ORNL/TM-2022/1837

Electrodeposition Method Development for Californium Rare Isotope Breeder Upgrade Source

Jisue Moon
Kristian Myhre
Susan Hogle

February 2022

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Radioisotope Science and Technology Division

ELECTRODEPOSITION METHOD DEVELOPMENT FOR CALIFORNIUM RARE ISOTOPE BREEDER UPGRADE (CARIBU) SOURCE

Jisue Moon
Kristian Myhre
Susan Hogle

February 2022

Prepared by
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, TN 37831
managed by
UT-BATTELLE LLC
for the
US DEPARTMENT OF ENERGY
under contract DE-AC05-00OR22725

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ACRONYMS

ATLAS	Argonne Tandem Linear Accelerator System
CARIBU	Californium Rare Isotope Breeder Upgrade
Cf	Californium
CP	Chronopotentiometry
ICP	Inductively coupled plasma
REDC	Radiochemical Engineering Development Center
SEM-EDS	Scanning electron microscopy–energy dispersive x-ray spectroscopy
Sm	Samarium

ABSTRACT

Electrodeposition is one of the most widely utilized techniques for thin-film production with high yields. Although it is used extensively, this technique still presents several issues limiting its usefulness, including excess thickness and frequent cracking of deposited films. Efforts to improve thin-film production recently undertaken at Oak Ridge Laboratory by the authors have included studying operating conditions and applied electrolytes. Samarium was used as a surrogate material for californium, and yield was compared with different deposition condition. The final procedure from the optimized condition will be applied during production of the californium-252 source for the Californium Rare Isotope Breeder Upgrade (CARIBU) project, and the fission fragment will be analyzed.

1. INTRODUCTION

The Argonne Tandem Linear Accelerator System (ATLAS) at Argonne National Laboratory can accelerate heavy ions at energies in the range of the Coulomb barrier. Beams of nearly all stable isotopes, ranging from a proton to uranium to a number of in-flight produced light radioactive beams can be studied in ATLAS [1]. The Californium Rare Isotope Breeder Upgrade (CARIBU) has been added to ATLAS to supply heavier neutron-rich beams that are used to produce and study neutron-rich nuclei similar to those produced by the astrophysical rapid neutron capture process—believed to be responsible for a large fraction of heavy elements in the universe [1],[2]. The Radiochemical Engineering Development Center (REDC) located at Oak Ridge National Laboratory was asked to produce the californium-252 (Cf-252) source needed for the CARIBU project.

The source has been produced by electrodeposition onto a stainless-steel substrate, which required material free from excess mass for efficient deposition. Traditionally, electrochemical deposition has been performed from both aqueous and organic matrices. This method is also referred to as molecular plating for its tendency to deposit the molecular form of the element rather than the metallic form. The method chosen to produce this source has used ammonium acetate or ammonium sulfate electrolyte as an inorganic solution base and isobutanol or isopropanol as an organic solution base. Both types of matrices require purification of the analyte fractions before electrodeposition, as trace amounts of organic or metal containment can limit the yield. Additionally, contaminants in the deposition can lead to unwanted spectral shifts and peak broadening when a radiation source is being produced. The use of aqueous solution requires low voltage (i.e., 10–20 V) to achieve quantitative depositions, whereas the organic matrices require voltages in excess of ~100 V for successful deposition. This high voltage additionally requires application of a cooling system to the electrodeposition cell to maintain a lower operating temperature. Thus, oftentimes an aqueous solution with ammonium acetate as an electrolyte is preferred as the system, which can simplify the handling and the unit.

The largest electrodeposition of ^{252}Cf ever produced at the REDC included 6.3 ± 0.5 GBq (1.7 ± 0.1 Ci) ^{252}Cf [3],[4], but the source was thicker than desired for the application, which resulted in reduced overall fission fragment yield. This is believed to originate from the electrodeposition procedure, in which the organic substituents from the ammonium acetate electrolyte solution are deposited along with ^{252}Cf. Additionally, issues resulting in unwanted fission fragments from the thin film need to be addressed. To understand what caused the abnormal thickness and unwanted organic matter in the thin film, this work focused on elucidating the role of each step in the electrodeposition of lanthanide, especially using samarium (Sm) as a representative lanthanide and a surrogate for Cf. The effect of the pH of the initial solution, current density, electrodeposition time, and treatment after electrodeposition will be elucidated throughout this study. The effect of each variable on the deposited film morphology, thickness, chemical composition, and stability of the thin film will be discussed.

2. EXPERIMENTAL METHODS

2.1 ELECTRODEPOSITION SETUP FOR CALIFORNIUM-252 TARGET

The electrodeposition unit used in this work (Figure 1) featured a well-type configuration with a copper base housing a stainless steel cathode as the deposition substrate with a deposition area of 7.2 cm^2. The well was machined with transparent acrylic so the solution could be observed during deposition. A silicon gasket was used to complete the seal between the well and substrate. The anode was a platinum plate containing slits to prevent gas accumulation between the electrodes. The anode was suspended in the well to maintain a distance of 1 cm between the deposition surface and cathode. Electrodepositions with the natural Sm surrogate were carried out using a BioLogics SP-300 potentiostat with 2A booster channel.

