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The Solvent Extraction of Heptavalent Technetium and Rhenium by Tributyl Phosphate

D. J. Pruett

CONSOLIDATED FUEL REPROCESSING PROGRAM

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THE SOLVENT EXTRACTION OF HEPTAVALENT TECHNETIUM AND RHENIUM BY TRIBUTYL PHOSPHATE

D. J. Pruett

Chemical Technology Division

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THE SOLVENT EXTRACTION OF HEPTAVALENT TECHNETIUM AND RHENIUM BY TRIBUTYL PHOSPHATE

D. J. Pruett

ABSTRACT

The literature pertaining to the solvent extraction of heptavalent technetium and rhenium from aqueous solution by tributyl phosphate (TBP) has been compiled, critically evaluated, and supplemented with new data in some areas. The effects of adding mineral acids, alkali metal nitrates, alkali metal chlorides, uranyl nitrate, thorium nitrate, and plutonium(IV) nitrate to these systems were also examined. Discussions of the possible nature of the organic-phase complexes are presented, along with values of ΔG, ΔH, ΔS, and the equilibrium constant for the extraction reaction in several systems.

Mathematical models correlating the distribution behavior over a wide range of conditions were also developed. The distribution coefficients for the extraction of Re(VII) or Tc(VII) from 0.2 to 4 M HCl by 0.339 to 2.90 M TBP at 298 to 333 K may be calculated using the following equations:

$$\ln D_{Tc} = -23.9 + 7462/T + 1.03 \ln a_{\pm}^{HCl} + 3.26 \ln [\overline{TBP}] ,$$

$$\ln D_{Re} = -22.1 + 6684/T + 1.04 \ln a_{\pm}^{HCl} + 3.28 \ln [\overline{TBP}] ,$$

where T is the absolute temperature, $a_{\pm}^{HCl}$ is the mean molar aqueous HCl activity, and $[\overline{TBP}]$ is the concentration of TBP in the organic phase. Under the same conditions, but with HNO_3 (instead of HCl) present in the aqueous phase, the distribution coefficients may be calculated from:

$$\ln D_{Tc} = -2013 + 6013/T + 0.865 \ln [HNO_3] + 2.91 [\overline{TBP}]_f ,$$

$$\ln D_{Re} = -25.88 + 7534/T + 0.944 \ln [HNO_3] + 3.19 \ln [\overline{TBP}]_f ,$$

where $[HNO_3]$ is the stoichiometric, equilibrium HNO_3 concentration in the aqueous phase and $[\overline{TBP}]_f$ is the equilibrium concentration of "free" (uncomplexed) TBP in the organic phase.

For initial aqueous solutions containing 0.01 to 4.6 M HNO_3 and 0.2 to 1.0 M $UO_2(NO_3)_2$ or 0.05 to 0.20 M $Th(NO_3)_4$, D_{Tc} or D_{Re} at equilibrium may be calculated using the following equations:

$$\ln D_{Tc} = -0.548 - 54.4/T + 0.641 \ln [\overline{U}] - 0.879 \ln [NO_3^-] ,$$

$$\ln D_{Re} = -0.980 - 66.4/T + 0.523 \ln [\overline{U}] - 0.907 \ln [NO_3^-] ,$$

$$\ln D_{Tc} = -0.560 + 1398/T + 0.921 \ln [\overline{Th}] - 0.780 \ln [NO_3^-] ,$$

$$\ln D_{Re} = -2.03 + 1408/T + 0.814 \ln [\overline{Th}] - 0.806 \ln [NO_3^-] ,$$

where $[NO_3^-]$ is the total, stoichiometric aqueous nitrate concentration at equilibrium and $[\overline{U}]$ and $[\overline{Th}]$ are the equilibrium organic-phase concentrations of the heavy metals.

Extensive tables of the solvent extraction data used to derive these equations are included in the appendix. Miscellaneous data obtained under more restricted conditions from other solvent extraction systems are also discussed and correlated.

1. INTRODUCTION

In the nuclear fuel reprocessing industry, there is growing concern about impurities that are not completely removed from uranium and plutonium by traditional reprocessing technology. Technetium is one such impurity that, when sent back through the fuel cycle, can cause a variety of problems. For example, when technetium enters a gaseous diffusion plant along with reprocessed uranium, it can escape as a volatile fluoride and subsequently be found in low levels in the vegetation surrounding the plant. Incomplete technetium removal also leads to higher radiation levels throughout the fuel cycle. As part of the overall effort to understand this behavior, the chemistry of technetium (and a nonradioactive model element, rhenium) in the fuel cycle has been the subject of several studies in this laboratory.[1-4].

Historically, the study of the extraction of technetium and rhenium is almost 25 years old. The first major systematic investigation of the solvent extraction of heptavalent technetium was made by Boyd and Larson,[5] who used a wide variety of extractants, including TBP. At approximately the same time, Siddall[6] was examining the behavior of heptavalent technetium in the Purex process, giving special attention to the coextraction of technetium with uranium by TBP. Cambell[7] made a brief, laboratory-scale study of the extraction of pertechnetate under the conditions of a typical Purex flowsheet. A few years later, Spitsyn,[8] Zaitsev,[9] and their co-workers published studies of technetium extraction which included some data for TBP systems. Most recently, Macasek[10] and Lieser and his co-workers[11] have examined the solvent extraction behavior of technetium, both alone and in the presence of uranium. The range of conditions used in all of these studies was limited, and no comprehensive mathematical models were developed to correlate the distribution behavior as a function of acidity, TBP concentration, and temperature.

Early investigations of the solvent extraction of perrhenate from acid solution by TBP were carried out by Kertes and Beck,[12] Colton,[13] and Whitney and Diamond.[14] These investigators, working under diverse conditions, reached somewhat different conclusions concerning the solvation number of rhenium in the organic phase. Again, the range of parameters studied was limited in these studies, and no correlations of the data were developed.

It is the purpose of this report to tabulate and critically review the solvent extraction data on technetium and rhenium in TBP systems as found in the literature. Some new data are also included. As many of the actual experimental results as possible have been assembled as tables in the Appendix. Many of the results are also included in graphical form in the main body of the text in order to facilitate visual comparison and analysis. Mathematical models and correlations are developed whenever possible, noting that improved models will be developed as more activity coefficients become available and as new theories of solvent extraction behavior are proposed. The

objective of this report is to provide a convenient summary and compilation of the available data so that those in need of raw experimental results can obtain them quickly.

2. EXPERIMENTAL

2.1 ORIGINAL DATA

All of the experimental and analytical methods, procedures, and reagents used to study the solvent extraction behavior of rhenium and technetium in this laboratory have been fully described in previous publications.[1-4,15] Only a summary of some of the more critical points is presented here.

Acid solutions of KXO_4 (X = Tc or Re) were prepared by diluting an aliquot of a stock KXO_4 solution, along with an aliquot of 10.0 M HCl or HNO_3 sufficient to produce the desired final KXO_4 and acid concentrations, to exactly 25 mL with distilled water. When HCl was used to acidify the aqueous phase, it was diluted to a concentration below 6 M to avoid reduction of the metal anions. A 5-mL aliquot of each aqueous solution was retained for analysis, while the remainder (20 mL) was emulsified with an equal volume of TBP—n-dodecane (TBP—NDD) that had been prequilibrated with a solution of the same type and concentration of acid and contained no potassium perrhenate or pertechnetate.

Equilibrations were carried out 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 anytime after 10 min. In the experiments containing alkali nitrate or chloride salts, aliquots of a standard solution or weighed amounts of the dried salts were added to the volumetric flask before dilution.

It should be noted that small changes in volume, and hence the molar TBP concentration, occur when the organic phase is preequilibrated with HNO_3. This effect can be substantial at high TBP and HNO_3 concentrations. However, under the conditions of the experiments reported

here, the volume change, as calculated from the data in ref. 16, was almost always less than 2%; a maximum of about 5% was reached only when the HNO_3 concentration exceeded 2 M and the TBP concentration exceeded 60 vol %. These changes were neglected in the discussion that follows. For HCl solutions, the effect is even smaller since essentially no acid is extracted under the conditions reported here.

2.2 LITERATURE DATA

Unfortunately, the experimental conditions under which other investigators have examined the solvent extraction of technetium and rhenium have not always been fully documented. These conditions are described as completely as possible in the tables, figures, and text in which the data appear, with special note being made when key parameters (e.g., temperature) were not reported. When a comparison is made between sets of data that are not fully documented, the assumptions involved in making the comparison are stated explicitly in the text.

3. RESULTS AND DISCUSSIONS

3.1 GENERAL APPROACH TO THE ANALYSIS OF THE SOLVENT EXTRACTION DATA

Traditionally, the stoichiometry of a solvent extraction reaction has often been determined in the following way. Consider a general reaction,

$$aA + mM + sS \rightleftarrows [A_aM_m \cdot sS]_{org} \ , \qquad (1)$$

where S is the extractant, M is the ion being extracted (frequently a metal cation or anion), and A is any counterion or other species that may be involved in the organic-phase complex. The equilibrium constant expression for Eq. (1) may be written as

$$K = \frac{[A_aM_m \cdot sS]_{org}}{[A]^a[M]^m[S]^s} \cdot Q = \frac{D_M}{[A]^a[S]^s} \cdot Q \ , \qquad (2)$$

where Q is the quotient of the appropriate activity coefficients, D is the distribution coefficient ($D_M = [M]_{org}/[M]_{aq}$), and square brackets indicate molar concentration. Rearranging and taking logarithms yields

$$\log D_M = a \log [A] + s \log [S] + \log [K/Q] \ . \qquad (3)$$

If a number of distribution measurements are made under conditions such that the activity coefficients are close to 1 or are at least constant, and if the concentrations of all reagents except one are held constant, a plot of log D_m vs for example, log [S], will be linear with a slope of s. In practice, activity effects are often ignored because the activity coefficients have not been measured. In addition, the method may break down if an extracted species forms complexes of more than one stoichiometry in the organic phase. Nonetheless, when these plots are linear, they have proved to be a relatively simple, accurate way to determine the stoichiometries of extracting complexes. Indeed, this method frequently provides insight into extraction reactions even under conditions where the assumptions used in the method are clearly incorrect. The utility and weaknesses of this procedure will be illustrated throughout the following discussion.

