

Mixing Commercially Available Extraction Resins for Minor Actinide and Lanthanide Separation

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Abstract

Two extractants (Tetraoctyldiglycolamide, TODGA, and 2-ethylhexylphosphonic acid mono-2-ethylhexyl ester, HEH[EHP]) used in Actinide Lanthanide SEParation (ALSEP) are analogous to Eichrom® Technologies' commercially available resins DGA-n and LN2, respectively. This work aimed to understand the translatability of a solvent extraction system to a multi-modal resin chromatography system. The results demonstrated the benefits and limitations of the ALSEP process via column chromatography when conducted on the radioanalytical scale. The column utilizing ALSEP extractants yielded over 80% of the Am with less than 3% Ce or Nd overlap, demonstrating a possible technique for rapid separation of minor actinides from mixed activation product/ mixed fission product samples.

Introduction

In recent years, there has been emphasis on research and development for cleaning up and/or reprocessing used nuclear fuels.¹ PUREX (Plutonium Uranium Reduction EXtraction) has been used for decades to purify plutonium and uranium through manipulation of oxidation states with subsequent solvent extraction. Through years of research many different separation schemes meant to follow a PUREX separation have been developed, e.g., Advanced TALSPEAK (Trivalent Actinide Lanthanide Separation with Phosphorus-Reagent Extraction from Aqueous Komplexes), GANEX (Group ActiNide EXtraction), TRUEX (TRansUranium EXtraction), TALSPEAK-MME (TALSPEAK – Mixed Monofunctional Extractants), ALSEP (Actinide Lanthanide SEParation), amongst others. Most of these processes have focused on reducing the number of steps in existing separation schemes to reduce the overall cost of reprocessing. One way this has been achieved is by combining two separation schemes.^{1–5} One of these schemes, ALSEP, utilizes two organic phase extractants, an acidic cation-exchanging extractant, HEH[EHP] which also acts as a phase modifier, and a neutral diglycolamide, such as TODGA, shown in Fig. 1, combining ARTIST and TALSPEAK separations.⁶ HEH[EHP] is known to complex with trivalent elements at lower acid concentrations, while TODGA is known to complex at higher acid concentrations. These two extractants work together in tandem at different acid and background electrolyte concentrations to separate the lanthanides (Ln), La through Er plus Y, from the minor actinides (MAs), Am and Cm, when employing different acid concentrations. They also effectively separate the Ln from fission and activation products.²

Systems such as PUREX and ALSEP are intended for separations of bulk-scale metals using solvent extraction as the primary mode of separation. For separations on the industrial scale, sometimes thousands of kg of feed, solvent extraction is more cost-effective than resin-based separations as the solvent can be regenerated and reused.² In nuclear forensics, typical sample sizes are small compared to those used in nuclear fuel reprocessing. This small sample scale is because nuclear forensics is concerned with understanding a sample containing radioactive material, not recovering a product. Typically, in a soil sample, the total analyzed matrix may be on the order of a few grams. Without a matrix (i.e., a neat solution of fissions), samples can easily be smaller than 0.1 grams.^{7–9} For such a small

sample, a column separation can provide a quicker, easier, more cost-effective separation with no organic solvent waste.

Such a separation scheme, with multiple resins, can be performed sequentially utilizing a stack of different resins.^{10–16} This scheme is beneficial when analyzing complex mixtures of isotopes that cannot be differentiated by analytical measurement methods (e.g., overlapping peaks in alpha or gamma spectroscopy or isobars in mass spectrometry) because it allows the separation of one sample into multiple analytes. Separation schemes are also beneficial when sample limited. In this work, the resins were added into the same column to form a mixed-modal resin system. The extractant molecules within resin are loosely bound thus mixing two resins may allow for different metal-extractant interactions, changing the separation and allowing for multiple analytes to be separated from a single column. This work looked at separating Am from an irradiated uranium target using ALSEP-inspired extractants bound on commercially available Eichrom resins. The aim of this work is to form a system similar to ALSEP but differ fundamentally from solvent extraction because extractant interaction is limited in their degrees of freedom due to being bound to a solid phase. However, we aim to exploit the fundamental extraction capability of each extractant at conditions that are beneficial for our goals. This separation was meant to separate Am from both mixed activation products (MAP) and mixed fission products (MFP).

Experimental

3.1 Materials: Resins purchased from Eichrom Technologies (Lisle, IL), LN2 and DGA-n, were both 50–100 µm particle diameter bulk resin and used as received. Fisher (Waltham, MA) Optima grade nitric acid was used as received. Diethylenetriamine pentaacetic acid (DTPA) and ammonium citrate were purchased from Acros Organics (Geel, Belgium) as 98+% and ACS reagent grade, respectively and used without further purification.

