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Synergistic Extraction of Plutonium(IV) from Nitric Acid Medium by Mixtures of TOPO and HTTA

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Abstract

Synergistic solvent extraction of Pu(IV) from nitric acid medium by mixtures of thenoyltrifluoroacetone (HTTA) and tri-*n*-octylphosphine oxide (TOPO) in benzene was investigated by a method developed for such studies. The species involved in the extraction were identified as $Pu(NO_3)_4 \cdot 2TOPO$, $Pu(NO_3)_3(TTA) \cdot 2TOPO$, $Pu(NO_3)_2(TTA)_2 \cdot TOPO$, and $Pu(NO_3)(TTA)_3 \cdot TOPO$. The concentration equilibrium constants for the extraction of all the suggested species from 1.0 *M* nitric acid were calculated from the data obtained, and the concentration equilibrium constants for their formation in the organic phase were estimated.

INTRODUCTION

Irving and Edgington have studied the extraction of Th(IV), Np(IV), and Pu(IV) from aqueous nitric acid by mixtures of HTTA with some neutral donors (S). They reported (*1*) that mixed NO_3^-–TTA^-–S complexes of the metal ions are responsible for the observed synergism. They, however, did not observe the formation of the species of the type $M(TTA)_4 \cdot S$ that was reported for Th(IV) by Healy (*2*). Healy and Ferraro have attempted to verify the formation of mixed NO_3^-–TTA^-–S complexes but with no success (*3*). While there are exhaustive data reported in

the literature (*4*) on the formation of simple adducts of metal β-diketonates with neutral donors, such data on the formation of mixed ligand adducts are sporadic. In our earlier work (*5*) on the extraction of Np(IV) from nitric acid medium by mixtures of HTTA and TBP, it was shown that mixed as well as normal adduct species are involved in the extraction. It is felt that the normal method of evaluating the extracted species by slope analysis of the log-log plots is inadequate to deal with the systems involving the extraction of a multiple number of species. An attempt was therefore made to develop an alternate method which would give the stoichiometry of various species extracted along with their equilibrium constant values. The extraction of Pu(IV) from nitric acid medium by mixtures of TOPO and HTTA using this method was investigated and the results obtained are reported here.

EXPERIMENTAL

Materials

The origin and the purity of most of the materials used were the same as described earlier (*6*). TOPO supplied by E. Merck was dried over P_2O_5 under vacuum prior to use.

Procedure

Plutonium in 8 *M* nitric acid was adjusted to Pu(IV) by adding a few drops of H_2O_2, the excess of which was destroyed by heating. The oxidation state of Pu(IV) was ascertained by the HTTA extraction method (*7*). A desired aliquot of Pu(IV) from this stock was added to 1 *M* nitric acid solution so as to have the concentration of Pu(IV) in the resulting solution ~ 0.5 μg/mL. About 10^{-4} *M* vanadium(V) was kept in the solution as holding oxidant for Pu(IV). Equal volumes (3 mL each) of this solution and a benzene solution of TOPO containing a varying concentration of HTTA were added to ground-glass stoppered equilibration tubes. The rest of the procedure used for the measurement of distribution ratios was the same as used earlier (*6*).

RESULTS AND DISCUSSION

Extraction of Pu(IV) from Nitric Acid by TOPO

The extraction of Pu(IV) from nitric acid medium by TOPO dissolved in an inert diluent such as benzene is given by (*8*)

$$Pu^{4+}_{(aq)} + 4NO^{-}_{3(aq)} + 2TOPO_{(org)} \overset{K_B}{\rightleftharpoons} Pu(NO_3)_4 \cdot 2TOPO_{(org)} \quad (1)$$

for which the equilibrium constant K_B is given by

$$K_B = \frac{[Pu(NO_3)_4 \cdot 2TOPO]}{[Pu^{4+}][NO_3^-]^4[TOPO]^2} \quad (2)$$

$$= \frac{D^\circ F}{[NO_3^-]^4[TOPO]^2} \quad (3)$$

where

$$D^\circ = [Pu(NO_3)_4 \cdot 2TOPO]_{(org)}/[Pu(IV)]_{(aq)} \quad (4)$$

is the observed distribution ratio of Pu(IV) and

$$F = \left(1 + \sum_1^n \beta_n [NO_3^-]^n\right)$$

is the complexing factor that accounts for the fraction of Pu(IV) that was present in the aqueous phase in the form of its nitrate complexes. The factor F can be computed from the stability constant values (β_n) for the nitrate complexes of Pu(IV). For Pu(IV) in 1.0 M nitric acid medium, $F = 14.7$ was reported earlier (*9*). As TOPO extracts nitric acid by the equilibrium shown in

