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The Electrochemical Separation of Curium and Americium: Quarterly Report April - June, 2005

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**DOE/TRP Quarterly Report
Funding Year 2005
April 2005– Jun 2005**

Title: The Electrochemical Separation of Curium and Americium

This is a list of the personnel working on the project 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

Sujani Gamage

Ph.D. Candidate Chemistry

Results

- We have completed the electrochemical investigation in of the Ce^{3+}/Ce^{4+} redox couple and have determined the optimum experimental conditions.
- Computer modeling of the cerium using the *JChess* speciation-modeling program has been completed for the Ce redox couple. Traditional complexing ligands such as EDTA, oxalate, NTA, phosphate, acetate, and sulfate have been purchased and will be used to initiate the complexation and electrochemical characterization.
- Electrochemical investigations have continued on the Eu^{2+}/Eu^{3+} redox system in $HClO_4$ supporting electrolyte at a glassy carbon working electrode. The redox couple has been electrochemically resolved using cyclic voltammetry and square wave voltammetry. The data suggests that the couple is stable with reversible oxidation/reduction occurring.
- Complex formation has been initiated and theoretical calculations regarding the stability of species has been used to target the solution conditions required to view the oxidation/reduction processes.

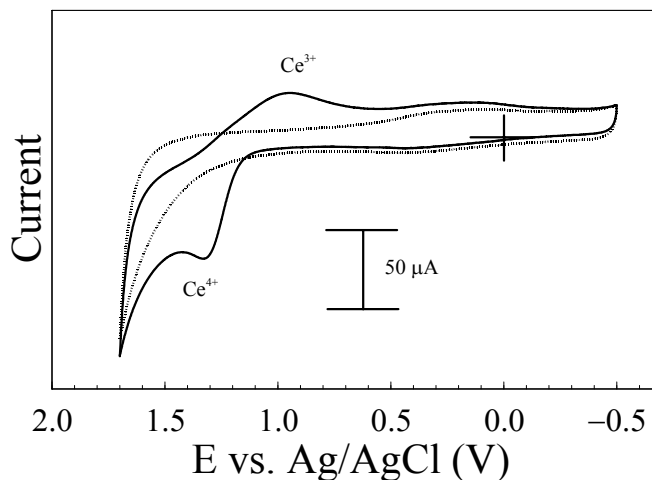
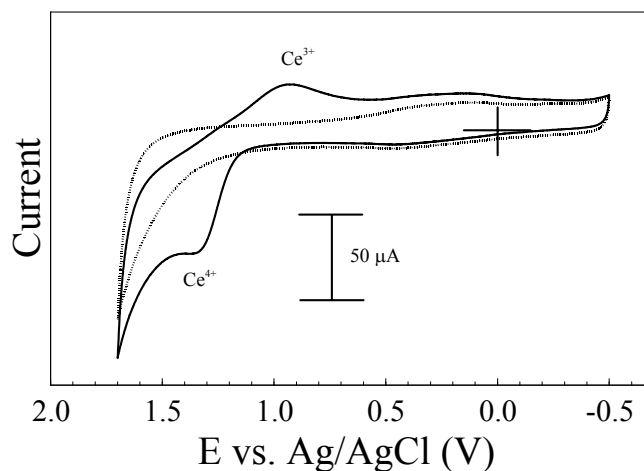
Discussion

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. We have been actively performing research on this project for six months and are currently on schedule for the proposed timelines.

The electrochemical characterization of the Ce^{3+}/Ce^{4+} redox couple as a function of working electrode material, pH and ionic strength has been completed. A manuscript outlining the data and results will be submitted in August. The work has shown that the

choice of electrode material determines the ability to resolve the redox couple under very different solution conditions. In the study optimum conditions for redox couple resolution at all three electrodes were realized in 0.1 M sulfuric acid solution. A decline in redox peak resolution, magnitude, and reversibility was observed as acid concentration was increased. The substitution of nitric acid for sulfuric acid yielded similar, though less resolved results. The influence of pH on the redox properties was illustrated using buffered solutions of incrementally increasing pH, while holding ionic strength constant. The results indicate that a pH greater than 2 was detrimental to the resolution of the cerium redox couple. Experimental results at the glassy carbon electrode differed significantly from those obtained at either Au or Pt, and revealed that glassy carbon was least affected by changes in pH. Glassy carbon also allowed successful resolution of the cerium redox couple in solution conditions that precluded its observation at either Au or Pt. Figures 1 through 7 are presented below.

We have also made progress on the $\text{Eu}^{2+}/\text{Eu}^{3+}$ system and a figure of the cyclic voltammetry and square wave voltammetry for the redox couple is provided in Figure 8.



