EXTRACTION OF COMPLEXES OF LANTHANIDES AND ACTINIDES WITH ARSENAZO III IN AN AMMONIUM SULFATE–POLY(ETHYLENE GLYCOL)–WATER TWO-PHASE SYSTEM


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The distribution of lanthanides and actinides in a two-phase liquid system obtained by mixing an aqueous solution of poly(ethylene glycol) and ammonium sulfate has been studied as a function of pH. Conditions are reported which provide the heterogeneity of the system suggested. It is shown that thorium and plutonium can be separated from transplutonium elements and lanthanides. Conditions have been chosen for quantitative group extraction of actinides and lanthanides.

Introduction

Organic reagents are widely applicable in various methods of separation and determination of actinides and lanthanides. Water-insoluble reagents and solvents are usually used for separation of these and other elements by liquid-liquid extraction. Meanwhile, a number of water-soluble organic reagents exhibit significant complexing ability and selectivity with respect to metal ions that makes it possible to use them for masking, photometric determination and other purposes. Such reagents could substantially extend the sphere of application of liquid-liquid extraction to separation and concentration of elements. Complexes of metals with water-soluble reagents, containing sulfonic groups, can be extracted by adding bulky hydrophobic cations.1–3

Arsenazo III, which belongs to reagents of such kind, is an effective photometric reagent for some elements including lanthanides and actinides.3,4 It is necessary to use alcohols as solvents and introduce bulky organic cations to extract metals with Arsenazo III and other reagents containing two or more sulfonic groups.3

Another approach is based on the usage of heterogeneous liquid systems, whose both phases contain significant amounts of water so that to decrease the effect of hydration on the transfer of complexes with water-soluble reagents from one phase to another. It is also of interest to use liquid-liquid extraction system without organic solvent which are often volatile, flammable, or explosive. Such systems have been
suggested in paper\textsuperscript{5} where unequal distribution of metal complexes has been shown to occur between two phases formed due to phase separation of aqueous solution of poly(ethylene glycol) (PEG) in the presence of some inorganic salts.

The present paper describes the extraction of complexes of some actinides in an ammonium sulfate - PEG - water system. Proper choice of salt, PEG and water concentrations may provide the formation of a heterogeneous system, where metal ions and their complexes with a water-soluble reagent distribute unequally between the two phases.

**Experimental**

**Reagents, radionuclides and apparatus**

The inorganic salts and acids were of chemical purity grade. PEG from Loba Chemie with a molecular mass of 2000 was used in all the extraction experiments. Arsenazo III was synthesized in the Vernadsky Institute by a procedure described in paper.\textsuperscript{6} Unless otherwise stated, the reagent concentration in the initial salt solution was $1 \cdot 10^{-3}$ mol/l.

Radiochemically pure radionuclides $^{241}$Am, $^{243}$Cm, $^{249}$Bk, $^{249}$Cf, $^{239}$Pu, $^{233}$U, $^{152}$-155Eu, $^{144}$Ce, $^{234}$Th were used. The distribution of $^{241}$Am, $^{243}$Cm, $^{249}$Cf, $^{152}$-155Eu was estimated radiometrically from $\gamma$-activity using a scintillation counter with NaI(Tl) crystal and a $\gamma$-Avtomat NRA-603 (Tesla) counter; the distribution of $^{239}$Pu, $^{233}$U from $\alpha$-activity and that of $^{240}$Bk from $\beta$-activity on a $2\pi$ proportional counter. Measurements of salt solution pH were made on LPU-01 pH meter using a microcell. The phase composition was studied by procedures described in paper.\textsuperscript{5} The distribution of sulfate ion was estimated gravimetrically by precipitation of BaSO$_4$, that of Arsenazo III on an SF-14 spectrophotometer at 540 nm.

**Extraction procedure**

To 0.5 ml of 40 mass % solution of ammonium sulfate (pH was adjusted by sulfuric acid or ammonium hydroxide aqueous solution) a radionuclide aliquot and 0.12 ml of 0.1% aqueous solution of Arsenazo III were added and the solution was diluted with water to a volume of 0.95 ml. The solution was mixed and shaken with 0.63 ml of 40 mass % PEG solution for 3 min. (The preliminary experiments had shown that the equilibrium is attained faster than 2 min). After phase separation and centrifuging, equal volumes of both phases were withdrawn and subjected to radiometric assay. The distribution coefficients were calculated as a ratio of radionuclide