Electrodeposition of Actinide Compounds from an Aqueous Ammonium Acetate Matrix: Experimental Development and Optimization*

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Abstract

Electrodeposition is a technique routinely employed in nuclear research for the preparation of thin solid films of actinide materials used in accelerator beam bombardments, irradiation studies, or as radioactive sources. This study investigates the deposition of both lanthanides and actinides from an aqueous ammonium acetate electrolyte matrix. Electrodepositions were performed primarily on stainless steel disks, with yield analysis evaluated using γ -spectroscopy. Experimental parameters (run time, current density, voltage, electrolyte concentration, and initial analyte mass) were studied and modified to optimize the uniformity and adherence of the deposition while maximizing yield. The procedure utilized samarium as the plating material, both with and without a radioactive tracer. Surface characterization studies were performed by scanning electron microscopy, electron microprobe analysis, radiographic imaging, and x-ray diffraction.

Keywords: electrodeposition, electrolysis, actinides, lanthanides, samarium, californium, ammonium acetate, molecular plating, source fabrication, radioactive tracer

Introduction

Electrodeposition is a technique that has been used in radiochemistry as a means to prepare thin solid films of actinide elements with applications ranging from source preparation for α -spectrometry to target fabrication for accelerator bombardments. Here a technique is described by which quantitative deposition of lanthanide and actinide elements onto a stainless steel cathode can be accomplished from an aqueous solution of ammonium acetate at low (10–20 V) applied voltages. Historically, electrochemical deposition has been performed from both aqueous and organic matrices. The technique is also referred to as "molecular plating" for its tendency to deposit the molecular form of the element, rather than the metallic. Electrochemical deposition from both organic matrices (e.g., isopropanol and isobutanol [1]) and aqueous solutions [e.g., ammonium sulfate or ammonium acetate (AmmAc)] requires meticulous purification of the analyte fractions before electrodeposition, as trace amounts of organic or metal contaminants can limit the yield. [2] Contaminants in the deposition can lead to unwanted spectral shifts and peak broadening when producing a radiation source, particularly an α -source. Ion-exchange chromatography is often used to achieve the necessary purity of the materials. It is also important that the cathode, anode, and deposition cell be thoroughly cleaned with acid and/or alcohol to remove remaining contaminants. High-purity reagents must also be used throughout the electrodeposition process. The use of ammonium acetate as an electrolyte provides a neutral-pH buffer as well as an inexpensive matrix that requires low voltage (10–20 V) to achieve quantitative depositions. Organic matrices are unique in their resistance to decomposition and/or bubbling during electrolysis. However, organics can require voltages in excess of ~ 100 V for successful deposition. [3] These high voltages can require the addition of a cooling system to the electrodeposition cell to maintain a lower operating temperature. Cells with AmmAc as the electrolyte do not require cooling or high-voltage shielding, simplifying handling and unit

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fabrication.

The use of AmmAc as a matrix for electrodeposition has received very little attention in the literature. Roman [4] explored the electrodeposition behavior of Ra, Th, U, and Po from 0.35 M AmmAc solutions onto stainless steel and found the plating behavior of the actinide fractions to be strongly dependent on pH. In this study, small amounts of HCl were used to dissolve the analyte as well as for pH control, while maintaining an operating pH between 3 and 5.

A surrogate material for Cf was used to allow experimentation outside a glove box or hot cell. The redox solution chemistry of Sm^{3+} and Cf^{3+} ions should be similar, as indicated by standard reduction potentials to the ground state of -2.3 V and -1.94 V, respectively (although it should not be assumed that the lanthanide/actinide elements can be as easily electrodeposited as ground-state metals). [5] Even though Dy is the electronic homolog to Cf, and Gd is closer to Cf in cationic radius, it has been suggested that Sm, like Cf, has a (transient) divalent state in acid solution between the trivalent and ground states. [6,7] It has also been shown via co-precipitation studies in various solvents that the reduction behavior of Cf will closely follow Sm. [8] This reduction behavior is an important aspect in electrodepositions.

