Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Romanovskii, Vadim V. and Wester, Dennis W.(1999) 'Effect of $Al(NO_3)_3$ on Extraction of Am, Cm, Eu, and Ce(III) by 18-Crown-6 and Mixtures of 18-Crown-6 and Chlorinated Cobalt Dicarbolliede in Nitrobeneze', Separation Science and Technology, 34: 11, 2141 — 2151

To link to this Article: DOI: 10.1081/SS-100100761

URL: http://dx.doi.org/10.1081/SS-100100761

SEPARATION SCIENCE AND TECHNOLOGY, **34**(11), pp. 2141–2151, 1999

Effect of $Al(NO_3)_3$ on Extraction of Am, Cm, Eu, and Ce(III) by 18-Crown-6 and Mixtures of 18-Crown-6 and Chlorinated Cobalt Dicarbollide in Nitrobenzene

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ABSTRACT

The extraction of Am, Cm, Eu, and Ce(III) tracers from nitric acid in the presence of $Al(NO_3)_3$ by 18-crown-6 (18C6) or mixtures of 18C6 and chlorinated cobalt dicarbollide (ChCoD) in nitrobenzene solutions was investigated. The partitioning of 18C6 between nitrobenzene and the aqueous phase was also investigated. In the presence of $Al(NO_3)_3$, 18C6 is selective for Ce over Am, Cm, and Eu. The extraction of these same metals by ChCoD is suppressed by $Al(NO_3)_3$. The extraction of these metals is a complicated function of the 18C6-ChCoD concentration ratio. The probable formation of a 1:1 18C6:ChCoD complex and the effects of ionic strength and pH on the extraction are discussed.

INTRODUCTION

The extraction selectivity of the crown ether 18-crown-6 (18C6) for light lanthanides and Am under different conditions (1–4) conforms to the stability sequence of lanthanide(III) chloride complexes with 18C6 in CH_3OH (5). The selectivity of 18C6 can be used in extraction technology based on chlorinated cobalt dicarbollide (ChCoD) for remediation of spent nuclear fuel (6).

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The crown ether 18C6 could replace linear polyoxa compounds that are added to ChCoD as a synergist and could enable the separation of lanthanides and transuranic elements (TRU) from radioactive high-level wastes (HLW). The main goal of the separation is to minimize the quantity of HLW and TRU waste destined for geologic storage. Implementation of new radionuclide separations and processes might result in large cost savings from treatment and disposal of HLW and TRU waste.

The extraction mechanism of metals by crown ethers has a solvation character. The species $M^{z+}A_3^-L_n$ transfers to the organic phase, where M^{z+} is a metal cation, A^- is an anion, and L is a crown ether (7).

$$M^{z+}_{(aq)} + sL_{(aq)} \rightleftharpoons ML^{z+}_{s(aq)} \qquad (K_{ML_s^{z+}}) \qquad (1)$$

$$sL_{(aq)} \rightleftharpoons sL_{(o)} \qquad (K_L) \qquad (2)$$

$$ML^{z+}_{s(aq)} + zA^-_{(aq)} \rightleftharpoons ML_sA_{z(o)} \qquad (K_{ML_sA_z}) \qquad (3)$$

$$M^{z+}_{(aq)} + zA^-_{(aq)} + sL_{(o)} \rightleftharpoons ML_sA_{z(o)} \qquad (K_{ex}) \qquad (4)$$

If the ionic strength is constant and the extractant is in large excess over the metal, the activity coefficients could be considered to be constant, and the concentration extraction constant would be

$$\bar{K}_{ex} = [ML_sA_z]_{(o)}/[M^{z+}_{(aq)}][A^-_{(aq)}]^z[L_{(o)}]^s \qquad (5)$$

The influence of the anion on the extraction is important. The transfer of the $M^{z+}L$ complex to the organic phase causes the transfer of the counterion through charge conservation. Hydrophobic anions such as picrate, tetraphenylborate, and dipicrylaminate favor metal extraction whereas hydrophilic anions such as chloride and sulfate interfere (8–10). Indeed, the synergistic behavior of crown ethers and hydrophobic anions is well established (11).