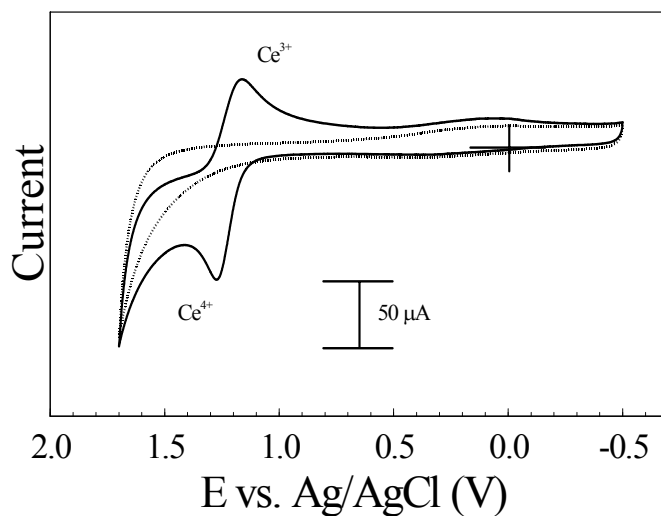


Fig. 1. Cyclic voltammetry performed on the Ce(III)/Ce(IV) redox couple in 0.1, 1.0, and 2.0 M (top) H₂SO₄ concentration at the glassy carbon working electrode ($v = 100$ mV/s).

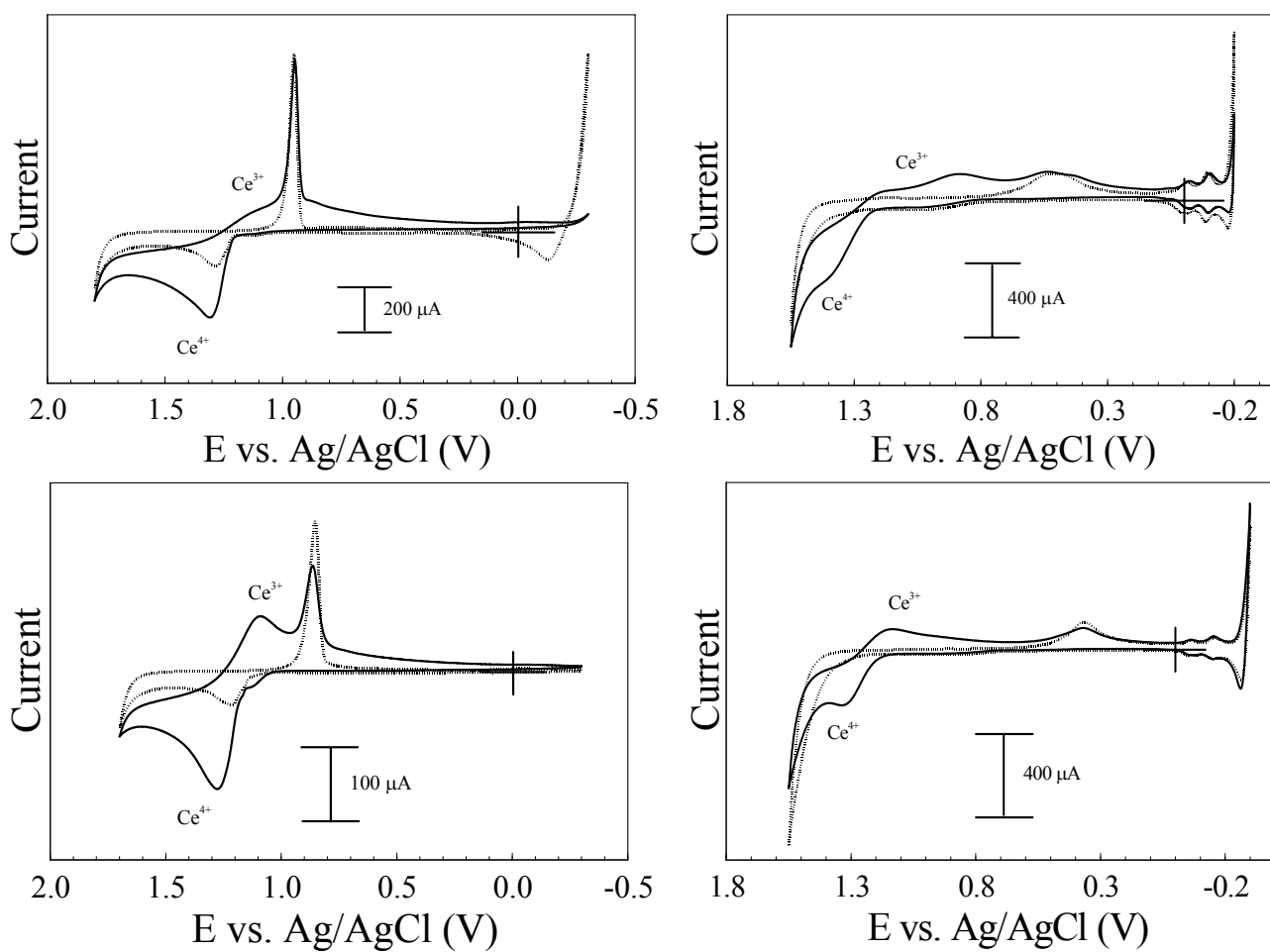
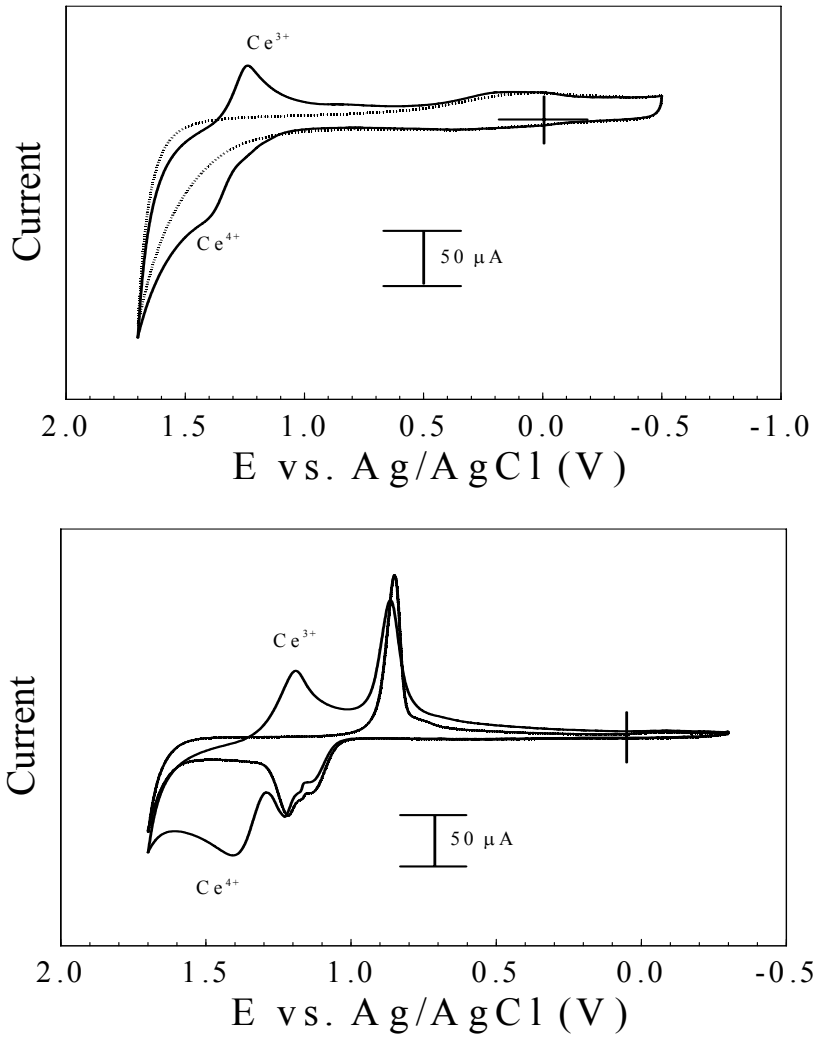