$$H^+_{(aq)} + NO^-_{3(aq)} + TOPO_{(org)} \overset{K_H}{\rightleftharpoons} HNO_3 \cdot TOPO_{(org)} \quad (5)$$

the equilibrium concentration of TOPO will be different from its initial concentration. The free TOPO values were calculated using Eq. (5) and $\log K_H = 0.83$ [reported (*10*) for benzene as the diluent]. The values of K_B were then calculated using Eq. (3) and by measuring the D° values using 1.0 M nitric acid and different concentrations of TOPO, the average value being $(1.2 \pm 0.1) \times 10^{10}$.

Extraction of Pu(IV) from Nitric Acid by Mixtures of HTTA and TOPO

As stated earlier, Pu(IV) is extracted from aqueous nitric acid by solutions of TOPO as $Pu(NO_3)_4 \cdot 2TOPO$. When a β-diketone such as HTTA is added to a solution of $Pu(NO_3)_4 \cdot 2TOPO$, it is expected that each nitrate ion associated with the complex will be successively replaced by TTA ions as the latter is more strongly complexing. Such a replacement may keep the 2TOPO molecules intact in the new complexes formed, or it may decrease their number because the introduction of bulky TTA ions

may impose steric hindrance, thereby decreasing the coordination number of Pu(IV). It is known that $Pu(TTA)_4$ forms 1:1 adducts with TBP (*6*) and DBBP (*11*), and hence it is certain that it can form the adduct $Pu(TTA)_4 \cdot TOPO$. Hence the mixed $TTA^- - NO_3^-$ complexes may be associated with either 1 or 2 molecules of TOPO. Thus when HTTA and TOPO are used together for the extraction of Pu(IV) from nitric acid medium, the species that are likely to be involved in the extraction are $Pu(NO_3)_3(TTA) \cdot y\text{TOPO}$, $Pu(NO_3)_2(TTA)_2 \cdot y\text{TOPO}$, $Pu(NO_3)(TTA)_3 \cdot y\text{TOPO}$, in addition to $Pu(NO_3)_4 \cdot 2\text{TOPO}$ and $Pu(TTA)_4 \cdot TOPO$, the value of y being either 1 or 2. Thus the distribution ratio D of Pu(IV) obtained with the mixtures of HTTA and TOPO can be given as

$$D = \frac{[Pu(NO_3)_4 \cdot 2\text{TOPO}] + [Pu(NO_3)_3(TTA) \cdot y\text{TOPO}] + [Pu(NO_3)_2(TTA)_2 \cdot y\text{TOPO}] + \text{etc.}}{[Pu(IV)]_{(aq)}} \quad (6)$$

If D and D° are obtained by using the same concentration of TOPO and if the interaction between TOPO and HTTA, at the concentrations used, is ignored, Eq. (7) follows from Eqs. (4) and (6):

$$\left(\frac{D}{D^\circ} - 1\right) = \frac{[Pu(NO_3)_3(TTA) \cdot y\text{TOPO}]}{[Pu(NO_3)_4 \cdot 2\text{TOPO}]} + \frac{[Pu(NO_3)_2(TTA)_2 \cdot y\text{TOPO}]}{[Pu(NO_3)_4 \cdot 2\text{TOPO}]} + \text{etc.} \quad (7)$$

