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The Solvent Extraction of Heptavalent Technetium by Tributyl Phosphate

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ABSTRACT

The solvent extraction of heptavalent technetium from aqueous nitric or hydrochloric acid by tributyl phosphate in n-dodecane (TBP-NDD) has been studied over a wide range of TBP and acid concentrations at 25, 50, and 60°C. The extraction was found to proceed according to the reaction $3TBP + H^+ + TcO_4^- \rightarrow (HTcO_4 \cdot 3TBP)$. A discussion of possible reaction mechanisms is presented, along with values for ΔG, ΔH, ΔS, and the equilibrium constant for the extraction reaction. Finally, evidence for the coextraction of technetium by uranyl ions is discussed.

INTRODUCTION

Technetium-99 is a high yield [6.06% from thermal neutron fission of ^{235}U] (1), long-lived ($T_{1/2} = 2.12 \times 10^5$ y) fission product that, along with ^{129}I, makes up a dominant portion of the radioactive nuclides remaining in nuclear wastes after decay periods of 200 to 1000 y. A knowledge of the solvent extraction behavior of the element may suggest ways to isolate it and perhaps simplify the waste problem. The solvent extraction chemistry of technetium has been studied under a variety of conditions (1-5), but relatively little work has been done in systems resembling those used in treating nuclear fuel. In one investigation, carried out under conditions similar to those used in the Purex process (6), tributyl phosphate (TBP) was used to extract technetium from an aqueous UO_2^{2+}-TcO_4^--HNO_3 solution. However, only a limited range of conditions was studied. Although TBP has been used as an extractant for technetium (e.g., ref. 2) in a few other reported instances, there has been no comprehensive study of this system.

The present work was undertaken to examine the use of tributyl phosphate in n-dodecane (TBP-NDD) as an extractant for aqueous, heptavalent technetium under a wide range of conditions. Distribution coefficients as a function of temperature and the concentrations of TBP, HNO_3, HCl, and $UO_2(NO_3)_2$ were determined and analyzed. These data were used to establish a probable stoichiometry for the extraction equilibrium reaction and to calculate certain approximate thermodynamic properties of the solvent extraction process.

EXPERIMENTAL

Technetium

Technetium-99 was obtained as solid NH_4TcO_4 from Isotopes Sales, Oak Ridge National Laboratory. The salt was purified and converted to the potassium salt by dissolution in warm, ammoniacal H_2O_2, followed by the addition of KOH and heating to drive off NH_3 and precipitate $KTcO_4$. The solution was chilled in an ice bath, and the fine white crystalline precipitate was filtered and washed with cold absolute ethanol. Finally, the product was vacuum dried at 80°C in an Abderhalden drying apparatus.

Tributyl Phospate

Solutions of TBP-NDD were prepared as described previously (7). All TBP-NDD solutions were saturated with water during their preparation and remained so during their analysis and use.

Distribution Measurements

Nitric acid solutions of $KTcO_4$ were prepared by diluting 2.00 mL of 0.0495 $\underline{M}$ $KTcO_4$, along with an aliquot of 10.0 $\underline{M}$ HNO_3 sufficient to produce the desired final acid concentration, to exactly 25 mL with distilled water. A 20-mL volume of each solution was emulsified with an equal volume of TBP-NDD that had been preequilibrated with nitric acid of the same concentration, but containing no technetium. Note that when the water-saturated TBP-NDD solution was equilibrated with an aqueous solution of nitric acid, a small change in the volume of the TBP phase occurred as HNO_3 was transferred into and H_2O was transferred out of the organic phase. At high TBP and HNO_3 concentrations, this effect can produce a significant change in the volume of the organic phase, and hence in the TBP concentration. However, under the conditions of the experiments reported here, a maximum volume change of about 5% (usually much less) occurs. These small changes were neglected, and all TBP concentrations cited refer to the initial, water-saturated TBP-NDD solution before it was equilibrated with HNO_3 and $KTcO_4$. If desired, the final concentration of TBP in the organic phase may be calculated using the data of Davis (8-10).

Equilibrations were carried out in a water bath at 25, 40, and 60 ± 0.5°C for 20 min to ensure thermal and chemical equilibrium. Preliminary experiments had shown that identical results were obtained for samples withdrawn any time after 10 min. In those experiments containing uranyl nitrate, aliquots of a stock solution prepared by dissolving reagent-grade $UO_2(NO_3)_2 \cdot 6H_2O$ (Alfa-Ventron) in distilled water were added to the volumetric flask before dilution.

Studies of the $KTcO_4$-HCl-H_2O-TBP system were carried out as described above, except that HCl was substituted for HNO_3. It was necessary to dilute the stock solution of HCl to a concentration of less than 6 M before adding $KTcO_4$ to prevent reduction of the pertechnetate ion.

Analyses

TBP was analyzed by a modification of the dielectric constant method originally developed by Kelly et al. (11). Samples were washed once with 0.01 M Na_2CO_3 followed by two washings with distilled water. The dielectric constant of the sample was then measured and the TBP concentration determined by comparison with a calibration curve prepared by measuring the dielectric constants of a series of standard TBP-NDD solutions and fitting the data to a quadratic equation. The estimated error of the method is ±0.1 vol % TBP.