Fig. 2. Cyclic voltammetry showing Ce(III)/Ce(IV) redox behavior at Au (left column) and Pt working electrodes. These experiments were conducted at 0.1 M (lower figure in both columns) and 2.0 M H_2SO_4 concentration ($v = 100$ mV/s).



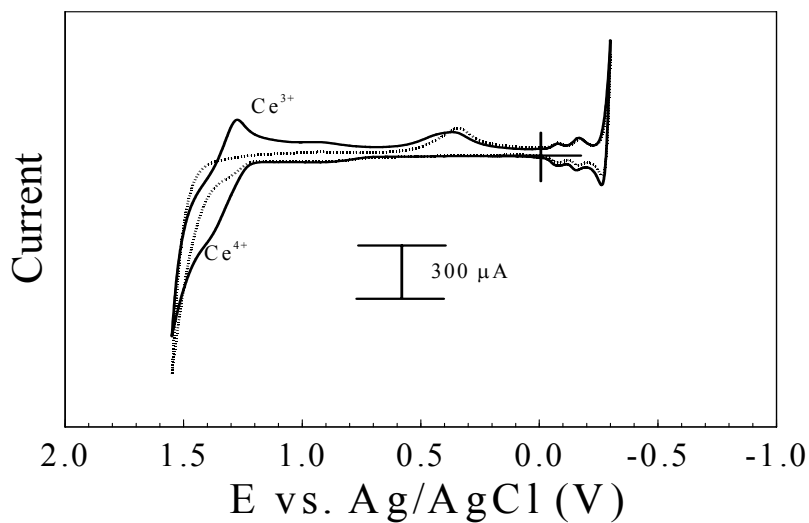
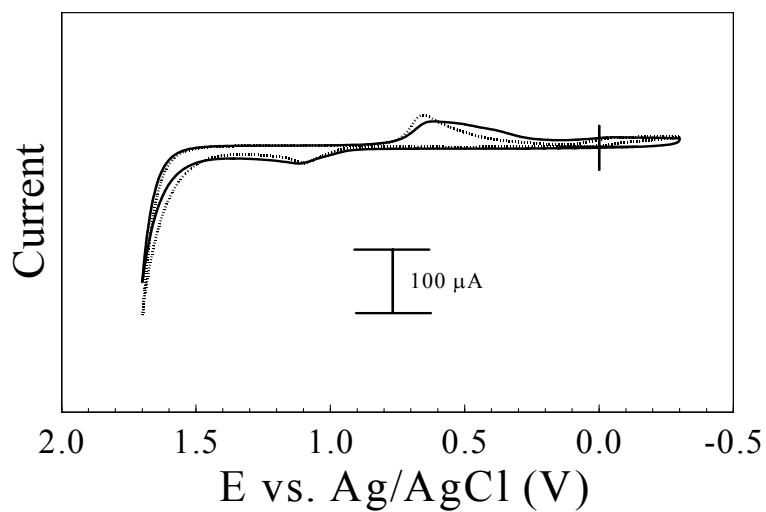
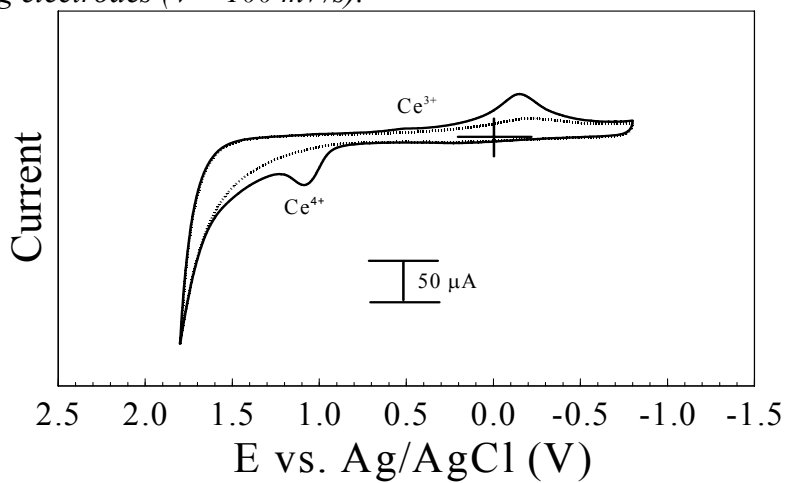


Fig. 3. Comparison of the Ce(III)/Ce(IV) redox couple in 0.1 M HNO₃ at GC (top), Au, and Pt working electrodes ($v = 100$ mV/s).



