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# Evaluation of three electrodeposition procedures for uranium, plutonium and americium



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## HIGHLIGHTS

- Four different electrodeposition procedures were performed and compared.
- Temperature changes were monitored with time and different currents.
- pH changes were monitored with time and different currents.
- Percentage deposition was monitored with time and different currents.

## ARTICLE INFO

Available online 27 November 2013

### Keywords:

Electrodeposition  
Actinides  
Alpha-particle spectrometry  
Uranium  
Plutonium  
Americium

## ABSTRACT

While both mass spectrometry and alpha-particle spectrometry have been widely used for measuring alpha activities, the former is preferred since many laboratories are not equipped with ICP-MS and MC-ICP-MS systems. In this study, three electrodeposition techniques using ammonium chloride, ammonium oxalate and ammonium sulphate as electrolyte solutions were applied and evaluated for the preparation of uranium, plutonium and americium sources for alpha-particle spectrometry. Changes in pH and temperature throughout the electrodeposition process were measured every 15 min, together with percentage deposition every 30 min. The percentage deposition in each method was checked at 300, 400 and 500 mA, and the optimised time and current were determined.

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## 1. Introduction

One of the methods for the determination of alpha-emitting nuclides in environmental samples involves radiochemical separation, followed by electrodeposition onto stainless-steel, silver, copper, nickel or platinum discs. It is essential to ensure a thin, uniform and clean deposit on the disc to obtain high-resolution alpha-particle spectra. While platinum discs can yield better-quality sources than other material discs, some of which partially dissolve and redeposit, the platinum discs are less commonly used because they are relatively expensive. A number of electrolytes have been employed, which include  $\text{NH}_4\text{Cl}/\text{HCl}$  (Mitchell, 1960),  $\text{NH}_4(\text{COO})_2/\text{HCl}$  (Bains, 1963),  $\text{NH}_4\text{Cl}/(\text{COOH})_2$  (Morgan, 1971),  $\text{NH}_4(\text{COO})_2/\text{NH}_4\text{Cl}$  (Puphal and Olsen, 1972),  $\text{NaHSO}_4/\text{Na}_2\text{SO}_4$  (Kressin, 1977),  $(\text{NH}_4)_2\text{SO}_4/\text{EDTA}$  (Lally and Eakins, 1978),  $\text{Na}_2\text{SO}_4/\text{H}_2\text{SO}_4$  (Hallstadius, 1984) and  $(\text{NH}_4)_2(\text{COO})_2-\text{H}_2\text{SO}_4-\text{HCl}$  (Liu et al., 1988). The most commonly used method for electrodepositing alpha-emitting nuclides is that introduced by Talvitie (1972), in

which sulphuric acid and ammonia gas are employed to adjust pH. The disc is usually positioned horizontally in these techniques, but it can also be placed vertically to prevent impurities settling on it (Mascanzoni, 1988). All of these techniques claim more than 90% recovery. However, only two techniques, one reported by Bains (1963) and the other by Morgan (1971), require currents lower than 500 mA. The other techniques require currents between 1 and 6 A and electrodeposition times ranging between 15 and 360 min. Electrodeposition also generates heat, and hence, a cooling jacket may be necessary when employing higher currents.

The best electrodeposition method should produce good-quality sources which leads to high deposition yields, save time, be sufficiently straightforward, and for simplicity, should not require any additional equipment such as a jacketed cell. Element of interest is also one of the factors determining the choice of a suitable method. Each method has advantages and disadvantages in terms of the current and electrodeposition time required. In this study, four electrodeposition techniques that use ammonium chloride (Mitchell, 1960), ammonium oxalate (Bains, 1963), sulphuric acid (Talvitie, 1972) and ammonium sulphate as the electrolyte solutions are tested and compared.

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## 2. Materials and methods

### 2.1. Equipment and reagents

#### Electrolyte solution

1. 20 g of ammonium chloride dissolved in 500 mL Milli-Q<sup>®</sup> water
2. 20 g of ammonium oxalate dissolved in 500 mL Milli-Q<sup>®</sup> water
3. 1.5% sulphuric acid adjusted to pH 2 (approx.) with ammonia solution
4. 15 g of ammonium sulphate dissolved in 500 mL Milli-Q<sup>®</sup> water

Milli-Q<sup>®</sup> water

Ammonia solution

10% Hydrochloric acid

10% Nitric acid

Traceable <sup>232</sup>U, <sup>241</sup>Am, and <sup>242</sup>Pu tracers supplied by AEA Technology, Harwell, UK.

EG&G Octete-PC alpha-particle spectrometer fitted with 450 mm<sup>2</sup> ruggedised low-background ion-implanted detectors.

