrix Constituents*

2018 Oral presentation at the American Chemical Society conference on *Effect of Matrix Constituents on the separation of Plutonium and Americium from Bone Samples*

2018 Oral presentation at the Mid-Year Health Physics Society conference on *Effect of Matrix Constituents on the separation of Plutonium and Americium from Bone Samples*

2016 Poster presentation at the Radiation Research Society on *The potential use of Electron Paramagnetic Resonance (EPR) on wild boar teeth to measure radiation doses in Fukushima prefecture.*

2014 Nguyen, N. *Paralysis, The Brain, Nervous System, and Its Diseases: An Encyclopedia of Neuroscience and Neurology.*pp. 800-805. Santa Barbara, California: Greenwood, 2014.

COMMUNITY SERVICE & EXTRACURRICULAR ACTIVITIES:

2018-Present **Vice President –** Student body for the Health Physics Master's Program

- ❖ Coordinated social events for the department of Environmental and Radiological Health Sciences
- ❖ Coordinated tours and housing for prospective students

2018 **CSU Mentor** - Volunteered with the Certified Health Physics online review class

- ❖ Monitored students' progress and their primary point of contact.

2017 **Sponsor** - Volunteered with CSU student exchange ambassador program, which focused on radiation research.

2015 **Camp Counselor** - Volunteered with the Muscular Dystrophy Association (*Denver, Colorado*)

- ❖ Worked one-on-one with campers, providing around-the-clock care, close supervision for children with neuromuscular disease through physical and emotional support

2007-2015 **Youth Group Leader -** Vietnamese Eucharistic Youth Leadership (*Wheat Ridge, Colorado*)

- ❖ Provided catechism and cultural education for 6-18 year old students
- ❖ Tutored general math and science for middle and high school students for 4 hours every Saturday
- ❖ Organized community service events, caroling at nursing homes, and canned food drives

2015 Volunteered with Dentistry from the Heart (*Littleton, Colorado*)

- ❖ Assisted Dentists to provide free dental care

2012-2014 Member of Tri-Beta Biology Honors Society (*CU Denver Downtown Campus*)

QUALIFICATIONS:

2018 Completion of Radiation Safety Training - Colorado State University

2017 Completion of Monte Carlo N-Particle Training - Los Alamos National Laboratory