

Mutual separation of U, Np, Pu and Am with ion exchange resin in chloride media

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1. Introduction

Numerous methods have been investigated for separating uranium and transuranium elements. The analytical methods for the determination of Am, Pu, Np and U from a nitric acid solution by an anion exchange resin can be found in literature[1-6]. Chilton[1] reported on the isolation of plutonium in chloride media by an anion exchange resin and N. Shinohara and N. Kohno[3] also studied the chemical separation of Np by an anion exchange method from a neptunium oxide material and the analytical method for Np and Am in irradiated fuels based on ion exchange and oxidation. Although many researchers have studied transuranium elements separation by an anion exchange resin from nitric acid solutions, but it is still limited related to experimental results in hydrochloric acid solutions. In this paper, we describe briefly the results about the mutual separation and purification of the Am, Pu, Np and U in a chloride media.

2. Experimental

2.1 Preparation of anion exchange resin column

The column was an integral unit consisting of three parts, a plastic funnel, 60 mm(L.) by 4.7 mm(I.D.) plastic syringe tube and a one-way plastic stopcock. A disk plug instead of glass wool was used in the bottom and in the upper part of the resin bed. Bio-rad AG 1x4 (100-200 mesh chloride form, analytical grade) was soaked in 1M HCl for 24 hour and the resin was continuously washed with distilled water until the wash was neutral. The column was packed with washed resin carefully. Before use, the resin column was washed with 10ml of 9M HCl-0.1M HNO₃.

2.2 Tracer solution

The tracer solution was made with ²⁴¹Am, ²³⁹Pu, ²³⁷Np and ²³³U nuclides. The individual activity of ²⁴¹Am, ²³⁹Pu, ²³⁷Np were approximately 80 Bq and ²³³U is much more than 80 Bq in 10ml of 0.1M HCl. For the control of the oxidation state of the nuclides in the tracer solution, 1 ml of 1M NH₂OH-HCl was added into the tracer solution and dried under an infrared lamp and then dissolved with 2ml of 9M HCl-0.1M HNO₃. Finally we obtained the Am(III), Pu(IV), Np(IV) and U(VI) oxidation state.

2.3 column operations procedure

Resin as a slurry in water is added to the plastic column until a resin bed about of 5cm in length (ca. 0.8 ml) is formed. The column is placed in a column supporter and it is then pretreated with 10ml of 9M HCl-0.1M HNO₃ and 0.5ml of the tracer solution is added.

Flow rate is controlled to about 0.42 ml/min. After the sample has passed into the resin bed, 3ml of 9M HCl-0.1M HNO₃ is added at the top of the bed. The tracer solution has passed into the bed and Am is eluted by passing it through of 6ml of 9M HCl-0.1M HNO₃. The effluent is collected in a plastic vial for the analysis. Pu is eluted by passing 9ml of 9M HCl-0.1M HI through the resin bed. The effluent is collected in a plastic vial. Np is removed with 9ml of 4M HCl solution by passing it through the resin bed. The effluent collected in a plastic vial. Finally, the uranium is removed with 5ml of 0.1M HCl by passing it through the resin bed. The effluent collected in plastic vial. All the effluents is collected in plastic vials were analyzed by alpha spectrometry or LSC measurements.

2.4 Electrodeposition

An electrodeposition system consisting of a deposition cell, Pt electrode, planchet, cell holder was installed in connection with a constant power supplier. For the pretreatment before the electrodeposition, the samples were dried with a 0.1MNaHSO₄-0.53MNa₂SO₄ buffer solution with an infrared lamp. Optimum condition of an electrodeposition was 1200 mA and 60 minutes in a 0.1MNaHSO₄-0.53MNa₂SO₄ buffer solution. This method was applied to the determination of the ²⁴¹Am, ²³⁹Pu, ²³⁷Np and ²³³U with an alpha spectrometry.

2.5 Alpha spectrometry and LSC Measurements

A multi-channel pulse height analysis system with a 300 mm² silicon surface-barrier alpha counter (EG&G/ORTEC Co., Alpha-King Module) was used to analyze the alpha spectra of ²⁴¹Am, ²³⁹Pu, ²³⁷Np, and ²³³U. Liquid scintillation analyzer (Packard model 2500TR/AB) was used to analysis the total activity of the elution samples.

3. Results and discussion

3.1 principle concept of sequential separation of Am, Pu, Np and U in chloride media

Distribution coefficients of the actinides on an anion exchange resins in a HCl solution and the flow sheet for the Am, Pu, Np and U separation by an anion exchange chromatographic technique are shown in Fig. 1 and Fig. 2.

3.2 Elution profile of Am, Pu, Np and U

Fig. 3 is the elution profile of Am(III), Pu(III), Np(IV) and U(VI) in the anion exchange separation column. A mixture of ²⁴¹Am, ²³⁹Pu, ²³⁷Np and ²³³U in 10ml of 9M HCl-0.1M HNO₃ was prepared as a stock solution. A

0.5ml aliquot of the stock solution was added to an anion exchange resin column, pretreated with a 9M HCl-0.1M HNO₃ solution. After the sample had drained into the resin bed, an elution was carried out with 9ml of 9M HCl-0.1M HNO₃ which was sufficient enough to removed the Am(III), but Pu(IV), Np(IV) and U(VI) remained adsorbed in the anion exchange resin column.