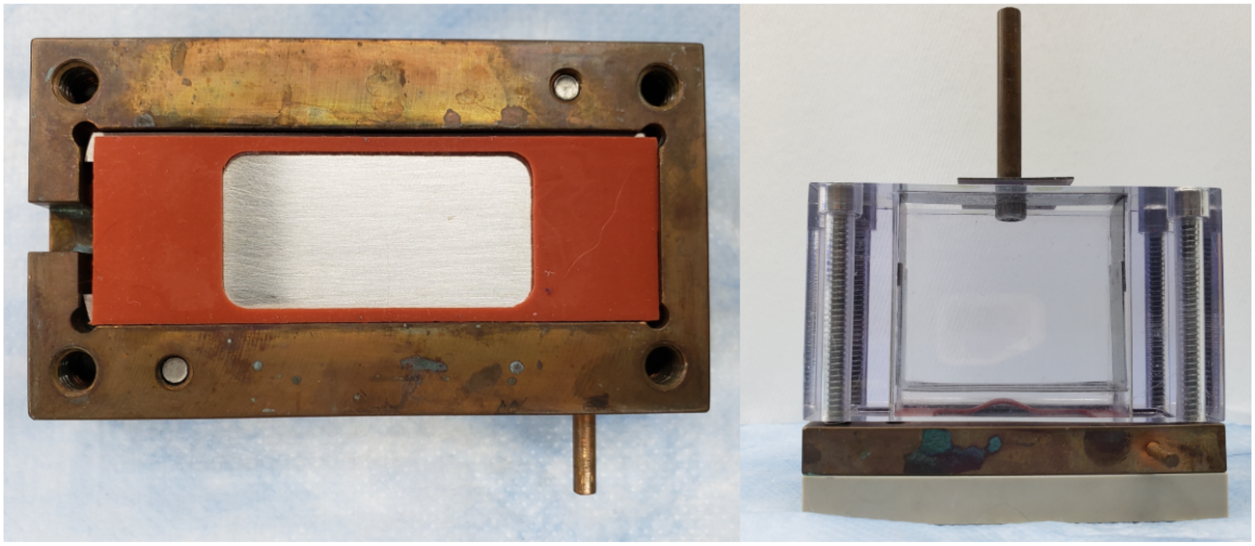


Figure 1. Electrodeposition cell. (left) Stainless steel substrate inserted copper cathode with silicon liner, and (right) assembled electrodeposition cell.

Prior to each deposition, the electrodeposition unit was washed with 0.1 M HNO_3, ethanol, and deionized water. It was then assembled and leak-tested with deionized water for a minimum of 1 h. Upon passing the leak test, the unit was set up for the electrodeposition run. Samarium Inductively coupled plasma(ICP) standard solution (10,000 mg/L, dissolved in 2–3% HNO_3) was used. Three hundred ppm of Sm was dissolved in the electrolyte solution and then added to the electrodeposition unit. The final electrolyte volume was 20 mL. The anode was then submersed in the electrolyte solution, and both cathode and anode were connected to potentiostat. The potentiostat was then energized, and constant current and different currents were applied to understand the role of current. After deenergizing the system, the solution was carefully collected without disturbing the formed thin film and analyzed using inductively coupled plasma optical emission spectrometry to obtain deposition yield. Additionally, the electrodeposition time was also varied from 30 min to 4 h. After removing the solution, the formed thin films were dried. Drying temperature was also studied to understand its effect. Table 1 summarizes the variables that have been tested in this study.

Table 1. Variables studied with detailed information of each.

Variables	Tested condition	Notes
Electrolyte	Ammonium acetate, ammonium nitrate, and mixture of ammonium acetate and ammonium nitrate	Concentration was tested at 0.1 M of each electrolyte
pH of initial solution	1.0, 4.0, 7.0	pH > 7.0 caused the precipitation of Sm before electrodeposition
Sm amount	300, 1,000 ppm	6 µg and 20 µg Sm in 20 mL solution
Applied current	100, 250, 500, 1,000 mA	Corresponding current densities are 13.9, 34.7, 69.4, 138.9 mA/cm^2
Deposition time	30 min; 1, 2, 4 h	Time was optimized for the mixed electrolyte system with 1 A system
Electrolyte concentration	0.1 M ammonium acetate with 0.03 M ammonium nitrate 0.1 M ammonium acetate with 0.1 M ammonium nitrate 0.05 M ammonium acetate with 0.05 M ammonium nitrate	Electrolyte concentration of was tested for mixed system to optimize the ammonium nitrate concentration in addition to the ammonium acetate
HCl effect	With and without HCl addition	HCl was used in previous CARIBU source production to dissolve Cf in solution
Heating effect after deposition	Room temperature, 80^{o}C	Substrate finished electrodeposition was heated on the hot plate

3. RESULTS AND DISCUSSION

3.1 EFFECT OF ELECTROLYTE

The use of ammonium acetate as an electrolyte provides a neutral-pH buffer as well as an inexpensive matrix that requires low voltage to obtain depositions. In this work, ammonium acetate was used as a main electrolyte for Cf source electrodeposition. A previous study by Myhre et al.[5] showed, through X-ray photoelectron spectroscopy analysis, that the carbon is also co-deposited within the Sm thin film. Ammonium nitrate was selected as an alternative electrolyte and tested at the same condition with ammonium acetate.