Various analytical techniques were used to study and characterize the physicochemical properties of the deposit, including the spatial distribution of activity (radiographic imaging), basic microstructure and thickness (scanning electron microscopy), crystallinity (XRD), and the chemical composition of the plated material (wavelength-dispersive spectroscopy). Gamma spectroscopy was performed to quantify the ^{153}Sm and ^{140}La radioactive tracers ($t_{1/2} = 1.946$ d, $E_\gamma = 103$ keV for ^{153}Sm and $t_{1/2} = 1.678$ d, $E_\gamma = 487$ keV for ^{140}La) to determine electrodeposition yields. [9]

Experimental

Reagents and Materials

The following reagents were used.

- 1.) 0.35 M, 0.7 M, 0.175 M ammonium acetate ($\text{NH}_4\text{CH}_3\text{COO}$) solution(s) at neutral pH, prepared by dissolving anhydrous ammonium acetate solid (certified ACS reagent-grade $\text{NH}_4\text{CH}_3\text{COO}$ purchased from Fischer Chemical) into deionized water.
- 2.) 0.1 M Sm^{3+} solution, prepared by dissolving $\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ into 0.1 M HNO_3 (trace, purchased from Fischer Chemical) to give 0.1 M Sm^{3+} solution in 0.1 M HNO_3 .
- 3.) Concentrated acids and bases: concentrated, reagent-grade (12.1 M) HCl purchased from EMD Chemicals®. Concentrated (14.5 M) NH_4OH certified ACS reagent purchased from Sigma-Aldrich.
- 4.) ^{153}Sm tracer ($t_{1/2} = 1.95$ d) obtained from irradiation of 99.2 wt % $^{152}\text{Sm}_2\text{O}_3$ in the High Flux Isotope Reactor (HFIR) at an unperturbed thermal neutron flux of 4.05×10^{14} neutrons/cm²/s. Between 1–10 μCi radioactivity was used per electrodeposition experiment.
- 5.) ^{140}La tracer ($t_{1/2} = 1.6780$ d) obtained as the daughter of ^{140}Ba ($t_{1/2} = 12.746$ d). Separation of daughter La(III) from parent Ba(II) was achieved using cation exchange chromatography with a disposable column of 1.0 mL bed volume and BioRad® AG-50 X4 analytical-grade cation exchange resin.

Electrodeposition Unit Design

Many electrodeposition cell designs explored in the literature have been based on the “chimney” style with the cathode placed at the bottom of the well. [2,3] The unit fabricated for this study is more comparable to an “aquarium” with the plating cathode fastened to the side of the chamber (*Figure 1*). The basic design involves the machining of a Teflon block with external dimensions 3.00 in. x 2.00 in. x 1.50 in. (length x width x height). The electrodeposition well is a 1.71 in. x 0.50 in. x 1.60 in. (length x width x depth) rectangular opening cut into the Teflon block, into which the electrolyte solution was transferred. The well contains a circular, 1.5 in. (3.81 cm), outer-diameter hole for securing the stainless steel cathode and a 0.50 in. (1.27 cm) cavity within this hole that serves as the active deposition area. The entire Teflon block is fastened to a stainless steel base, and electrical current is directed into a copper piece that screws onto the surface of the cathode to keep it firmly attached to the unit. A small Teflon piece separates the copper piece from the grounded stainless steel base, and a silicone rubber gasket seals the solution chamber from leaking around the cathode window. A flat round platinum disk, anode, was suspended in the aquarium ~0.25 in. (~0.64 cm) from the cathode, once the unit was assembled.

Figure 1: Schematic diagram of the electrodeposition unit.

Experimental Procedure

Experimental variables including run time, AmmAc concentration, solution pH, voltage/current, and mass of Sm dissolved were varied independently while keeping other variables constant. This allowed for testing of the system's response to adjustment of one variable at a time. The experimental procedure below attempts to optimize and maximize deposition yield. One 1.5 in. (3.81 cm) unabraded stainless steel disk with a 0.197 in² (1.27 cm²) active deposition area was first wiped down with ethanol and a lint-free cloth to remove any trace oils. The disk was then rinsed thoroughly using deionized (DI) water and placed into the electrodeposition cell as the cathode. The unit was assembled, sealed, treated with weak HNO₃ solution, and washed with DI water. The electrodeposition unit was then “conditioned” with the electrolyte solution before adding the feed solution. The conditioning step involved filling the assembled unit with AmmAc solution for 1–5 minutes and then discarding the conditioning solution, with the electrodes in place. Although optional, the conditioning step is thought to leach contaminants out of the unit that would otherwise be mobile during the deposition process.