In the present work we examined the effect of 18C6 on the extraction of representative lanthanides and actinides by ChCoD in nitrobenzene. In addition we determined their extraction behavior in solutions with a high ionic strength created by the presence of $Al(NO_3)_3$.

EXPERIMENTAL

The compounds 18C6 (99.5+% pure) and nitrobenzene (99+% pure) were obtained from Aldrich (USA) and used as received. The cesium salt of ChCoD was obtained from Dzerzhinsk Experimental Plant (Russia) and was converted into the acid by extraction. A stock solution of the cesium salt in nitrobenzene was successively shaken 15 times with an equal volume of 10 N HNO_3, once with distilled water, and once with 0.1 N HNO_3. The concentrations of ChCoD acid were determined by titration using a Brinkman Model 720 titrator and Brinkman Model 655 digital buret.

Radiochemically pure ^{241}Am, ^{244}Cm, ^{152,154}Eu, and ^{141}Ce were used to study the extraction of the respective metal ions. Curium was assayed by alpha liquid scintillation counting. Americium, Eu, and Ce were measured as the gamma emitters by using equipment employing a semiconductor detector. The extraction experiments were performed in glass vials at 25 ± 1°C. The volumes of the organic and aqueous phases were 2 mL each. The shaking time was 90 minutes. Aliquots of the two phases were taken for liquid scintillation or gamma counting. Before liquid scintillation counting, Cm was backextracted from the organic phases by 0.2 M 1-hydroxyethane-1,1-diphosphonic acid to avoid quenching effects. The distribution coefficients (*D*) were calculated as the ratio of the activity in the organic phase to that in the aqueous phase. The concentrations of 18C6 in the aqueous phase and nitrobenzene were determined by gas chromatography with mass spectrometric detection (Hewlett-Packard GC/MS HP589011/HP5989A fitted with a 30 meter J&W DB-5MS column).

RESULTS AND DISCUSSION

Extraction of 18C6

The distribution of 18C6 between the aqueous and organic phases is plotted in Fig. 1 as a function of [Al(NO_3)$_3$] in order to reveal the change in be-

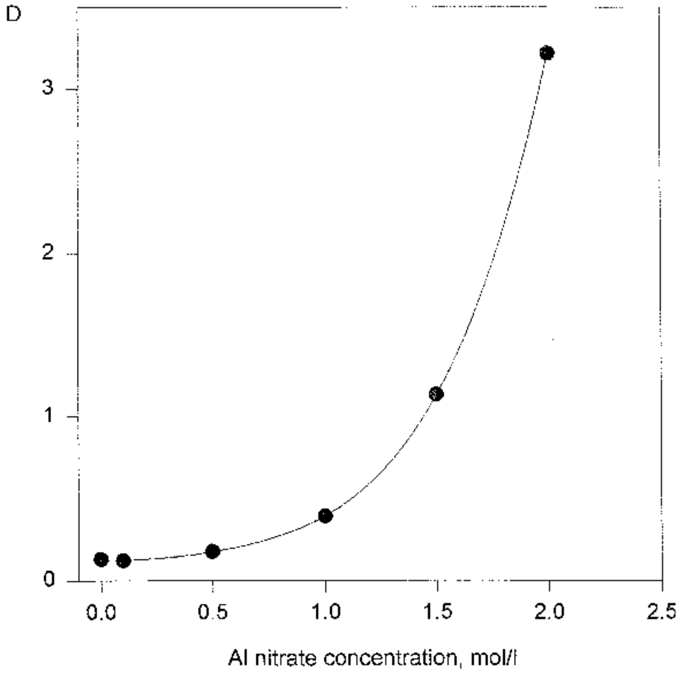

FIG. 1 Distribution coefficients of 18C6 (initially 0.2 M in the organic phase) between nitrobenzene and 0.5 M HNO_3 as a function of [Al(NO_3)$_3$].

havior of 18C6 on going to solutions with high ionic strength. For 0.5 M HNO_3, the *D* value is 0.133, i.e., 18C6 is poorly extracted into the aqueous phase. The *D* value increases as $[Al(NO_3)_3]$ in 0.5 M HNO_3 increases. A plot of log(*D*) vs log $[Al(NO_3)_3]$ exhibits no simple linear relationship. In all instances, negligible amounts of $Al(NO_3)_3$ are extracted. Thus, the extraction of 18C6 into acidic aqueous solutions of $Al(NO_3)_3$ is probably a complex function of the concentration and activity of the hydronium ion. Thus, it is likely that under otherwise constant conditions significant quantities of 18C6 will be lost to the aqueous phase from solutions with high ionic strength. Thus, industrial processes using crown ethers should utilize those with more lipophilic properties.