Electrodeposition was performed using chronopotentiometry (CP), which controls current and monitored voltage change. Voltages of 100, 250, and 500 mA were applied to the system, corresponding to current densities of 13.9, 34.7, and 69.4 mA/cm^2$_,$ respectively. First, two individual tests using 0.1 M of ammonium acetate and 0.1 M ammonium nitrate solution were conducted at 500 mA (Figure 2). For ammonium acetate, the gas production (H_2 in this case) from the Kolbe reaction [6], which involves the oxidation of carboxylate at the anode followed by decarboxylation and dimerization of the alkyl radical, was observed. After deposition, the color of deposited area turned to a black/brown color (Figure 2b); observed morphology was the typical mud crack shape with some empty spots caused by bubble generation (Figure 2c). The ratio of Sm and oxygen from weight percentage from scanning electron microscopy–calibrated energy dispersive x-ray spectroscopy (SEM-EDS) analysis was close to 3, suggesting the formation of $Sm(OH)_3$. Likewise, ammonium nitrate also showed significant gas formation, but the bubble size observed during the deposition was bigger than the bubble size with ammonium acetate. There was no discoloration on the substrate surface, and the morphology of the deposited Sm compounds was bulk chunks with bigger empty spaces. For ammonium nitrate, two additional current levels were tested to minimize gas generation close to the level of ammonium acetate and get the well-distributed thin film over the substrate at lower current. As the current decreased, deposits appeared as flattened mud crack shape with thicker deposits on the surface, but significant lift-up at the edge was observed for the thicker pieces.

An additional problem often discussed in the literature is the redissolving of extracted metal ions on the substrate after deenergization. A liquid sample was collected before and after deenergization to compare the yield and dissolution effect before and after power-off (Table 2). Before powering off the potentiostat, the average yield was found to be close to 80%, with the highest yield with ammonium nitrate. However, after power-off, the deposition dropped a maximum of ~8% for both ammonium acetate and ammonium nitrate. This indicated that the dissolution effect exists after the potentiostat powers off.