Once the electrodeposition unit was assembled, cleaned, and conditioned, the deposition material was prepared. Samarium masses of 300, 600, and 900 µg were used as they reflect the low concentrations of radioisotopes often encountered during radiochemical operations. A radioactive tracer (¹⁵³Sm or ¹⁴⁰La) was combined with the Sm material and dried. The dried combination was then dissolved in 0.25 mL of

concentrated HCl, and the resulting solution was transferred into the well with four 3.5 mL rinse aliquots of AmmAc, bringing the total solution volume to approximately 14.25 mL. The “serial rinses” minimize loss of the radionuclide tracer during transfer.

Figure 2: The electrodeposition unit in operation.

The power was turned on and increased gradually during the first 5 minutes and then set at the desired constant current or constant voltage for a total of 4 hours. *Figure 2* shows a deposition cell in use, and *Figure 3* shows a close-up view of the post-deposition cathode plate. No post-experimental conditioning or surface treatment was performed beyond a water wash. Electrodeposited samples were stored in sealed plastic petri dishes. Post-deposition characterization was performed using x-ray diffraction, radiographic imaging, scanning electron microscopy/electron microprobe analysis, and wavelength-dispersive spectroscopy, with electrochemical yield analysis performed using γ -spectroscopy with the aid of a radioactive tracer (^{153}Sm or ^{140}La).

Figure 3: Photo of electrodeposited material after 4 hours electrolysis at 12 V constant voltage, 0.175 M AmmAc, and 600 μg Sm. Visible asymmetry in deposit due to hydrogen bubble

formation at cathode.

Results and Data Analysis

Quantitative electrodeposition was achieved with yields as high as 75% with a constant current density of 137.5 mA/cm^2 and yields over 90% with a constant applied voltage of 12 V. Thus, better electrochemical yields were achieved under conditions of constant applied voltage. *Figure 4* represents electrochemical yields at constant voltage, with no other changes.

Figure 4: Electrochemical yield data for constant-voltage runs at 0.35 M AmAc, 600 μg total Sm and a 4-hour runtime.

Quantitative analyses were performed via gamma (γ)-spectroscopy using a calibrated high-purity germanium detector (HPGe) with an active detector volume of $\sim 100 \text{ cm}^3$ and PC-based multichannel analyzer (MCA) from Canberra Industries (Meriden, CT). The detector has an energy resolution of 0.8 keV at 5.9 keV, 1.0 keV at 123 keV, and 1.9 keV at 1332 keV. Relevant nuclear data were obtained from Browne and Firestone. [9] All calibrations were determined using standard γ -ray sources traceable to the National Institute of Standards and Technology (NIST). Analysis was based on the observed activities of the ^{153}Sm or ^{140}La tracers. By measuring the activity of the test solution at regular 1-hour intervals during the electrolysis and comparing these decay corrected values to the activity of the initial solution and the stainless steel plate before and after the electrolysis, a simple relationship was established for determining the electrodeposition yield:

$$Y = \frac{A_{\text{plate}}}{A_{\text{feed}}} \approx \frac{A_{\text{feed}} - A_{\text{end}}}{A_{\text{feed}}} \quad (1)$$

where A_{plate} is the activity of the electrodeposited material on the plate after 4 hours of electrodeposition, A_{feed} is the activity of the test solution at the beginning of the experiment, and A_{end} is the activity of the test solution after the 4-hour electrodeposition is complete. The difference between the two quantities in *Equation 1* is correlated to the precision in yield measurements. For tracer-level work, both calculation methods, ($A_{\text{plate}}/A_{\text{feed}}$)

and $(A_{feed}-A_{end}/A_{feed})$, were able to be performed and correlated. However, when higher activity is utilized, the final electrodeposited plate will be too radioactive to gamma count directly and only the second method can be used.

Constant-Current and Constant-Voltage Experiments

For constant-current experiments, it was observed that electrochemical yields would increase with an applied current density up to a maximum of 137.5 mA/cm², after which yields began to decrease. The minimum current density tested was 50 mA/cm², with a maximum of 200 mA/cm². No electrodepositions were achieved in experiments with current densities lower than 50 mA/cm², and electrochemical yields did not reach greater than ~75–80% when the current density exceeded 137.5 mA/cm². In electrodepositions with constant voltage, the range of useful applied voltages was narrow (10–15 V), as only minimal deposition could be achieved using voltages under 10 V, and severe issues with bubbling were experienced once the voltage exceeded 15 V. Constant-voltage electrodepositions, consistently offering higher yields and fewer problems with reproducibility, comparatively, made them the primary focus when preparing samples for surface characterization.