Extraction of Trivalent Am, Eu, Ce, and Cm Tracers by 18C6

The *D* values for extraction of trivalent Am, Eu, Ce, and Cm by 0.2 M 18C6 in nitrobenzene from 0.5 M HNO_3 are plotted in Fig. 2 as a function of $[NO_3^-]$. The nitrate ion (in addition to the 0.5 M supplied by HNO_3) was supplied by adding $Al(NO_3)_3$ to the aqueous solution. The *D* values are high enough for

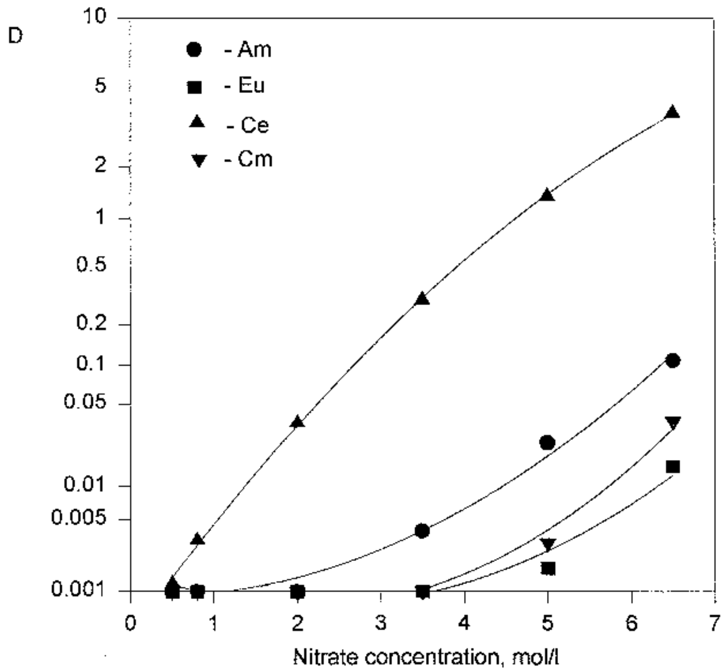

FIG. 2 Extraction of trivalent Am, Eu, Ce, and Cm tracers by 0.2 M 18C6 in nitrobenzene from 0.5 M HNO_3 and $Al(NO_3)_3$ solution. Values on the abscissa have activities in the organic phase that are below the detection limit and represent insignificant extraction.

TABLE 1
Extraction of Trivalent ^{241}Am, ^{152}Eu, and ^{141}Ce by ChCoD from HNO_3

[ChCoD], M	D_{Am}	D_{Eu}	D_{Ce}
From 0.1 M HNO_3			
0.01	34.0	59.4	104
0.05	60.0	99.4	197
0.1	56.2	75.1	173
0.15	124	132	357
0.2	173	137	273
0.25	274	231	414
0.3	432	366	496
From 0.5 M HNO_3			
0.01	0.030	0.050	0.083
0.05	0.020	0.031	0.063
0.1	0.070	0.056	0.090
0.15	0.27	0.21	0.32
0.2	0.70	0.52	0.85
0.25	1.5	1.2	1.7
0.3	3.1	2.6	5.3

effective extraction only for the light lanthanide Ce when the aqueous phase contains appreciable concentrations of $Al(NO_3)_3$. As noted above, the *D* value for 18C6 also increases as a function of $[Al(NO_3)_3]$ (Fig. 1).

Extraction of Trivalent Am, Eu, and Ce Tracers by ChCoD

The presence in the extraction system of so hydrophobic an anion as ChCoD could facilitate the transfer of metal species into the organic phase. The metal extraction can be considered as an ion exchange:

$$ML^{3+}_{(aq)} + 3H^{+}_{(o)} + 3ChCoD^{-}_{(o)} \rightleftharpoons ML^{3+}_{(o)} + 3H^{+}_{(aq)} + 3(ChCoD)^{-}_{(o)} \qquad (6)$$

Data describing the extraction of trivalent Am, Eu, and Ce tracers from 0.1 and 0.5 M HNO_3 as a function of [ChCoD] are presented in Table 1. As expected according to Eq. (6), increasing $[H^+]$ suppresses the extraction of the tracers. The effect is highly magnified by the fact that the suppression should depend on $[H^+]^3$. For this reason the effect of 18C6 as a modifier on the extraction of the trivalent tracers was examined (see below).

