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JAN 25 1968

THE DETERMINATION OF AMERICIUM
WITH DI-N-BUTYL-N-BUTYLPHOSPHONATE

I. INTRODUCTION

The high specific activity of americium makes feasible the determination of very small quantities of this element by alpha counting. Quantities as small as 6×10^{-19} grams (2 d/m) have been detected.¹⁶ However, americium is produced in a mixture of transuranium elements and fission fragments from successive nuclear reactions in a chain-reacting pile. Nearly all the elements produced in such a mixture are radioactive. Many are alpha and gamma emitters, as is americium. Thus, the simple determination of americium by alpha or gamma counting is impossible. Alpha energy analysis (AEA) and measurements on gamma spectra can be applied to some extent.³ However, they suffer from serious drawbacks due to interfering particles, especially when americium is present in small concentrations. In such cases, a preliminary chemical separation can be employed to eliminate or reduce these interferences.

The chemical treatments of americium samples reported previously fall into four principal categories:

A. Solvent Extraction

This is by far the most popular category at present. Many of the organo-phosphorous compounds have been reported to be good extractants for actinides and lanthanides.^{5,32} Among the organic reagents reported are: tri-n-butylphosphate (TBP)²⁸, di (2-ethylhexyl) orthophosphoric acid (D2EHPA)^{6,12}, thenoyltrifluoroacetone (TTA)^{19,23},^{24,28,31}, tri-n-octyl phosphine oxide (TOPO)⁴, triisooctylamine (TiOA)²⁰, ethyl ether²⁴, tricaprylmethylammonium nitrate²², and cupferron.²⁴

B. Ion Exchange

This is the classical method for the separation of lanthanides and actinides.^{25,26} It is now losing favor due to the simpler and more quantitative liquid-liquid extractions.

C. Precipitation

Precipitation is also a classical method. Coprecipitation of insoluble fluorides is probably the most common procedure. Previously reported methods involve: calcium fluoride¹¹, lanthanum fluoride^{16,17,21}, cerium fluoride^{7,18,27}, zirconium and phenylarsonic acid², and bismuth phosphate.¹⁶

D. Electrophoresis

Only one method has been reported in this new and interesting category.⁹

Several of the above methods involve the peculiar oxidation-reduction characteristics of the actinide and lanthanide series and Am in particular.^{7,8,18,27,31}

A comprehensive review of Am chemistry up to 1960 is given in a paper by Penneman and Keenan.²⁵ That work also summarizes the quantitative analyses for Am up to 1960. A similar paper of smaller scope, due to Gift, is a review of analytical methods for Am up to 1954.¹⁰

II. DISCUSSION

This paper deals specifically with the use of di-n-butyl-n-butylphosphonate for the determination of Am. It has been described as an organic extractant for Pu and U.^{5,32} However, no work prior to this has been reported on its use as an analytical reagent. The isotope Am²⁴¹ was used exclusively throughout this experimental work. The principles given here may be extended to other Am isotopes such as Am²⁴³ by alpha energy analysis (AEA).

DBBP extraction coefficients in the pH range 0 to 6 have been given for HNO_3 and various actinides and lanthanides.^{1,13,14,15,29,30} Few data have been reported for acid concentrations greater than 2.0 M. Many of the data deal with process streams which have a high salt content. In this study, the salt content was kept very low in order to avoid self-absorption of the alpha radiation by the sample. Since the salting-out effect is of considerable importance^{1,15,29}, not all of the previous data is applicable to this work.