The extraction equilibria involved for the mixed species conceived can be given as

$$Pu^{4+}_{(aq)} + 3NO^-_{3(aq)} + HTTA_{(org)} + y\text{TOPO}_{(org)} \rightleftharpoons Pu(NO_3)_3(TTA) \cdot y\text{TOPO}_{(org)} + H^+_{(aq)} \quad (8)$$

$$Pu^{4+}_{(aq)} + 2NO^-_{3(aq)} + 2HTTA_{(org)} + y\text{TOPO}_{(org)} \rightleftharpoons Pu(NO_3)_2(TTA)_2 \cdot y\text{TOPO}_{(org)} + 2H^+_{(aq)} \quad (9)$$

$$Pu^{4+}_{(aq)} + NO^-_{3(aq)} + 3HTTA_{(org)} + y\text{TOPO}_{(org)} \rightleftharpoons Pu(NO_3)(TTA)_3 \cdot y\text{TOPO}_{(org)} + 3H^+_{(aq)} \quad (10)$$

$$Pu^{4+}_{(aq)} + 4HTTA_{(org)} + TOPO_{(org)} \overset{K_4}{\rightleftharpoons} Pu(TTA)_4 \cdot TOPO_{(org)} + 4H^+_{(aq)} \quad (11)$$

By subtracting Eq. (1) from Eqs. (8), (9), (10), and (11), Eqs. (12), (13), (14), and (15), respectively, are obtained:

$$Pu(NO_3)_4 \cdot 2TOPO_{(org)} + HTTA_{(org)} \overset{B_1}{\rightleftharpoons} Pu(NO_3)_3(TTA) \cdot yTOPO_{(org)} + H^+_{(aq)} + NO^-_{3(aq)} + (2 - y)TOPO_{(org)} \quad (12)$$

$$Pu(NO_3)_4 \cdot 2TOPO_{(org)} + 2HTTA_{(org)} \overset{B_2}{\rightleftharpoons} Pu(NO_3)_2(TTA)_2 \cdot yTOPO_{(org)} + 2H^+_{(aq)} + 2NO^-_{3(aq)} + (2 - y)TOPO_{(org)} \quad (13)$$

$$Pu(NO_3)_4 \cdot 2TOPO_{(org)} + 3HTTA_{(org)} \overset{B_3}{\rightleftharpoons} Pu(NO_3)(TTA)_3 \cdot yTOPO_{(org)} + 3H^+_{(aq)} + 3NO^-_{3(aq)} + (2 - y)TOPO_{(org)} \quad (14)$$

$$Pu(NO_3)_4\ 2TOPO_{(org)} + 4HTTA_{(org)} \overset{B_4}{\rightleftharpoons} Pu(TTA)_4 \cdot TOPO_{(org)} + 4H^+_{(aq)} + 4NO^-_{3(aq)} + TOPO_{(org)} \quad (15)$$

Rearranging Eq. (7) by using Eqs. (12), (13), (14), and (15), it follows that

$$\left(\frac{D}{D^\circ} - 1\right) \Big/ [HTTA] = \frac{B_1}{[H^+][NO_3^-][TOPO]^{(2-y)}} + \frac{B_2[HTTA]}{[H^+]^2[NO_3^-]^2[TOPO]^{(2-y)}} + \frac{B_3[HTTA]^2}{[H^+]^3[NO_3^-]^3[TOPO]^{(2-y)}} + \cdots \quad (16)$$

The data obtained on the extraction of Pu(IV) from 1 M nitric acid into a benzene solution of TOPO containing a varying concentration of HTTA are given in Table 1. From these data $((D/D^\circ) - 1)/[HTTA]$ were calculated, and they are plotted against [HTTA] in Fig. 1. The values of the parameters $B_1/[H^+][NO_3^-][TOPO]^{(2-y)}$, $B_2/[H^+]^2[NO_3^-]^2[TOPO]^{(2-y)}$, etc. which give the best fit for the curves in Fig. 1 were calculated by a least squares procedure using a computer program and they are given in Table 2. The best fit was obtained with three constants, thereby indicating only three species are involved in the system in the concentration range of HTTA used. It is seen that the values of $B_1/[H^+][NO_3^-][TOPO]^{(2-y)}$ obtained for both concentrations of TOPO agree reasonably well with each other, thereby showing that they are independent of TOPO concentration. Hence the value of y in Eq. (12) and Eq. (8) equals 2, suggesting that the first mixed species formed is $Pu(NO_3)_3(TTA) \cdot 2TOPO$. The values

TABLE 1

Variation of the Distribution Ratio of Pu(IV) with HTTA Concentration:[a] $[HNO_3]_{(aq)} = 1.0\ M$

