ANALYSIS OF AMERICIUM
IN TRANSPLUTONIUM PROCESS SOLUTIONS

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INTRODUCTION

One of the more difficult analyses in the transplutonium field is the determination of americium at trace levels in a complex matrix such as a process discharge solution. These solutions have gross alpha and gamma radiations in the range of $10^3$ dis/min/ml with accompanying high beta and neutron background. These radiations, contributed by americium, are extremely small by comparison. Because of these conditions a highly selective separation must precede the measurement of americium. The separation technique should be mechanically simple to permit remote operation with master-slave manipulators. For subsequent americium measurement by the mass spectroscopic radioanalytical technique, photonics and certain interferences must also have been removed.

DISCUSSION AND RESULTS

F. L. Moore's [1] method that uses the "Aliquat-336-SCN" in xylene for the separation of actinides from lanthanides was modified to separate americium and uranium from fission products, iron, mercury, aluminum and plutonium. Americium was separated fromcurium and californium by means of a potassium peroxysulfate oxidation and HDDEP (diethylene triamine pentaacetic acid)-orthophosphoric acid chromatographic column elution similar to the techniques of Moore [2] and Horwitz [3].

The two main variables affecting the extraction of americium at constant acidity (0.06 N HNO₃) by "Aliquat-336-SCN"/xylene are the concentrations of ammonium thiocyanate and aluminum nitrate in the aqueous phase (figure 1). The extraction of americium from solutions with low acid and without aluminum nitrate increases with increasing thiocyanate concentration. Figure 1 shows that at an acid concentration of 0.04 N thiocyanate was selected for maximum americium recovery in process samples containing from 0 to 2.5 N aluminum nitrate.

Americium recovery and decontamination factors are presented in table 1. Decontamination factors for the lanthanides are the highest so far reported in the literature. To optimize the extraction of americium from the lanthanides and plutonium, the following steps were developed and evaluated:

1. An aqueous wash of the "Aliquat-336-SCN"/xylene phase with 0.44 N ammonium thiocyanate was performed.

2. The dilute nitric acid solution used to strip americium from the xylene phase was made 0.0007 N with sodium nitrate. This modification increased the plutonium decontamination factor from 8 to 100.

Other elements normally encountered in process samples (iron, aluminum, mercury, zinc and fission products) were separated from americium to non-interfering levels.

Figure 2 shows that DTPA (diethylene triamine pentaacetic acid) which is used in some processes to separate the transplutonium elements into individual species prevents the extraction of americium. It was found that the addition of aluminum nitrate to the samples prior to extraction eliminates the interfering effect of DTPA and makes unnecessary the time consuming sodium hydroxide precipitation of americium normally required in this type of sample.

Americium is separated from curium and californium on a chromatographic column of 8% HDEEP with inert diatomaceous earth support. Americium (III) is oxidized with solid potassium peroxysulfate to americium (VI) which self-reduces to americium (V). The nonsorbable americium (V) elutes rapidly in dilute acid, while curium (III) and californium (III) are held by the column. Curium can be separated from californium by increasing the nitric acid strength of the eluant. The elution profile is shown in figure 3 and decontamination factors are given in table 2. For mass spectroscopic analysis americium is separated from neptunium and the sulfate salt of oxidation. This is accomplished by reduction with hydroxylamine nitrate and separation on another HDEEP column.

Results of the americium analyses are shown in table 3. The independent analyses were performed by a complex separation using fluoride, oxalate and hydroxide precipitations followed by high pressure chromatographic column separation.

* The efficiency of oxidation of americium with solid potassium peroxysulfate is normally 90%. When a freshly prepared solution of ammonium peroxysulfate was used the oxidation was never more than 15%.

REFERENCES