The behavior of organic DBBP solutions toward aqueous salting agents can be rationalized theoretically. Sheppard²⁹ determined that the extraction reaction for Am by DBBP is

where the subscripts denote the aqueous (a) or organic (O) phases. The physical quantity of interest in this system is the extraction coefficient, defined by equation (1).

$$E_{\text{a}}^{\text{O}} = \frac{\text{concentration in organic phase}}{\text{concentration in aqueous phase}} \quad (1)$$

This quantity can be related to the equilibrium constant for reaction (a) in the following way: The expression for the equilibrium constant for reaction (a) is

$$K_1 = \frac{[\text{Am}(\text{NO}_3)_3 \cdot 3\text{DBBP}(\text{O})]}{[\text{Am}^{3+}(\text{a})][\text{NO}_3^-(\text{a})]^3[\text{DBBP}(\text{O})]^3} \quad (2)$$

From equation (1)

$$E_{\text{a}}^{\text{O}} = \frac{[\text{Am}(\text{NO}_3)_3 \cdot 3\text{DBBP}(\text{O})]}{[\text{Am}^{3+}(\text{a})]} = K_1 [\text{NO}_3^-(\text{a})]^3 [\text{DBBP}]^3 \quad (3)$$

Theoretically, E_{a}^{O} for Am^{3+} should vary directly as the third power of the aqueous nitrate ion concentration and directly as the third power

of the organic DBBP concentration. This strong dependence of $E \frac{O}{a}$ on the nitrate ion concentration is the well-known "salting-out" effect. In practice, a strong dependence of $E \frac{O}{a}$ on organic phase DBBP concentration is observed. However, it is not the same dependence predicted by equation (3). Rather, an optimum DBBP concentration occurs at about 1.0 - 1.2 M DBBP.^{14,29} This corresponds to a 30% solution of DBBP in an organic diluent.

It is important here to point out that $[DBBP(O)]$ refers to the molar concentration of free, or uncomplexed, DBBP in the organic phase. Nitric acid forms a 1:1 complex with DBBP¹⁵ which will effect this important variable considerably. The extraction reaction for HNO_3 can be represented as

The expression for the equilibrium constant is

$$K_2 = \frac{[HNO_3 \cdot DBBP(O)]}{[H^+(a)][NO_3^-(a)][DBBP(O)]} \quad (4)$$

However, in these experiments,

$$[H^+(a)] \approx [NO_3^-(a)] \quad (5)$$

since the concentration of nitrate salts was kept very low. The actual ratio of the acid nitrate molarity to the salt nitrate molarity was $> 7 \times 10^3$. Substituting equation (5) into equation (4),

$$[DBBP(O)] \approx \frac{1}{K} \frac{[HNO_3 \cdot DBBP(O)]}{[H^+(a)]^2} \quad (6)$$

Therefore, for the conditions of this experiment, the free DBBP concentration in the organic phase should be dependent upon the inverse of the square of the aqueous phase acidity.

Am (III) has been reported to form a nitrate complex in acid media.¹⁵ The formula given for the complex is $\text{HAm}(\text{NO}_3)_4$. It is entirely possible that other complex ions can exist in solution and that equilibria can exist between these ions and the other components of the aqueous phase. The position of these equilibria would be affected by the acidity and nitrate ion concentration of the aqueous phase. A general formula for these equilibria is:

where n denotes the coordination number for Am (III) and x and y are integers with the restrictions: $0 \leq x \leq n$, $x + y = n$. Such a reaction would be expected to proceed stepwise, involving several equilibria at once.

The formation of the neutral complex $\text{Am}(\text{NO}_3)_3 \cdot 3\text{DBBP}$ in organic media²⁹ suggests that $n = 6$. Equation (a) may then be rewritten as:

According to Kingsley and Hopkins, the acid complex $\text{HAm}(\text{NO}_3)_4$ is inextractable into 30% DBBP.¹⁵ Their study showed that the concentration of this complex becomes appreciable at acid concentrations greater than 1 M. The formation of this inextractable complex was evident by a sharp decrease in $E_{\frac{O}{A}}$ for Am with increasing acidity. A possible formation reaction for this complex from the neutral, extractable species is:

Bray and Benedict¹ have shown that $E_{\frac{O}{A}}$ for Am has a very low value at $\text{pH} > 3$, reaches a maximum at $\text{pH} 1.0 - 1.5$, and decreases once again at $\text{pH} < 1$. This behavior is in fairly good agreement with that found

by Kingsley and Hopkins.¹⁵ A reasonable explanation of these phenomena is that the presence of the inextractable complex $[\text{Am}(\text{NO}_3)_4(\text{H}_2\text{O})_2]^-$ is appreciable at $\text{pH} \leq 1$, that the neutral complex $[\text{Am}(\text{NO}_3)_3(\text{H}_2\text{O})_3]$ is in appreciable concentration at $\text{pH} 1.0 - 1.5$, and that another equilibrium involving another inextractable species becomes significant at $\text{pH} > 3$. For example, reaction (f) may be displaced to the left at $\text{pH} > 3$.