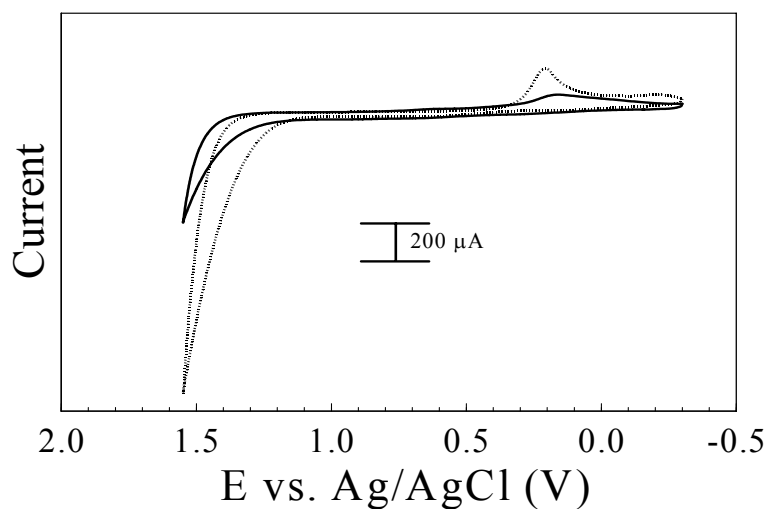
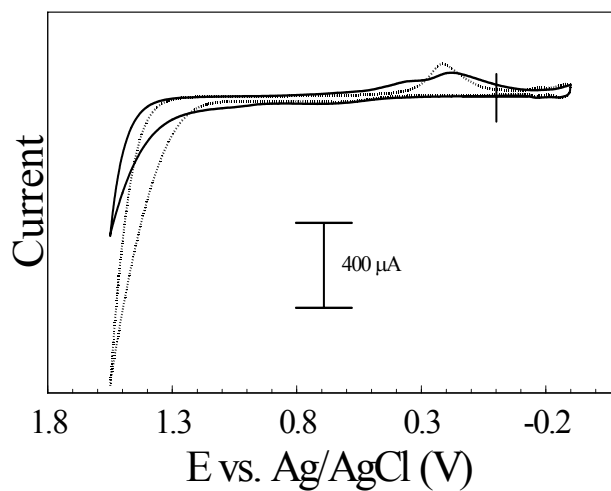
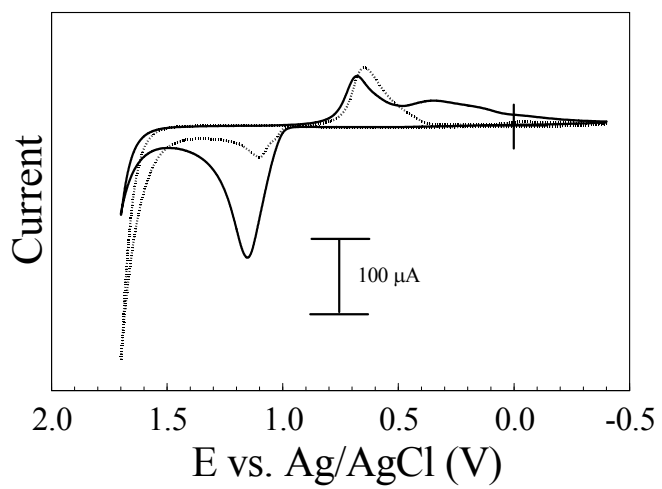
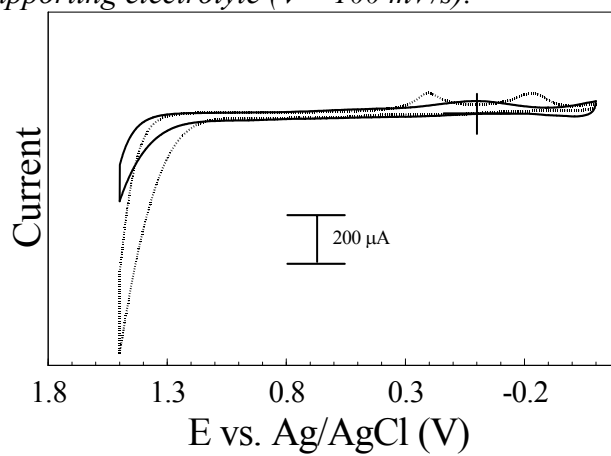
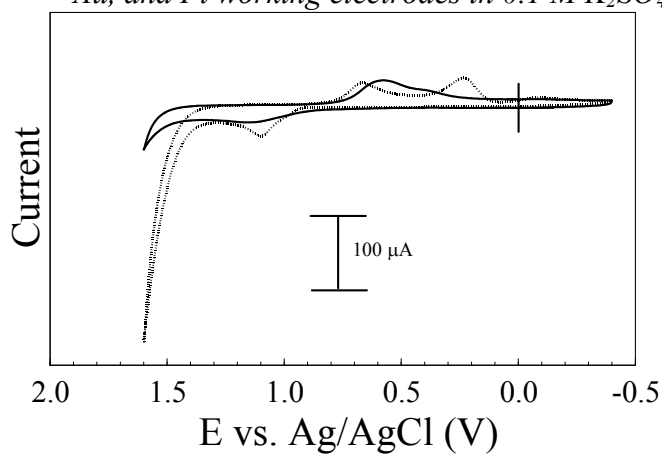


Fig. 4. The Ce(III)/Ce(IV) redox behavior examined by cyclic voltammetry at GC (top), Au, and Pt working electrodes in 0.1 M K_2SO_4 supporting electrolyte ($v = 100$ mV/s).