Technetium concentrations in aqueous and organic solutions were determined by beta counting using a Packard Model 2425 liquid scintillation spectrometer. Counting efficiency was greater than 80%. Most solutions required dilution prior to beta counting; acid solutions were diluted with water, and organic solutions were diluted with 30% TBP-dodecane. Quenching was minimized by counting small aliquots of the dilution. The liquid scintillation medium was Packard Instagel.

Uranium was determined by neutron activation analysis.

RESULTS AND DISCUSSIONS

Effect of Nitric Acid Concentration

Figures 1-3 show plots of D_{Tc} [i.e., $(Tc)_{organic}/(Tc)_{aqueous}$] as a function of nitric acid concentration at 25, 40, and 60°C. For any given TBP concentration, the distribution coefficient increases rapidly with increasing acidity until a maximum is reached between 0.6 and 1.0 M HNO_3. Above 1.0 M HNO_3, D_{Tc} falls off rapidly as competition from the significant amounts of nitric

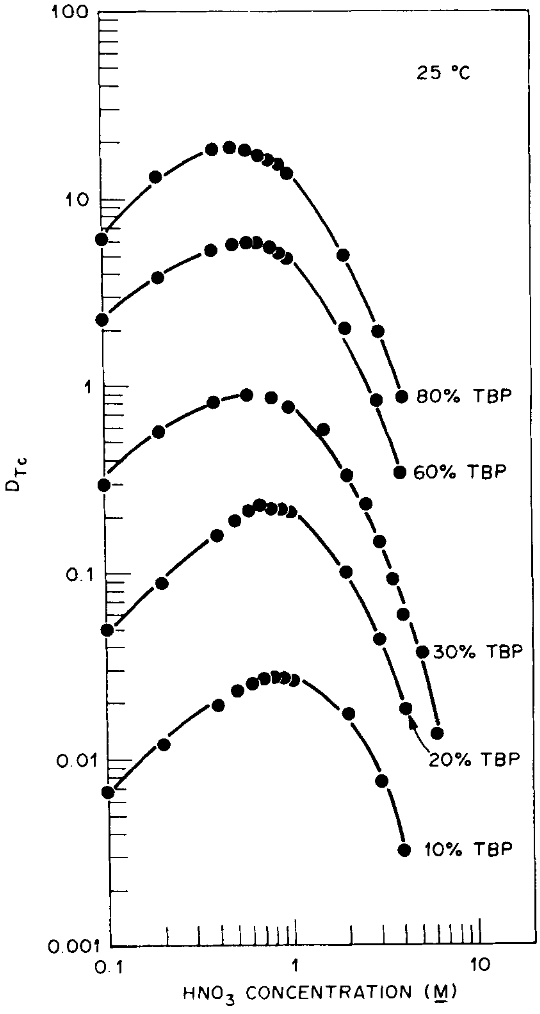


FIGURE 1. D_{Tc} as a function of HNO_3 concentration at 25°C.

acid present in the organic phase reduces the amount of free TBP available to extract technetium. The maximum seems to occur at slightly lower acidities as the TBP concentration increases and at slightly higher acidities as the temperature increases. These trends, while not very pronounced, follow the qualitative changes

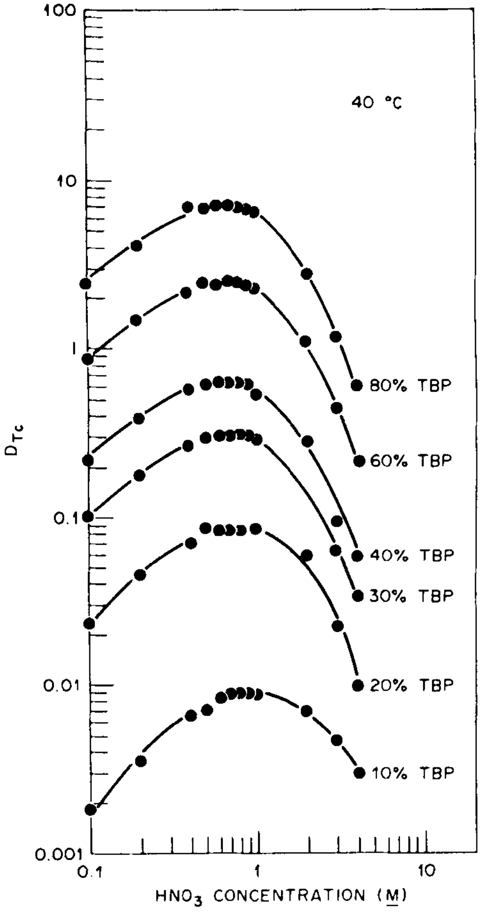


FIGURE 2. D_{Tc} as a function of HNO_3 concentration at 40°C.

in the distribution coefficient of nitric acid and hence the concentration of free TBP. However, the effect of temperature on the distribution coefficient is substantial. D_{Tc} decreases steadily as the temperature is increased.