Distribution coefficients of actinides on anion exchange resins in HCl							
Metal	Molarity of HCl						
	1	2	4	6	8	10	12
Am(III)	←		No adsorption		→		
U(III)	←		No adsorption		→		
U(IV)	<1	<1	<1	1	50	>100	>100
U(VI)	~2	~10	~180	800	1000	1000	800
Np(III)	No adsorption						→
Np(IV)	<1	<1	<1	20	700	>1000	>1000
Np(VI)	<1	<1	2	200	800	>1000	>1000
Np(VI)	Strongly adsorbed from HCl > 6M						→
Pu(III)	No adsorption						→
Pu(IV)	<1	<1	<1	1	50	>100	>100
Pu(VI)	Strongly adsorbed from HCl > 6M						→

Fig. 1 Distribution coefficients of actinides on anion exchange resins in HCl solution

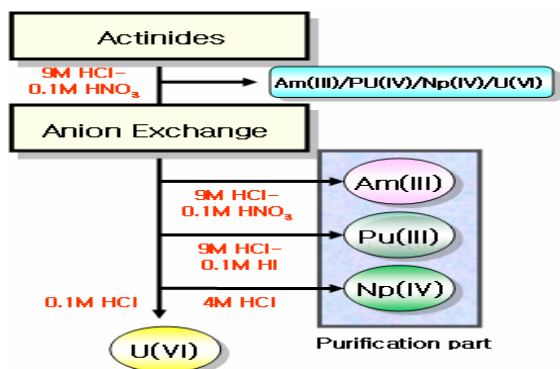


Fig. 2 Flow sheet for separation Am Pu, Np, and U

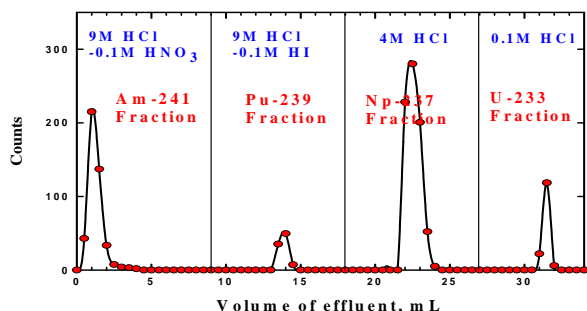


Fig. 3 Elution profile of Am, Pu, Np and U on a column packed with Bio-rad (100-200 mesh)

Plutonium was removed with a 9ml 9M HCl-0.1M HI solution. Pu(IV) was removed with a 9ml 4M HCl solution and uranium with a 5ml 0.1M HCl solution. All of the eluent was added by 0.5ml in one time. When the

eluent has passed into the bed, an additional 0.5 ml of the eluent is added and collected in a scintillation plastic vial. The eluents were analyzed by the LSC. In the elution of Pu, the HCl concentration should not exceed 10 M, otherwise a partial reduction of U(VI) by iodide can occur and the Np fraction can become contaminated with U.

3.3 Mutual separation of Am, Pu, Np and U in HCl

The mutual separation of ²⁴¹Am, ²³⁹Pu, ²³⁷Np and ²³³U was carried out by the anion exchange chromatographic method according to the flow sheet in Fig. 2. The procedure of the mutual separation is similar to that of the elution profile of Am, Pu, Np and U. The plutonium fraction and Neptunium fraction need more repeated purifications as described in the same procedure of the mutual separation. Alpha spectra the of americium fraction, plutonium fraction, neptunium fraction and uranium fraction are shown in Fig 4(A), Fig 4(B), Fig 4(C) and Fig 4(D), respectively.

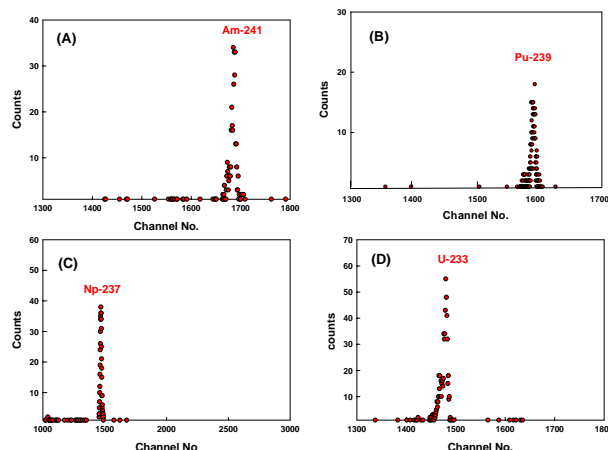


Fig. 4 Alpha spectra of Am fraction(A), Pu fraction(B), Np fraction (C) and U fraction (D)

4. Conclusion

This study could be used as a rapid method for the mutual separation of actinide elements, especially Am, Pu, Np, and U. The developed procedure was applied to a nuclear waste sample analysis which contained the Am, Pu, Np, and U nuclide. This method also can be applied to the purification of actinide elements.

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