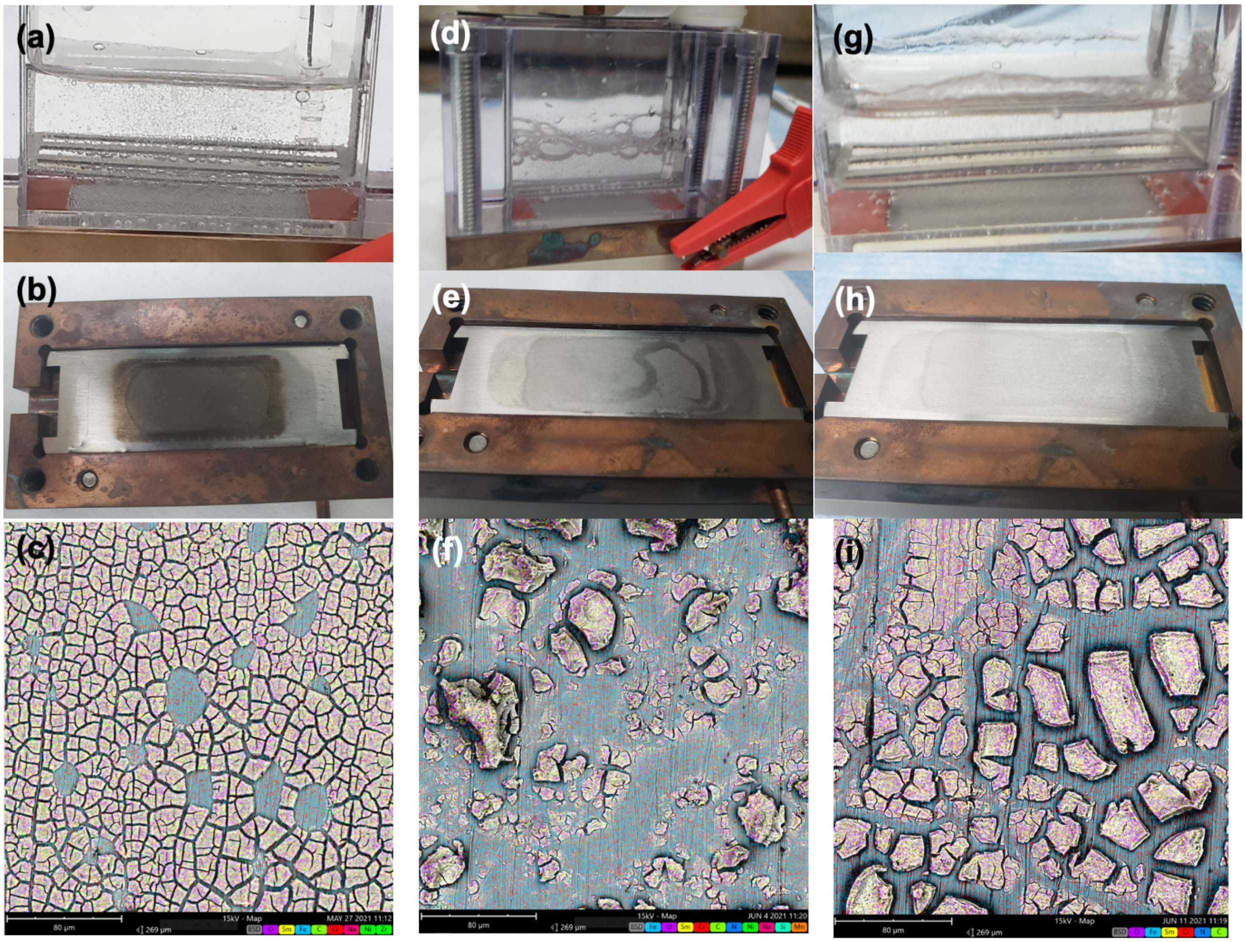


Figure 2. Electrodeposition using 0.1 M ammonium acetate and 0.1 M ammonium nitrate with CP mode. (a) Electrodeposition unit after deposition for 2 h; (b) anode and substrate after electrodeposition for 4 h; (c) SEM-EDS results for thin-film surface after electrodeposition for 4 h using 0.1 M ammonium nitrate with CP mode (500 mA); (d) electrodeposition unit during the deposition for 2 h; (e) anode and substrate after electrodeposition for 4 h, showing disassembled well and cathode; (f) SEM-EDS results for the thin-film surface after electrodeposition for 4 h using 0.1 M ammonium nitrate with CP mode (100 mA) for 4 h; (g) electrodeposition unit during the deposition for 2 h; (h) anode and substrate after electrodeposition for 4 h; (i) SEM-EDS results for the thin-film surface after electrodeposition for 4 h using 0.1 M ammonium nitrate with CP mode (100 mA) for 4 h. Note that all SEM images have same scale of 80 µm.

Table 2. Electrodeposition conditions with deposition yield.

Solution	Current	Current density (mA/cm^2)	Sm (ppm)	Deposition yield before power-off (%)	Deposition yield after power-off (%)
Ammonium acetate	500	69.5	300	79.07	76.91
Ammonium nitrate	500	69.5	300	96.27	86.23
Ammonium nitrate	250	34.7	300	79.18	71.34
Ammonium nitrate	100	13.9	300	77.29	73.93

Adhesion of thin-film coating was assessed using standard test methods (i.e., ASTM D3359 method) and the tape listed in the standard. Tape was applied over the film and then removed. Adhesion was assessed by comparing the weight before and after the tape test. The film obtained by deposition using ammonium acetate at 500 mA showed stronger adhesion, removing 0.0005 g of material from the surface compared to 0.0013 g removed from the deposit formed by deposition using ammonium nitrate at 500 mA. The test was repeated for three times. The weight loss from the tape test was within a 2% margin of error. The results suggest that ammonium acetate–generated thin film is more strongly adhered on the substrate and ammonium nitrate can deposit more materials on the substrate, but the dissolution issue for both ammonium nitrate and acetate electrolyte must be solved to obtain better yield.

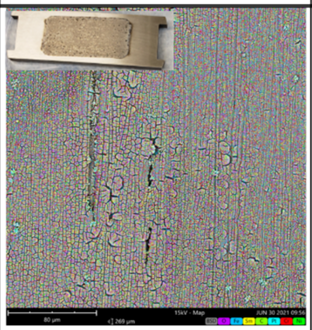
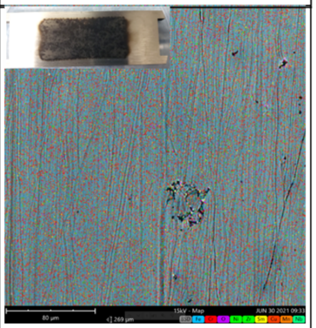
3.2 EFFECT OF PH OF INITIAL SOLUTION ON THE DEPOSITION

Electrodeposition requires a specific pH to make sure the targeted lanthanide or actinide element remains in the solution before the electrodeposition and that it is evenly deposited on the substrate during the deposition process. Solutions can be acidic or basic, but the wrong pH results in unwanted particles on the target or unwanted reactions.

To examine how pH affects lanthanide/actinide electrodeposition, pH values for the initial solution of ~1, 4, and 7 were chosen as variables. pH values over 7 were not selected in this study because they would precipitate targeted lanthanide and actinide before the electrodeposition as a form of hydroxide and the particles would stay in the solution and not become involved in electrodeposition.

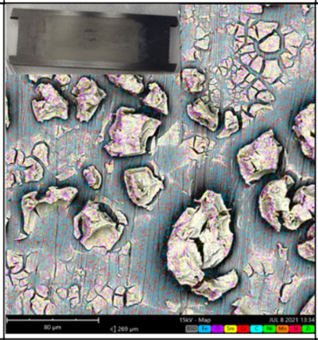
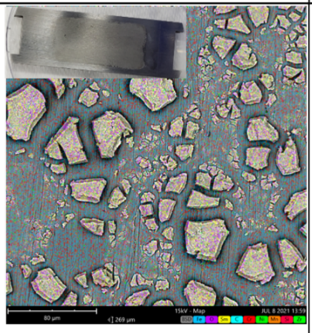
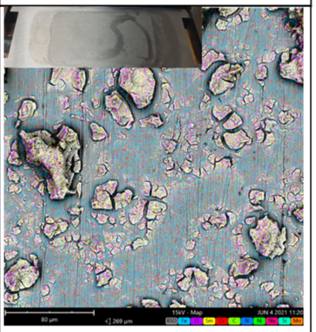
As a result, for ammonium acetate (Table 3), more materials were deposited on the substrate at a pH between 6 and 7. However, SEM results showed that most of the deposit at higher pH levels was carbon decomposed from acetate and did not show any significant Sm deposits on the surface, with only 28% yield. For lower pH levels (i.e., 1.03 and 3.73), lighter discoloration significant Sm deposition were seen on the substrate compared to higher pH solution. The highest yield and strongest adhesion were observed at pH levels between 3 and 4.