Effect of Hydrochloric Acid Concentration

In contrast, Figs. 4-6 show the distribution behavior of technetium when hydrochloric acid is substituted for nitric acid

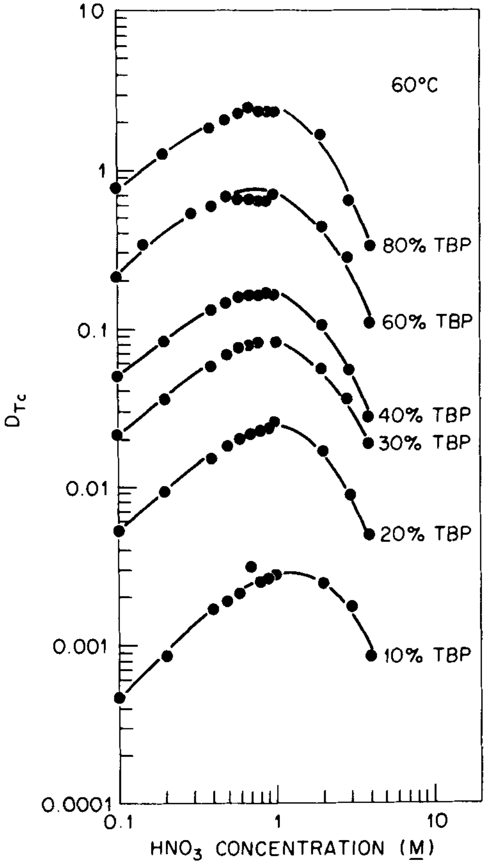


FIGURE 3. D_{Tc} as a function of HNO_3 concentration at 60°C.

under the same conditions of temperature and TBP concentration. The distribution coefficient again decreases as the temperature is increased but increases smoothly as the HCl or TBP concentration is increased. Since hydrochloric acid does not extract appreciably under these conditions, corrections for the amount of TBP complexed by the mineral acid are negligible.

Figure 7 shows that plotting D_{Tc} as a function of the mean molar activity of hydrochloric acid (12) at 25°C yields linear

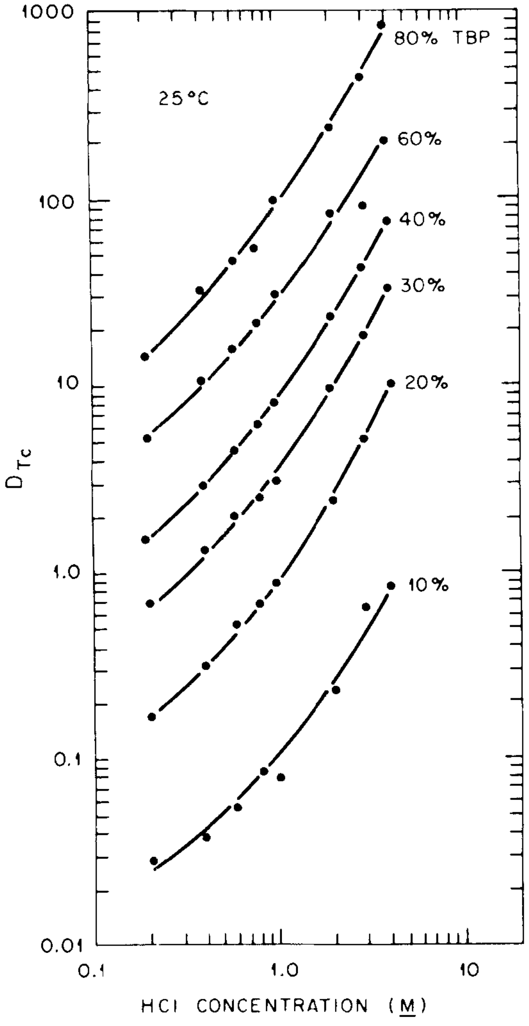


FIGURE 4. D_{Tc} as a function of HCl concentration at 25°C.

curves with a slope of 1 under all conditions studied. This suggests that a single proton is involved in the extraction reaction and that, under these conditions, competition between $HTcO_4$ and HCl for TBP is negligible. Implicit in this treatment is the assumption that the ratios of the activity coefficients of

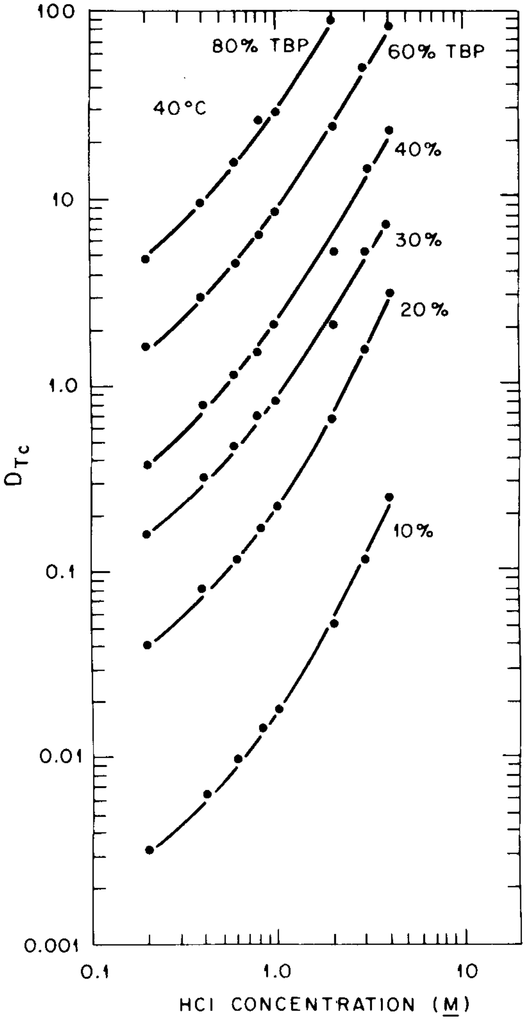


FIGURE 5. D_{Tc} as a function of HCl concentration at 40°C.

the aqueous and organic technetium species are constant in these solutions. The activity coefficient of TBP at a given concentration is also assumed to be unchanged by the extraction of small amounts of technetium and HCl.