3.2 EXTRACTION FROM HYDROCHLORIC ACID

3.2.1 Acid Dependence

Tables A.1-A.3 (the "A" prefix refers to the Appendix) summarize the distribution data obtained in this laboratory for rhenium and technetium between aqueous hydrochloric acid and TBP-NDD at 25, 40, and 60°C. Figures 1-6 show log-log plots of these distribution coefficients as a function of the mean molar hydrochloric acid activity ($a_{\pm} = [HCl]\gamma^{\pm}$). Mean molar activity coefficients (Table A.4) were calculated from the mean molal activity coefficients,[17] using published density data[18] and the measured molar concentrations of HCl. The distribution coefficients of both technetium and rhenium are seen to increase linearly with HCl activity, with a slope of very close to 1.

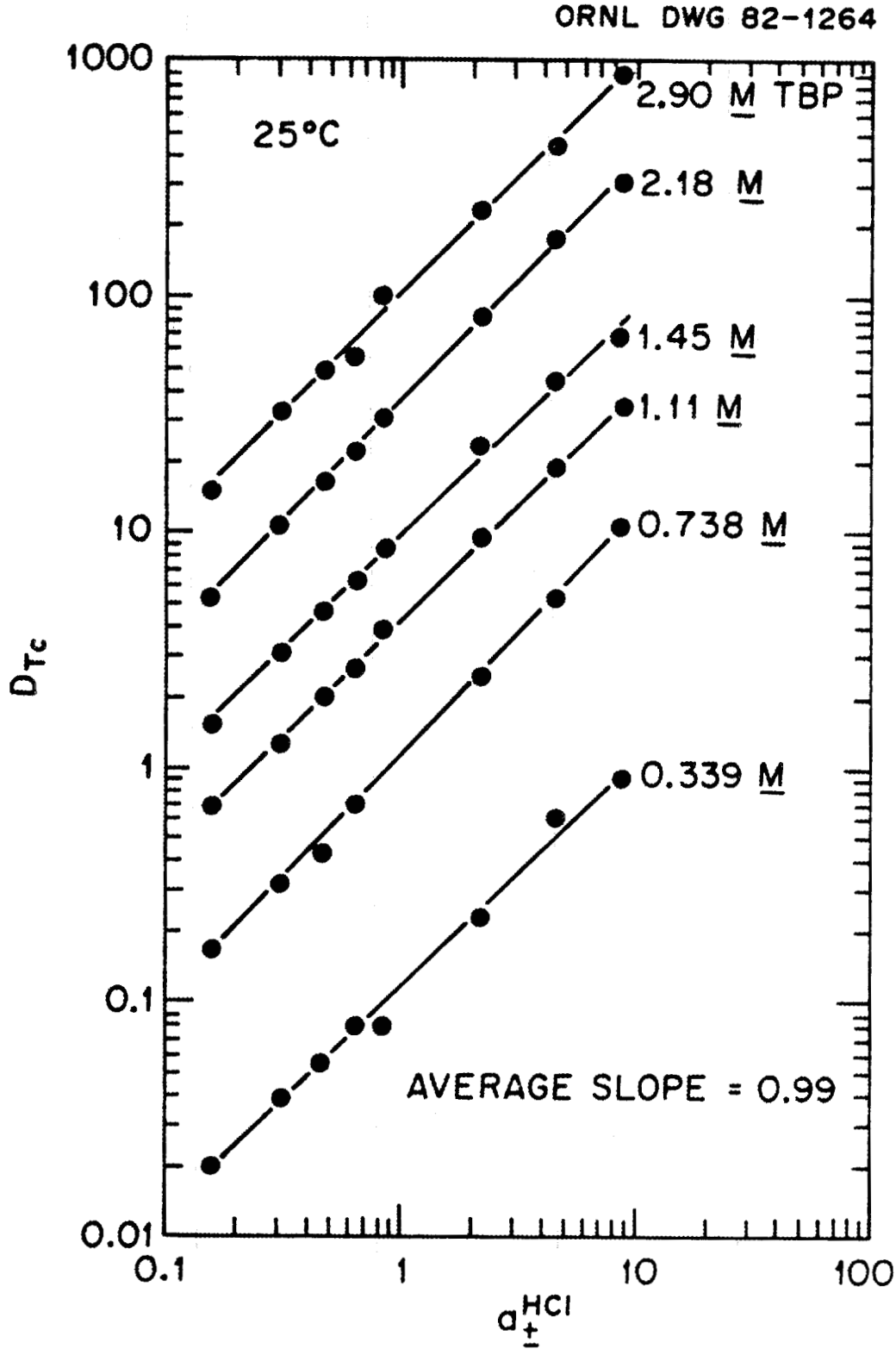

Fig. 1. Distribution of pertechnetate between aqueous HCl and TBP-NDD at 25°C.

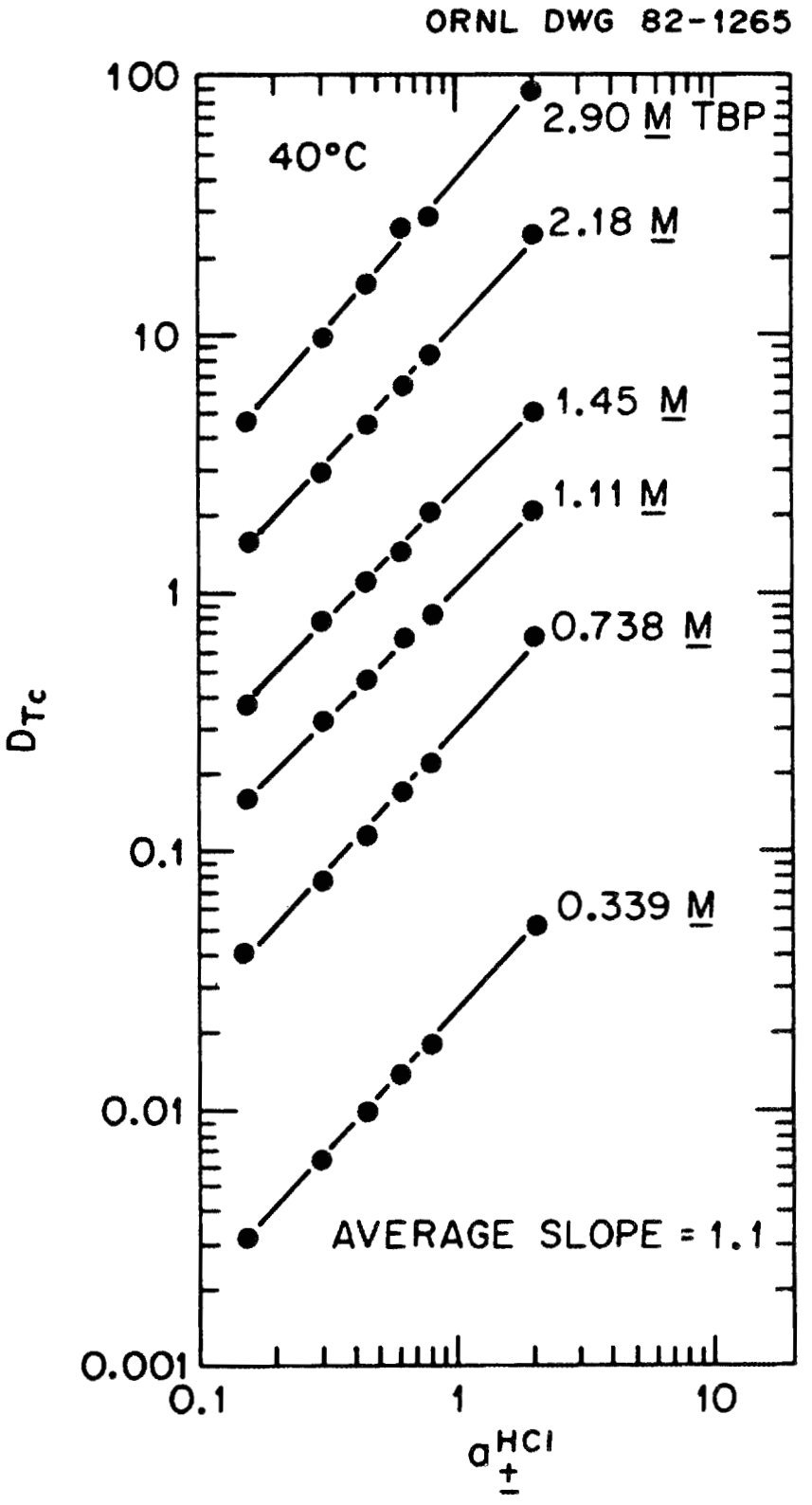

Fig. 2. Distribution of pertechnetate between aqueous HCl and TBP-NDD at 40°C.