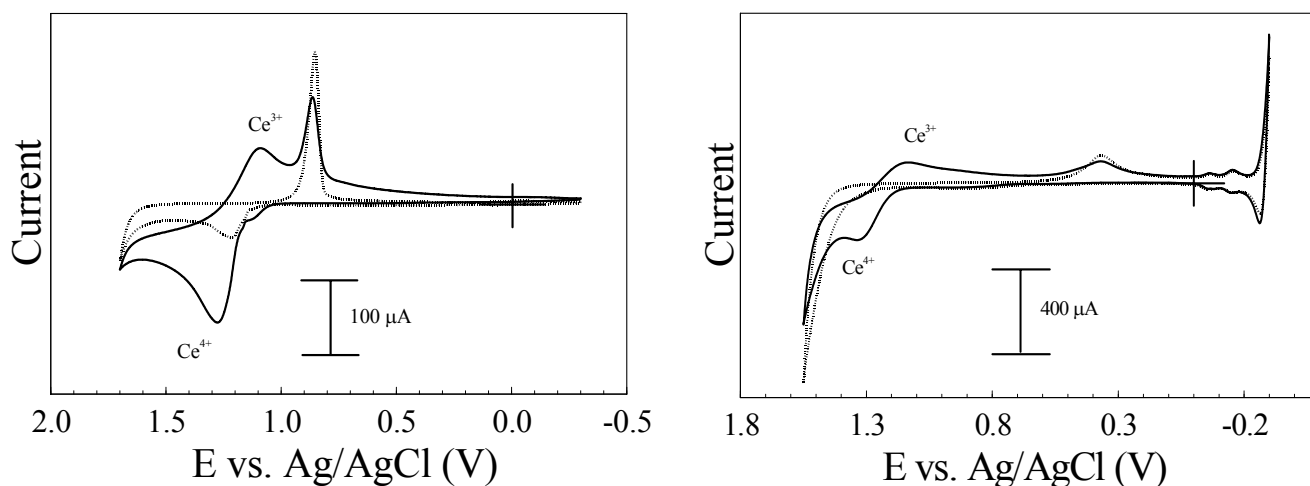
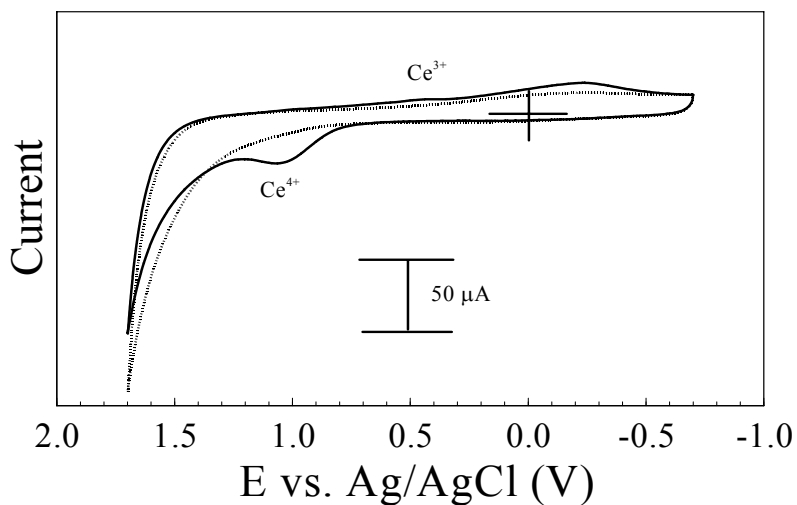


Fig. 5. Ce(III)/Ce(IV) electrochemistry at Au (left column) and Pt working electrodes. pH was controlled by means of $\text{HSO}_4^-/\text{SO}_4^{2-}$ buffered solutions. The figure at the top of each column shows cyclic voltammetry conducted at pH 4.7, followed by pH 3.4, compared to 0.1 M H_2SO_4 (lower figure in each column) at pH 1.0. ($v = 100 \text{ mV/s}$).



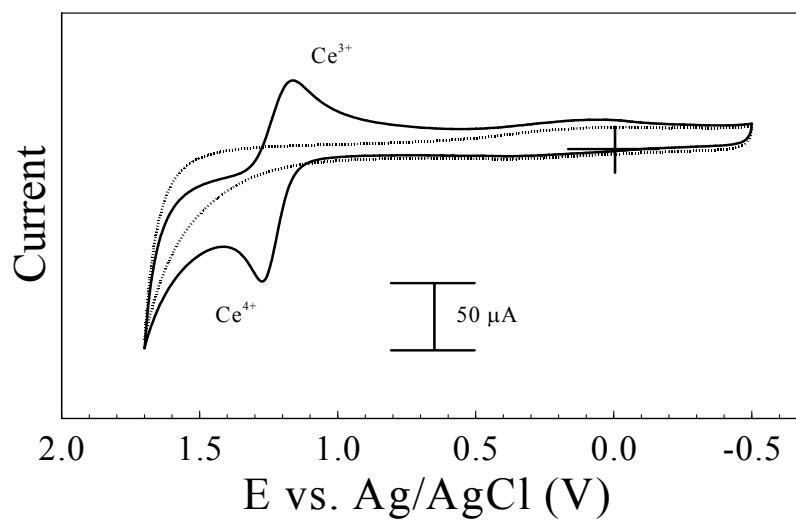
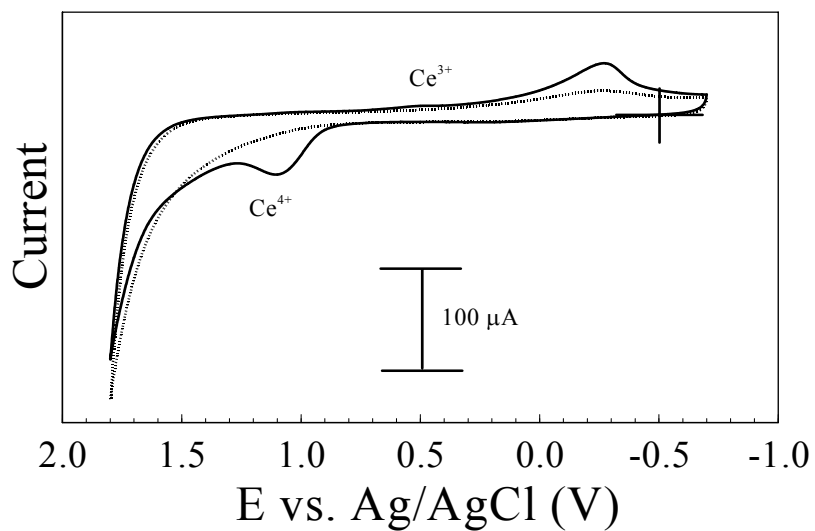