Metal tracer solutions were prepared based upon the ability to be analyzed via Inductively Coupled Plasma – Optical Emission Spectroscopy (ICP-OES). Two custom mixes of elemental tracers, custom cocktail 1 (CC-1) and custom cocktail 2 (CC-2), and single element standards Ce, Eu, Nd, Pr, Sm, and Tb were obtained from Inorganic Ventures (Christiansburg, VA). CC-1 contained 1000 µg/g Ce, Ga, Na, Nd, Pb, Pr, Sm, Tb, and Tl. CC-2 contained 1000 µg/g Ag, Ba, Cd, Co, Cr, Cu, Eu, Fe, La, Lu, Ni, Y, and Zn. The elements in the custom cocktails were specifically chosen to ensure no spectral interferences between individual elements when analyzed via ICP-OES.

15 mM DTPA, 200 mM ammonium citrate solutions were prepared as follows: DTPA solutions were prepared by fully deprotonating the ligand with small additions of 100 mM NH₄(OH) until complete dissolution (slow process). The working solutions were corrected to the desired pH using 100 mM HNO₃ and 100 mM NH₄(OH). In the presence of ammonium citrate, the amount of NH₄(OH) needed to be added and the time for full dissolution of DTPA were smaller. For the entire preparation, pHs were monitored using a Beckman Coulter Elect GP/Epoxy 6X150 mm pH electrode attached to a Thermo Scientific Orion Star 2115203 Dual Star pH / ISE benchtop meter.

A 200 mg 93% Highly Enriched Uranium (HEU) metal foil was irradiated at the Washington State University Dodgen Research facility for two hours. This foil was dissolved in 8 M HNO₃ and brought up to a final volume of 12 mL and concentration of 3 M HNO₃. When used in experiments, 0.1 mL were aliquoted into samples and weights were taken gravimetrically for yielding purposes.

3.2 Methods: All resins, typically 0.5 grams of LN2 and 0.05 grams of DGA-n, were dispensed gravimetrically and conditioned in 3 M HNO₃ overnight while being shaken at 300 rpm on a shaker table. When both LN2 and DGA-n resins were used in the same column they were weighed out into the same vessel and conditioned together. The 1:10 LN2:DGA-n ratio, although not the typical ratio of HEH[EHP]:TODGA in ALSEP, was used to simulate ALSEP concentrations that have been used with centrifugal contactors.^{2, 17-23} One hundred micrograms of each metal tracer were loaded onto the gravity columns in 3 M HNO₃ and successively rinsed off in different acid concentrations of 0.015 M DTPA/0.2 M ammonium citrate. The sequential separation is shown in Table 1. Collected fractions were weighed and diluted in 2% HNO₃ for analysis via ICP-OES on a Thermo iCAP7600 Duo or a Thermo iCAP PRO.

Table 1
Final separation used

Eluent	Total Fraction Size/ Increment of Collection
3 M HNO ₃	1 mL Load, 5 mL rinse/ Total of 6 mL combined collection
0.015 M DTPA/0.2 M ammonium citrate – pH 2	10 mL/Five 2 mL collections
0.015 M DTPA/0.2 M ammonium citrate – pH 4	10 mL/ Five 2 mL collections

When ²⁴¹Am was added it was added as a 5 µL spike from a 0.99 µCi/g tracer and analyzed via Mirion HPGe systems utilizing Canberra Genie 2000 V3.4.1 and APEX software, or by NaI detector with a PerkinElmer Wizard2 system. HPGe detectors were calibrated utilizing NIST traceable multi-element standards from Eckert and Ziegler. Unless otherwise stated, all elements reported were analyzed via ICP-OES on a Thermo iCAP7600 Duo or a Thermo iCAP PRO. The instrument was calibrated prior to each set of samples and calibration verification check samples were run after calibration and each set of 10 samples to confirm calibration and monitor instrument drift. A list of experiments is given in Table 2.

Table 2
Experimental conditions studied in this work

Experiment Number	Conditions of Experiment
1-1 DGA-n only 1-2 LN2 only	100 µg of the metals in CC-2 tracer with 0.05 grams DGA-n, elution profile used given in Table 1 except that the load and rinse were 1 mL each and collected separately, and the two rinse solutions were collected as one sample each of 10 mL, for a total of 4 fractions.
2 DGA-n and LN2 in series	100 µg of the metals in CC-2 tracer with 0.05 grams DGA-n eluting into another column of 0.5 grams LN2, elution profile used given in Table 1 except that the load and rinse were 1 mL each and collected separately, and the two rinse solutions were collected as 10 mL each, for a total of 4 fractions.
3 DGA-n and LN2 mixed - non-rad	100 µg of the metals in CC-2 tracer with 0.5 grams LN2/0.05 grams DGA-n, elution profile used given in Table 1 except that the load and rinse were 1 mL each and collected separately, and that the two rinse solutions were collected as one sample each of 10 mL, for a total of 4 fractions.
4 DGA-n and LN2 mixed - HEU	100 µg of the metals in CC-2 tracer, a spike of Am ²⁴¹ , and 100 mg of an aged solution of 3 M HNO ₃ containing irradiated HEU with 0.5 grams LN2/0.05 grams DGA-n, elution profile used given in Table 1.
5 DGA-n and LN2 mixed – Ln's	100 µg of Ce, Eu, Nd, Pr, Sm, and Tb in solution with 0.5 grams LN2/0.05 grams DGA-n, elution profile used given in Table 1.
6 DGA-n and LN2 mixed – non-rad, no NH ₄ -citrate	100 µg of the metals in CC-1 tracer with 0.5 grams LN2/0.05 grams DGA-n, elution profile used given in Table 1 with the adjustment of no ammonium citrate being added.
7 DGA-n and LN2 mixed – no NH ₄ -citrate + ²⁴¹ Am	The same as Experiment 6, but with an ²⁴¹ Am spike added.