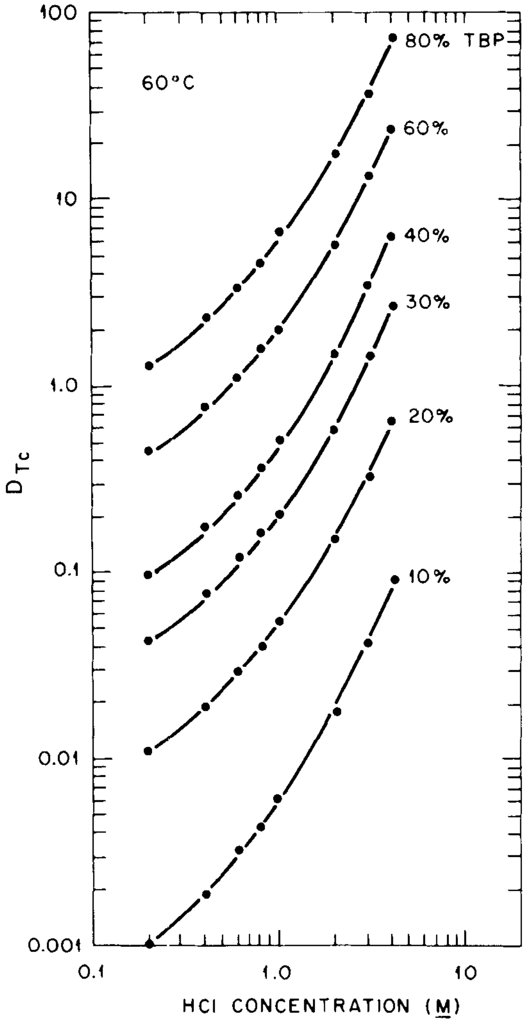


FIGURE 6. D_{Tc} as a function of HCl concentration at 60°C.

Effect of TBP Concentration

Typical plots of D_{Tc} as a function of TBP concentration in the nitric acid system are shown in Fig. 8. The slopes of these and similar lines that may be constructed from the data in Figs. 1-3 have an average value of 3.1 ± 0.2, suggesting that three TBP

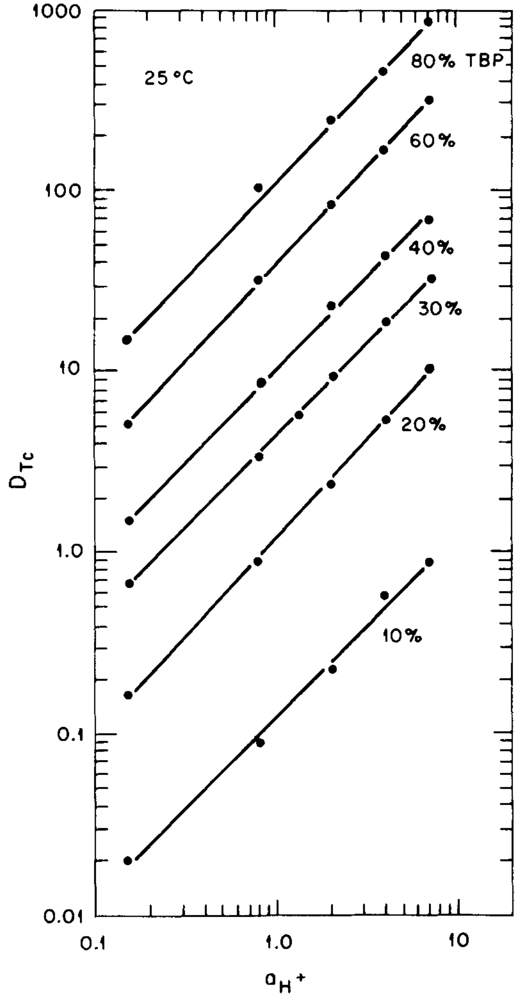


FIGURE 7. D_{Tc} as a function of the mean molar activity of HCl at 25°C.

molecules are involved in the extraction of pertechnetate. If the distribution data for the hydrochloric acid system (Figs. 4-6) are treated in the same way, a similar family of curves is generated with an average slope of 3.2 ± 0.1.