The effect of $Al(NO_3)_3$ on the extraction of the tracers was determined by measuring *D* values between an aqueous phase containing 0.5 M HNO_3 and 2 M $Al(NO_3)_3$ and an organic phase containing 0.3 M ChCoD in nitrobenzene. In all instances, the *D* values in the presence of $Al(NO_3)_3$ are well below unity, ranging from 0.00561 for Eu to 0.00868 for Ce. These values are three orders of magnitude less than those obtained in the absence of $Al(NO_3)_3$.

Extraction of Trivalent Am, Eu, Ce, and Cm Tracers by 18C6–ChCoD Mixtures

The dependences of the *D* values have a complex character as a function of the aqueous and organic phase composition for extraction by a mixture of 18C6 and ChCoD. Figure 3 shows the *D* values for the tracers from 0.5 M HNO_3 into an organic phase in which the sum total concentration of 18C6 and ChCoD was 0.3 M. For the solution without ChCoD, the *D* values are consistent with extraction of the tracers by 18C6 alone (Fig. 2). As the mole fraction of ChCoD increases, the *D* values begin to increase, especially after the mole fraction exceeds 0.5. As expected, the *D* values for the solution without 18C6 are the same as those found in the last line of Table 1. The plots in Fig. 3 provide data that is useful in determining whether synergism or antagonism is exhibited by the 18C6–ChCoD mixtures. A comparison of the *D* values for extraction by 18C6 alone (0.2 M), ChCoD alone (0.2 M), and the 18C6–ChCoD mixture (0.2 M ChCoD) in Figs. 2 and 3 and Table 1 suggests that the pres-

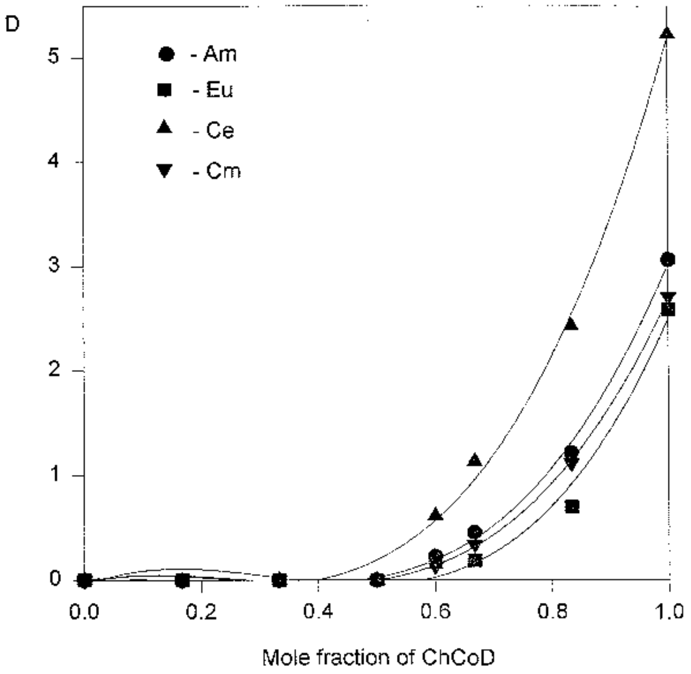

FIG. 3 Extraction of trivalent Am, Eu, Ce, and Cm tracers by 18C6–ChCoD mixtures (0.3 M total) in nitrobenzene from 0.5 M HNO_3. Values on the abscissa have activities in the organic phase that are below the detection limit and represent insignificant extraction. The slightly positive curves below 0.5 mole fraction of ChCoD are artifacts of the plotting program.