Fig. 6. Cyclic voltammetry showing cerium redox behavior at a GC electrode at pH 4.7 (top), 3.4 (middle), and in 0.1 M H_2SO_4 ($v = 100$ mV/s).

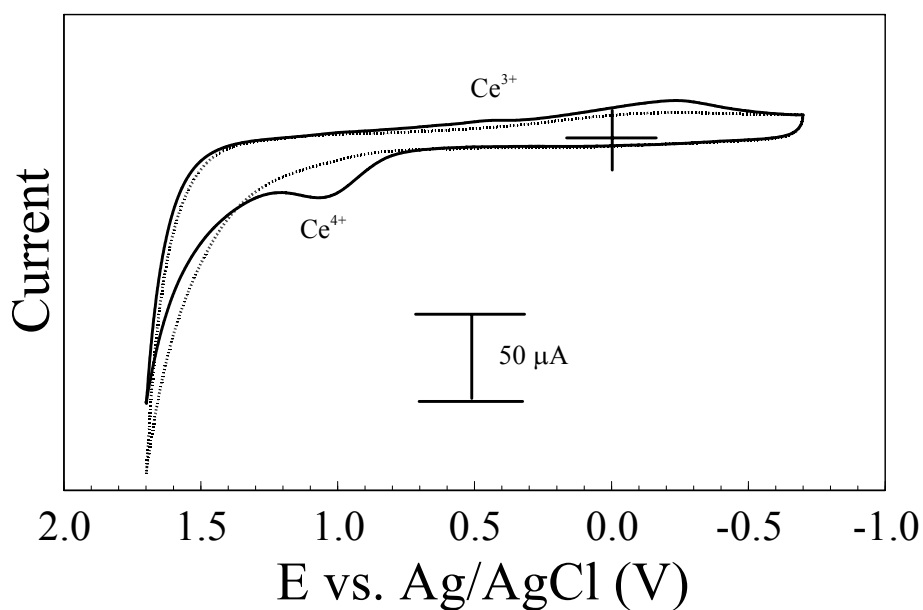
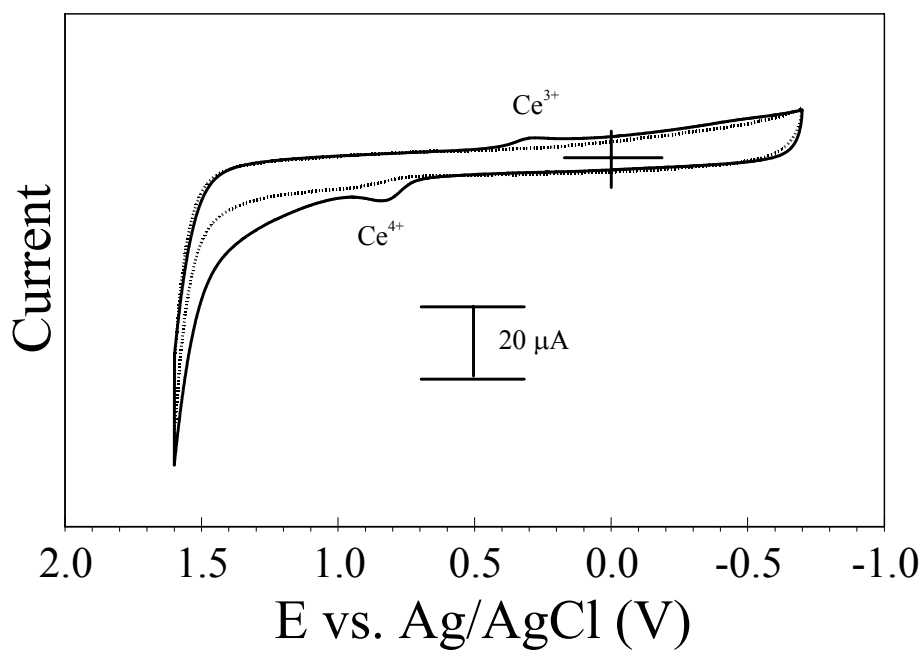


Fig. 7. Cyclic voltammetry showing cerium redox couple at a GC electrode in pH 4.7 acetate buffer (top) compared to pH= 4.7 in sulfate buffer ($v = 100 \text{ mV/s}$). Apart from the nature of the buffer system, both voltammograms were obtained under the same experimental conditions.