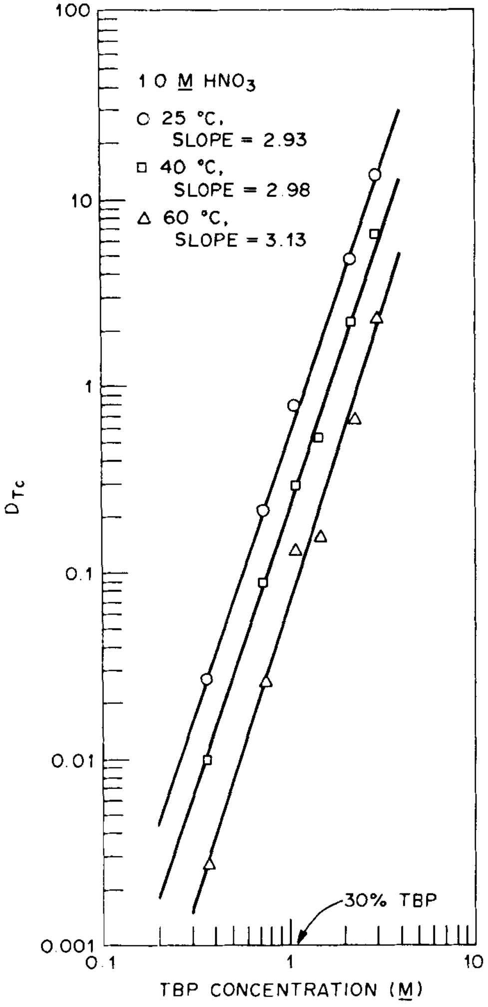


FIGURE 8. D_{Tc} as a function of TBP concentration.

Extraction Reaction Stoichiometry

The data presented above suggest that the following equilibrium predominates in the extraction of pertechnetate ion from acid solution by TBP:

$$H^+ + TcO_4^- + 3TBP \rightarrow (HTcO_4 \cdot 3TBP)_{organic} \quad . \qquad (1)$$

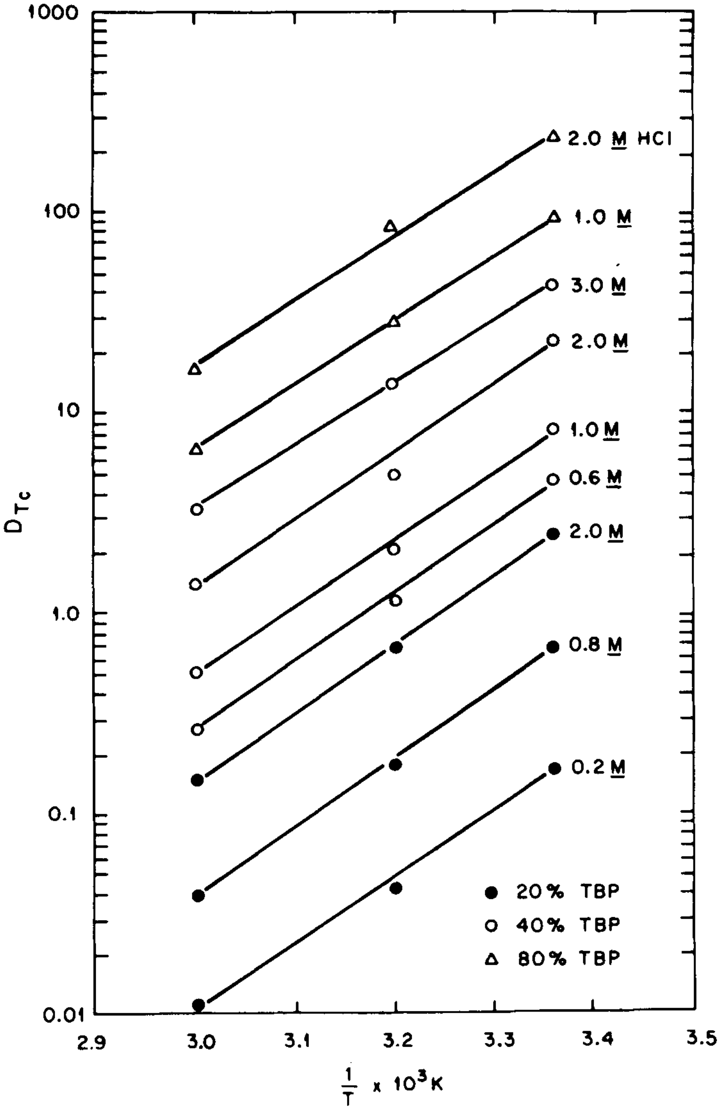


FIGURE 9. D_{Tc} as a function of the inverse absolute temperature in the HCl system.

