

Separation of americium in marine samples by extraction chromatography. A new proposal for the americium-rare earth separation.

A.DELLE SITE, V.MARCHIONNI
CNEN, FUNZ-CSN Casaccia, Rome, Italy

Abstract

The procedure for the determination of americium-241 in sediment samples requires thirteen working days, four of which are necessary for the americium-rare earth separation step by anion exchange. The separation can also be achieved by extraction chromatography, using Aliquat-336 in xylene supported on microporous polyethylene. This method, which requires only one day, assures a good purification of americium and a satisfactory recovery.

Résumé

La méthode de détermination de l'américium-241 dans les sédiments exige treize jours de travail, dont quatre sont nécessaires pour la séparation américium-terres rares par échange anionique. Cette séparation peut être aussi accomplie par chromatographie d'extraction, en utilisant Aliquat-336 en xylène, supporté sur polyéthylène microporeux. Ce système, qui exige un jour seulement pour une séparation complète, assure un bon degré de purification de l'américium et une récupération satisfaisante.

The procedures for the determination of americium-241 in environmental samples, and especially in sediment and soil, involve a series of separation steps which have the main aims: a) to separate americium from the matrix and from iron; b) to achieve a good decontamination from thorium, uranium and plutonium, which interfere in the alpha spectrum; c) to separate americium from rare earths, which are electroplated at high yield and reduce the americium counting efficiency.

The method adopted at CNEN (1) for the analysis of americium in sediments is reported in Fig. 1. Decontamination from thorium, uranium, and plutonium is obtained by extraction chromatography using 0.3M tri-n-octylphosphine oxide (TOPO) in cyclohexane supported on microporous polyethylene (Microthene-710). Two Ca-oxalate co-precipitation steps are used to separate transuranics from iron and a chromatographic separation of residual

thorium and plutonium is achieved with 30% di-(2-ethylhexyl)phosphoric acid (HDEHP) in toluene supported on Microthene. The final purification of americium is obtained by a two step anion-exchange separation in thiocyanate medium.

This chemical procedure requires a total of 13 working days (Fig. 1). The most time-consuming steps are the co-precipitation and the anion-exchange separation, which require 3 and 4 days respectively. Therefore it was decided to investigate the possibility to simplify the procedure, starting from the americium-rare earth separation step.

The methods of separation of transplutonium elements were reviewed by Stary (2) in 1966. Solvent extraction with 30% Aliquat-336 (tricaprylmethylammonium salt) in xylene is one of the most efficient methods, which allows a lanthanide-actinide (III) group separation factor higher than 100. Moreover the same organic phase can be favourably employed in column extraction chromatography to obtain good americium-rare earth separations (3). The main advantage of this technique over the solvent extraction is the possibility to avoid time-consuming phase separation. A series of experiments were carried out to test the application of this system to the determination of americium in environmental samples. The feeding solution was prepared by addition of 10^4 dpm (β) Eu-152,154, 10^4 dpm (α) Am-241, and 50 mg neodymium to 1 ml ammonium thiocyanate. Elution was carried out until the first fraction of americium emerged. Then 1M hydrochloric acid was passed through to collect americium very rapidly. An eluant composition of 0.02 M sulphuric acid-2M ammonium thiocyanate gave a satisfactory separation (Fig. 2) at a flow-rate of 0.25 ml/min. The change in flow-rate from 0.25 ml/min to 1 ml/min resulted in a worse exchange equilibrium, which anticipated the elution of small amounts of americium. However, when the flow-rate ranged between 0.25 ml/min and 0.5 ml/min, the americium recovery was about 95%, the decontamination factor from europium was about 1000, and the time required for the complete separation one day.

References

- 1) A. Delle Site, V. Marchionni, Methods used at CNEN for the analysis of plutonium and americium-241 in marine samples, Meeting on Behaviour of Transuranics in the Aquatic Environment and Sediment-Water Exchanges; Techniques for Identifying Speciation, Ispra, Italy, 24-28 March, 1980.
- 2) J. Stary, Separation of transplutonium elements, *Talanta*, 13, 421 (1966).
- 3) P.G. Barbano, L. Rigali, Separation of americium from rare earths by reversed-phase partition chromatography, *J. Chromatog.* 29, 309 (1967).