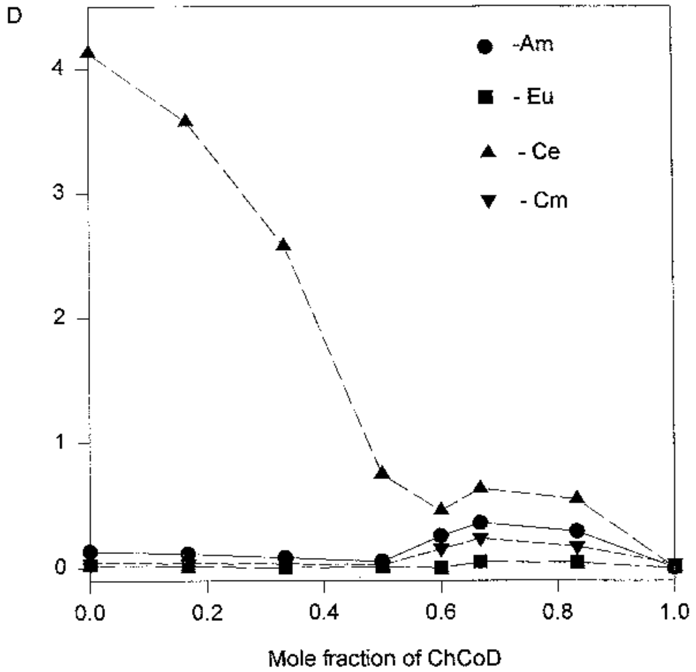

FIG. 4 Extraction of trivalent Am, Eu, Ce, and Cm tracers by 18C6–ChCoD mixtures (0.3 M total) in nitrobenzene from 0.5 M HNO_3 and 2 M $Al(NO_3)_3$. Values on the abscissa have activities in the organic phase that are below the detection limit and represent insignificant extraction.

ence of 18C6 slightly suppresses the extractions of Am and Eu and slightly enhances the extraction of Ce. The difference in the *D* values, which determines whether synergism or antagonism is present, is positive for Ce and negative for Am and Eu. For Cm, the lack of data for extraction by pure ChCoD prohibits such a comparison.

The extraction of trivalent Am, Eu, Ce, and Cm tracers in the presence of 2 M $Al(NO_3)_3$ by 18C6–ChCoD mixtures with a sum total concentration of 0.3 M was also studied. The *D* values are plotted in Fig. 4 as a function of the mole fraction of ChCoD. For the solution without ChCoD, the *D* values are the same as those farthest to the right in Fig. 2, where the total nitrate concentration is 6.5 M. Conversely, the *D* values for the solution without 18C6 are well below unity, as seen above. The behaviors of the curves in the intermediate range are very interesting. In all instances the curves show a broad maximum that stretches from about 0.5 to 1.0 mole fraction of ChCoD. This anomaly is observed only when the aqueous phase has a high nitrate concentration, 2 M $Al(NO_3)_3$ (Fig. 4), unlike when the aqueous phase contains only 0.5 M HNO_3 (Fig. 3).

The effect of 18C6 on the extraction of the tracers in 0.5 M HNO_3 by 0.3 M ChCoD in nitrobenzene was studied by gradually increasing [18C6]. The re-