Table 3. Results of electrodeposition using ammonium acetate with different initial pH levels.

pH	1.03	3.73	6.24
Experiment condition: Ammonium acetate 300ppm Sm 500 mA, CP mode			
Deposited mass	3.5 mg	3.2 mg	4.3 mg
Sm deposition yield (%)	86 %	90 %	26 %
Adhesion test mass loss (mg)	0.5 mg	0 mg	0.3 mg

For ammonium nitrate (Table 4), a significant amount of Sm was deposited on the substrate, with the highest yield at pH levels between 1 and 2, whereas ammonium nitrate showed weak adhesion compared to ammonium acetate. The morphology is bigger particles rather than thin film.

Table 4. Results of electrodeposition using ammonium nitrate with different initial pH levels.

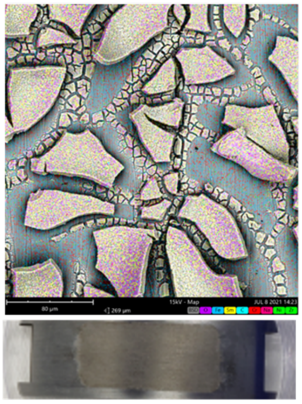
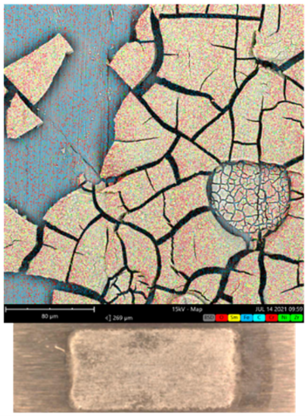
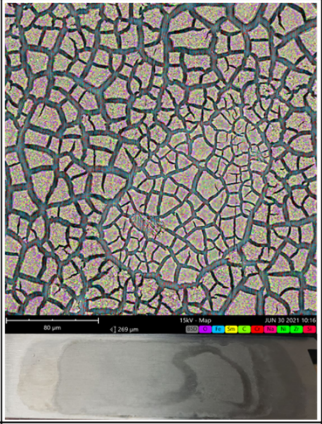
pH	1.40	3.54	6.37
Experiment condition: Ammonium nitrate 300ppm Sm 500 mA, CP mode			
Deposited mass	150 mg	141 mg	50 mg
Sm deposition yield (%)	98 %	71 %	17 %
Adhesion test mass loss (mg)	3.3 mg	1.6 mg	1.3 mg

Considering the fact that ammonium acetate showed good adhesion and ammonium nitrate showed improved yield, the two electrolytes can be mixed and tested with different concentrations of ammonium nitrate to optimize yield and adhesion. When a lower amount of ammonium nitrate (0.035 M) than ammonium acetate (0.1 M) was applied in the system (Table 5), pH affected morphology significantly and deposited mass. When the initial pH was 3.87, the largest amount of Sm was deposited, showing the highest yield. At the lowest pH, lift-up of the edge of thin film was observed with most of the larger pieces. At a higher pH of 7.03, the deposition had a mud crack shape, but the individual cracked particles are smaller than the thin-film deposits formed at a pH of 3.87. Interestingly, a pH of 3.87 improves the deposited mass significantly as well as the morphology of thin film but did not improve adhesion. The highest yield was shown at a pH of 3.87.

When the concentration of ammonium nitrate was increased to the same level (0.1 M) of ammonium acetate (Table 6), the adhesion was improved greatly, and more Sm was deposited on the substrate than ammonium acetate. Additionally, the morphology was more uniform and formed fewer crack than ammonium nitrate. Sm yield was the highest at the lowest pH and exhibited strong adhesion.

For lower concentrations of the mixture (0.05 M ammonium nitrate and 0.05 M ammonium acetate), at a pH of 1.31, the yield was decreased by 81%, and deposited mass was 5.7 mg but showed similar adhesion. The morphology is also similar with higher concentrations (Figure 3).

Table 5. Results of electrodeposition using mixture of 0.1 M ammonium acetate and 0.03 M ammonium nitrate with different initial pH levels.

pH	**1.75**	**3.87**	**7.03**
Experiment condition: 0.1M Ammonium acetate 0.035M Ammonium nitrate 300ppm Sm 500 mA, CP mode			
Deposited mass(mg)	10.7	133.50	5.90
Sm deposition yield (%)	60	97	75
Adhesion test mass loss (mg)	3.9	6.2	6.0

* Note that the scale of each SEM images is same (80 μm).

Table 6. Results of electrodeposition using mixture of 0.1 M ammonium acetate and 0.1 M ammonium nitrate with different initial pH levels.