By analogy with the charged complex formed by equation (e), the charged complex formed by equation (f) may be inextractable into an organic solvent. This conclusion is supported by the nature of the organic phase. A neutral complex should be more soluble in a non-polar solvent than in a polar one.

The effect of the aqueous phase acidity on the concentration of extractable Am (III) is a very complex one. However, if a strongly acidic aqueous phase were employed, all the equilibria would be shifted toward the more highly nitrated complexes. These highly charged complexes would be expected to be inextractable into an organic phase. For this reason, $E \frac{O}{a}$ for Am should decrease as the aqueous phase acidity increases above 1 M.

In summary, the overall behavior of $E \frac{O}{a}$ for Am as a function of aqueous phase acidity is: 1) an increase occurs as the pH decreases from high values and approaches 1.5, 2) a maximum is approached at $\text{pH} 1.0 - 1.5$, and 3) a decrease occurs as the pH decreases beyond 1. A reasonable rationalization of these effects is: 1) the concentration of $[\text{Am}(\text{NO}_3)_3(\text{H}_2\text{O})_3]$ and the salting-out effect increase as the pH decreases to 1.5, 2) optimum conditions for the formation of $[\text{Am}(\text{NO}_3)_3(\text{H}_2\text{O})_3]$ and the

salting-out effect occur at pH 1.0 - 1.5, 3) the formation of $[\text{Am}(\text{NO}_3)_4(\text{H}_2\text{O})_2]^-$ and other inextractable complexes and the acid complexing of DBBP become significant at pH ≤ 1 .

The preceding discussion was limited to Am (III). Similar considerations apply to other ions, such as Pu (IV). The effects discussed above have been observed for Am, Cm, Eu, and Ce, the maximum $E \frac{O}{A}$ occurring at about pH 1.¹ In contrast, no such maximum has been observed for Pu, Th, U or Np.³⁰ This is probably due to a greater salting-out effect for these higher-charged ions and, perhaps, the lack of formation of inextractable nitrate complexes. It seemed advantageous to study $E \frac{O}{A}$ at higher aqueous phase acidities in order to detect the optimum acidity for Am decontamination by DBBP extraction.

III. EXPERIMENTAL

A. Preparation of Reagents

DBBP was obtained through Van Waters and Rogers Co. from Mobil Chemical Co., Charleston, S. C. Soltrol 170 (Petroleum Diluent) was obtained from Phillips Petroleum Co.

A 30% by weight solution of DBBP in Soltrol 170 was prepared. The solution was then washed with an equal volume of 5% sodium carbonate solution and rinsed three times with equal volumes of 1 M nitric acid. This procedure is necessary to remove impurities in the DBBP. No other purification steps were taken.

Nitric acid solutions in concentrations from 1-16 M were prepared by accurately measuring calculated volumes of concentrated nitric acid (15.99 M by analysis) into calculated, accurately measured volumes of distilled water.

Analytical reagent grade xylene, purchased from the Mallinckrodt Chemical Co., was used as a rinse solvent.

The plutonium-americiium solution used in the extraction coefficient study was a laboratory sample which had been analyzed for Am^{241} by the accepted TBP-TTA method.²⁸ It contained 19.41 g/l total Pu and 501.5 ppm Am^{241} relative to total Pu. The other components of the sample are listed in Table 1.