[HTTA], M	Initial [TOPO] $= 2 \times 10^{-4}\ M$		Initial [TOPO] $= 2.5 \times 10^{-4}\ M$	
	D	$\frac{\left(\frac{D}{D^\circ} - 1\right)}{[HTTA]}$	D	$\frac{\left(\frac{D}{D^\circ} - 1\right)}{[HTTA]}$
0	3.61	—	0.772	—
0.001	4.13	145	0.886	148
0.002	4.73	155	1.09	206
0.004	6.35	190	1.55	252
0.006	8.18	211	2.11	289
0.008	10.7	247	3.10	377
0.010	13.6	277	3.31	458
0.012	18.3	339	6.04	569
0.014	22.6	376	8.25	692
0.016	28.4	429	11.0	829
0.018	35.5	491	14.1	957
0.020	45.0	573	18.3	1135

[a] $D = D^\circ$ when [HTTA] = 0.

of $B_2/[H^+]^2[NO_3^-]^2[TOPO]^{(2-y)}$ and $B_3/[H^+]^3[NO_3^-]^3[TOPO]^{(2-y)}$ obtained at the two concentrations of TOPO differ and are almost inversely proportional to the TOPO concentration used, thereby suggesting the composition of the second and the third complexes to be $Pu(NO_3)_2(TTA)_2 \cdot TOPO$ and $Pu(NO_3)(TTA)_3 \cdot TOPO$, respectively. From these values the values of concentration equilibrium constants B_1, B_2, and B_3 for the equilibria represented by Eqs. (17), (18), and (19) were calculated and are included in Table 2.

$$Pu(NO_3)_4 \cdot 2TOPO_{(org)} + HTTA_{(org)} \overset{B_1}{\rightleftharpoons} Pu(NO_3)_3(TTA) \cdot 2TOPO_{(org)} + H^+_{(aq)} + NO^-_{3(aq)} \quad (17)$$

$$Pu(NO_3)_4 \cdot 2TOPO_{(org)} + 2HTTA_{(org)} \overset{B_2}{\rightleftharpoons} Pu(NO_3)_2(TTA)_2 \cdot TOPO_{(org)} + 2H^+_{(aq)} + 2NO^-_{3(aq)} + TOPO_{(org)} \quad (18)$$

$$Pu(NO_3)_4 \cdot 2TOPO_{(org)} + 3HTTA_{(org)} \overset{B_3}{\rightleftharpoons} Pu(NO_3)(TTA)_3 \cdot TOPO_{(org)} + 3H^+_{aq)} + 3NO^-_{3(aq)} + TOPO_{(org)} \quad (19)$$