Results and Discussion

The first three experiments were conducted to determine the viability of mixing the two different resins. The first experiment tested the elution profile with DGA or LN2 resin alone. Experiments two and three used both resins. Experiment 2 utilized the DGA resin followed by the LN2 resin in series. Experiment 3 utilized the resins mixed together in a single column. The results for Experiments 1, 2, and 3 are shown in Table 3, Table 4, Table 5, and Table 6.

Table 3

Results from Experiment 1–1. Load and rinse were 3 M HNO₃. This column contained only 0.05 grams of DGA-n resin. The pH 2 and pH 4 solutions were both 0.015 M DTPA and 0.2 M ammonium citrate. Uncertainty is reported as $\pm 1\sigma$ of the compounded error from the ICP-OES of the experiment run in triplicate. Boxes with a dash mean the element was not detected in that fraction.

Ag	2 mL Load + Rinse	10 mL pH 2	10 mL pH 4	Total
	100.3% $\pm$ 0.3%	-	-	100.3% $\pm$ 0.3%
Cd	101.7% $\pm$ 0.2%	0.60% $\pm$ 0.02%	-	102.3% $\pm$ 0.2%
Co	101.5% $\pm$ 0.3%	-	-	101.5% $\pm$ 0.3%
Cr	101.5% $\pm$ 0.2%	-	-	101.5% $\pm$ 0.2%
Cu	101.1% $\pm$ 0.3%	-	-	101.1% $\pm$ 0.3%
Eu	9.8% $\pm$ 0.1%	8.6% $\pm$ 0.1%	77.6% $\pm$ 0.5%	96.0% $\pm$ 0.6%
La	16.1% $\pm$ 0.1%	25.9% $\pm$ 0.1%	55.9% $\pm$ 0.2%	97.9% $\pm$ 0.4%
Lu	6.5% $\pm$ 0.1%	1.60% $\pm$ 0.07%	86.1% $\pm$ 0.4%	94.2% $\pm$ 0.4%
Ni	101.8% $\pm$ 0.2%	-	-	101.8% $\pm$ 0.2%
Y	7.4% $\pm$ 0.1%	2.20% $\pm$ 0.01%	85.4% $\pm$ 0.3%	94.9% $\pm$ 0.3%
Zn	101.6% $\pm$ 0.3%	-	-	101.6% $\pm$ 0.3%

Table 4

Results from Experiment 1–2. Load and rinse were 3 M HNO₃. This column contained only 0.5 grams of LN2-n resin. The pH 2 and pH 4 solutions were both 0.015 M DTPA and 0.2 M ammonium citrate. Uncertainty is reported as $\pm 1\sigma$ of the compounded error from the ICP-OES of the experiment run in triplicate. Boxes with a dash mean the element was not detected in that fraction.

Ag	2 mL Load + Rinse	10 mL pH 2	10 mL pH 4	Total
	103.6% $\pm$ 3.3%	-	-	103.6% $\pm$ 3.3%
Cd	104.1% $\pm$ 3.3%	-	-	104.1% $\pm$ 3.3%
Co	102.7% $\pm$ 3.4%	-	-	102.7% $\pm$ 3.4%
Cr	102.6% $\pm$ 3.5%	-	-	102.6% $\pm$ 3.5%
Cu	102.0% $\pm$ 3.3%	-	-	102.0% $\pm$ 3.3%
Eu	104.2% $\pm$ 3.3%	-	-	104.2% $\pm$ 3.3%
La	102.5% $\pm$ 3.3%	-	-	102.5% $\pm$ 3.3%
Lu	101.4% $\pm$ 3.5%	-	-	101.4% $\pm$ 3.5%
Ni	103.1% $\pm$ 3.5%	-	-	103.1% $\pm$ 3.5%
Y	103.7% $\pm$ 3.3%	-	-	103.7% $\pm$ 3.3%
Zn	103.9% $\pm$ 3.4%	-	-	103.9% $\pm$ 3.4%

Table 5

Results from Experiment 2. This experiment had two columns stacked upon one another, 0.05 grams of DGA-n were followed by 0.5 grams of LN2. The pH 2 and pH 4 solutions were both 0.015 M DTPA and 0.2 M ammonium citrate. Uncertainty is reported as $\pm 1\sigma$ of the compounded error from the ICP-OES of the experiment run in triplicate. Boxes with a dash mean the element was not detected in that fraction.

Samples with < 30% recovery are bolded and italicized.