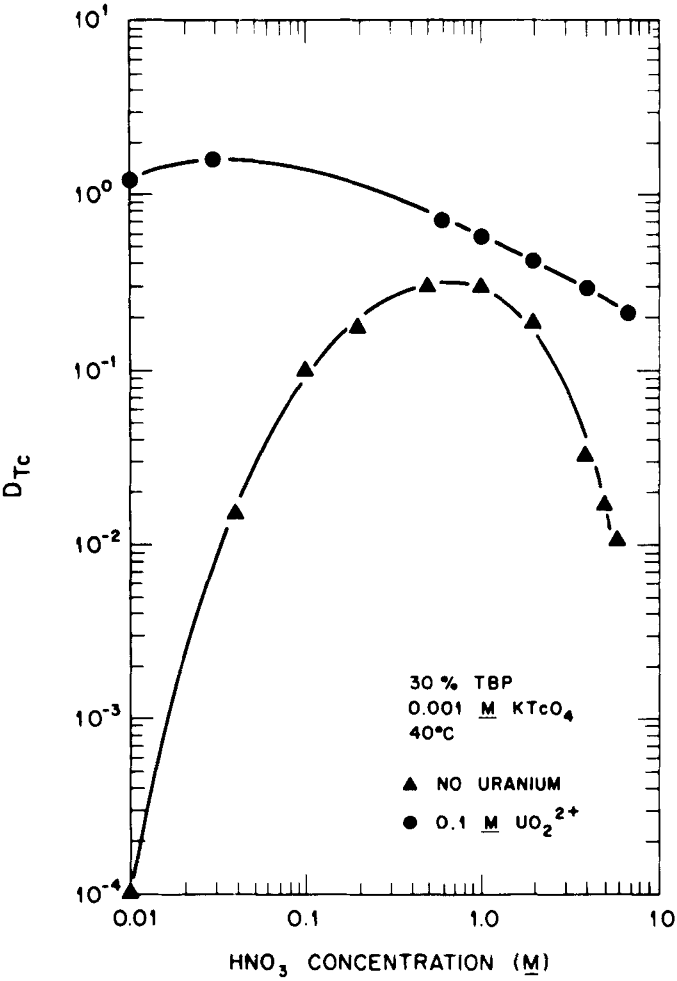
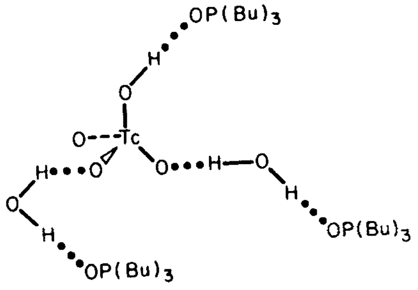


FIGURE 10. The effect of uranyl ion on D_{Tc}.

This stoichiometry has been suggested for pertechnetate (6), perrhenate (13), and perchlorate although Kertes (14) reported fourth-order TBP dependence for perrhenate and up to a fourth-order dependence for perchlorate (15). The large, negative entropy calculated for this reaction (see below) indicates that several water molecules are also involved in the organic phase complex. In the organic phase, both TBP and TBP monohydrate are available to form coordination complexes. Consideration of these facts and an examination of molecular scale models suggest that the organic phase complex may be similar to:



Note that the fourth oxygen atom of $HTcO_4$ is also available to coordinate to a TBP monohydrate complex. Although this site is somewhat obscured in the tri-TBP complex, it can be reached; and the resulting $HTcO_4 \cdot TBP \cdot (TBP \cdot H_2O)_3$ complex, while somewhat more crowded, does not appear to be impossible. Indeed, the existence of this higher TBP solvate may account for the fact that plots of D_{Tc} versus TBP concentration in the HCl system produce lines whose slopes are consistently greater than 3.0 (3.08 to 3.48). Similar complexes may account for the reported $HReO_4 \cdot 4TBP$ and $HClO_4 \cdot 4TBP$ complexes.

Calculation of Equilibrium Quotients

Nitric acid system - The concentration equilibrium quotient expression for Eq. (1) may be written as:

$$K_c = \frac{[HTcO_4 \cdot 3TBP]}{[TcO_4^-][TBP]^3[H^+]}, \qquad (2)$$

where $[HTcO_4 \cdot 3TBP]$ is equated with the measured concentration of technetium in the organic phase. Unfortunately, the available activity coefficient data needed to calculate the true, ther-

TABLE 1

Calculated Values of K^{α}_{c} for the Extraction of $HTcO_4$ from Nitric Acid (10 vol % TBP, 25°C)

$[HNO_3]$(<u>M</u>)	K^{α}_{c}	$[HNO_3]$(<u>M</u>)	K^{α}_{c}
0.1	1.27	0.8	0.89
0.2	1.16	0.9	0.86
0.4	1.02	1.0	0.82
0.5	0.97	2.0	0.77
0.6	0.95	3.0	0.81
0.7	0.91	4.0	0.89

modynamic equilibrium constant for this system are quite limited, especially at temperatures other than 25°C. Nonetheless, a reasonably constant value for K_c can be obtained by inserting the known stoichiometric concentrations of each species into Eq. (3). Further, an improved expression can be developed by taking advantage of the known activity coefficients of nitric acid and hydrochloric acid.

In the nitric acid system at 25°C, Eq. (2) can be modified to take into account the degree of dissociation (α) of nitric acid (16) and to correct for free TBP effects (17):

$$K^{\alpha}_{c} = \frac{[HTcO_4 \cdot 3TBP]}{[TcO_4^-][TBP]^3_{free}\alpha[HNO_3]} \quad (3)$$

Table 1 shows some typical values of K^{α}_{c} calculated from Eq. (3). For a given TBP concentration, the values of K^{α}_{c} are somewhat higher at low acid concentration, pass through a minimum near 2.0 <u>M</u>, and then rise again slightly.