It is seen that the values of B_1, B_2, and B_3 obtained for the two sets of

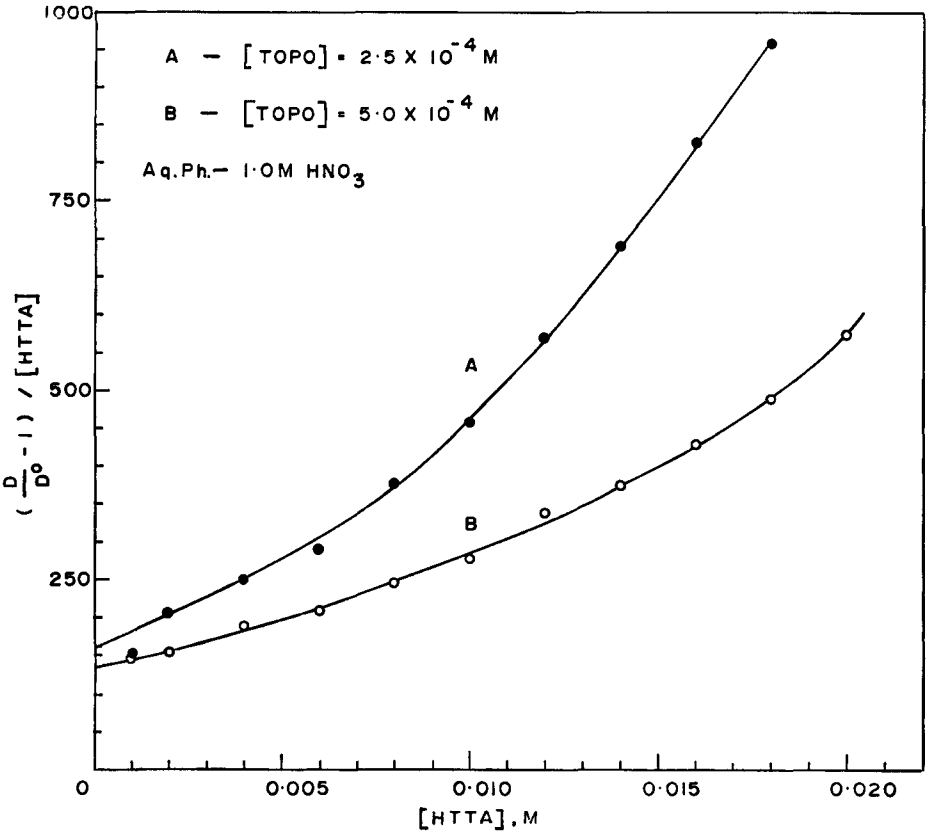

FIG. 1. Variation of $((D/D^\circ) - 1)/[\mathrm{HTTA}]$ with HTTA concentration.

data are fairly close to each other, thus confirming the validity of the composition of the complexes inferred.

The equilibria involved for the extraction of different mixed NO_3^-–TTA^-–TOPO complexes of Pu(IV) can now be obtained by adding Eq. (1) to each of Eqs. (17), (18), and (19), and these are given below:

$$Pu^{4+}_{(aq)} + 3NO^-_{3(aq)} + HTTA_{(org)} + 2TOPO_{(org)} \overset{K_1}{\rightleftharpoons} Pu(NO_3)_3(TTA)\cdot 2TOPO_{(org)} + H^+_{(aq)} \quad (20)$$

$$Pu^{4+}_{(aq)} + 2NO^-_{3(aq)} + 2HTTA_{(org)} + TOPO_{(org)} \overset{K_2}{\rightleftharpoons} Pu(NO_3)_2(TTA)_2\cdot TOPO_{(org)} + 2H^+_{(aq)} \quad (21)$$

TABLE 2

The Values of Parameters and Equilibrium Constants (Eqs. 16–19)
$[HNO_3]_{(aq)} = 1.0\ M$

Parameter	Equilibrium [TOPO] $= 6.44 \times 10^{-5}\ M$	Equilibrium [TOPO] $= 3.22 \times 10^{-5}\ M$	Equilibrium constant	Equilibrium [TOPO] $= 6.44 \times 10^{-5}\ M$	Equilibrium [TOPO] $= 3.22 \times 10^{-5}\ M$
$\frac{B_1}{[H^+][NO_3^-]}$	141 ± 6	154 ± 12	B_1	141 ± 6	154 ± 12
$\frac{B_2}{[H^+]^2[NO_3^-]^2[TOPO]}$	$(7.43 \pm 1.45) \times 10^3$	$(1.36 \pm 0.27) \times 10^4$	B_2	0.478 ± 0.093	0.438 ± 0.087
$\frac{B_3}{[H^+]^3[NO_3^-]^3[TOPO]}$	$(6.89 \pm 0.68) \times 10^5$	$(1.76 \pm 0.13) \times 10^6$	B_3	44.4 ± 4.4	56.7 ± 4.2

TABLE 3

Values of Equilibrium Constants for the Equilibria Involved in the Extraction of Different Complexes of Pu(IV) by Mixtures of HTTA and TOPO
(Aqueous phase = 1.0 *M* HNO_3; diluent, benzene)

Equilibrium	Log *K*
$Pu^{4+}_{(aq)} + 4NO_3^-{}_{(aq)} + 2TOPO_{(org)} \overset{K_B}{\rightleftharpoons} Pu(NO_3)_4 \cdot TOPO_{(org)}$	10.08 ± 0.04
$Pu^{4+}_{(aq)} + 3NO_3^-{}_{(aq)} + HTTA_{(org)} + 2TOPO_{(org)} \overset{K_1}{\rightleftharpoons} Pu(NO_3)_3(TTA) \cdot 2TOPO_{(org)} + H^+_{(aq)}$	12.25 ± 0.06
$Pu^{4+}_{(aq)} + 2NO_3^-{}_{(aq)} + 2HTTA_{(org)} + TOPO_{(org)} \overset{K_2}{\rightleftharpoons} Pu(NO_3)_2(TTA)_2 \cdot TOPO_{(org)} + 2H^+_{(aq)}$	9.74 ± 0.10
$Pu^{4+}_{(aq)} + NO_3^-{}_{(aq)} + 3HTTA_{(org)} + TOPO_{(org)} \overset{K_3}{\rightleftharpoons} Pu(NO_3)(TTA)_3 \cdot TOPO_{(org)} + 3H^+_{(aq)}$	11.78 ± 0.06

