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Separation of neptunium, plutonium, americium and thorium by anion exchange

The separation of Np(IV) and Pu(III) by anion exchange from nitric acid[1–3] as well as Pu(IV), U(VI) and Am(III)[4] has been reported. Because Np(VI), like U(VI), does not form strong anionic complexes, the separation of Np(VI) and Pu(IV) can be carried out by using the agents which hold the neptunium in hexavalent and plutonium in tetravalent state. The difference in the distribution coefficients for Pu(IV) and Th(IV) in nitric acid solutions[5–7] gives the possibility for their separation, too.

This paper describes investigations carried out on the separation of Np(VI) and Th(IV) from Pu(IV) by anion exchange from nitric acid solution. Americium has also been included in the separation scheme.

TABLE I

SEPARATION OF Am(III), Np(VI), Th(IV) AND Pu(IV)

Column: 15 cm × 0.11 cm². Exchanger: Dowex 1 X4, 100–200 mesh. Temperature: 50°. Flow rate: 1 ml/min/cm².

Element	*No. of samples*	*Percent of elements eluted in fractions*			
		7.2 M HNO_3 (5 ml)	*7.2 M HNO_3 (10 ml)*	*4.0 M HNO_3 (25 ml)*	*0.35 M HNO_3 (10 ml)*
^{241}Am	9	100.1 ± 1.2	⩽0.01	[a]	[a]
^{237}Np	9	[a]	99.3 ± 1.4	⩽0.05	⩽2
^{234}Th	9	~0.02	~0.04	100.3 ± 1.3	[a]
^{239}Pu	9	[a]	⩽0.06	⩽0.09	100.0 ± 0.7

[a] The activity of these radionuclides could not be detected. About 30% of ^{233}Pa elutes in the neptunium fraction and 70% in thorium fraction.

Experimental

Reagents and radionuclides. The exchanger Dowex 1 X4 (100–200 mesh) in nitrate form was used.

The purification and the preparation of the Am and Pu solutions were carried out as described previously[4]. The isolation of ^{234}Th from $UO_2(NO_3)_2 \cdot 6H_2O$ was done as described earlier[8]. ^{237}Np was in the form of nitrate in HNO_3. The amount of ^{241}Am and ^{234}Th used were at the tracer levels. From $4.2 \cdot 10^{-3}$ to 0.45 mg of ^{237}Np and $1.6 \cdot 10^{-3}$ to 1.7 mg of ^{239}Pu were used. All chemicals were of p.a. purity.

Determination of radionuclides. ^{239}Pu, ^{237}Np and ^{241}Am were measured with a Tracerlab Model P-12 alpha scintillation detector. The ^{234}Th was measured with a Nuclear Chicago GM counter. In the presence of the alpha-emitters mentioned, it is possible to measure ^{234}Th over an absorber of 8 mg/cm² aluminium. However, because of the presence of beta-emitters of ^{233}Pa (which is produced by alpha decay of ^{237}Np), ^{234}Th–^{234}Pa were measured over an absorber of 99.6 mg/cm² of aluminium.

The purity of ^{239}Pu, ^{237}Np and ^{241}Am was checked, *i.e.* the content of each one in the other two was determined by alpha spectrometric analysis using a Tracerlab,