For the data in Fig. 1, the average value of K^{α}_{c} is found to be 0.97 ± 0.45. Large deviations from this value occur at high TBP concentrations and low acid concentrations, resulting in a large standard deviation on the value for K^{α}_{c}. For example, in 0.2 <u>M</u> HNO_3 using 80 vol % TBP, a value of 2.51 is obtained for K^{α}_{c}. These deviations, which are similar to those observed in a study of the ReO_4^- system (14), are apparently due to changes in the activity coefficients of the various components of the system.

Unfortunately, no data are available to correct for the concentration of free TBP or the degree of dissociation of HNO_3 at elevated temperatures. However, such corrections should be small at nitric acid concentrations below 0.8 <u>M</u>, and using uncorrected values for [TBP] and [H^+] in Eq. (2) yields values of 0.42 ± 0.16 and 0.11 ± 0.05 for K_c at 40 and 60°C.

Hydrochloric acid system - In the HCl system, free corrections are negligible, and values of K_c can be calculated directly from Eq. (2) by equating [H^+] with the stoichiometric hydrochloric acid concentration. Average values of K_c at 25, 40, and 60°C were 3.4 ± 1.8, 0.89 ± 0.49, and 0.23 ± 0.14, respectively. As in the nitric acid system, large deviations from the average value occur at high acid and TBP concentrations.

For those concentrations where activity data are available, a mixed concentration-activity equilibrium constant, K'_c, may be calculated by substituting the mean molar hydrochloric acid activity for the stoichiometric hydrochloric acid concentration in Eq. (2). The average value of K'_c obtained in this way is 3.4 ± 0.8 at 25°C. Apparently, the variation in the values of K_c that lead to the large standard deviation is due mainly to activity effects, since using even a limited amount of activity data in Eq. (2) results in the much more constant value of K'_c. A typical set of values for K_c and K'_c is shown in Table 2 for 30% TBP at 25°C.

Similar results are obtained when K_c and K'_c are calculated at 40 and 60°C. These values are summarized in Table 3, along with average values of ΔG obtained from the relationship $\Delta G_c = -RT \ln K_c$ (or $\Delta G'_c = -RT \ln K'_c$).

Enthalpies and Entropies

Hydrochloric acid system - From the general relationship $\ln K = -(\Delta H/RT) + C$, it can be easily shown that when [TBP] and [H^+] are

TABLE 2

Calculated Values of K_c and K'_c for the Extraction of $HTcO_4$ from Aqueous HCl (30% TBP, 25°C)

HCl	K_c	K'_c
0.2	2.67	3.49
0.4	2.51	--
0.6	2.57	--
0.8	2.54	--
1.0	2.61	3.23
2.0	3.72	3.69
3.0	4.99	3.79
4.0	6.54	3.71

TABLE 3

Average Values of K_c, K'_c, ΔG_c, $\Delta G'_c$, ΔS_c, and $\Delta S'_c$ for the Extraction of $HTcO_4$ from Aqueous HCl

T (°C)	K_c	ΔG_c (kJ/mole)	ΔS_c (J/mole K)	K'_c	$\Delta G'_c$ (kJ/mole)	$\Delta S'_c$ (J/mole K)
25	3.4±1.8	-0.66±0.24	-47.8±0.8	3.4±0.8	-0.68±0.17	-47.6±0.5
40	0.89±0.49	0.17±0.34	-48.1±1.0	0.87±0.32	0.13±0.23	-48.0±0.7
60	0.23±0.14	1.1±0.34	-47.9±1.0	0.23±0.07	1.0±0.20	-47.8±0.6

held constant, $\ln D_{Tc} = -(\Delta H/RT) + C'$, and semilogarithmic plots of log D_{Tc} versus 1/T such as those shown in Fig. 9 should be linear with slope $-(\Delta H/2.3\ R)$. The data in Figs. 4-6 may be used to construct 48 such plots, which yield an average ΔH value of -62.3 ± 4.2 kJ/mole. Using this average value of ΔH, ΔS_c (or $\Delta S'_c$) may be calculated from the relationship $\Delta S_c = (\Delta H/T + R \ln K_c$ [or $\Delta S'_c = (\Delta H/T + R \ln K'_c)$]. The average values of ΔS_c and $\Delta S'_c$ calculated in this way are shown in Table 3.

It should be noted that use of the average value of K_c or K'_c to calculate ΔG_c or $\Delta G'_c$ (and subsequently ΔS_c and $\Delta S'_c$) gives

TABLE 4

Summary of Values for K_c, ΔH, and ΔS in the HCl System

T (°C)	K_c	ΔH (kJ/mole)	ΔS (J/mole K)
25	0.97 ± 0.45^a	0.075 ± 0.8	-197 ± 25
40	0.42 ± 0.16^b	2.3 ± 1.4	193 ± 13
60	0.11 ± 0.05^b	6.1 ± 1.7	193 ± 14

$^aK^{\alpha}_c$ from Eq. (3).
^bK_c from Eq. (2).

slightly different values for the free energy and entropy changes than are shown in Table 3. This difference arises because taking the logarithm of each calculated value of K and then averaging is not mathematically equivalent to taking the logarithm of an average value for K. In an ideal data set, this difference would not occur.

It should also be noted that the average values of the various parameters are essentially the same when calculated with or without the use of the available activity coefficient data. However, the scatter in the values, as measured by the standard deviation of the average, is substantially reduced when the activity coefficients are used.