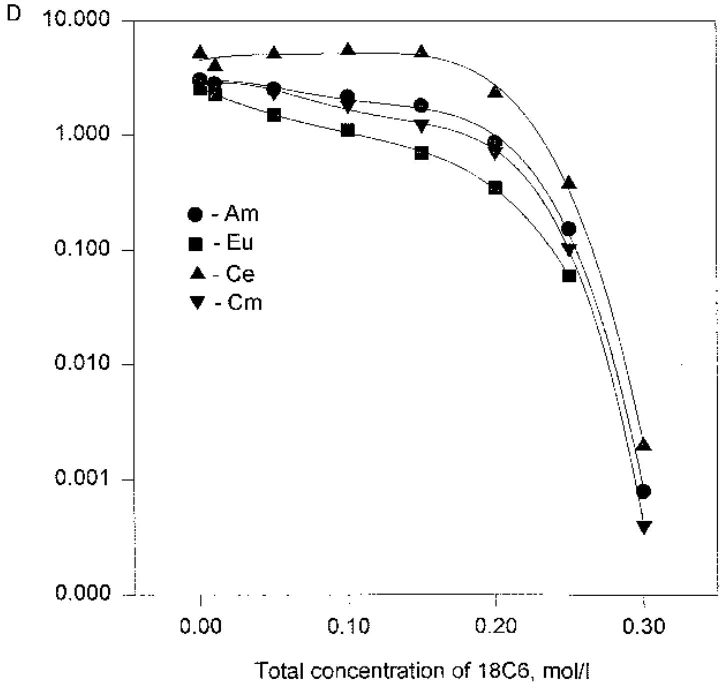

FIG. 5 Extraction of trivalent Am, Eu, Ce, and Cm tracers by 0.3 M ChCoD from 0.5 M HNO_3 as a function of total [18C6].

sults are plotted in Fig. 5. The *D* values slightly decrease until [18C6] reaches ~0.1 M, especially for Ce, and then begin to decrease more rapidly. At [18C6] = 0.3 M, where the concentrations of 18C6 and ChCoD are equal, the tracers are essentially not extracted. This confirms the suggestion by the data in Fig. 3 and Table 1 that 18C6 suppresses the extraction of these trivalent metals with ChCoD.

Formation of 18C6–ChCoD Complex

The results suggest the formation of a 1:1 complex in nitrobenzene between 18C6 and ChCoD. For example, Fig. 3 shows that the *D* values for the tracers remain negligible until the mole fraction of ChCoD reaches 0.5. The *D* values then rise very rapidly until the mole fraction of ChCoD reaches 1.0. Such behavior is consistent with the formation of a complex of formula $[(18C6)H_3O]^+$ $ChCoD^-$ since until the mole fraction of ChCoD exceeds 0.5, the extractant is entirely consumed by complex formation and is not available to form an ion pair with the metal cation.

The rapid decrease in the *D* value of Ce and the inhibition of the extraction of Am, Eu, and Cm that are seen in Fig. 4 are consistent with the formation of a 1:1 complex of 18C6 and ChCoD. As ChCoD is added to the solution, it is bound into the complex with 18C6 until [ChCoD] exceeds [18C6].

The paths of the curves that are plotted in Fig. 5 are also consistent with the formation of a complex between 18C6 and ChCoD. As 18C6 is added to the solution, ChCoD is gradually bound into the complex and becomes unavailable as the counterion for the extraction. This effectively blocks the extraction where the concentrations of 18C6 and ChCoD are equal.

The literature also contains data that are consistent with the formation of a 1:1 18C6–ChCoD complex (4). The reported log *D* vs log[18C6] curves bear a striking resemblance to those obtained in the present work. For example, the reported curve for Ce with [ChCoD] = 0.0485 M and $[HNO_3] = 0.395$ can be compared with our curve for Ce with [ChCoD] = 0.3 M and $[HNO_3] = 0.5$ M. Both curves exhibit a rapid decrease of *D* as [18C6] approaches [ChCoD] $[\log(0.0485) = -1.3]$. It is also notable that the *D* values are lower for the solution with the lower [ChCoD].

Finally, gas chromatographic (GC) analysis of 18C6 in 18C6–ChCoD mixtures provided convincing evidence for the formation of a 1:1 complex. For solutions of pure 18C6, GC results were in excellent agreement with the expected values. However, 18C6 could not be detected in 1:1 18C6–ChCoD mixtures. For 2:1 18C6–ChCoD mixtures, only about 50% of the 18C6 was detected. These results are consistent with the formation of a nonvolatile complex with the probable formula $[(18C6)H_3O]^+\ ChCoD^-$.

Effect of Ionic Strength

The salt $Al(NO_3)_3$ was chosen to adjust the ionic strength of the solutions for several reasons. First, Al^{3+} has a small ionic radius and should not effectively compete with the lanthanides and actinides for 18C6 or the ChCoD counterion. This was confirmed by examining the extraction of Al^{3+}, which showed that no detectable extraction occurred, as mentioned above. Second, the high charge on Al^{3+} imparts a greater ionic strength to its solutions than would be produced by a unipositive ion, e.g., Li^+. Finally, reprocessing solutions can be expected to contain large concentrations of Al^{3+} from dissolved cladding or fuel or both.