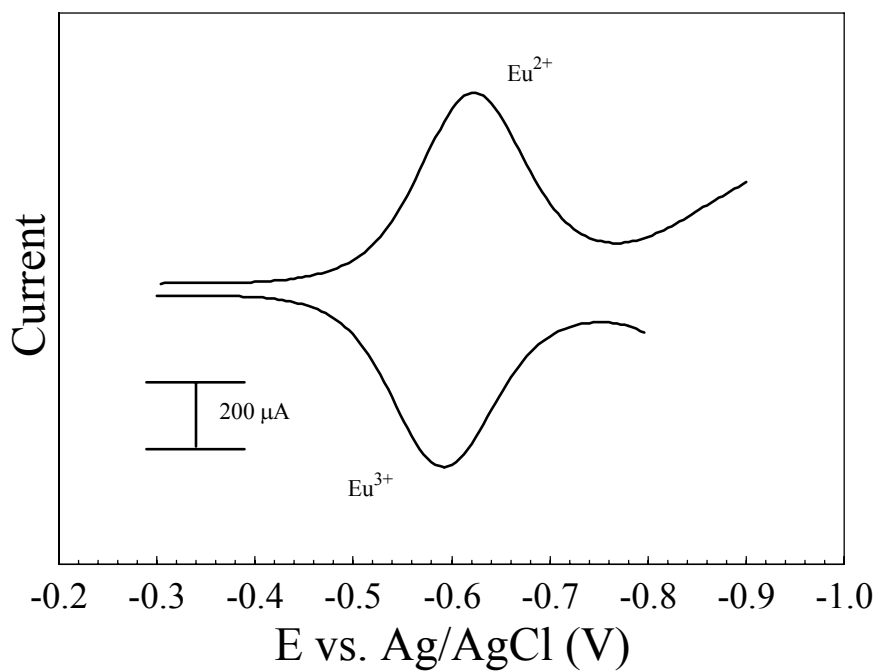
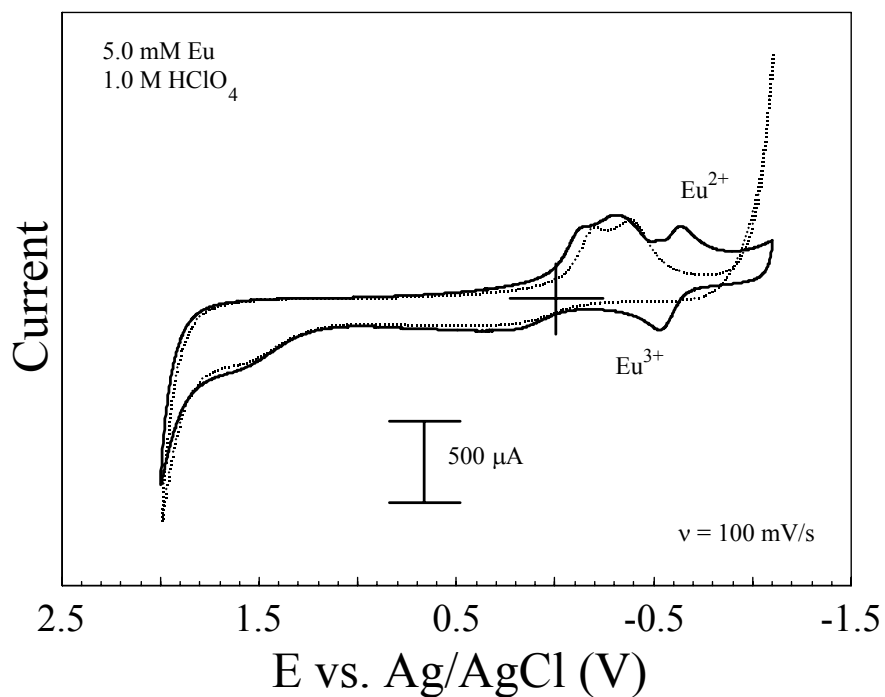


Fig 8. Cyclic voltammetry showing Europium redox couple at a GC electrode in 1M HClO₄ (top) containing 5mM Eu³⁺ ($v = 100$ mV/s). Square wave voltammetry for the forward and reverse scan (bottom) obtained under the same experimental conditions.