Ag	2 mL Load + Rinse	10 mL pH 2	10 mL pH 4	Total
	73.7% $\pm$ 0.3%	26.1% $\pm$ 0.1%		99.8% $\pm$ 0.4%
Cd	71.4% $\pm$ 0.2%	30.4% $\pm$ 0.1%	-	101.9% $\pm$ 0.3%
Co	75.7% $\pm$ 0.2%	26.4% $\pm$ 0.1%	-	102.1% $\pm$ 0.3%
Cr	75.6% $\pm$ 0.2%	25.9% $\pm$ 0.1%	-	101.5% $\pm$ 0.2%
Cu	75.1% $\pm$ 0.3%	26.0% $\pm$ 0.1%	-	101.1% $\pm$ 0.3%
Eu	3.71% $\pm$ 0.02%	0.63% $\pm$ 0.01%	4.55% $\pm$ 0.03%	8.89% $\pm$ 0.06%
La	8.05% $\pm$ 0.04%	7.34% $\pm$ 0.03%	5.12% $\pm$ 0.02%	20.51% $\pm$ 0.09%
Lu	-	-	-	-
Ni	76.0% $\pm$ 0.2%	26.0% $\pm$ 0.1%	-	102.1% $\pm$ 0.3%
Y	2.11% $\pm$ 0.01%	0.85% $\pm$ 0.01%	-	2.95% $\pm$ 0.01%
Zn	74.9% $\pm$ 0.2%	26.6% $\pm$ 0.1%	-	101.5% $\pm$ 0.3%

Table 6

Results from Experiment 3. This experiment had a single column with 0.05 grams of DGA-n mixed with 0.5 grams of LN2. The pH 2 and pH 4 solutions were both 0.015 M DTPA and 0.2 M ammonium citrate. Uncertainty is reported as $\pm 1\sigma$ of the compounded error from the ICP-OES of the experiment run in triplicate. Boxes with a dash mean the element was not detected in that fraction. Samples with < 30% recovery are bolded and italicized.

Ag	2 mL Load + Rinse	10 mL pH 2	10 mL pH 4	Total
	73.8% $\pm$ 0.3%	26.7% $\pm$ 0.1%	1.06% $\pm$ 0.01%	101.5% $\pm$ 0.4%
Cd	73.7% $\pm$ 0.2%	28.5% $\pm$ 0.1%	1.19% $\pm$ 0.01%	103.4% $\pm$ 0.3%
Co	75.2% $\pm$ 0.2%	27.4% $\pm$ 0.1%	1.09% $\pm$ 0.01%	103.4% $\pm$ 0.3%
Cr	74.9% $\pm$ 0.2%	27.4% $\pm$ 0.1%	0.95% $\pm$ 0.01%	103.0% $\pm$ 0.2%
Cu	74.1% $\pm$ 0.2%	26.9% $\pm$ 0.1%	1.14% $\pm$ 0.01%	102.2% $\pm$ 0.3%
Eu	-	-	23.1% $\pm$ 0.2%	23.1% $\pm$ 0.2%
La	-	-	-	-
Lu	-	-	-	-
Ni	74.9% $\pm$ 0.2%	27.2% $\pm$ 0.1%	1.07% $\pm$ 0.01%	103.2% $\pm$ 0.3%
Y	-	-	-	-
Zn	74.4% $\pm$ 0.2%	27.4% $\pm$ 0.1%	1.35% $\pm$ 0.01%	103.1% $\pm$ 0.3%

Although experiments 1–3 did not contain any minor actinides (MAs, specifically Am and Cm here), as europium is a tri-valent f-elements similar in size as Am, it was used to predict the behavior of the trivalent minor actinide ions. These results demonstrate that, although a 1 mL rinse was sufficient for the 0.05-gram DGA-n column to strip non-adhering ions, it was not enough for the 0.55-gram columns. Based on these experiments, 5 mL rinses, which corresponded to approximately 2.5 column volumes, were used for the remaining experiments. Even with the increased rinse volumes, Eu was still retained, showing almost full separation from the other elements tested. Around 10% of the Eu came off the DGA-n only column during the load/rinse steps. Close to 5% of the Eu also eluted from the DGA-n/LN2 stack during the load/rinse as well. It is believed this loss occurred due to a combination of the small resin volume of the DGA-n and low retention of Eu on LN2 resin in 3 M HNO₃.²⁴ The low retention of the lanthanide series on LN2 in 3 M HNO₃ is also shown by the experiment with just the LN2 resin, as was expected from previous literature.²⁴

