



Evaluation of several electrolyte mixture-cathode material combinations in electrodeposition of americium radioisotopes for alpha-spectrometric measurements



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HIGHLIGHTS

- Three electrodeposition procedures were compared and optimized for Am electroplating.
- Three cathode materials were used in combination with selected electrolyte solutions.
- Optimal electrolyte-cathode combinations for high yield/resolution are provided.
- Sources with highest resolution were prepared on stainless steel and silver discs.
- Ammonium oxalate-chloride electrolyte gave highest yields and spectral resolution.

ARTICLE INFO

Keywords:

Alpha-particle spectrometry
Electrodeposition
Americium
Cathode material
Spectral resolution

ABSTRACT

Three different types of electrolytes, subsequently modified and adjusted, in combination with three cathode materials used as source backings were analysed for electrodeposition of americium isotopes for alpha-spectrometric measurements. The obtained results are discussed in terms of electrodeposition yield and source quality (source homogeneity and spectral resolution, FWHM). The optimal conditions for source preparation are provided.

1. Introduction

Preparation of alpha-emitting sources for alpha-spectrometric measurements is the final step of any radiochemical analysis. In order to obtain highly resolved peaks in the alpha spectrum, the source should be prepared as pure as possible, free of any stable or radioactive interferences, and mounted onto a stable solid backing. Otherwise the interpretation of alpha spectra would be questionable due to overlapping of poorly resolved peaks. Many different source preparation procedures for alpha-particle spectrometry have been developed over the last few decades, such as direct evaporation (Denecke et al., 2000; Štastná et al., 2010), electro-spray (Bruninx and Rudstam, 1961; Hernegger and Chalupka, 1989), vacuum sublimation (Lally and Glover, 1984), micro-coprecipitation (Kikunaga et al., 2009; Nilsson et al., 2001; Sill and Williams, 1981; Sill, 1987), electrodeposition (Hallstadius, 1984; Janda et al., 2010; Lee and Pimpl, 1999; Talvitie, 1972), selectively adsorbing thin films (Surbeck, 2000), etc. The first

three mentioned methods are primarily used in metrology, while others are more suitable for the preparation of alpha sources in determination of low-level activities of alpha emitters in environmental samples. An overview of preparation of radioactive sources for radionuclide metrology is summarized elsewhere (Sibbens and Altitzoglou, 2007; Crespo, 2012).

After introducing electrodeposition of alpha-emitting radionuclides as a semi-quantitative source preparation technique in mid-20th century many studies were focused on actinides' mounting on different cathode surfaces either from aqueous solutions of electrolytes, such as $\text{NH}_4\text{Cl-HCl}$ (Mitchell, 1960; Oh et al., 2014), $(\text{NH}_4)_2\text{SO}_4\text{-H}_2\text{SO}_4$ (Oh et al., 2014; Talvitie, 1972), $(\text{NH}_4)_2\text{C}_2\text{O}_4\text{-NH}_4\text{Cl}$ (Mirashi and Aggarwal, 2009; Puphal and Olsen, 1972), $(\text{NH}_4)_2\text{C}_2\text{O}_4\text{-NH}_4\text{Cl-NH}_2\text{OH-HCl-DTPA}$ (Puphal et al., 1984), $\text{NaHSO}_4\text{-Na}_2\text{SO}_4$ (Glover et al., 1998; Kressin, 1977; Plionis et al., 2009; Trdin et al., 2012), $\text{Na}_2\text{SO}_4\text{-H}_2\text{SO}_4$ (Beesley et al., 2009; Hallstadius, 1984; Méndez et al., 2010), $(\text{NH}_4)_2\text{C}_2\text{O}_4\text{-H}_2\text{SO}_4\text{-HCl}$ (Liu et al., 1988), $(\text{NH}_4)_2\text{C}_2\text{O}_4\text{-(NH}_4)_2\text{SO}_4\text{-(NH}_3\text{OH)}_2\text{SO}_4\text{-}$

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DTPA (Janda et al., 2010; Lee and Pimpl, 1999), $(\text{NH}_4)_2\text{C}_2\text{O}_4\text{-HCl}$ (Klemenčič and Benedik, 2010; Oh et al., 2014), or from organic solutions like isopropanol (Jobbágy et al., 2013; Parker and Falk, 1962), propanol, ethanol, acetone (Parker et al., 1964), DMSO (Handley and Cooper, 1969), ammonium acetate (Torrico et al., 2015), ionic liquids (Novikov et al., 2014; Sankhe et al., 2015), etc., also known as molecular plating. Electrodeposition is certainly the most extensively studied source preparation method in alpha-particle spectrometry due to several advantages: i) it allows almost quantitative actinide deposition from the electrolyte solution under optimized conditions of the proposed method, depending on the radionuclide of interest; ii) it is relatively simple and can be performed in any radioanalytical laboratory; iii) prepared sources are thin, quite homogenous and chemically stable, with as low as possible self-absorption of the emitted radiation; iv) spectral resolution is superior to all above mentioned methods used for alpha source preparation. On the other hand, there are a few disadvantages as electrodeposition can be rather time-consuming (which is a serious drawback when fast source preparation is required) and the materials used as cathodes can be quite expensive (e.g. platinum). Furthermore, higher applied currents can produce high temperatures, which can consequently (if the cell is not introduced in the cold water bath) cause spatter and potential loss of electrolyte solution. In such cases the solution pH (which is one of the most important parameters in source preparation by electrodeposition) will be changed and can consequently degrade the quality of the prepared sources. Therefore, optimization of the method in order to obtain uniform, high quality alpha-sources with minimum surface degradation, in a short period of time, preferably under low current conditions, using relatively inexpensive materials with as minimal consumption of chemicals as possible, and easy-to-decontaminate cell equipment is more than necessary.