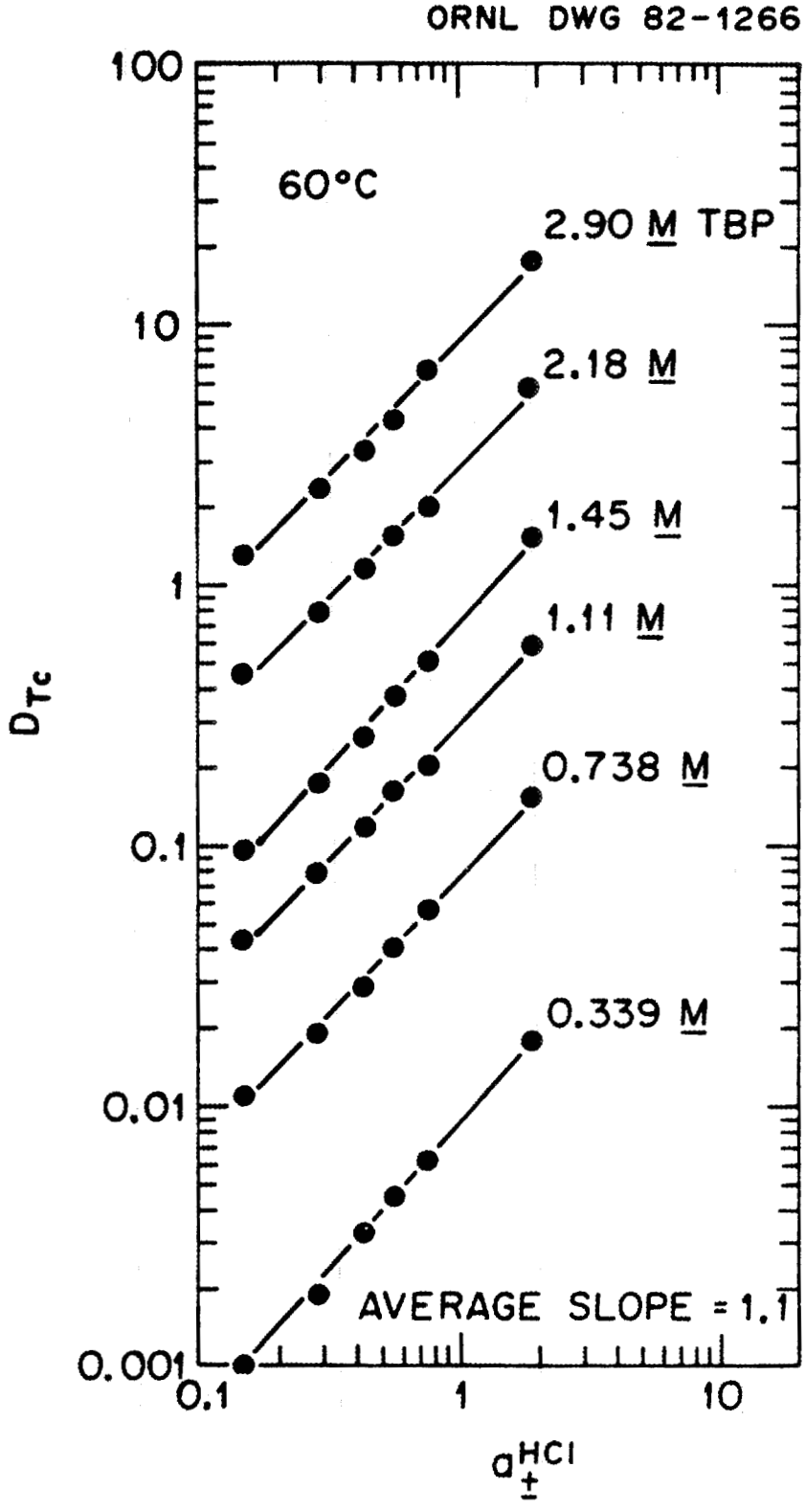

Fig. 3. Distribution of pertechnetate between aqueous HCl and TBP-NDD at 60°C.

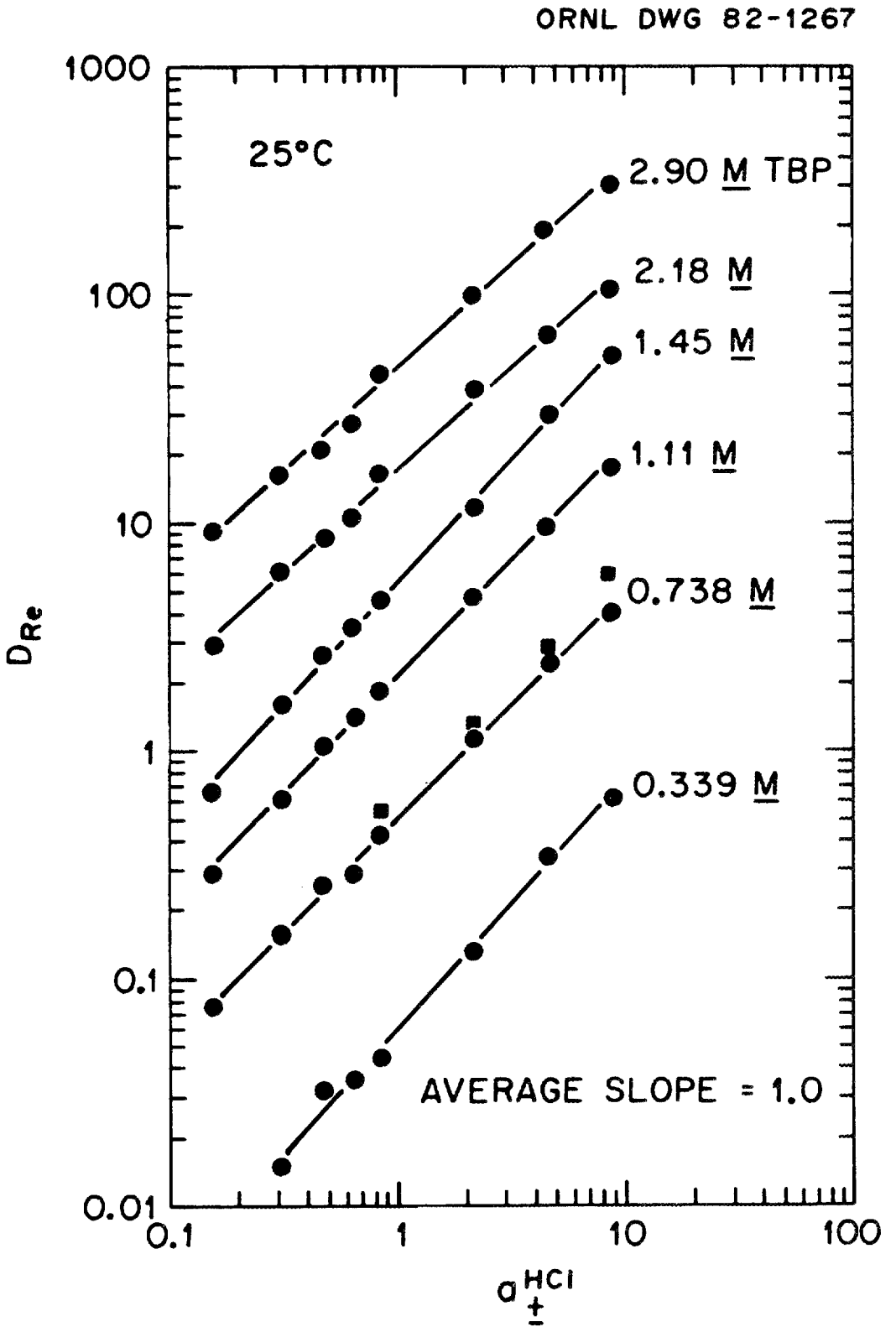

Fig. 4. Distribution of perrhenate between aqueous HCl and TBP-NDD at 25°C. = this work; = ref. 13.

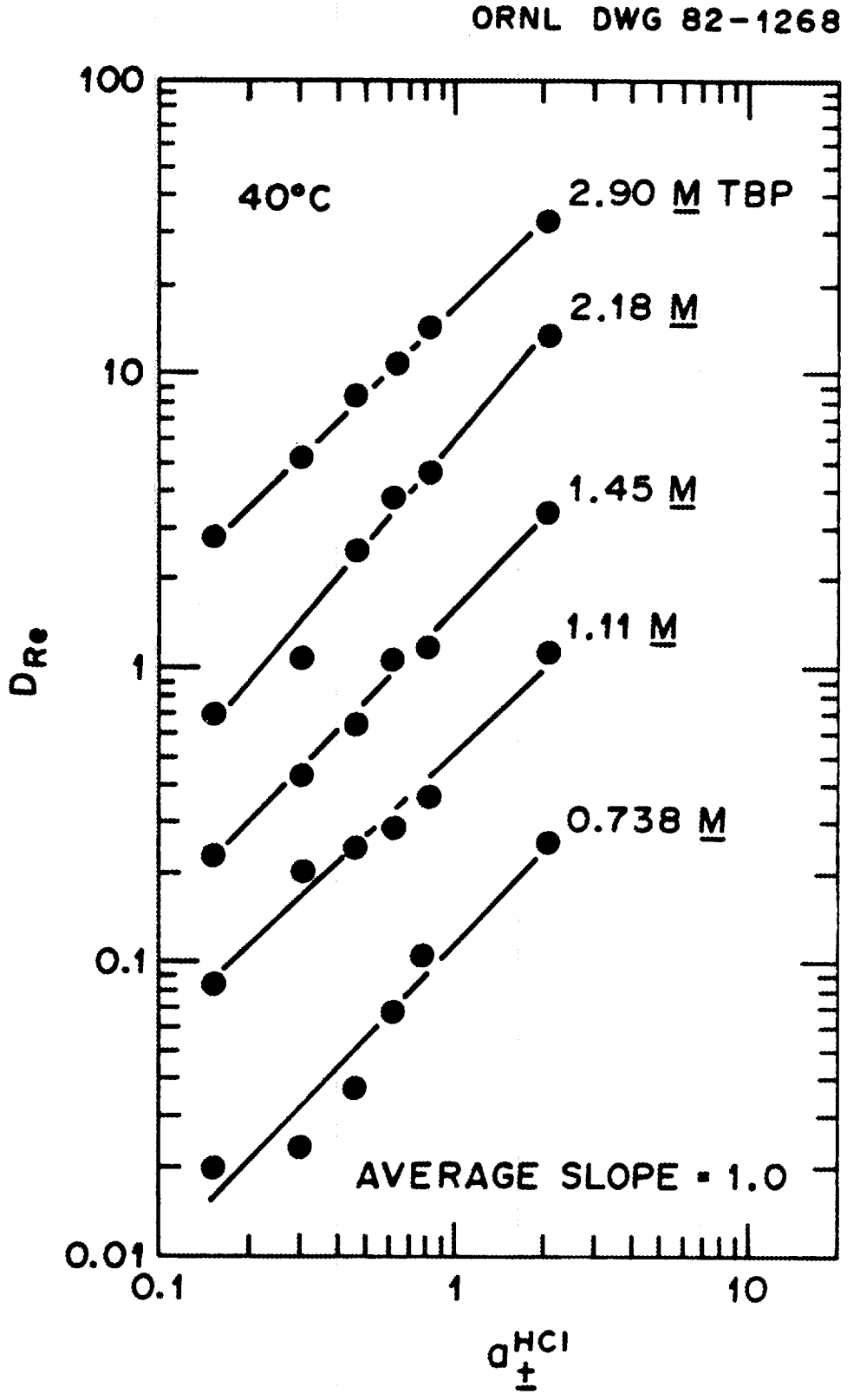

Fig. 5. Distribution of perrhenate between aqueous HCl and TBP-NDD at 40°C.