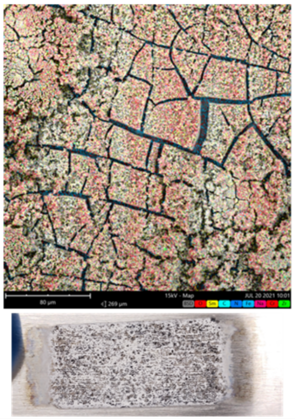
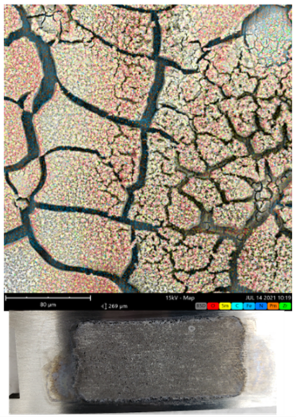
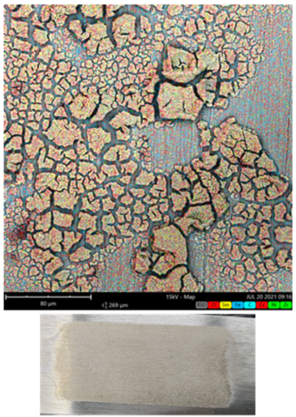
pH	1.20	3.12	7.71
Experiment condition: 0.1M Ammonium acetate 0.1M Ammonium nitrate 300ppm Sm 500 mA, CP mode			
Deposited mass(mg)	7.3	10.7	8
Sm deposition yield (%)	92	84	71
Adhesion test mass loss (mg)	0.9	3.2	2.2

Figure 3. SEM results from electrodeposition using mixture of 0.05 M ammonium acetate and 0.05 M ammonium nitrate with a pH of 1.31.

3.3 APPLIED CURRENT AND DEPOSITION TIME EFFECT WITH MIXTURE OF AMMONIUM ACETATE AND AMMONIUM NITRATE

Traditionally, the Cf source for CARIBU research is made using ammonium acetate performed at ~12 V, which occurs at a higher current density of ~138 mA/cm^2. This applied current is higher than the current applied in this work, and few studies have been performed to understand how the current and deposition time affects the properties of thin film. Three different currents for CP mode using the mixture of ammonium acetate and ammonium nitrate (i.e., 350, 500, 1,000 mA) were applied for different amounts of time. When electrodeposition was started at any current, the solution had a low pH (between 1 and 2) that turned into a turbid solution, especially above the anode (Figure 4, left panel); the solution then turned clear.

When 350 mA was applied to the system, a clear solution was achieved at 3 h. As the current was increased to 500 mA and 1 A, it took 1 h and 30 min, respectively. The yields at the point where the solution turned clear at each current (350 mA, 500 mA, and 1 A) were 53, 88, and 79%, respectively (Table 7). For the 500 mA test, three different timeframes were applied (1, 2, and 4 h); the test showed that 92% of deposition yield occurred at 2 h with the wider crack gap between deposits, but the substrate was still filled with $Sm(OH)_3$, and the substrate element is barely visible in the SEM images (Figure 5). As deposition times become longer than 4 h, black spots were visible on the plate, and those spots were concentrated with extra carbon.

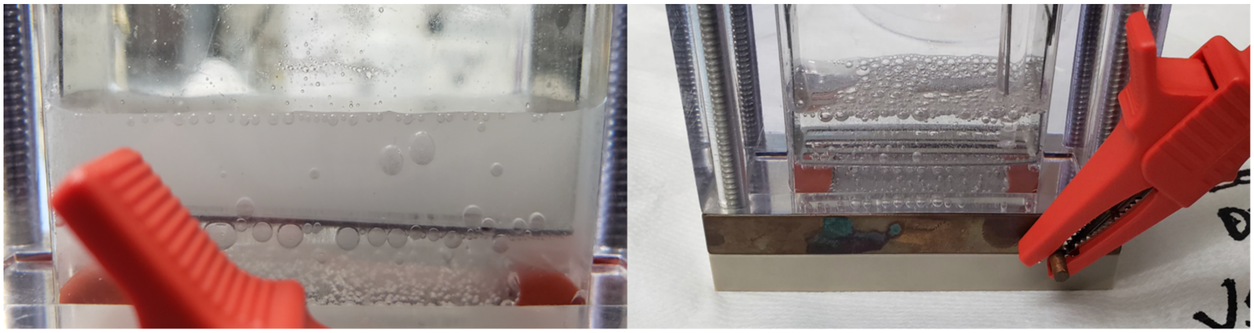


Figure 4. (left) Solution above the anode turned into a turbid solution 5 min after energizing the system. (right) After a varying number of hours, the solution turned clear.

Table 7. Current and final deposition yield for mixture of ammonium nitrate and ammonium acetate.

Electrolyte	Current	Current density (mA/cm^2)	Electrodeposition (time)	Final deposition yield (%)
Ammonium acetate and ammonium nitrate	350	48.65	3 h	53
	500	69.5	1 h	88
	500	69.5	2 h	95
	500	69.5	4 h	92
	1,000	139.04	30 min	79
	1,000	139.04	1 h	99