B. Radiochemical Analyses

Radioactive solutions were mounted on stainless steel disks, flamed to redness, covered with a thin collodion film, and counted in a calibrated Simpson alpha proportional counter (ASP) for total alpha activity. They were then analyzed according to alpha particle energies with a gold barrier solid state detector and a calibrated 400-channel analyzer (AEA) to separate activities due to specific nuclides.

C. $E \frac{O}{a}$ for Pu and Am vs. HNO_3 Molarity

$E \frac{O}{a}$ for Pu was measured as a function of nitric acid molarity before equilibration by use of equation (1) and measurements of Pu concentrations in the aqueous and organic phases. An organic to aqueous volume ratio of 2:1 was used in this study. No rinse was employed. Contact time was 20 minutes. These results are shown in graphical form as the upper curve in Figure 1.

$E \frac{O}{a}$ for Am was measured semi-quantitatively by the following method: When the Pu was extracted into DBBP, most of the Am was left in the aqueous phase. The alpha activity of Am in the aqueous phase was sufficient to permit its measurement to a precision of a

few per cent. However, the Am in the organic phase could not be measured by alpha counting, since essentially all of the alpha activity in the organic phase was due to Pu. Therefore, the approximation was made that any Am not detected in the aqueous phase was in the organic phase. A concentration was then calculated for the organic phase on the basis of this assumption. These concentrations were used to calculate $E \frac{O}{a}$ for Am. The experimental data are shown in graphical form as the lower curve in Figure 1.

D. Recovery of Am

A better measure of the efficiency of the method is the amount of Am recovered in the aqueous phase, a quantity which can be measured quantitatively. This quantity is expressed as a per cent of the accepted concentration of 501.5 ppm and is listed as a function of nitric acid molarity in Table 2.

E. Decontamination Factor

As far as the analytical procedure is concerned, the variable of critical importance is the decontamination factor. In this experiment, this quantity is defined as the ration of $E \frac{O}{a}$ for Pu to $E \frac{O}{a}$ for Am and is a measure of how efficient the extraction works for the separation of Am from Pu. The decontamination factor is plotted as a function of nitric acid molarity in Figure 2.

F. Effect of Time on Extraction Efficiency

The total ASP counts in a 100 μ aliquot of the aqueous phase and the percent of alpha activity due to Am²⁴¹ were measured as a function of contact time. The results are displayed in Table 3. These data agree with previous observations.^{1,14} The effect of time is small.

G. Procedure for the Determination of Am by 30% DBBP and Alpha Counting

Since one contact with the organic phase was insufficient to remove the Pu from the Am quantitatively, a procedure was developed which would decontaminate the Am completely. This procedure is outlined in Figures 3-9. Figure 3 is a flowsheet diagram of the steps in the procedure. These steps are described in more detail below. The numbers listed in Figure 3 for this analysis procedure are suggested for their safety and ease in manipulations. Figure 4 shows the hood set-up for handling Pu-Am samples.

Step 1. A sample aliquot is pipeted into a known volume of 12 M HNO_3 in a dilution vial (Figure 5). The dilution is stirred.

Step 2. An aliquot of the dilution from step 1 is pipeted into a sample vial. An equal volume of 30% DBBP in some suitable organic solvent (eg. Soltrol or xylene) is pipeted into the small vial. The two phases are thoroughly mixed for a sufficient time (about two minutes) to insure that equilibrium has been reached. The phases are allowed to separate and the organic phase is discarded.

Step 3. A second equal volume of 30% DBBP is pipeted into the small vial, the phases are mixed and the organic phase is discarded.

Step 4. An equal volume of xylene is pipeted into the small vial, the phases are mixed, and the organic phase is discarded.

Step 5. A suitable aliquot of the resulting aqueous phase is pipeted onto a stainless steel counting disk (Figure 6). The disk is dried under an infrared heat lamp and flamed to redness with a Bunsen burner (Figure 7). It is then covered with one or two drops of collodion, counted in a proportional counter for total alpha activity (Figure 8), and counted in an alpha energy analyzer (AEA) to separate and determine the alpha activities due to the various alpha emitters (Figure 9).