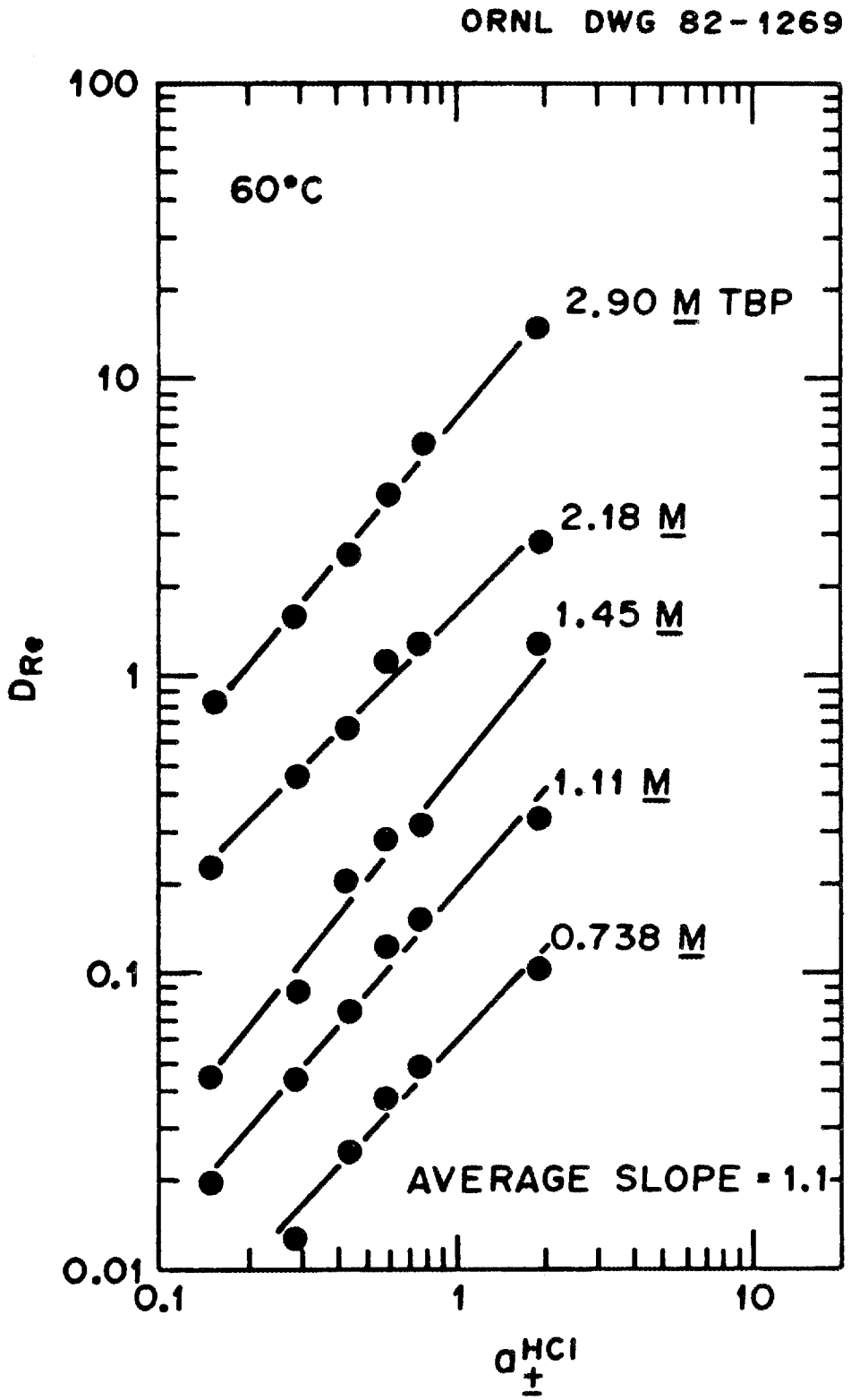

Fig. 6. Distribution of perrhenate between aqueous HCl and TBP-NDD at 60°C.

These data, obtained over a very wide range of conditions, confirmed the earlier conclusion[6,12] that perrhenate and pertechnetate extract from acid solution as their respective peracids, $HReO_4$ and $HTcO_4$. The same results were obtained, regardless of whether the anions were added as an acid or as a potassium salt.

Because TBP is most often used to extract metal values from HNO_3 solution, there are relatively few additional studies of perrhenate and pertechnetate extraction from HCl. Only Colton[13] reports distribution data for rhenium from HCl (Table A.5), which are shown in Fig. 4. Although no temperature was given for these data, the agreement with the other data in Fig. 4 is good, if if it can be assumed that the work was done at a room temperature of slightly less than 25°C. For technetium, only Boyd and Larson[5] report data on extraction from aqueous acid phases other than HNO_3 (Tables A.6-A.8). Their single data point for the extraction of pertechnetate from 1 M HCl with pure TBP at room temperature is consistent with the data in Tables A.1-A.3.

3.2.2 Tributyl Phosphate Dependence

Previous reports on the extraction of $HReO_4$ and $HTcO_4$ by TBP have presented conflicting values for the dependence of the distribution coefficient on TBP concentration. Most of these studies were done in HNO_3 systems, and generally a third-order[6,8,9,10,13,14] or fourth-order[11,12,19,20] dependence on the TBP concentration was reported. This ambiguity is at least partially a result of the difficulties that arise in interpreting the variation of D_{Re} and D_{Tc} as a function of TBP concentration in the presence of HNO_3, which itself competes strongly for coordination by the extractant. The effect of this competition on the amount of TBP available to extract perrhenic or pertechnic acid must be properly accounted for in order to successfully apply the method for determining solvation numbers described in Sect. 3.1. An acceptable method of making this correction is described in Sect. 3.3.1. There are several cases in the literature where these corrections have been made improperly[11,20] or not at all[12,19].

On the other hand, HCl is very poorly extracted by TBP, at least at low (<4 M) concentrations, and offers essentially no competition to $HReO_4$ oθ →ψ)$_4$ over a wide range of conditions. Thus, interpretation of the solvent extraction data is greatly simplified. Figures 7 and 8 show plots of log D_{Tc} and log D_{Re} vs log [TBP] from hydrochloric acid at 25°C (Table A.1). The plots show good linearity, with slopes very close to 3. This finding supports the work of previous investigators who claimed that a trisolvated pertechnetate or perrhenate complex was formed in the organic phase.

Unfortunately, the situation becomes less clear when the 40°C and 60°C data (Tables A.2 and A.3) are considered. Table 1 summarizes the slopes of the curves obtained from log D-vs-log [TBP] plots of these data. All of the slopes are significantly greater than 3, especially for the rhenium data, and approach 4 in some cases. The fact that so many of the values lie about halfway between 3 and 4, along with the inherent limitations of this method for determining solvation numbers, makes it difficult to unequivocably state the solvation number of these complexes on the basis of this analysis.

In reviewing all of these available data on $HReO_4/HTcO_4$ extraction, it becomes clear that the work of Whitney and Diamond (Fig. 9)[14] was carried out under conditions where this method of determining solvation numbers is most likely to yield a reliable result. Using pure solutions of $HReO_4$ (and $HClO_4$, which behaves similarly), low TBP concentrations, and low organic-phase loadings, they demonstrated that the extraction of $HReO_4$ (and $HClO_4$) clearly shows a third-order dependence on the organic-phase TBP concentration. These authors specifically sought evidence of a second- or fourth-order dependence but none was found. Considering all of the available data, it seems most likely that $HReO_4$ and $HTcO_4$ are extracted by TBP as a trisolvated complex. Variation in this result, based on simple slope analysis, is probably due to the inherent limitations of the technique and complications that arise when other extracting species are present.

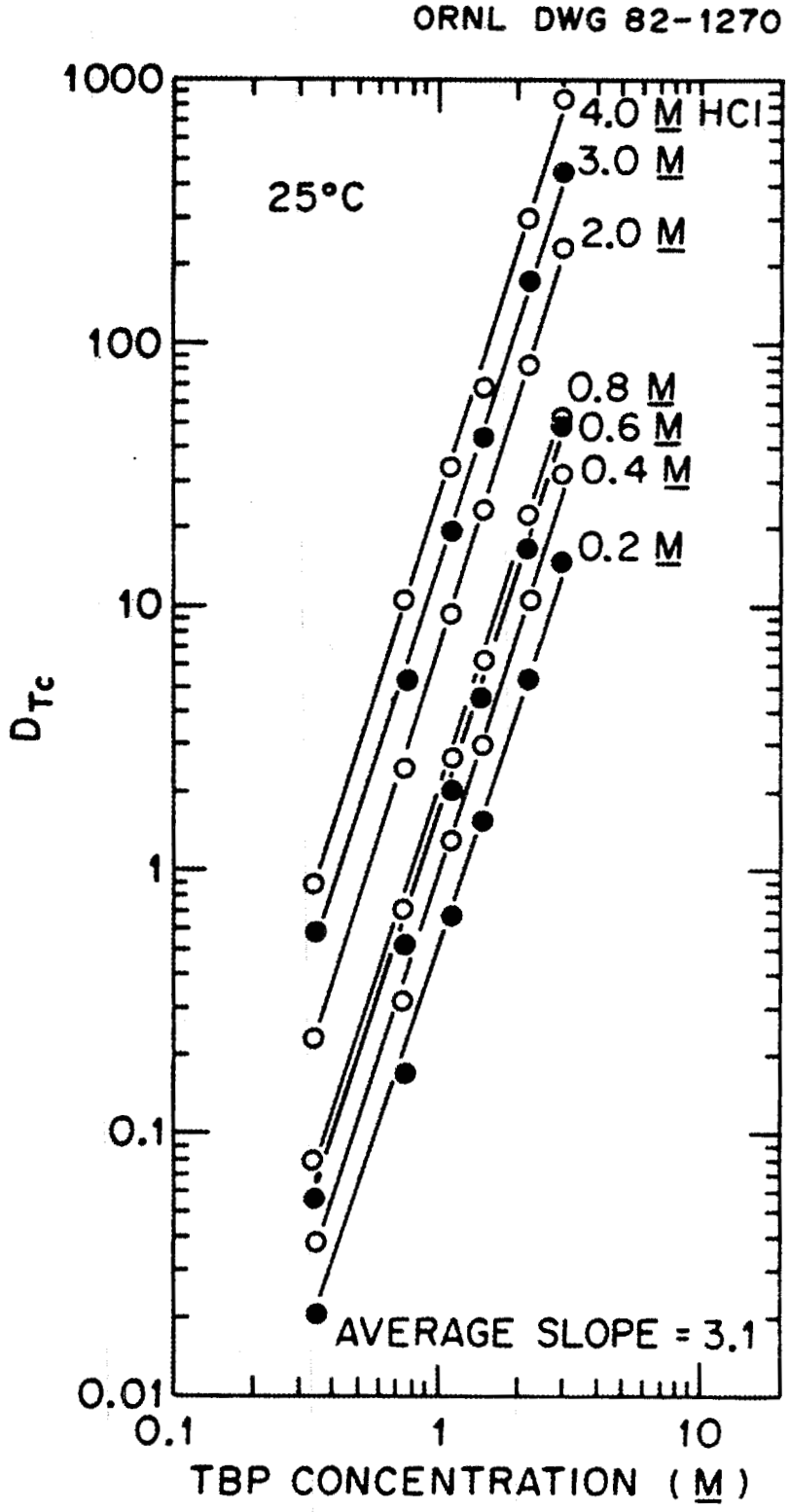

Fig. 7. Distribution of pertechnetate between HCl and TBP-NDD as a function of TBP concentration at various HCl concentrations at 25°C.