The results for Experiment 4 are shown in Fig. 2. This experiment utilized an aliquot dissolved from an irradiated HEU foil to emulate separations with a more complex distribution of stable and radioactive isotopes. Elements from both ICP-OES and HPGe analysis are shown, representative elements were chosen in place of showing all of them. Elements included in the figure represent transition metals (Co and Ru), alkaline Earth metals (Ba), lanthanides (Eu and Nd), actinides (Am), and metals (Ag). Many of

the lanthanides were not fully recovered, Eu and Nd specifically being shown. These elements, however, could possibly be eluted with a greater volume of the pH 4 0.015 M DTPA/0.2 M ammonium citrate solution. A large fraction of Am, ~ 55%, was collected in the first 2 mL of the pH 4 0.015 M DTPA/0.2 M ammonium citrate solution along with ~ 15% of the Nd. Of the elements shown, only the lanthanides and Am had an affinity for the column under the conditions tested with nearly 100% recovery of other elements including silver, barium, cobalt, and ruthenium in the initial load and rinse. It has been shown by McAlister and Horwitz that, at least in the configuration of HEH[EHP] being the extractant on LN2 resin, does not bind particularly well to trivalent Lns or MAs in high molarity acids.^{1-2, 25-26} TODGA, the extractant in DGA-n resin, readily complexes with the trivalent Lns resulting in > 80% retention initially on the resin.^{1-2, 25-26}

The first four experiments showed that Ln and MAs were the only elements significantly retained on, and able to be eluted off, the column. The 5th experiment was designed to look specifically at the lanthanide elution profile on the mixed resin. These results are shown in Fig. 3 with the results of Am from Experiment 4 included as the elution profile was the same. As in Experiment 4 only 15% of the Nd was found in the fraction that Am was previously eluted in. This time, however, 10% of the Ce was also eluted in the Am fraction. Meanwhile, both smaller (Eu, Sm, Tb) and larger (Pr) trivalents are eluted after, or not at all in the case of Tb, this Am fraction. Both the ionic radii and the relative covalency of the minor actinides compared to the lanthanides facilitates the separation observed.²⁷⁻²⁸ We believe that this system is behaving closer to the individual extractant expectations rather than some mixed behavior. In the elution conditions utilized, HEH[EHP] is the dominant extractant being exploited and the system is acting more like an Advanced TALSPEAK system.^{1-3, 22, 24 - 25}

The final two experiments (#6 and 7) were conducted to determine if a citrate buffering system was necessary for the separation. The results for these two experiments are shown in Fig. 4 and Fig. 5. Forgoing the citrate buffer did not change the elution of the non-trivalent elements. The lanthanides tested, however, noticeably shifted due to their complexation with citrate. More than 60% of the Nd and 40% of the Ce were in the fraction that previously contained the Am. When looking at results for Experiment 7's, there is a significant shift in when the elements eluted, even compared to Experiment 6. This is attributed to full dissolution of the DTPA, see the experimental section.

The separation obtained from Experiment 7, see Fig. 5, yielded 80% of the Am in fractions 4–8 with less than 3% of any of the lanthanides tested in the same experiment. This separation, despite being reported in previous literature multiple times, is still remarkable due to the fact that Am and Nd under these conditions are both trivalent and have effective ionic radii that differ by only 0.8 picometers.^{1, 30}

Conclusions

A process has been developed for the rapid isolation of Am from a mixed activation product, mixed fission product sample using commercially available resins from Eichrom Technologies. It was found that a mixed column of DGA-n/LN2 was more effective at isolating Am³⁺ and the lanthanides than either

DGA-n being used alone, or a stacked approach of DGA-n followed by LN2. It was also found that in this context an ammonium citrate buffer is not needed for the separation. This separation provides a rapid route for the analysis of Am in nuclear forensics.

Declarations

Funding/Conflicts of Interest:

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Author Contribution

Matthew is the primary author of the manuscript and work presented therein. Staci, Evan, Nic, and Hilary assisted with editing and process development. Ean, Bethany, Grant, and Carson assisted with data acquisition and analysis. Chelsie and Brienne provided oversight throughout the entirety of the work.

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Figures

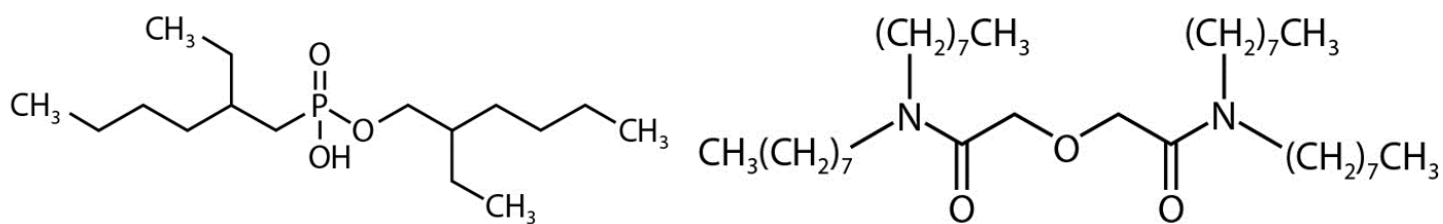


Figure 1

Structures of HEH[EHP], left, and TODGA, right.