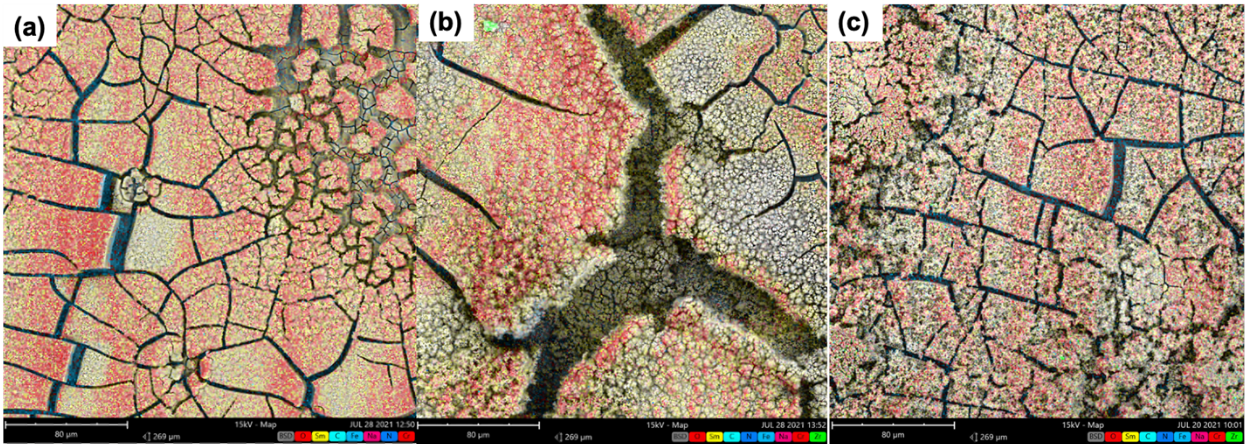
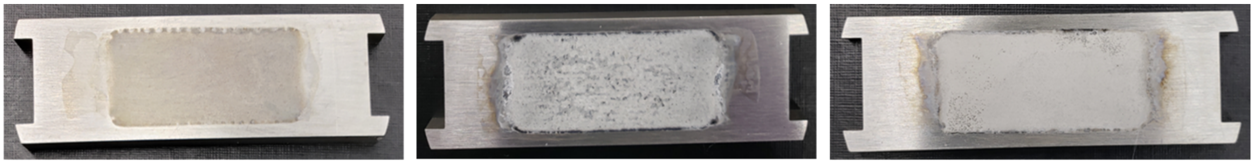


Figure 5. SEM-EDS images for thin film synthesized at 500 mA electrodeposition with different times: (a) 1 h (b) 2 h, and (c) 4 h. The scale of SEM images is identical.

3.4 CONCENTRATION OF SAMARIUM

Throughout this study, 300 ppm (~6 mg in 20 mL solution) of Sm was applied. Although 6 mg is more than enough to obtain targeted activity for the Cf target, a higher amount (1,000 ppm; 20 mg in 20 mL solution) of Sm was also tested to elucidate the relationship between deposition time and initial concentration of targeted deposit element. Result discussed in section 3.3, it showed that the highest yield (99%) was achieved at 1 A for 1 h of electrodeposition. For the higher Sm amounts using 1,000 ppm, 1 h deposition showed 87% yield, and the resulting solution was still turbid, suggesting the electrodeposition was not fully finished. As the applied time was increased to 2 h and 3 h, the yield was 97.8 and 99.5, respectively. The resulting plate from 2 h deposition (Figure 6, middle panel) showed lighter dotted spots on the substrate that were not fully covered by the Sm deposits, but as deposition time increased, the surface of the thin film was smoother with no voids. This result showed that the electrodeposition time is proportional to the target amount to achieve the same yield from different concentrations of initial target in the solution.



1A 1hr with 300 ppm Sm **1A, 2hr** with 1000ppm Sm **1A, 3hr** with 1000ppm Sm

Figure 6. Resulting thin film after electrodeposition with different amount of Sm and time for 1 A electrodeposition. (left) after 1 h with 300 ppm Sm, (middle) after 2 h with 1,000 ppm Sm, (right) after 3 h with 1,000 Sm.

3.5 EFFECT OF HCl

In previous electrodeposition to produce the CARIBU target, Cf was dissolved in an HCl solution. However, when chloride is present in the solution, it works as electrolyte; with electrolysis, chloride had a corrosive effect on stainless steel, which led to thick and nonadherent deposits [7].

In earlier studies, we showed that the mixture of ammonium nitrate and ammonium acetate showed good adhesion and yield, but no detailed studies have been conducted to examine how HCl affects thin-film structure or yield when it is synthesized using a mixture or ammonium acetate along. Thus, it is important to elucidate the role of HCl with ammonium acetate first. To understand the changes resulting from HCl, the resulting deposition yield, morphology, and atomic analysis over the thin film was compared by applying a corresponding amount of HCl in the solution. Two systems including 0.17 M ammonium acetate solution with and without 600 µL of HCl was used for electrodeposition with 500 mA for 3 h.

The surface of the substrate also showed discoloration; a thin layer of brown residue was observed on the side of the substrate after electrodeposition, which indicates corrosion of the stainless steel substrate. The morphology of the surface deposited using the solution containing HCl did not exhibit any differences, but carbon co-deposit in addition to Sm thin film was increased from 2% to 5%, and an additional 1% of chlorine was also observed. This suggests that HCl tends to decompose acetate and forms more carbon radicals, which can be deposited with the target material. In addition, the yield of deposition decreased from 76% to 54% when HCl was added to the system (Figure 7).

Figure 7. SEM images after electrodeposition with HCl.

3.6 EFFECT OF HEATING AFTER ELECTRODEPOSITION

In previous work, the target was air-dried after electrodeposition. To understand the effect of heat applied during this step, a hot plate was used (80°C) to compare the morphology and mud cracking before and after heating. For the thin film dried without heat treatment, oxygen was concentrated at the edge of each piece with slight lift-up. This suggests that air-drying may cause water to escape between the layer and the substrate, causing the lift-up and weak adhesion. However, this oxygen-concentrated edge disappeared after heating the plate to up to 80°C. More uniform distribution of all elements was observed as the size of the broken pieces increased. This can be explained by the fact that heating allows water to penetrate by increasing its solubility in the hydroxide structure and to evaporate at the surface of the thin film instead of escaping between the thin film and substrate. Thus, heat treatment during the air-drying step can improve adhesion of the thin film after electrodeposition.