Varying Initial Dissolved Samarium Mass and/or Electrolyte Concentration at Constant Voltage or Current Density

With the ideal current density of 137.5 mA/cm² chosen, varying the initial mass of Sm was pursued. The results showed that at constant current, and maximum yields of just below 80% plating efficiency were achieved in relation to initial dissolved Sm, regardless of quantity. Tests were performed using 300, 600, and 900 µg Sm.

The effects observed as a result of changing electrolyte concentration were more pronounced. Ammonium acetate concentrations of 0.175 M, 0.35 M, and 0.7 M were used along with a constant voltage of 12 V, 600 µg Sm, 4-hour electrolysis time, ¹⁴⁰La or ¹⁵³Sm tracer, and 0.25 in. electrode spacing. One consequence of increasing electrolyte concentration is increased gas production accompanied by splattering of the solution. If working with costly, irreplaceable, or hazardous isotopes, splattering can result in losses and contamination. This was observed by covering the electrodeposition cell with a lid for a visual confirmation of splattering and condensation. It was also observed that depositions had a more uniform thickness when electrodepositions were run at lower AmAc concentrations. It is thought that this may be due to decreased gas production, resulting in fewer disturbances in the solution.

Sample Characterization

Scanning Electron Microscope Imaging

Imaging was performed using a Hitachi S-4800 Field-Effect scanning electron microscope (SEM), with magnifications ranging from X35 to X100, with all images taken at the 5.0 kV operating voltage. Only dry, nonradioactive samples were staged. Grain boundaries and “valleys” assumed to have resulted from the dehydration process are referred to as “mud-caking,” and are clearly visible, as are the unequal grain sizes. Nucleation is favorable in defects, where so-called “active sites” of lower surface energy promote the microstructures visible in both images shown in *Figure 5* [10] At higher magnifications, clusters become visible, although little to no crystallinity can be seen in the deposited material, as was verified by x-ray diffraction (XRD) analysis.

Figure 5: SEM (X800 magnification, 5.0-kV mode) shows amorphous deposition grains, a “columnar” microstructure, and the iconic “mud-caking” resulting from the drying process in air.

Radiographic Imaging and X-Ray Diffraction (XRD)

Continuous θ - 2θ scans were performed on the PANalytical Xpert Diffractometer in 60 min readings using Mo K α radiation ($\lambda=70.93$ pm) and the X'Celerator detector. All scans used $1/4^\circ$ fixed slits and $1/2^\circ$ anti-scatter slit. Only dry, nonradioactive samples were imaged. When deposition samples were run, a broad, amorphous hump of relatively low peak height was observed between 10° and 20° that does not appear when the steel backing, or substrate, was run (Figure 6). This broadening represents regions of low ($<10\%$) crystallinity, and the center lies in the region where the samarium sesquioxide (Sm_2O_3) peak should appear. The rest of the diffraction spectrum is dominated by the sharp, intense peaks from the steel substrate. Attempts were made to reduce this broadening by “firing” the plates at relatively high temperatures ($200\text{--}450^\circ\text{C}$) for 1 hour to decompose potential organic contaminants. However, no effect on the XRD spectrum was observed. Electron microprobe analysis (EMPA) of the fired plates indicated that carbon and oxygen were lost from the sample surface during the firing, leaving the Sm in place.