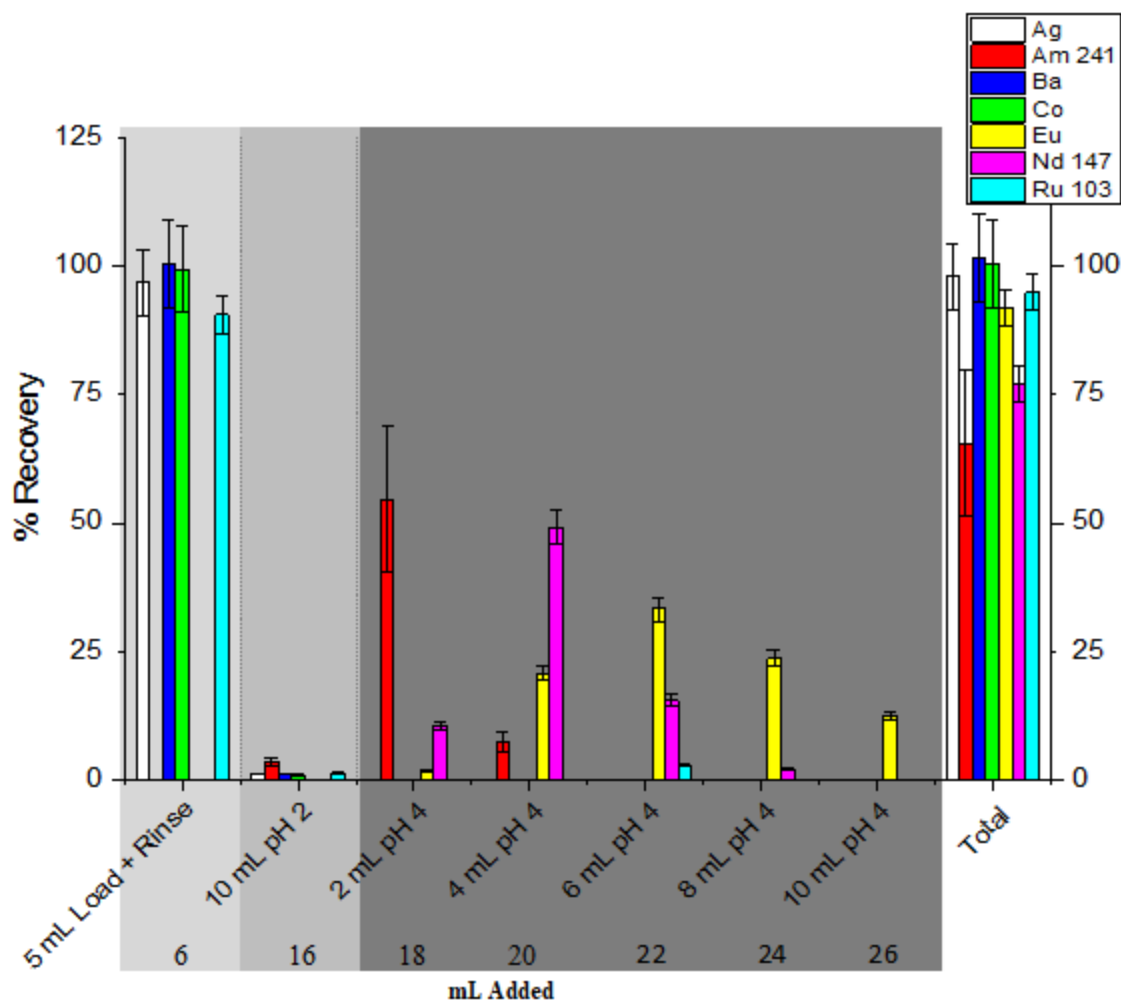


Figure 2

Combined HPGE and ICP-OES results from Experiment 4, sample consisted of 100 μg of the metals in CC2 tracer, a spike of ^{241}Am , and 100 mg of an aged solution of 3 M HNO_3 containing irradiated HEU. Separated with 0.5 grams LN2/0.05 grams DGA-n mixed in a single column. The pH 2 (medium gray block) and pH 4 (dark gray block) solutions were both 0.015 M DTPA and 0.2 M ammonium citrate. Uncertainty is reported as $\pm 1\sigma$ of the compounded error from the instrument utilized for the analysis of

the experiment run in triplicate. Isotopes listed in the legend were analyzed via HPGE whereas elements listed were analyzed via ICP-OES.