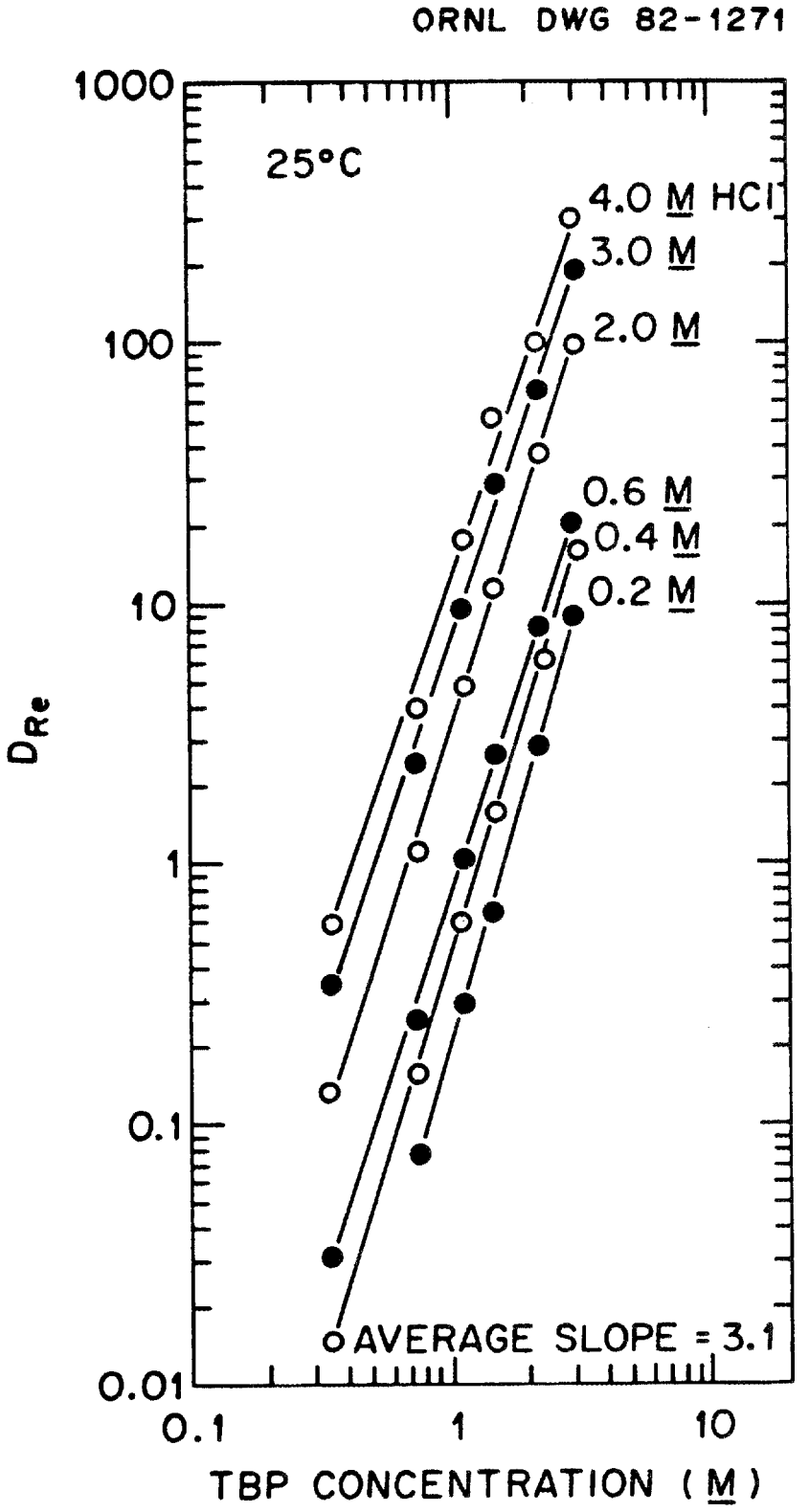

Fig. 8. Distribution of perrhenate between HCl and TBP-NDD as a function of TBP concentration at various HCl concentrations at 25°C.

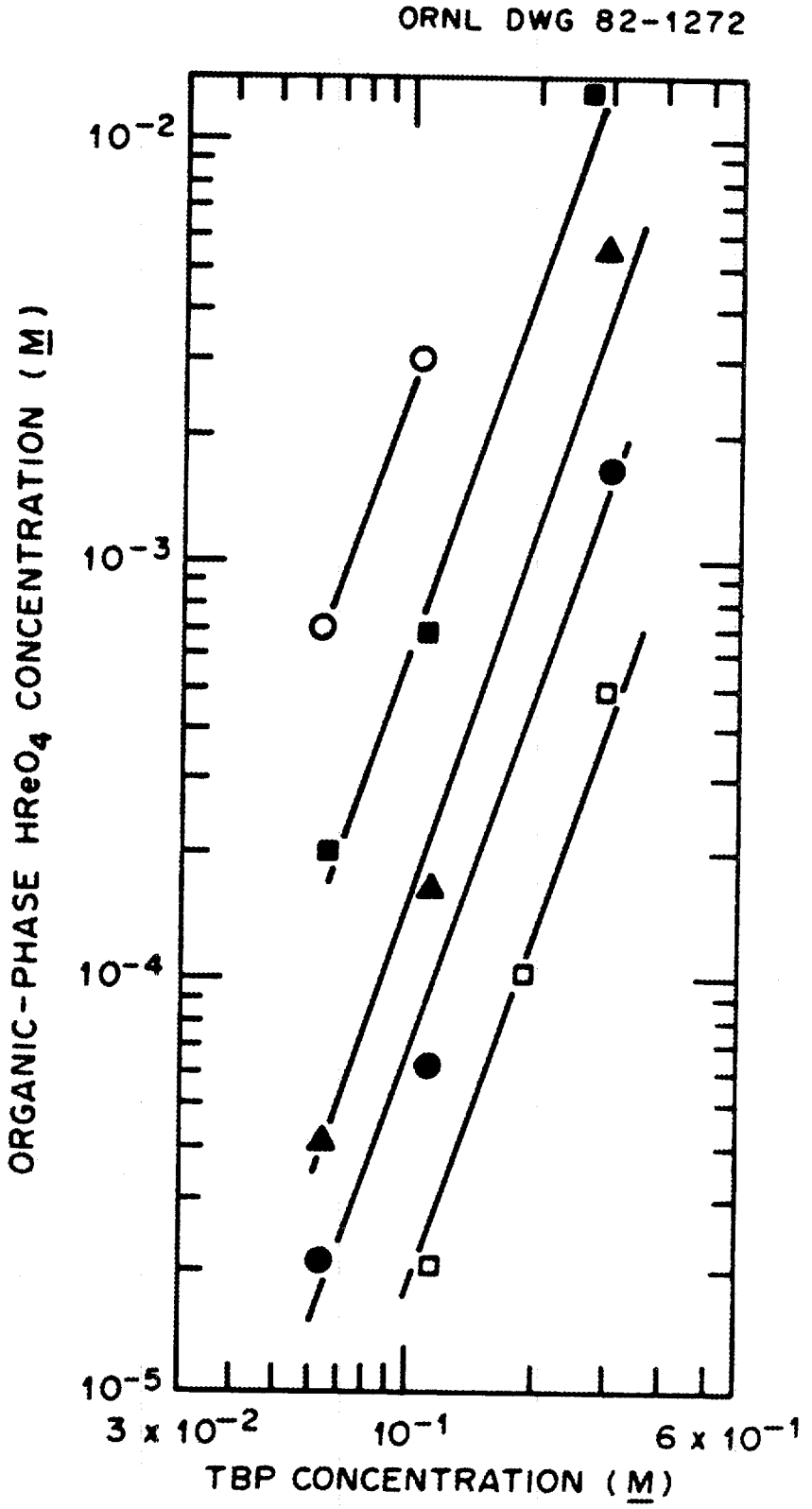

Fig. 9. Variation of perrhenic acid content of organic phase with equilibrium TBP concentration for aqueous $HReO_4$ concentrations of 0.109 M ( ), 0.217 M ( ), 0.43 M ( ), 0.87 M ( ), and 1.74 M ( ).

Table 1. TBP dependence of D_{Re} and D_{Tc} under various conditions as determined by the slopes of plots of log D–vs–log [TBP]

HCl concentration (M)	Temperature (°C)	Slope	
		Tc	Re
0.2	40	3.38	3.49
0.4	40	3.37	3.64
0.6	40	3.39	3.86
0.79	40	3.43	3.72
0.99	40	3.40	3.60
1.98	40	3.40	3.42
2.96	40	3.38	3.19
3.92	40	3.12	3.30
0.2	60	3.33	3.92
0.4	60	3.30	3.51
0.6	60	3.22	3.33
0.78	60	3.22	3.39
0.98	60	3.25	3.45
1.96	60	3.21	3.53
2.93	60	3.20	3.64
3.88	60	3.13	3.73

3.2.3 Effects of Chloride and Alkali Metal Ions

Previous studies have shown that Re(VII) is extracted from an acidic solution of its potassium salt only as perrhenic acid and that no potassium is found in the organic phase.[12] Further, when KCl (up to 3.0 M) is added to an $HCl-H_2O-HMO_4$-TBP solvent extraction system, the distribution coefficients of technetium and rhenium are essentially unchanged.[2,4] However, when univalent cations such as sodium or lithium, which themselves form moderately extractable pertechnetate and perrhenate salts,

are added to an aqueous HCl solution, D_{Tc} is increased (Fig. 10, Table A.9). The total distribution coefficient is simply the sum of D_{HTcO_4} and D_{MTcO_4}, measured at the same activity. On the other hand, multivalent cations, such as UO_2^{2+} and Th(IV), have very different effects on D_{Tc} and D_{Re}, as discussed in Sects. 3.5 and 3.6.

