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Electrodeposition of selected alpha-emitting nuclides from ammonium acetate electrolyte

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Abstract

The experimentally optimal conditions of the electrodeposition of selected alpha particle-emitting radionuclides, including ^{208}Po, ^{226}Ra, ^{228}Th, ^{238}U, ^{239}Pu, ^{241}Am and ^{243,244}Cm, with ammonium acetate electrolyte have been determined. This simple method could be used for the determination of the most important actinides in radiological waste and could be applicable to waste treatment. In addition, this method could be used for radium determination instead of the traditional radon emanation technique, which requires approximately 30 days.

1. Introduction

Reliable determinations of the alpha-emitting radionuclides require well-prepared sources of activity. Although these sources may be prepared by different techniques, electrodeposition has been widely used to obtain undegraded energy spectra and good chemical yields. Different electrolytes could be utilized for the electrodeposition of different elements. Ammonium sulfate has been commonly used for electrodepositing some actinides, such as Th, U and Pu [1], since it was proposed [2]. Other electrolytes, such as ammonium chloride–ammonium oxalate [3], had been proposed for electrodepositing various alpha-emitting nuclides. For our interest, some alpha-emitting radionuclides could not be satisfactorily electrodeposited with ammonium sulfate or mixed oxalate–chloride electrolytes. The purpose of this study is to find a simple and practical method to electrodeposit and determine selected alpha-emitting radionuclide activities in the environmental samples. These radionuclides include the isotopes of Po, Ra, Th, U, Pu, Am, and Cm.

2. Experimental details

The standard reference materials of the radionuclides, including ^{208}Po, ^{226}Ra, ^{228}Th, ^{238}U, ^{239}Pu, ^{241}Am, and ^{243,244}Cm, used in this study were obtained from the US Environmental Protection Agency, the Environmental Monitoring Systems Laboratory, Las Vegas, NV, and an NIST-traceable radioactive source manufacturer.

Disposable electrodeposition cells were constructed from 20 ml, linear-polyethylene, liquid scintillation vials. The anode was a platinum–iridium disk having six perforations and attached at the center to a platinum–iridium rod. A piece of stainless steel planchet served as the electrodeposition base and contacted the cathode.

In our laboratory, some conditions were designed to be fixed for the electrodeposition study. They included: (i) electrode spacing – 12 mm, (ii) ammonium acetate electrolyte volume – 10 ml, (iii) electrodeposition potential – 12 V, and (iv) current – 400 mA (if possible). However, a 400 mA current was adjusted or the maximal current was used if 400 mA could not be reached. The following variables were designed to be changed in order to search for the optimal conditions: (i) concentrations of ammonium acetate electrolyte – 0.175 M, 0.35 M, and 0.70 M, (ii) pH-values – 1–9, and (iii) electrodeposition periods – 1–4 h.

The procedure included the following steps: (i) take 10 ml of ammonium acetate solution with a known concentration and a pH-value; (ii) spike it with a known amount of radioactivity from an EPA- or NIST-traceable solution; (iii) adjust electrode spacing and current; (iv) electrodeposit for a chosen period of time; (v) count the source with an alpha spectrometer with a known counting efficiency; (vi) calculate the chemical yield and its associated uncertainty; and (vii) determine the

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TABLE 1. Summary of optimal conditions for electrodeposition of individual elements and the averaged yields

Element	Concentration (M)	pH-value	Time (h)	Yield (%)[a]
Po	0.175	1	2	90
Ra	0.35	8	4	78
Th	0.35	1	2	95
U	0.175	1–4	2	100
Pu	0.35	1–4	3	100
Am	0.175	1–4	3	90
Cm	0.175	1–2	2	100

[a]There is a less than 5% relative uncertainty (95% confidence level) associated with the yields.

optimal conditions of electrodeposition for individual elements and re-examine the chemical yields at those conditions.

3. Results and discussion

Among three different ammonium acetate concentrations, 0.175 M, 0.35 M, and 0.70 M, the experimental results show that higher chemical yields could be obtained from lower concentrations for all radionuclides. With a 0.70 M ammonium acetate solution, a 60–70% chemical yield could be reached at pH 1 for Po within 1 h, at pH 4 for Pu within 4 h, and at pH 1 for Am and Cm within 4 h also. An approximately 85% chemical yield could be reached for U at pH 1 within 1 h, and the chemical yield would decrease rapidly to 20% or less at pH 5 as pH values increased. For Ra and Th, generally a chemical yield greater than 50% would be difficult to obtain for pH 1–9 within 4 h.

For the 0.175 M concentration of ammonium acetate, an approximately 90% chemical yield from Po and Th could be obtained at pH 1 after 2 h. Ra could be electrodeposited with an approximately 70% chemical yield at pH 5 within 4 h. U, Pu, Am, and Cm show similar patterns and could be electrodeposited with fairly good chemical yields within a pH range of 1–4. For pH-values over 5, all chemical yields would decrease rapidly to approximately 20% or less.

For the 0.35 M concentration, chemical yields of U, Pu, Am, and Cm show similar variations as for 0.175 M. Lower chemical yields from Po and Th could be obtained than for 0.175 M at pH 1. At higher pH values, approximately 50% chemical yields could be obtained with 0.35 M, which are higher than those with 0.175 M. Ra could be electrodeposited with an approximately 78% yield with 0.35 M at pH 8 within 4 h. This condition could be applicable to the Ra determination and for obtaining the result within 1–2 days. The commonly used radon emanation technique [4] requires approximately 30 days.

The optimal conditions for the electrodeposition of those selected elements and the averaged yields from duplicate analyses are summarized in Table 1. There is approximately 3–4% relative uncertainty associated with most yields. The variations of the chemical yields from those radionuclides indicate that both Po and Th could be electrodeposited very efficiently at pH 1 only. There is a larger pH range, pH 1–3 or 4, for U, Pu, Am, and Cm, which is convenient for laboratory work. Only Ra could be electrodeposited in a basic condition with pH 8. Electrodeposition of mixtures of alpha-particle emitters is under investigation.

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