Alpha energy analysis showed that the aqueous phase resulting from this procedure contained only Am^{241} . The concentration of the standard used in this paper was determined by this procedure to be 515.5 ppm Am^{241} . This is a recovery of 102.8%, which is within experimental error. No correction was applied for the radioactive growth of Am^{241} in the sample by decay of Pu^{241} . Such a correction would be small.

IV. CONCLUSIONS

The greatest decontamination and the best recovery of Am occurred at 12 M HNO_3 , the decontamination factor being greater than 10^4 . In addition, $E \frac{O}{a}$ for Am at this acid concentration is less than 10^{-2} . These circumstances will permit the determination of very small quantities of Am in large excesses of Pu. Though Am^{241} was used exclusively in this work, other isotopes of Am, such as Am^{243} , can also be determined by suitable changes in the counting techniques.

The advantages of the method outlined in Figure 3 are: 1) it is fast, 2) it is quantitative, 3) only one dilution is involved, and 4) DBBP has a high tolerance for radiation.

Some of its disadvantages are: 1) the use of 12 M HNO_3 , which can be quite corrosive, 2) a very high salt concentration in the mounted phase interferes with alpha counting, and 3) on the basis of work by Bray and Benedict,¹ Cm, Eu, and Ce are expected to follow Am throughout the procedure. These elements were not investigated in this work. Fortunately, all of them can be separated from Am by other methods^{9,11,20,22,24,25,31} or simply detected and corrected for by alpha energy analysis.

Pu^{238} may interfere due to its extremely high specific activity. A slight trace of this isotope would cause a great difference in the total alpha activity. Unfortunately the alpha particle of Am^{241} cannot be distinguished from that of Pu^{238} by AEA. If Am^{241} is to be detected in Pu^{238} samples by this procedure, resort to a gamma counting method is recommended.³

When high salt concentrations are encountered in the phase to be counted, the Am may be extracted into 0.5 M TTA at pH 4.5 and subsequently stripped into a low-salt aqueous phase²⁸ to be analyzed by the DBBP procedure. This procedure has a high tolerance for salts, however, because of the dilution involved. As long as the Am concentration is sufficiently high, sample salt concentrations as high as 7 M can be tolerated. The high nitric acid concentration does not interfere with alpha counting due to its vaporization when the counting disk is dried.

The accuracy of the method was not measured precisely, principally due to lack of a suitable standard. On the basis of the results listed in Section III (G) and Table 2, it appears that they are at least as good as those for other methods. Agreement between several analyses was found to be from 1-5%. Recovery of Am^{241} at 12 M HNO_3 was $100 \pm 3\%$.

This work was intended specifically to develop a new method for determining Am in Pu samples with DBBP. Further work involving the calibration of the method with Th, Cm, Np, Ce, Eu, and Pu is suggested. The accuracy and precision of the method should be better determined.

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TABLE 1

Sample Impurities

<u>Impurity</u>	<u>Concentration</u>
C	175 ppm
F ⁻	<60 ppm
U	43 ppm
Np	84 ppm
Nb ⁹⁵	<.474 ppm
Ru ¹⁰³	<1.86 ppm
Ru-Rh ¹⁰⁶	<7.41 ppm

TABLE 2Percent Recovery of Am^{241}

HNO_3 Concentration, <u>M</u>	% Recovery of Am^{241}
1	61.6
2	62.3
3	69.5
4	75.4
5	85.9
6	90.7
7	92.6
8	93.6
9	95.6
10	97.3
11	97.2
12	98.8
13	93.7
14	90.5
15	85.6
16	68.6

TABLE 3

Effect of Time on Extraction Efficiency

Equilibration time, minutes	ASP c/m	% α Activity due to Am^{241}
1	130919	66.2
5	133105	70.0
11.5	132890	66.7
15	128874	69.3
20	129201	72.7

Figure 1

 E_{ox} for Americium and Plutonium as a Function of Aqueous Phase Acidity

Figure 2

Decontamination Factor as a Function of Aqueous Phase Acidity

Figure 3 Procedure for the Determination of Americium with DBBP

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

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