

Radiochemical separation of plutonium, americium and curium from environmental material, by solvent extraction A Preliminary Report

by

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Abstract

A preliminary report is presented for a possible chemical separation of americium and curium from plutonium using di-2-ethylhexyl phosphoric acid (HDEHP). Work to date with gamma emitting isotopes, Pu-237 and Am-241 indicate a clean separation with high chemical yields. Application to the analysis of transuranics in environmental samples is being studied.

Résumé

Un rapport préliminaire est présenté, étudiant la possibilité de séparer chimiquement l'américium et le curium du plutonium en utilisant l'acide phosphorique di-2-ethylhexyl (HDEHP). Les résultats obtenus jusqu'à ce jour avec les isotopes gamma émetteurs Pu-237 et Am-241 indiquent une séparation propre avec des rendements chimiques élevés. Une application de la méthode à l'analyse des transurani-ques dans des échantillons de l'environnement est étudiée.

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An analytical method is presented for the separation of plutonium (+3, +4, +6 forms) from americium (+3 form) and curium (+3 form) involving solvent extraction using di-2-ethylhexyl phosphoric acid (HDEHP) from a nitrate media, in the presence of iron. Application to environmentally contaminated samples is at present under investigation. The separation method was worked out using the gamma emitting isotopes plutonium-237 (100 KeV major peak energy) and americium-241 (60 KeV Major peak energy); their activity being measured by a NaI (TI) scintillation well crystal. Radiochemical separation of curium was measured by alpha spectrometry on reagent blank samples.

The radiotracer procedure adopted for sediments was as follows :

1. Ten grams of dried sediment is leached with aqua regia reagent. Tracer activities of plutonium-237 and americium-241 are added to give total count of 2000 cpm for each isotope. The acid solution is heated to between 90-100° C for 8 hours. The solution is then allowed to cool and the residue filtered through a glass fibre filter. The total volume is made up to 150 mls with distilled water and adjusted to pH 7 using ammonia hydroxide. At this pH iron hydroxide precipitates, carrying down plutonium and americium as well as many other ions. The precipitate is allowed to settle and then centrifuged, the aqueous phase is discarded.

2. The iron precipitate is dissolved in a minimum quantity of 16N nitric acid and the solution then made up to 8N with respect to nitric acid, the total volume being about 50 mls. This solution is then put into a 250 ml separating funnel and 20 mls 0.5M HDEHP in normal Heptane is added. The solutions are vigorously shaken for 2 minutes and the organic phase then allowed to separate. At this

acid concentration all forms of plutonium have been shown to extract leaving americium/curium and iron in the aqueous phase. The plutonium is then back extracted with 2 ml 1N ammonium iodide solution in 20 mls concentrated hydrochloric acid. The back back extraction of Pu has been shown to be about 85 % of the total spike added at the start of the separation. The separation of plutonium from americium under these conditions is shown to be greater than 98 %.

3. The extracted plutonium solution is evaporated to dryness, a few milliliters of concentrated hydrochloric and nitric acid are added to get rid of any ammonium iodide. This procedure is repeated twice. The plutonium, containing a small fraction of iron is then taken up in 30 ml of 8N nitric acid and the procedure of WONG [1971] followed for plutonium purification.

4. The aqueous solution containing americium and curium after extraction of the plutonium is evaporated almost to dryness. The solution is made 1N with respect to hydrochloric acid; The solution is then passed through a cation resin Dowex 50W-x8, 100-200 mesh. The americium and curium and lanthanides stay on the column but most of the silica and iron passes through. The column is washed with 1N hydrochloric acid and then eluted with 12N hydrochloric acid. This elutes americium and curium as well as a small quantity of iron, the other lanthanides are eluted much more slowly thus giving an effective method of separation.

5. The eluate is evaporated to dryness, made 1N with respect to nitric acid (20 ml vol.). The solution is gently heated for 1 hour and is allowed to cool and adjusted to pH 2 with ammonium hydroxide. 10 mls HDEHP are then added and the americium and curium extracted. The HDEHP is then washed with 0.075 HCl, two washes of 25 mls each and discarded. The americium and curium are then back extracted with 10 ml of 9N HCl. If at this stage any traces of ion remain, the americium and curium are passed through an anion exchange resin AG 1 -x8. The iron remains on the column while the americium and curium pass through. The solution is then evaporated to dryness, the evaporated residue is completely clean at this stage.

It is found that before electroplating the average recovery of plutonium-237 is about 60-70 % of the initially added activity. For americium-241 and curium-244 the recovery is 65-75 %.

Electroplating is carried from an oxalate media at a current density of about 1 ampere cm² at 10 volts, according to the method of PUPHAL & OLSEN [1972]. Electroplating time is 50 minutes and the resulting plates give excellent resolution of all alpha peaks.

The method has been applied to sea water, sediment and biological material with success and the advantage of the present analytical scheme would appear to be its rapidity with respect to the separation of americium and curium from plutonium, as well as its high chemical yields.

References

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