The electrodeposition cells were prepared by placing new 27 mm stainless-steel discs and polythene inserts at the bottom and top of the cells, respectively, and then finally tightening the top. The cells were checked for leakage using distilled water. Platinum anodes with a spiral shaped end were cleaned by boiling in concentrated hydrochloric acid. Aliquots of <sup>232</sup>U (62 mBq), <sup>241</sup>Am (45 mBq) and <sup>242</sup>Pu (49 mBq) tracers were evaporated to near dryness in a 50 mL beaker and were retained for electro-deposition. A schematic of the electrodeposition cell used is shown in Fig. 1. A separate electrodeposition cell with no tracers added but only with the electrolyte solution was prepared and used for the measurement of temperature and pH. Temperature and pH changes were measured every 15 mins by inserting a calibrated thermometer and a pH metre into the cell during electrodeposition. Eight individual electrodeposition cells were prepared and used to measure deposition yield. The same aliquot of tracers together with the chosen electrolyte solution was added and the electrodeposition started. The distance between stainless-steel disc and platinum anodes was approximately 5 mm. The alpha-particle counting source was prepared by disconnecting the current to the first cell after 30 min. The second cell was stopped after 60 min and this procedure was repeated up to 240 min. Each stainless-steel disc was counted using alpha-particle spectrometers already calibrated for their counting efficiencies. Five millilitres of 2% nitric acid were added to the tracer beakers and the acid rinse solution was transferred to a scintillation vial. Fifteen millilitres of GoldStar cocktail were added to the scintillation vial and measured on a Wallac 1220 Liquid Scintillation Counter (LSC) to confirm that all the tracer activities had been

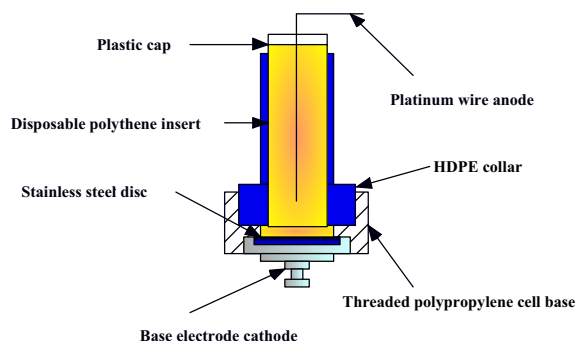


Fig. 1. Schematic of the EDP cell.

transferred to the electrodeposition cells. LSC results confirmed no residual activities in the rinse solution. For each method, three sets of eight electrodeposition cells were prepared and three different currents (300, 400 and 500 mA) were applied for each set. The power unit used for this experiment has 10 channels, and each channel can supply currents adjustable up to 550 mA.

### 2.2. Ammonium chloride technique

A solution of 10% hydrochloric acid (1 mL) was added to the tracer beaker, followed by 5 mL of 4% ammonium chloride solution. The resulting solution was transferred to the electrodeposition cell. The beaker was then rinsed with 5 mL of distilled water, and all the wash was again transferred to the electrodeposition cell. The cathode connection was attached, and the platinum anode was lowered into the solution. The electrodeposition cell was covered with a cap in order to reduce the loss of electrolyte solution by evaporation and spray that would cause cross-contamination. The current was adjusted and checked at regular intervals. At the end of electrodeposition, 4–5 drops of concentrated ammonia solution were added. Electrodeposition was continued for another 60 s, and then the power supply was disconnected. The cell was rinsed several times with distilled water, and the stainless-steel disc was removed. The disc was washed, dried on a hot plate and counted by alpha-particle spectrometry.

### 2.3. Ammonium oxalate technique

A solution of 10% hydrochloric acid (1 mL) was added to the tracer beaker, followed by 5 mL of 4% ammonium oxalate solution. The solution was transferred to the electrodeposition cell. The beaker was then rinsed with 5 mL of distilled water, and the wash was transferred to the electrodeposition cell. Electrodeposition was then performed as described for the ammonium chloride technique.

### 2.4. Sulphuric acid technique

The electrolyte solution was prepared in advance by adjusting 5% sulphuric acid to pH 2 (approx.) with ammonia solution. Five millilitres of the electrolyte solution were added to the beaker and then transferred to the electrodeposition cell. Another 5 mL of the electrolyte solution was added to rinse the beaker and transferred to the electrodeposition cell. Electrodeposition was then performed as described for the ammonium chloride technique.

### 2.5. Ammonium sulphate technique

A slightly modified version of the above technique was also tested. The electrolyte solution was prepared by dissolving 15 g of ammonium sulphate in 500 mL Milli-Q<sup>®</sup> water. One millilitre of 10% nitric acid was added to the tracer beaker, followed by 5 mL of the electrolyte solution. The solution was then transferred to the electrodeposition cell. Another 5 mL of the electrolyte solution was added to rinse the beaker and transferred to the electrodeposition cell. Electrodeposition was then performed as described for the above techniques.

## 3. Results and discussion

### 3.1. pH change

The pH changes observed in the case of all the electrodeposition techniques are shown in Fig. 2. No significant change in pH

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