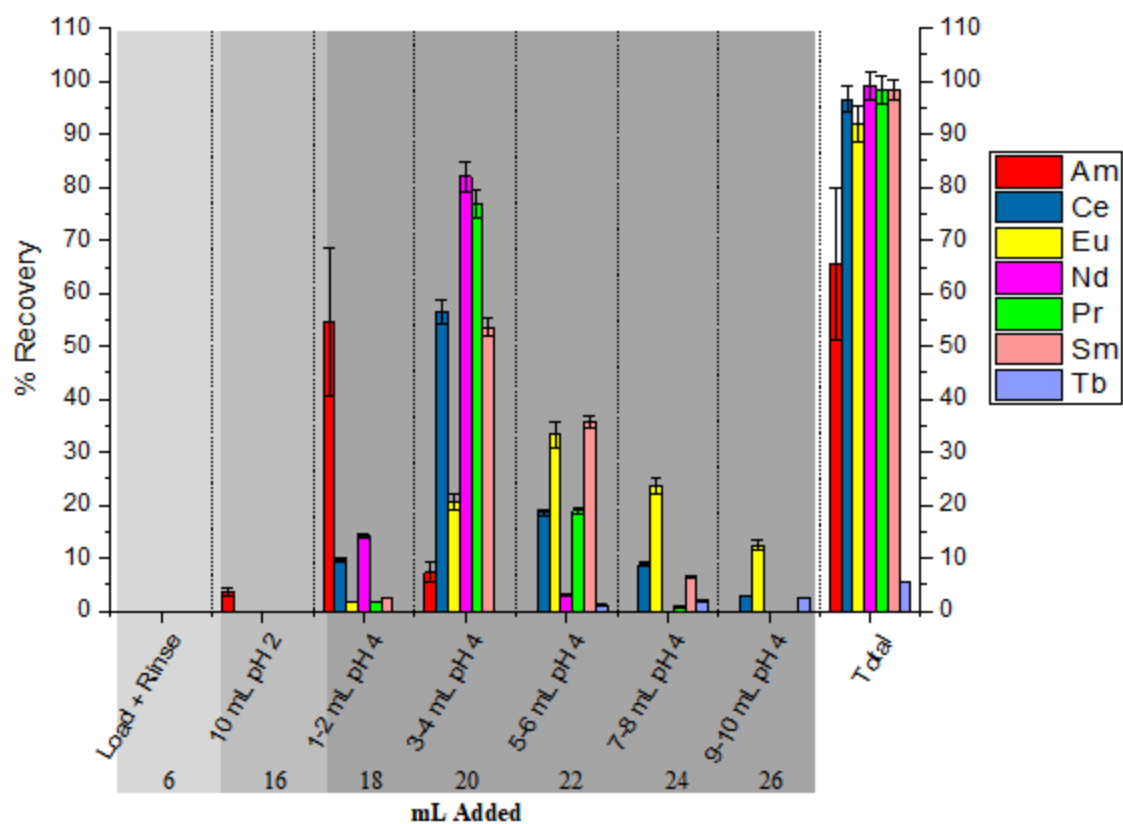


Figure 3

Results from Experiment 5, the elution of select lanthanides on a mixed column of 0.05 grams DGA-n and 0.5 grams of LN2. Am elution is taken from Experiment 4. The pH 2 (medium gray block) and pH 4 (dark gray block) solutions were both 0.015 M DTPA and 0.2 M ammonium citrate. Uncertainty is reported as $\pm 1\sigma$ of the compounded error from the ICP-OES of the experiment run in triplicate.