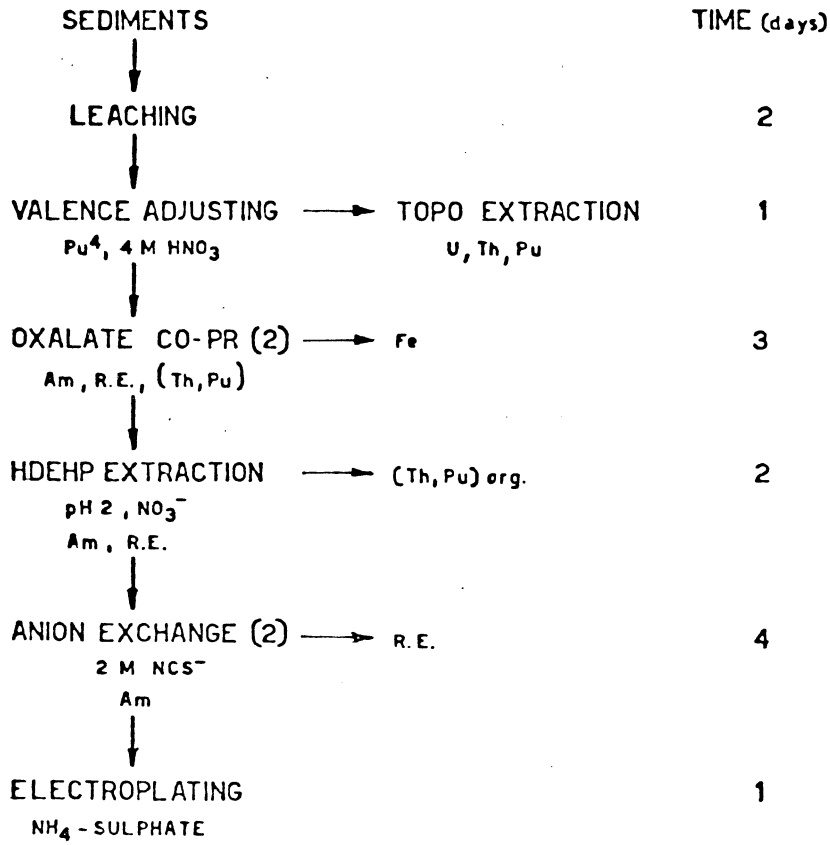


Fig.1 - Radiochemical separation of americium in 50g of dried sediments.

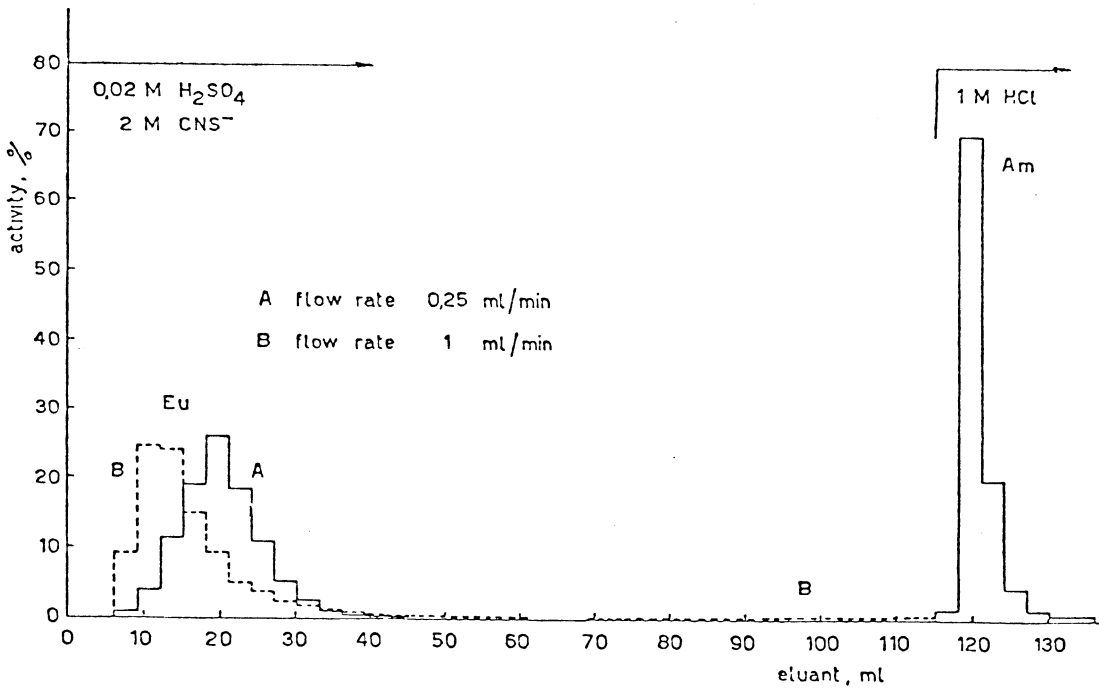


Fig.2 - Europium-americiu separation by extraction chromatography with 30% Aliquat-336.

DELLE SITE, A., MARCHIONNI, V.

*"Separation of americium in marine samples by extraction chromatography.
A new proposal for the americium-rare earth separation".*

Paper presented by A. Delle Site (Italy)

Discussion

C. PAPADOPOULOU: Can this method be applied to different compositions of sediments?

A. DELLE SITE: I think so. The pretreatment is so drastic that all transuranics should be in ionic form. Moreover the following chemical conditions are such that no great matrix effect should be expected.