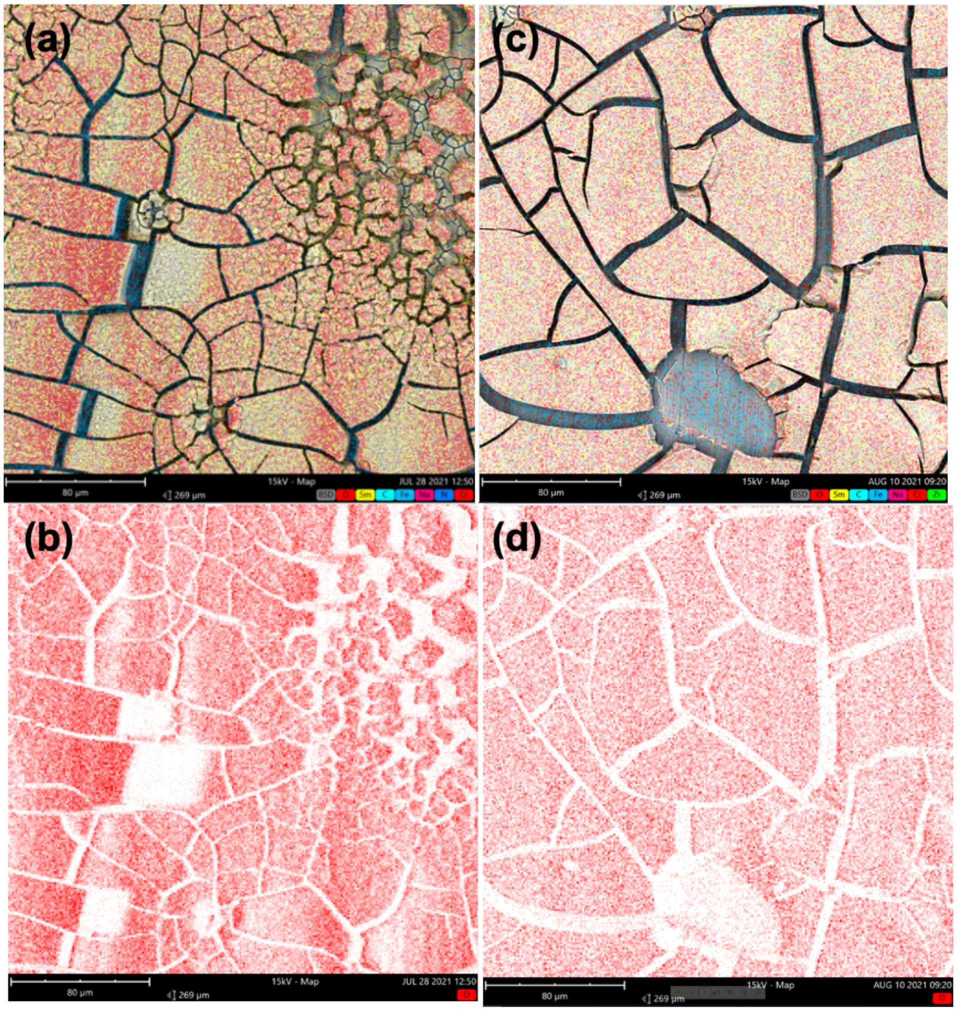


Figure 8. (a) SEM-EDS image for the obtained thin film after electrodeposition using ammonium acetate at 500 mA without heating, (b) oxygen elemental SEM-EDS image separated from (a), (c) SEM-EDS image for the obtained thin film after electrodeposition using ammonium acetate at 500 mA and heating at 80°C, and (d) oxygen elemental SEM-EDS image separated from (c).

4. CONCLUSIONS

In this work, Sm-based thin film were synthesized by electrodeposition as a surrogate of Cf under different electrodeposition conditions including pH, applied current, deposition time, initial concentration of Sm, the effect of additional acid (HCl) applied to the electrolytes, and the effect of heating after electrodeposition. Morphology of the resulting thin film, electrodeposition yield, and adhesion was observed, and electrodeposition conditions were optimized to obtain the best yield and most stable thin film over stainless steel substrate. Traditionally, ammonium acetate was only used as electrolyte for Cf electrodeposition in the presence of HCl, but it contained higher carbon deposits after Cf deposition and thicker deposition with weaker adhesion. The studies reported here have shown that ammonium acetate is required for electrodeposition to result in stronger adhesion, but the amount used for electrodeposition can be reduced by adding ammonium nitrate to the system. Ammonium nitrate also results in deposition of higher amounts of deposits on the substrate, but weaker adhesion was observed. In terms of deposition condition, the highest yield and strongest adhesion was observed at 1 A (current density ~139A/cm^2) for 1 h. The addition of HCl resulted in a complicated reaction with ammonium acetate as well as more carbon deposit and chlorine on the substrate, increasing the amount of unwanted element found with Cf during electrodeposition. The literature shows that HCl can react like ammonium chloride, causing unwanted thickening of the thin film and weak adhesion. Thus, it is important to minimize the usage of additional acid in the solution and ensure the reagent preparations are as pure as possible. This work also confirmed that the stability of thin film can be improved by air-drying the films at higher temperatures.

5. REFERENCES

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