The uniformity of electrodeposited material was analyzed by radiographic imaging. Samples were wrapped in polyethylene, smeared to confirm no contamination was present, placed in contact with a high-resolution phosphor screen (Perkin-Elmer®), and allowed to be exposed in darkness for 5–6 minutes. The phosphor screen was then placed in a Cyclone® phosphorimager for image analysis using the OptiQuant® software package. The OptiQuant® software provided two-dimensional contour plots; X and Y are spatial dimensions, with the radioactive intensity indicated by a color scale. The graphical software package ROOT® was used to transform these plots into three-dimensional graphs, where the z-axis is used to display radioactive intensity. The plots indicate that the distribution of activity is relatively uniform, but clusters around the center of the deposition (Figure 7). SEM thickness measurements, EMPA, back-scattered SEM imaging, and wavelength-dispersive spectrometry (WDS) were performed on both high-temperature fired (450°C , 1 hour) and “cold” (unfired) samples. Electron backscattering provided thickness measurements, and WDS was performed for elemental analysis (Figure 8). The unfired, Sm-containing sample had a plate-to-peak average thickness of 5.3 ± 0.1 μm , whereas the fired sample had a lower average thickness of 1.8 ± 0.1 μm . While both samples could be considered thin films, the attenuation properties of the fired sample are more desirable for radioactive source applications. The material “cracking” and mud-caking visible in the top-down SEM images become more striking in the cross-sectional views. In the case of the fired sample, cracks can be as deep as the thickness of the

deposited material. WDS and x-ray mapping reinforce the hypothesis that the deposited Sm takes the form of a molecular oxide (*Figure 8*, bottom). In the x-ray maps, O and Sm are located in the same regions of the deposit. The molecular hydroxide, $\text{Sm}(\text{OH})_3$, cannot be ruled out as WDS is not sensitive to the lowest-Z elements. Other atoms that may be ruled as contaminants include small amounts of alkali metals, Ca, and As. Iron and Cr exist in small amounts, likely due to their migration from the steel cathode, and Cl from the HCl seems to exist locally in the deposit. The reason for localization of the Cl is not known, but cluster-based growth of SmCl_3 is a possibility.

Figure 6: X-ray diffraction (XRD) crystallography shows an amorphous hump between 10° and 20° on deposited samples, representing the presence of Sm_2O_3 .

Figure 7: Radiographic imaging (RI) shows a relatively uniform distribution of radioactivity clustered around the center of the deposit. Slight asymmetry in the x-direction (more activity in the top left corner) may be attributed to the agglomeration of bubbles.

Figure 8: Cross-sectional SEM of electrodeposited material, thickness measurements (top), and associated elemental characterization via WDS and x-ray mapping (bottom). This sample did not undergo high-temperature firing prior to measurement. Abundance of O and Sm suggests molecular oxide form of electrodeposited material.

Discussion and Conclusions

The chemical behavior of the solution during electrolysis is complex, but it has been shown that many simultaneous reactions are occurring in the solution beyond the splitting of water and reduction of Sm^{3+} . The production of H_2 and O_2 gases accompanies the splitting of water in any aqueous electrochemical cell, but the presence of an organic electrolyte complicates analysis. A well-studied example of an electrolytic reaction involving the acetate ion is the Kolbe reaction, which involves the oxidation of carboxylate at the anode, followed by decarboxylation and dimerization of the alkyl radical [11]:

The Kolbe product R_2 is formed in solution assuming recombination of the intermediate radical, but interfering processes include disproportionation and attack of R^* on solvent molecules. Further analysis of the evolved gases would be required to differentiate between the gases released (e.g., H_2 , CH_4 , and C_2H_6). The gas production of these processes is problematic due to the generation of bubbles at the cathode, which can lead to brief voltage instabilities if the bubbles grow large enough. A large hydrogen bubble at the cathode surface will restrict conduction and prevent deposition. In addition, the tendency for bubbles to float to the top of the cathode well and become trapped has resulted in “half-moon” asymmetries in the distribution of electrodeposited material visible in *Figure 3*. Deposition is restricted in the region of the hydrogen bubble and instead accumulates at the lower half of the stainless steel plate.

A reproducible method for the preparation of radioactive lanthanide and actinide sources via electrodeposition onto stainless steel from a mildly acidic (pH between 3 and 5) ammonium acetate solution has been demonstrated. Gamma spectroscopy and a radioactive tracer allow for quantitative determination of the deposition yield, and SEM-based and radiographic imaging, XRD, and WDS spectroscopy clarify surface patterns, source thickness, and chemical composition of the deposit. The cracked, mud-caked appearance of the SEM images is highly characteristic of a so-called “columnar” microstructure common to the molecular plating technique. [10,12] Average deposit thickness, with and without post-electrolysis heat treatment, is in the range typically defined as a thin film. [13] Further analytical work focused on characterizing the gases produced during electrolysis is planned and will be taken into consideration for future unit designs to reduce problems related to bubbling and the concentration of material in specific areas of the stainless steel cathode.

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