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The Electrochemical Separation of Curium and Americium: Quarterly Report January - March, 2006

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**DOE/TRP Quarterly Report
Funding Years 2006
January 2006 – March 2006**

This is a list of the personnel working with foams on the UNLV campus.

Academic Faculty

David Hatchett, Assistant Professor, Department of Chemistry

Kenneth Czerwinski, Associate Professor, Department of Chemistry

Graduate Students

Sandra Elkouz

M.S. Candidate Chemistry

Sujanie Gamage

Ph.D. Candidate Chemistry

Patrick Houlihan

Undergraduate Chemistry

Title: The Electrochemical Separation of Curium and Americium.

- The electrochemical characterization of Ce and Eu complexed with EDTA, NTA, and Citrate has been completed.
- Synthesis of the polymer substrate and the chelating ligand is underway. Approximately 50 grams of disulfide has been produced to produce the chelating thiol group required for the last set of studies.
- Gold substrates have been prepared to perform the 2-D surface chelation using the chelating disulfide and thiol. The goal is to determine the potentials required for chelation.
- Polymer gold composite systems have been prepared and characterized for use with the chelating ligands.

Technical report:

This research report outlines the current status and progress associated with the electrochemical separation of Curium and Americium. The following pages outline the progress on our project to date. For two and a half years, research has been actively performed on this project, and is currently on schedule for the proposed timelines.

Data collection and analysis of the Ce^{3+}/Ce^{4+} redox couple complexes has been accomplished using EDTA, NTA and Citrate ions. All complex systems were investigated at Pt, Au, and Glassy Carbon working electrodes as a function of pH. Analysis of these data was accomplished by performing appropriate background subtractions to reveal net peaks due to Ce redox behavior. Successful identification of the Ce redox couple was achieved in many cases at pH values greater than two. This is well beyond the typically pH = 2 used to resolve the redox couple. Figure 1 shows the voltammetric response of Ce at pH = 2 and from pH = 2 to 12 with an equimolar concentration between EDTA. The redox couple for EDTA is centered at $E_{1/2} = 1.55V$ vs. Ag/AgCl. In contrast the redox couple shifts to $E_{1/2} = 0.85 V$ vs. Ag/AgCl when complexed with EDTA. This represents a 0.70 V shift to lower energy. In addition the

redox couple is clearly resolved at pH = 10. The Ce redox couple is not observed at pH = 12.

For comparison the redox couple for Eu is presented in Figure 2. The redox couple for Eu is observed at $E_{1/2} = -1.15$ V vs. Ag/AgCl. However, this redox couple is not observed at any pH for Pt electrodes in the absence of EDTA. The redox couple in complex form shifts the potential of the couple, which allows resolution to be achieved.

Figures 3 and 4 display the cyclic voltammetric response of graphite and Pt electrodes in solutions containing Ce and Citrate ion as a function of pH. The graphite electrode shows the best resolution for the uncomplexed and complexed couple at pH = 2. However, the voltammetry of the uncomplexed Ce is observed at $E_{1/2} = 1.45$ V. The complex with citrate is observed at $E_{1/2} = 0.75$ V. The complex shifts to 0.70 V lower potential. However, the process is kinetically limited above pH = 2 showing pseudo reversible behavior. For the Pt electrode the best resolution is observed at pH = 6. The data clearly demonstrates that complexation of the redox species can shift the potential significantly and produce stable voltammetry at pH values typically not accessible in acidic solutions. The complex keeps the Ce from precipitating at high pH values as a hydroxide.

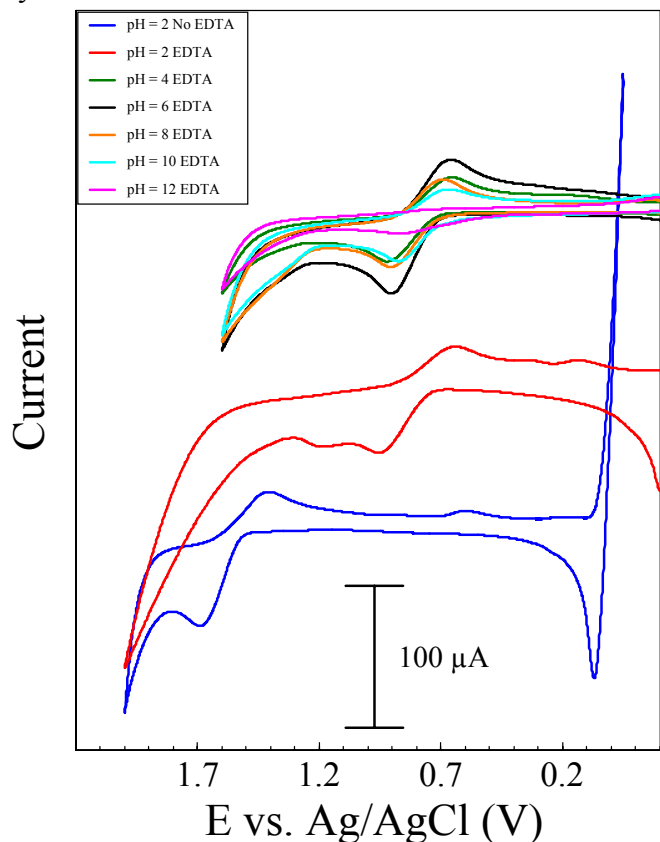


Figure 1. Cyclic voltammetric response of a Pt electrode for Ce uncomplexed and complexed with EDTA as a function of pH.