3.2.4 Reaction Stoichiometry and Mathematical Modeling

The available data suggest that the dominant reaction for the extraction of TcO_4^- or ReO_4^- from aqueous HCl by TBP may be written as

$$H^+ + XO_4^- + 3\overline{TBP} \rightleftarrows \overline{HMO_4 \cdot 3TBP} \ , \qquad (4)$$

where the bars are used to indicate organic-phase species. The equilibrium constant for Eq. (4) may be written as

$$K = \frac{[\overline{HXO_4 \cdot 3TBP}]}{[H^+][XO_4^-][\overline{TBP}]^3} \cdot Q \ , \qquad (5)$$

where Q is the quotient of the appropriate molar activity coefficients. Since both $HReO_4$ and $HTcO_4$ are very strong acids,[21] $[XO_4^-]$ is the stoichiometric X(VII) concentration. If all of the organic-phase HXO_4 is in the form $HXO_4 \cdot 3TBP$, then $D_M = [HXO_4 \cdot 3TBP]/[XO_4^-]$ and Eq. (5) can be rearranged to yield

$$D_X = \ln(K/Q) + \ln [H^+] + 3 \ln [\overline{TBP}] \ . \qquad (6)$$

Given the appropriate values for the activity coefficients and the equilibrium constants, Eq. (6) could be used to calculate the distribution behavior of technetium or rhenium under any conditions. Unfortunately, activity data for the various species in this two-phase system are not available over the wide range of conditions studied. In order to circumvent this difficulty, Q is set equal to 1, and the relationship

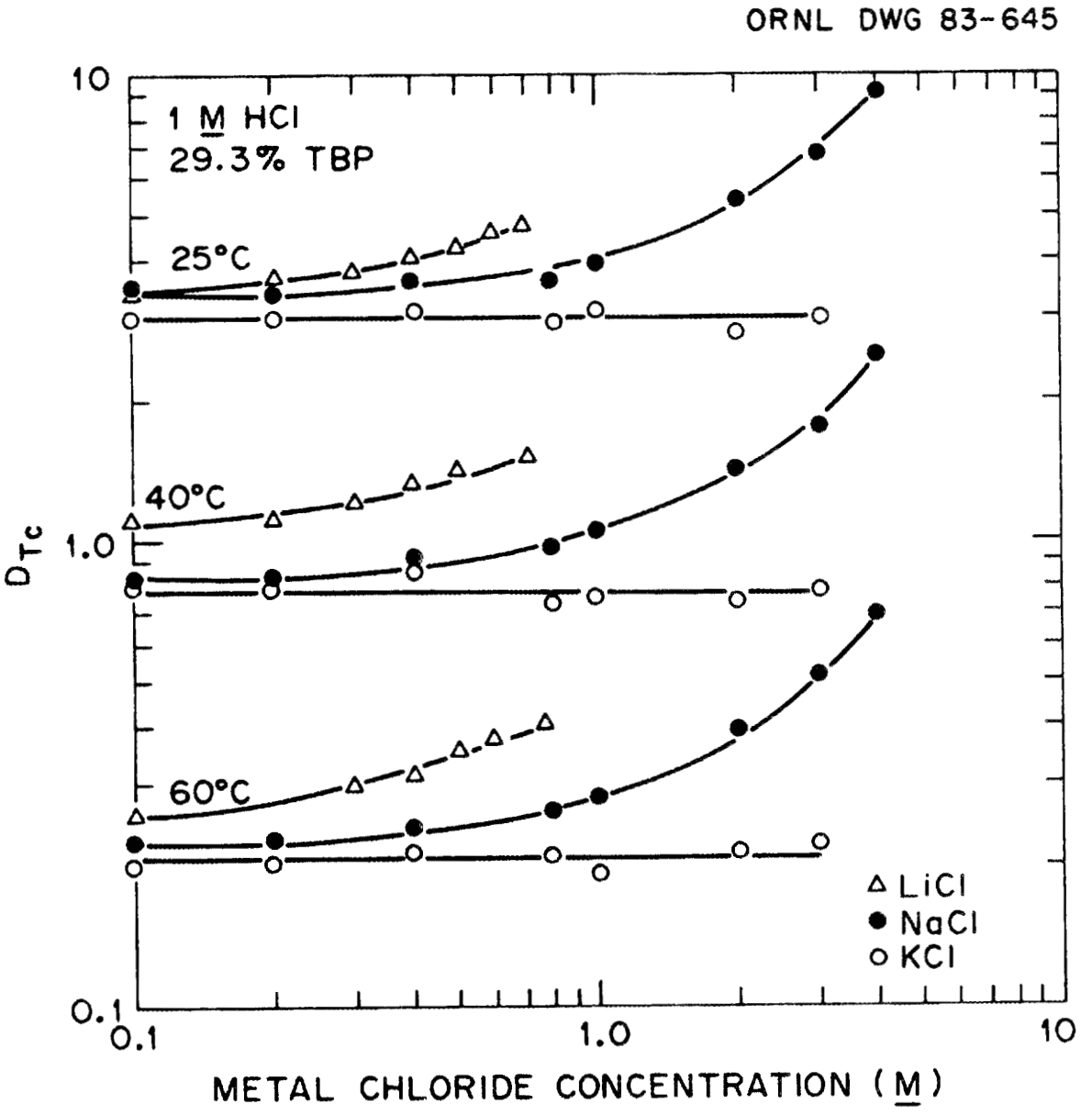

Fig. 10. Influence of alkali metal chlorides on pertechnetate extraction with HCl by 2.93 vol % (1.07 M) TBP in NDD at 25, 40, and 60°C.

$\ln K = -\Delta G/RT = \Delta S/R - \Delta H/RT$ is used to transform Eq. (6) to Eq. (7):

$$D_X = (\Delta S/R) - (\Delta H/RT) + \ln [H^+] + 3 \ln [\overline{TBP}] \ . \qquad (7)$$

Values of ΔS and ΔH can be estimated from the temperature dependence of the distribution coefficients of ReO_4^- or TcO_4^-; Eq. (7) can then be used to calculate values of D_X under any conditions. However, the assumption that $Q = 1$ is clearly an oversimplification, and it has already been shown that D_X does not show a simple, third-order dependence on the TBP concentration. Thus, values of D_X calculated from Eq. (7), while often surprisingly accurate, can differ from the experimentally determined values by factors of up to 10. In order to improve the agreement between the calculated and experimental values D_X, the mean molar HCl activity, $a_{\pm}^{HCl}$, was used instead of the stoichiometric hydrogen ion concentration. Experimental data were fit to an equation of the form of Eq. (7) by using a multiple linear regression analysis and allowing the values of $\Delta S/R$, $\Delta H/R$, and the coefficients of the $\ln a_{\pm}^{HCl}$, and $\ln [TBP]$ terms to vary until the best fit was achieved. This resulted in Eq. (8), which can be used to calculate the distribution coefficient of pertechnetate, and Eq. (9), which can be used to calculate the distribution coefficient of perrhenate in HCl-TBP-NDD systems:

$$\ln D_{Tc} = -23.9 + 7462/T + 1.03 \ln a_{\pm}^{HCl} + 3.26 \ln [\overline{TBP}] \ , \qquad (8)$$

$$\ln D_{Re} = -22.1 + 6684/T + 1.04 \ln a_{\pm}^{HCl} + 3.28 \ln [\overline{TBP}] \ . \qquad (9)$$

These equations are valid for temperatures between 25 and 60°C, acid concentrations of 0.2 to 4 M, and TBP concentrations of 0.339 to 2.90 M. The excellent agreement between the values of D_{Tc} calculated using Eq. (8) and the experimental values is shown in Fig. 11. However, a large amount of scatter occurs when calculated and experimental values

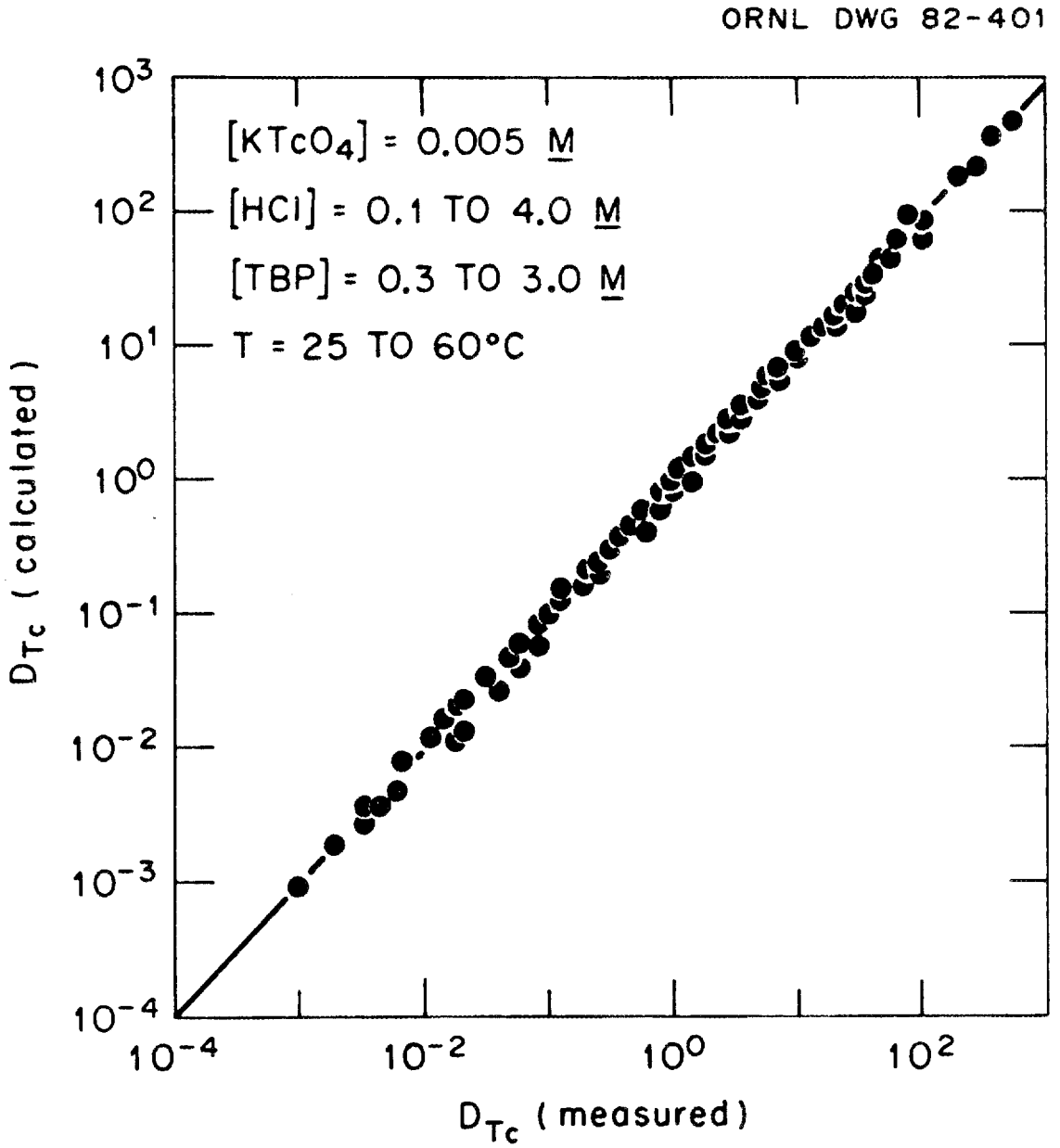

Fig. 11. Comparison of D_{Tc} as calculated from Eq. (8) with experimental results.

of D_{Re} are compared (Fig. 12). This scatter is due to the greater error in D_{Re} values that results when very low levels of rhenium are determined by x-ray fluorescence, as opposed to the very accurate D_{Tc} values that can be obtained by liquid scintillation counting of very low concentrations of technetium.