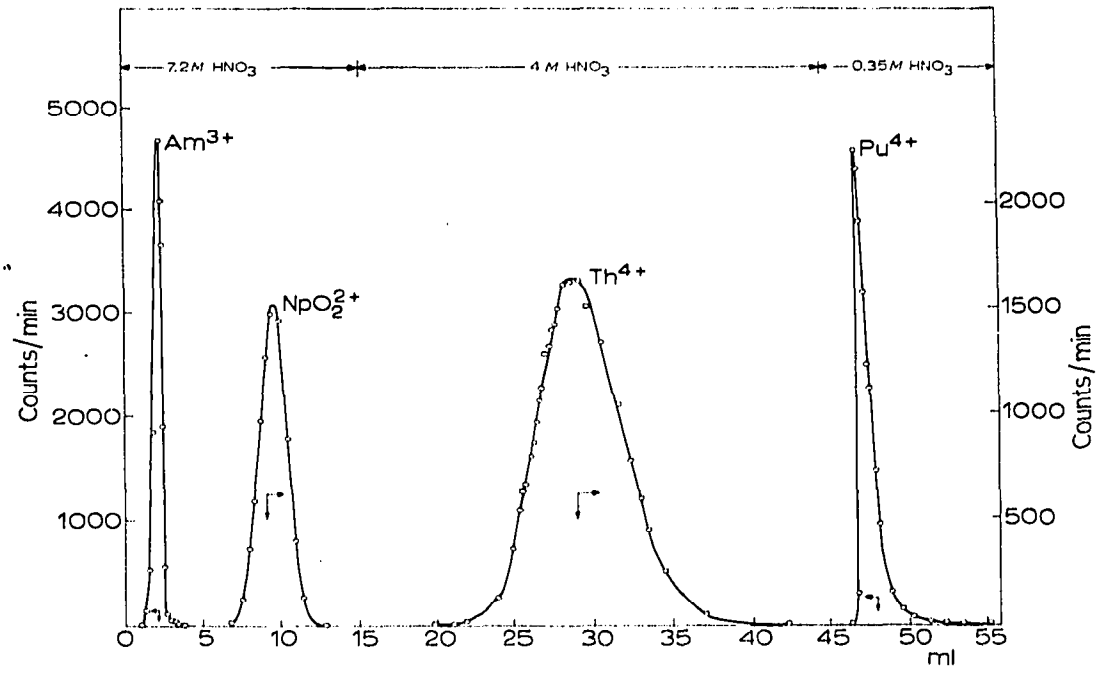

Fig. 1. Separation of Am(III), Np(VI), Th(IV) and Pu(IV) (tracer amounts of Am and Th, 40 μg of ^{237}Np and 1.5 μg of ^{239}Pu) on Dowex 1 X4(NO_3^-) 100–200 mesh, at 50°. Column: 15 cm × 0.11 cm^2. Flow rate: 1 ml/min/cm^2.

Model RLD-1, Frisch Grid chamber with a Model 20623 RCL 256 channel pulse height analyser.

Check of the valency state of Np in nitric acid solutions was made spectrophotometrically[9,10] with a Unicam SP-500 spectrophotometer.

Sample preparation. The solution of the radionuclides in 1–2 *M* HNO_3 were treated with $NH_2OH \cdot HCl$ (0.025–0.05 *M*) for 15–30 min and then $NaNO_2$ (0.05–0.10 *M*) was added. After 30 min the solution is adjusted to 7.2 *M* HNO_3, thermostated for 30 min at 50° and transferred to the column, pretreated with a 7.2 *M* HNO_3 and thermostated at 50°. The amount of sample transferred to the column (15 cm × 0.11 cm^2) should not exceed 1.6 ml.

Results and discussion

It was reported that Np(V) in nitric acid higher than 3 *M* in the presence of NO_2^- oxidises to Np(VI)[11]. Under the same conditions Pu exists as Pu(IV). The anion-exchange behaviour of neptunium in strong nitric acid containing $NaNO_2$ was identical to the behaviour of Np(VI) which was prepared by oxidising Np with Ag^{2+} or Ce^{4+}. Spectrophotometric analysis of a neptunium solution in 4–7 *M* HNO_3 containing 0.1 *M* $NaNO_2$ have shown that Np(V), at 50°, was oxidised to Np(VI).

Np(VI) elutes from the anion-exchange column approximately like U(VI). The distribution coefficients determined by the column method[12] for 7.2 *M* HNO_3 at 50° for Np(VI) and U(VI) are 10.4 and 7.0, respectively.

On the basis of these data as well as on the basis of the data reported[4], the separation of Am(III), Np(VI) and Pu(IV) was carried out (Fig. 1). The elution of

Th(IV) from the column has been achieved with 4 M HNO_3. Results are shown in Table I.

As it is seen from Table I satisfactory separation of given radionuclides is obtained. However, when a sample contained about 0.2 M H_2SO_4, only 90% of ^{237}Np was recovered.

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