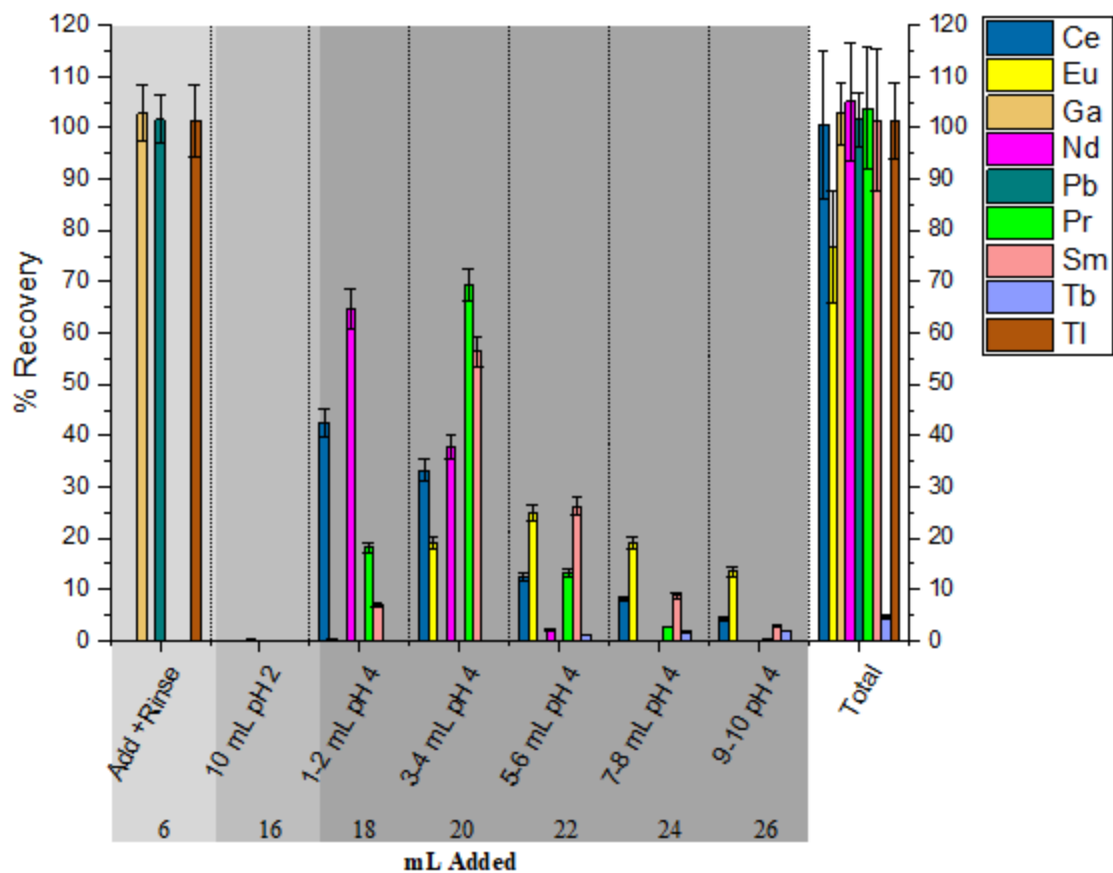


Figure 4

Results from Experiment 6, elution of 100 μg of metals in solution of CC1 from 0.05 grams of DGA-n resin mixed with 0.5 grams LN2 resin. The pH 2 (medium gray block) and pH 4 (dark gray block) solutions are 0.015 M DTPA. Uncertainty is reported as $\pm 1\sigma$ of the compounded error from the ICP-OES of the experiment run in triplicate.

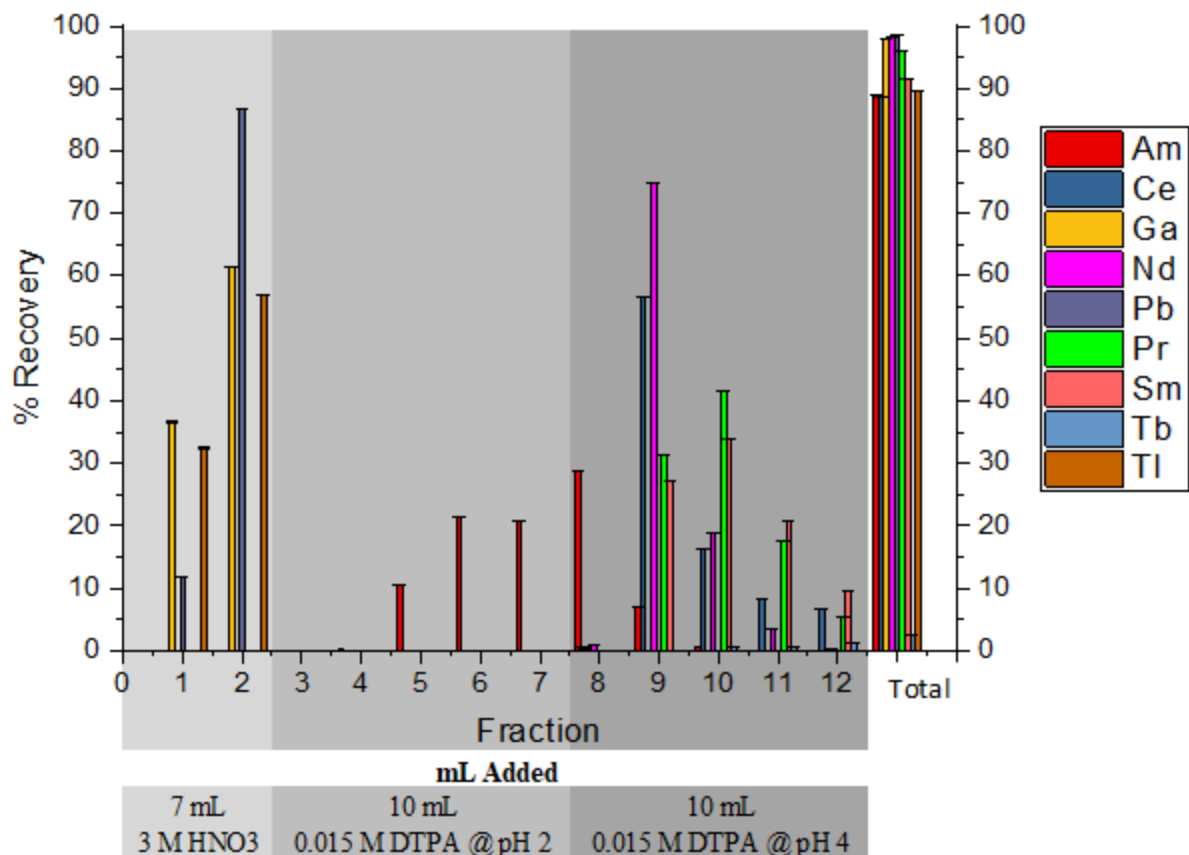


Figure 5

Results from Experiment 7, a mixed element sample on a column of 0.5 grams of LN2 resin mixed with 0.05 grams of DGA-n resin. Sample was loaded and rinsed with 3 M HNO₃ (light gray block), then eluted with 10 mL of 0.015 M DTPA at pH 2 (medium gray block) followed by 10 mL of 0.015 M DTPA at pH 4 (dark gray block). No ammonium citrate was included in this experiment. Uncertainty is reported as $\pm 1\sigma$ of the compounded error from the instrument utilized for the analysis of the experiment run in triplicate. Americium was analyzed via HPGE whereas the rest of the elements listed were analyzed via ICP-OES.