The increased extraction of 18C6 with increasing $[Al(NO_3)_3]$ is attributed to the increased activity of H_3O^+ as the ionic strength increases. This supposition is supported by the nonlinear plot of log *D* vs log $[NO_3^-]$. The slope continuously increases with increasing $[Al(NO_3)_3]$. For example, the slope between 0 and 0.5 M $Al(NO_3)_3$ is only 0.22, whereas that between 1.5 and 2.0 M $Al(NO_3)_3$ is 3.97.

Increasing $[NO_3^-]$ by adding $Al(NO_3)_3$ to solutions of trivalent Am, Eu, Ce, and Cm tracers in 0.5 M HNO_3 is responsible for the increasing *D* values. The data are of somewhat limited quality owing to the small *D* values. However, the slope of a plot of log D_{Ce} vs $\log[NO_3^-]$ is 3.16 with a correlation coefficient of 0.993. The fact that the *D* values for extraction of Ce are much greater than for the other metals is probably due to its larger stability constant with 18C6 (5).

Effect of pH

A comparison of our data with published data (3) for the extraction of Am and Eu by 18C6–ChCoD mixtures reveals that the pH of the aqueous phase is very important. For example, the *D* values for extraction by 0.03 M ChCoD and 0.2 M 18C6 in nitrobenzene vary from 90 ± 2 for Am and 33 ± 1 for Eu at pH 1.9 to 0.20 ± 0.006 and 0.078 ± 0.00, respectively, at pH 1.1. Similarly, at pH 0.3 the *D* values for extraction by 0.3 M ChCoD and 0.3 M 18C6 in nitrobenzene are 6.36 ± 0.18 and 2.15 ± 0.06, respectively, whereas the *D* values measured in the present work for these same metals by 0.15 M ChCoD and 0.15 M 18C6 are insignificant (Fig. 3). This large decrease of the *D* values with decreasing pH agrees well with the published results. The same effect is seen with 15-crown-5–ChCoD mixtures (3).

The effect of pH is also evident in the data in Table 1. The *D* values for all tested metals significantly decrease as the pH of the aqueous phases decreases.

Effect of Extractant Concentration

In the presence of 2.8 M $Al(NO_3)_3$, *D* values of 3.20 ± 0.20 and 0.51 ± 0.03 for Am and Eu, respectively, on extraction by 0.3 M ChCoD and 0.3 M 18C6 in nitrobenzene from 0.5 M HNO_3 are reported (3). The present work yielded *D* values of 0.0540 and 0.0113 for Am and Eu, respectively, on extraction by 0.15 M ChCoD and 0.15 M 18C6 in nitrobenzene from 0.5 M HNO_3 (Fig. 4). The differences in these *D* values are probably due to the lower extractant concentrations in the present work since the pH of the solutions and $[Al(NO_3)_3]$ are approximately the same.

In another study (2), *D* values of 0.89 ± 0.04 and 0.28 ± 0.01 for Am and Eu, respectively, on extraction by 0.2 M 18C6 and 0.3 M ChCoD from 0.5 M HNO_3 were found. Our work gave *D* values of 0.231 and 0.170 for Am and Eu, respectively, on extraction by 0.12 M 18C6 and 0.18 M ChCoD from 0.5 M HNO_3. The slightly lower *D* values in the present work are consistent with poorer extraction at lower extractant concentrations.

CONCLUSIONS

Addition of 18C6 to ChCoD is advantageous in separating actinides and lanthanides from HLW owing to the fact that Ce extraction is enhanced whereas that of Eu and Am is inhibited. Use of 18C6 in the ChCoD extraction technology would enable the extraction and separation of Cs, Sr, and transplutonium elements from HLW with one extractant. This would have a definite advantage for waste treatment. Data relating to the separation factors of Am and Eu in similar systems have been reported by this group (12).

ACKNOWLEDGMENTS

We are grateful to the US Department of Commerce and the Efficient Separations and Processing Integrated Program of the US Department of Energy for jointly supporting the Special American Business Internship Training program at Pacific Northwest Laboratory, through which V.V.R. obtained an internship, and to the Khlopin Radium Institute for suggesting and supporting the work. Pacific Northwest Laboratory is operated by Battelle Memorial Institute for the US Department of Energy under Contract DE-AC06-76RLO-1830.

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Received by editor February 9, 1995
Revision received September 6, 1995

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