With the aim to obtain quantitative sources of the highest quality, the present study was focused on investigation and evaluation of several electrolyte solution-cathode material combinations for the preparation of americium sources for alpha-spectrometric measurements. Electrolytes investigated in this study were chosen based on the frequency of their usage in the literature and were subsequently slightly modified either by concentration or by composition (addition of chelating agents to some solutions to prevent polymerization and hydrolysis during electrodeposition, reduction in components' concentrations to simplify the preparation step due to relatively poor solubility of some components). Three main electrolyte types chosen were sodium sulphate (Hallstadius, 1984), ammonium oxalate-hydrochloric acid mixture (Klemenčič and Benedik, 2010), and modified sulphate-oxalate mixtures based on the study by Lee and Pimpl (1999), in combination with three cathode materials: polished stainless steel, copper and silver discs as source backings (Trdin et al., 2012). For this purpose, the mixture of ^{241}Am and ^{243}Am was selected due to several properties: i) relatively long half-lives of both radioisotopes ($T_{1/2} (^{241}\text{Am}) = 432.6 \text{ y}$; $T_{1/2} (^{243}\text{Am}) = 7.4 \times 10^3 \text{ y}$), ii) alpha-particle energies of 5.5 MeV for ^{241}Am and 5.3 MeV for ^{243}Am being close enough to be resolved if the prepared sources for alpha-spectrometric measurement are of high quality, iii) no separation from radioactive progeny needed prior to electrodeposition, and iv) determination of ^{241}Am with addition of ^{243}Am as a yield tracer is a common practice. The results are presented in terms of electrodeposition yield, source homogeneity and spectrum quality (FWHM).

2. Materials and methods

2.1. Chemicals and reagents

The ^{241}Am and ^{243}Am tracer solutions used in the study were

Table 1

Electrolyte solution mixtures used in this study, with according components' concentrations (mol/dm^3) for electrodeposition of $^{241}\text{Am}/^{243}\text{Am}$; pH pre-adjusted to 1.7.

Electrolyte designation	c (mol/dm^3)				
	$(\text{NH}_4)_2\text{SO}_4$	$(\text{NH}_3\text{OH})_2\text{SO}_4$	$(\text{NH}_4)_2\text{C}_2\text{O}_4$	DTPA	EDTA
Mix	0.4	0.1	0.2	–	–
Mix 1:1	0.2	0.05	0.1	–	–
Mix 1:4	0.1	0.025	0.05	–	–
Mix-D	0.4	0.1	0.2	0.005	–
Mix-D 1:1	0.2	0.05	0.1	0.0025	–
Mix-D 1:4	0.1	0.025	0.05	0.0013	–
Mix-E	0.4	0.1	0.2	–	0.005
Mix-E 1:1	0.2	0.05	0.1	–	0.0025
Mix-E 1:4	0.1	0.025	0.05	–	0.0013

obtained from National Institute of Standards & Technology (NIST, Maryland, USA). The working mixture of tracer solutions was prepared by gravimetric dilution of standards to approximately 1 Bq/g of each radioisotope. All other chemicals and reagents used were of analytical grade. The electrolyte solutions for electrodeposition were prepared by dissolution of salts in deionized water, and the pH was adjusted by concentrated H_2SO_4 or concentrated NH_4OH addition if necessary. The composition of the electrolytes, along with their concentrations are given in Tables 1 and 3.

2.2. Alpha source preparation

Sources of ^{241}Am and ^{243}Am were prepared by electrodeposition at Ruđer Bošković Institute. The cell used for electrodeposition was designed and manufactured at Jožef Stefan Institute in Ljubljana, Slovenia, and is shown in Fig. 1. The electroplating cell was made of polypropylene (to prevent adsorption of actinides onto the cell walls), containing a closing lid and anode insertion opening at the top and a metal backing for the cathode at the bottom. Polished stainless steel, copper and silver discs, with an active area of 2.01 cm^2 , were used as



Fig. 1. Photography of the polypropylene electroplating cell manufactured at Jožef Stefan Institute, Ljubljana, Slovenia.

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