Nitric acid system - A similar treatment of the data from the nitric acid system (Figs. 1-3) can be carried out but is hampered by a lack of activity data and data to correct for the concentration of free TBP at elevated temperatures. Nonetheless, an approximate value for ΔH can be obtained in the same way as in the hydrochloric acid system. Linear plots of D_{Tc} versus $1/T$ are obtained for nitric acid concentrations up to 1.0 M and TBP concentrations up to 60 vol % (2.2 M), and an average value for ΔH of -58.2 ± 3.1 kJ/mole may be calculated. Using the average value of

0.97 for K^{α}_c at 25°C, a value of 75 ± 800 J/mole is obtained for ΔG, and ΔS is calculated to be -197 ± 25 J/mole°K. The large, negative entropy term obtained in each acid system is consistent with the large amounts of ordering that must occur as the five molecules and ions on the left-hand side of Eq. (1) condense into a single complex species and as the structure-breaking oxyanions are removed from the aqueous phase and allow the structure of the solvent to reform. Table 4 summarizes the values of the various parameters calculated for the HNO_3 system.

Derivation of a Correlative Equation for the Hydrochloric Acid System

Ideally, the true thermodynamic equilibrium constant for Eq. (1) could be calculated using Eq. (2) along with the appropriate activity coefficients. This constant would be independent of the type of acid used to supply the protons for the reaction. This is clearly not the case for the "constants" calculated here, as a comparison of the values in Tables 3 and 4 will show. However, this treatment does have considerable predictive value, especially for the less complicated HCl system. Substituting D_{Tc} for $[HTcO_4 \cdot 3TBP]/[TcO_4^-]$ in Eq. (3) and taking logarithms, it is easily shown that

$$\ln D_{Tc} = \ln K_c + \ln [H^+] + 3 \ln [TBP] \ . \qquad (4)$$

But since $\Delta G = -RT \ln K = \Delta H - T\Delta S$, then $\ln K = (\Delta S/R) - (\Delta H/RT)$ and, using the calculated values of ΔS and ΔH,

$$\ln D_{Tc} = -24.1 + 7500/T + \ln [H^+] + 3 \ln [TBP] \ . \qquad (4)$$

This equation can be used to calculate an approximate value for D_{Tc} at any given temperature, acid concentration, and TBP concentration in the hydrochloric acid system. However, an improved equation can be obtained by allowing ΔS, ΔH, and the power dependences of $[H^+]$ and $[TBP]$ in Eq. (2) to act as parameters in the general equation,

$$\ln D_{Tc} = a + b/T + c \ln [H^+] + d \ln [TBP] \ . \qquad (5)$$

The multiple linear regression program in the statistics package of a Hewlett-Packard 41C calculator was used to fit this equation

TABLE 5

Selected Calculated and Experimental Values for D_{Tc} in the HCl System

[HCl] (M)	[TBP] (M)	T (K)	$D_{expt.}$	$D^{a}_{calc.}$	Difference (%)
1.0	0.366	298	0.230	1.199	-10
3.0	0.366	298	0.401	0.403	+ 1
0.8	2.20	298	22.4	24.1	+ 8
0.2	2.93	298	14.9	6.8	-54
0.2	0.732	313	0.223	0.243	+ 9
2.0	2.20	313	24.6	25.1	+ 2
0.2	2.93	313	4.75	4.60	- 3
1.0	1.47	333	0.521	0.615	+18
0.2	2.93	333	1.31	1.15	-12

Average difference for all data: -4.8%

[a] From Eq. (7).

to the experimental values for D_{Tc}, TBP concentration, and the mean molar activity of hydrochloric acid, yielding eq.(6),

$$\ln D_{Tc} = -23.0 + (7160/T) + 1.05 \ln a_{\pm}^{HCl} + 3.37 \ln [TBP] \quad . \quad (6)$$

The values of a, b, c, and d are not drastically different from their "theoretical" values in Eq. (4) but generally yield estimates values of D_{Tc} much closer to the experimentally determined results. As shown by the examples in Table 5, the calculated values are often quite close to the experimental values, but can be substantially different at high acid or TBP concentrations. In extreme cases, the calculated D_{Tc} may be in error by as much as 60%, but more typically it will be within 5 to 15% of the experimental value.

Attempts to correlate the data from the nitric acid system were unsatisfactory except over very narrow ranges of conditions.

Effects of Adding Uranyl Nitrate

The final parameter affecting technetium extraction by TBP that was included in this study was the addition of uranyl nitrate to the aqueous phase. Figure 10 shows some values of D_{Tc} at various nitric acid concentrations when 0.1 M $UO_2(NO_3)_2$ is added to the aqueous phase compared to those with no uranium in the system. Under most conditions, D_{Tc} is substantially increased. Apparently, a pertechnetate anion is substituted for a nitrate anion in the extraction of the uranyl ion (6,18):

$$UO_2^{2+} + NO_3^- + TcO_4^- + 2TBP \rightarrow UO_2(NO_3)(TcO_4)\cdot 2TBP \quad . \qquad (7)$$

Efforts are currently being made to further quantify and explain this effect.

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