Equations (8) and (9) can be combined in order to derive a simple relationship between D_{Tc} and D_{Re}. Since both distribution coefficients have essentially the same dependence on a^{HCl} and [TBP], taking the exponentials of these equations and dividing the expression for D_{Tc} by the expression for D_{Re} yields

$$D_{Tc}/D_{Re} = e^{-1.8 + 778/T} \quad . \tag{10}$$

The similarity of Eqs. (8) and (9) indicates that rhenium is an excellent model compound for technetium in these solvent extraction systems, while Eq. (10) shows that D_{Tc} is about 2.2, 2.0, and 1.7 times D_{Re} at 25, 40, and 60°C, respectively.

3.2.5 Thermodynamic Parameters

Given the data in Tables A.1-A.3 and values for the activity coefficients of each of the species involved, a complete thermodynamic description of these solvent extraction systems could be written in the form

$$D_X = (\Delta S/R) - (\Delta H/RT) + \ln a^{H^+} + \ln a^{TBP} + \ln \gamma_X^{aq}/\gamma_X^{org} \quad , \tag{11}$$

where X refers to the rhenium or technetium anion in the system. Unfortunately, the data available only allow Eqs. (8) and (9), which are less rigorous, to be derived explicitly. Nonetheless, to the degree that the coefficients of the first two terms of Eqs. (8) and (9) can be equated with the values of $\Delta S/R$ and $\Delta H/R$ in Eq. (11), they can be used to calculate the approximate values of ΔH and ΔS shown in Table 2.

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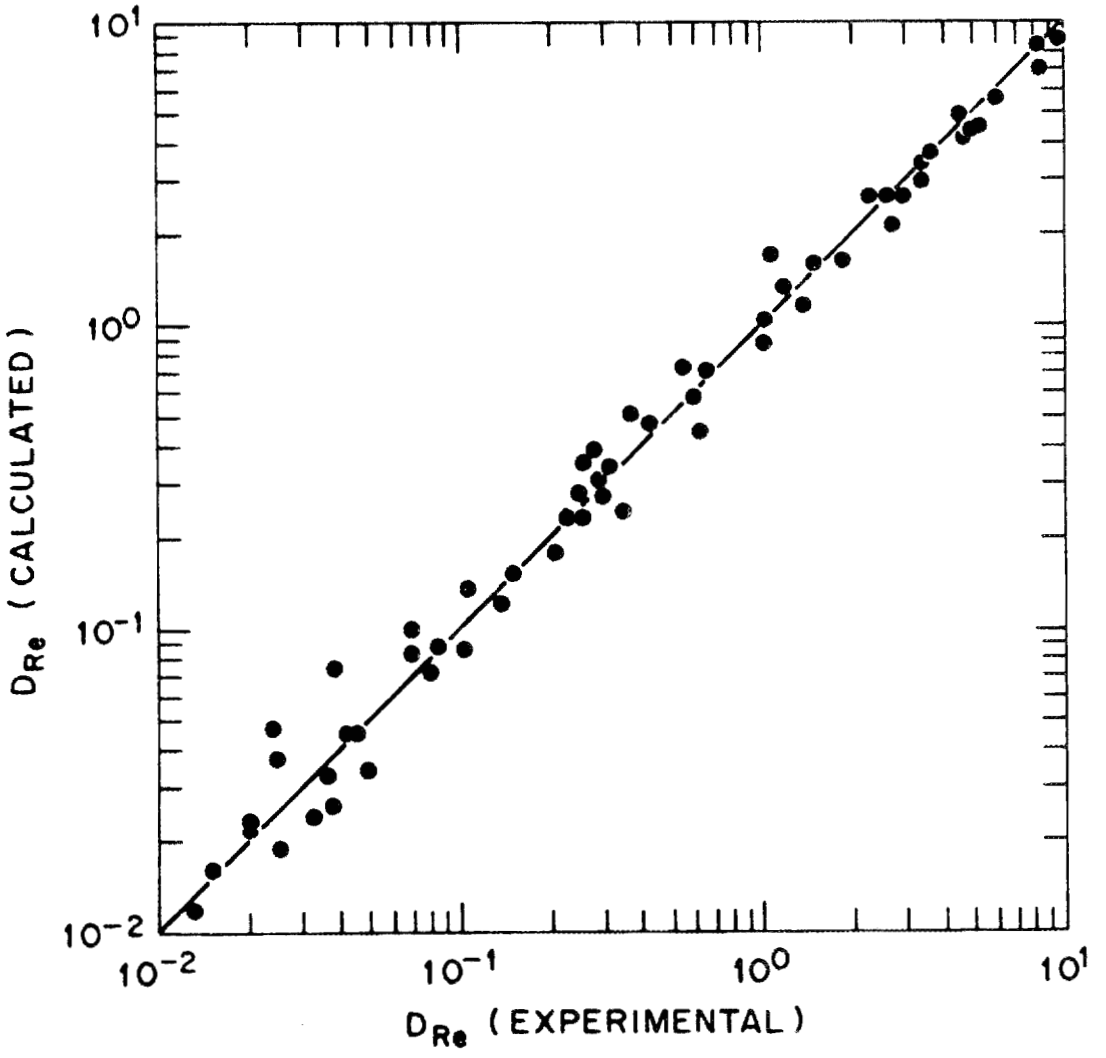

Fig. 12. Comparison of D_{Re} as calculated from Eq. (9) with experimental results.

Table 2. Thermodynamic parameters for the extraction of perrhenate and pertechnetate by tributyl phosphate

Metal species	ΔS (cal/mol·K)	ΔH (kcal/mol)	K_{eq}^{298} [a]
Technetium	-47.5 ± 1.4	-14.8 ± 0.4	3.0
Rhenium	-43.9 ± 2.4	-13.3 ± 0.7[b]	1.4

[a]Value of equilibrium constant at 298 K.
[b]Compare with value measured via calorimetry, -13.1 ± 0.8.

In order to determine whether the ΔH and ΔS values calculated in this way were close enough to the true values to be useful, a direct calorimetric determination of ΔH for the reaction $ReO_4^- + H^+$ 3TBP $HReO_4 \cdot 3TBP$ was carried out[22] using the calorimeter and techniques described elsewhere.[23] As shown in Table 2, the agreement between the measured value of ΔH and the value calculated from Eq. (9) is excellent. While there is no similar way to check the accuracy of the ΔS values their sign and magnitude are consistent with the ordering that must occur as two ions and three neutral molecules condense to form a single complex. The large, negative entropies also probably reflect the advantages of re-forming the particularly strong three-dimensional structure of water after the relatively large and poorly solvated ReO_4^- or TcO_4^- anions are removed from the aqueous phase.[24] By analogy to other solvent extraction systems,[25] these values may also indicate some involvement of water molecules in the organic-phase complexes.

3.3 EXTRACTION FROM NITRIC ACID

3.3.1 Acid and TBP Dependence

Tables A.10-A.12 and Figs. 13-18 summarize the data obtained in this laboratory for the distribution of rhenium and technetium between aqueous HNO_3 and TBP–NDD at 25, 40, and 60°C. Distribution data reported by previous investigators for perrhenate are shown in Tables A.13 and A.14, while Tables A.15-A.19 and Fig. 19 present similar data for pertechnetate. All of the data are in general agreement, but different and incompletely reported experimental conditions among the various reports make detailed, critical comparisons difficult.

A comparison of Figs. 1-6 with Figs. 13-18 shows that substituting HNO_3 for HCl in these solvent extraction systems results in large qualitative and quantitative differences in the distribution behavior of rhenium and technetium. These differences arise because of the greater tendency of HNO_3 to self-associate [Eq. (12)] and the subsequent extraction of molecular HNO_3 by TBP [Eq. (13)]:

$$H^+ + NO_3^- \rightleftarrows HNO_3 \ , \tag{12}$$

$$HNO_3 + \overline{TBP} \rightleftarrows [\overline{HNO_3 \cdot TBP}] \ . \tag{13}$$

It is obvious that the amount of uncomplexed, or "free," TBP available to extract perrhenate or pertechnetate cannot be varied independently of the acid concentration in the aqueous phase. Thus, the simple graphical methods used to determine the acid and TBP dependences in the HCl system cannot be used in the HNO_3 system, except over very small concentration ranges or at very low HNO_3 concentrations. For the data in Tables A.10-A.12, only the distribution coefficients measured for technetium from 0.1 M HNO_3 can be used to estimate the TBP dependence of the extraction reaction without taking Eq. (13) into account. An order of about 3 is indicated by these data.