TABLE 4

Equilibrium Constant Values for the Organic Phase Equilibria Derived from the Solvent Extraction Data: Diluent, Benzene[a]

Equilibrium	Log β
$Pu(NO_3)_4 \cdot 2TOPO_{(org)} + HTTA_{(org)} + TOPO_{(org)} \overset{\beta_1}{\rightleftharpoons} Pu(NO_3)_3(TTA) \cdot 2TOPO_{(org)} + HNO_3 \cdot TOPO_{(org)}$	3.00
$Pu(NO_3)_4 \cdot 2TOPO_{(org)} + 2HTTA_{(org)} + TOPO_{(org)} \overset{\beta_2}{\rightleftharpoons} Pu(NO_3)_2(TTA)_2 \cdot TOPO_{(org)} + 2HNO_3 \cdot TOPO_{(org)}$	1.32
$Pu(NO_3)_4 \cdot 2TOPO_{(org)} + 3HTTA_{(org)} + 2TOPO_{(org)} \overset{\beta_3}{\rightleftharpoons} Pu(NO_3)(TTA)_3 \cdot TOPO_{(org)} + 3HNO_3 \cdot TOPO_{(org)}$	4.19

[a]Errors in β values could not be given because errors in K_H (*10*) are not available.

$$Pu^{4+}_{(aq)} + NO^{-}_{3(aq)} + 3HTTA_{(org)} + TOPO_{(org)} \underset{}{\overset{K_3}{\rightleftharpoons}} Pu(NO_3)(TTA)_3 \cdot TOPO_{(org)} + 3H^{+}_{(aq)} \quad (22)$$

The equilibrium constants K_1, K_2, and K_3 are calculated from the relations $K_1 = K_B B_1$, $K_2 = K_B B_2$, and $K_3 = K_B B_3$, using the average B values and are given in Table 3.

The organic phase equilibria involving the stepwise replacement of nitrate in $Pu(NO_3)_4 \cdot 2TOPO$ by TTA^- can now be obtained by adding Eq. (5) to each of Eqs. (17), (18), and (19), and they are given in Table 4. The equilibrium constants representing these equilibria are obtained from the relations $\beta_1 = K_H B_1$, $\beta_2 = K_H{}^2 B_2$, and $\beta_3 = K_H{}^3 B_3$. It may be worthwhile verifying these organic phase equilibria by an independent method, and spectrophotometry appears to be a potential alternative.

Irving and Edgington have studied (*1*) the synergistic extraction of Pu(IV) from 1 *M* nitric acid by mixtures of HTTA and TBPO. Their data led them to infer that $Pu(NO_3)(TTA)_3 \cdot TBPO$ and $Pu(NO_3)_2(TTA)_2 \cdot 2TBPO$ are the species extracted in addition to $Pu(TTA)_4$. In the light of the present work, however, it is doubtful whether $Pu(NO_3)_2(TTA)_2 \cdot 2TBPO$ is formed at all. Apart from this, ignoring the aqueous phase nitrate complexing of Pu(IV) and the change in the concentration of HTTA in the organic phase due to the low distribution ratio of HTTA with cyclohexane as diluent (*12*) makes their values of different equilibrium constants uncertain.

It is felt that the participation of the anion from the aqueous medium in the synergistic mixed ligand complexes depends primarily on the extractability of the metal ion by the neutral donor alone from the aqueous medium used. When the neutral donor alone is able to extract the metal ion, a number of mixed ligand species would be involved in the extraction as by mixtures of TOPO and β-diketones. Because studies on such systems are meagre, we hope to continue investigating them.

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