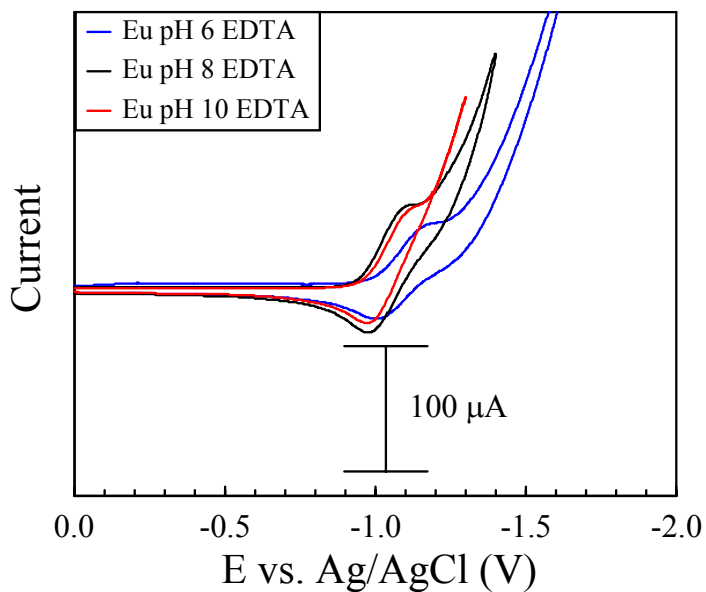


Figure 2. Cyclic voltammetric response of a graphite electrode for Eu complexed with EDTA as a function of pH.

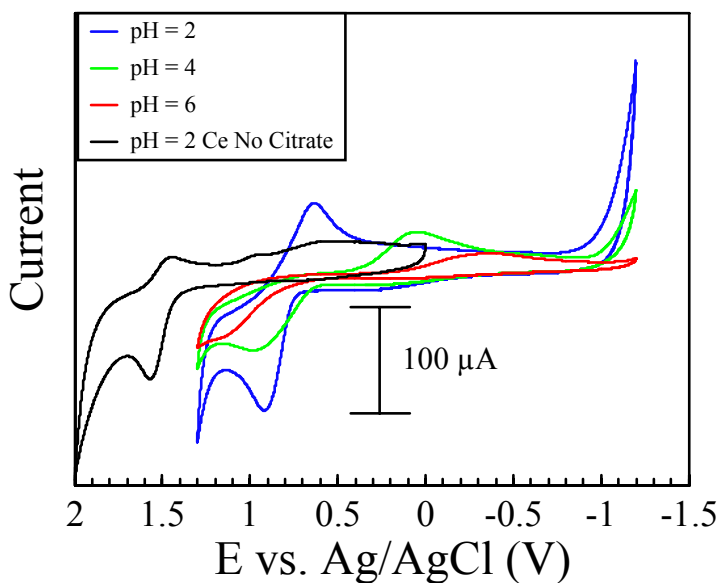


Figure 3. Cyclic voltammetric response of a graphite electrode for Ce uncomplexed and complexed with Citrate as a function of pH.

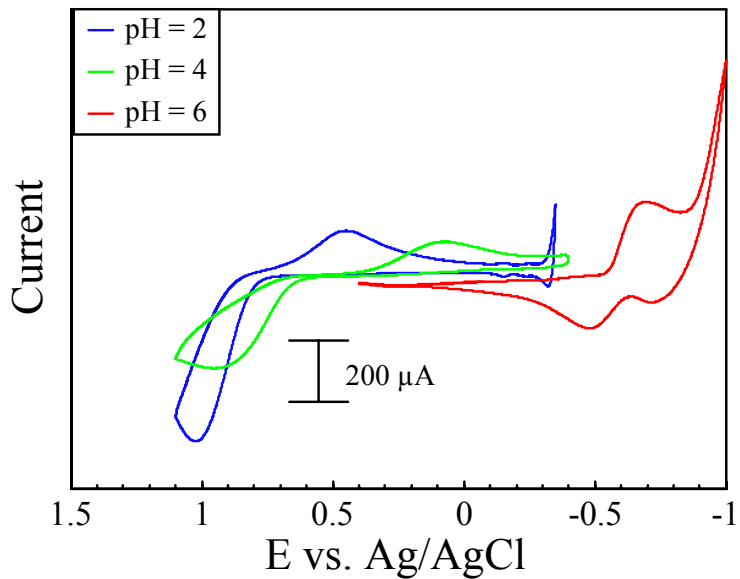


Figure 4. Cyclic voltammetric response of a Pt electrode for Ce uncomplexed and complexed with Citrate as a function of pH.

In the next six months the separation of Ce, Sm, and Eu will be attempted individually and from mixtures to determine if the potential mediated chelation using the chelating ligand dispersed in PANI/Au composite materials is possible.