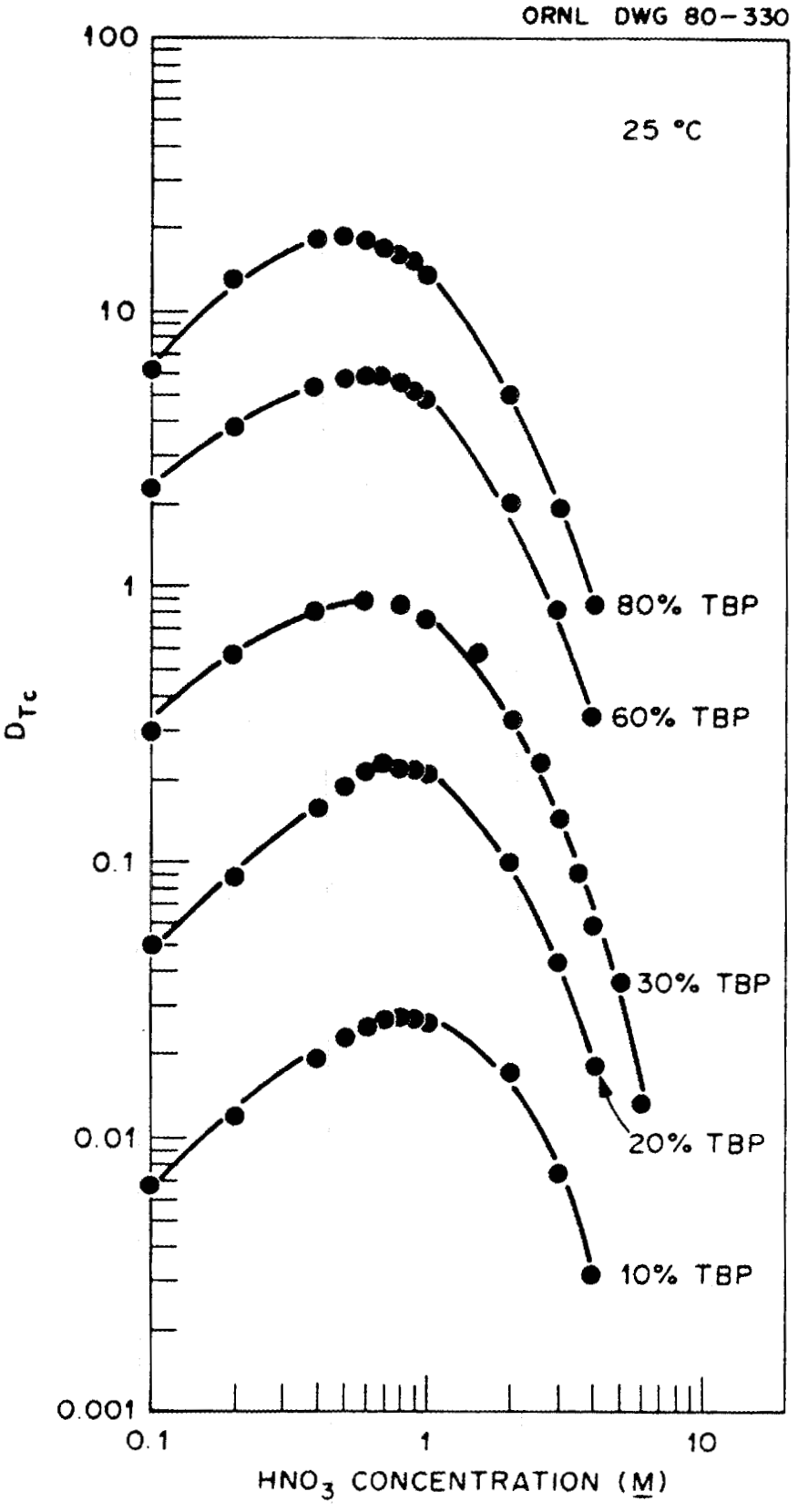

Fig. 13. Distribution of pertechnetate between aqueous HNO_3 and TBP-NDD at 25°C.

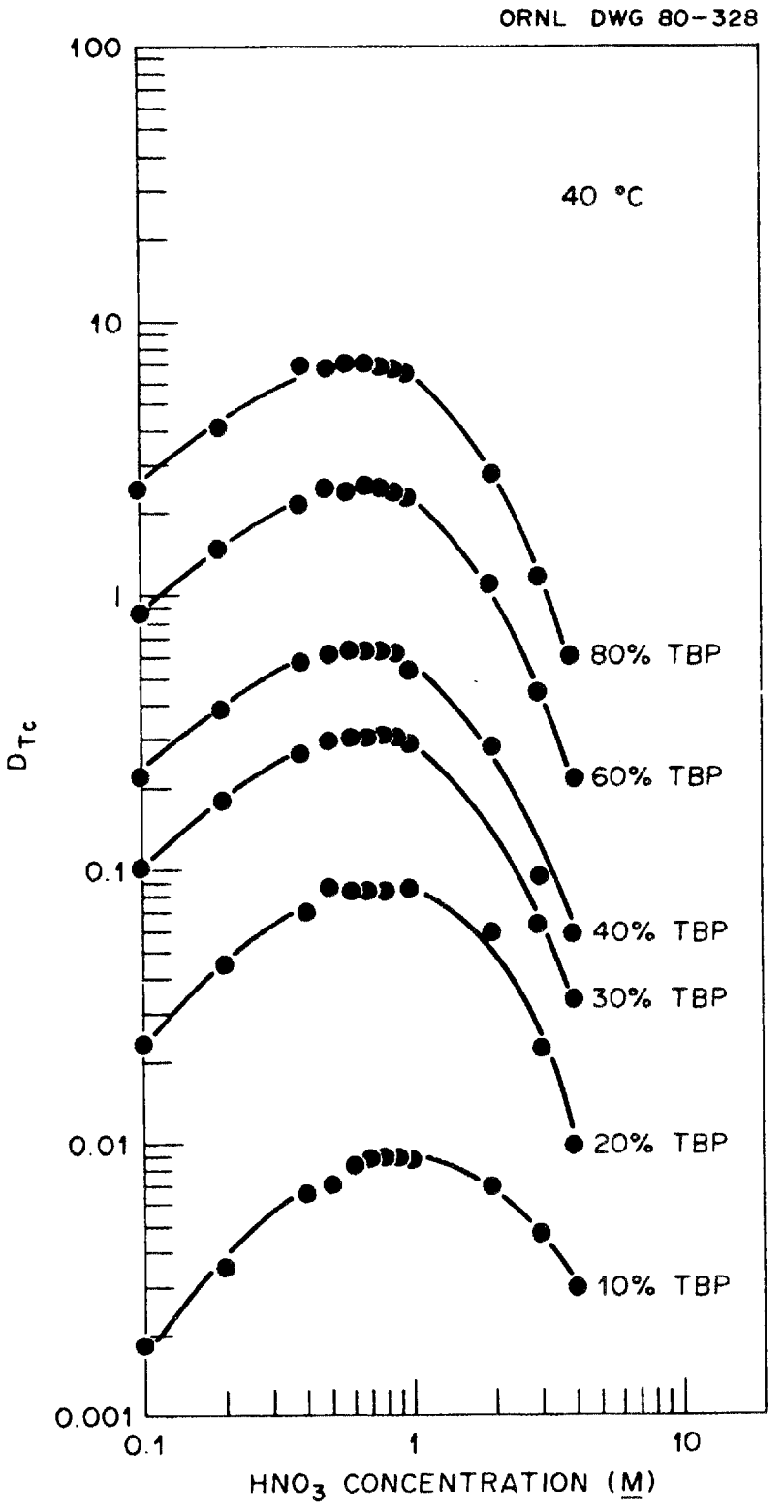

Fig. 14. Distribution of pertechnetate between aqueous HNO_3 and TBP-NDD at 40°C.

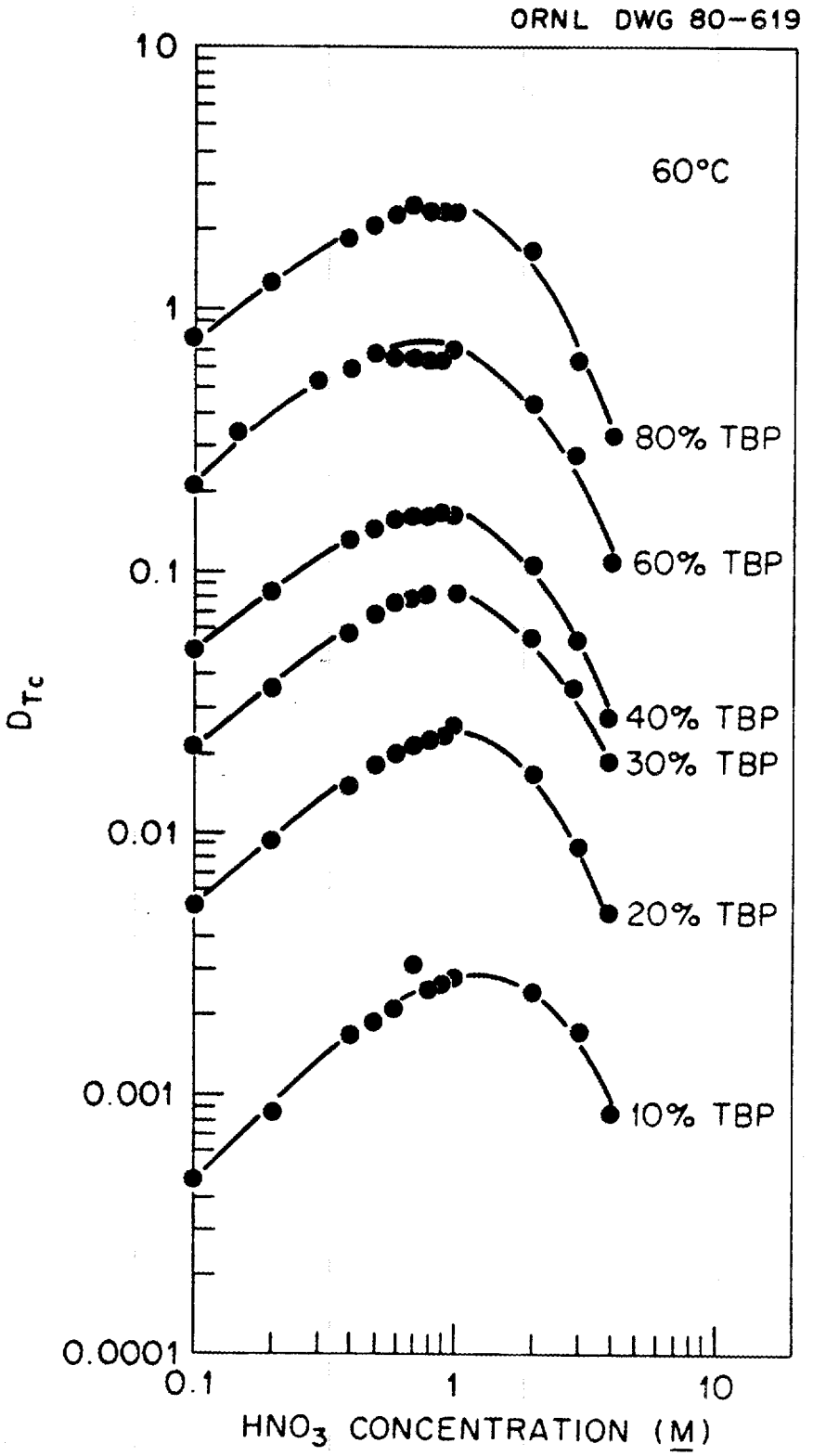

Fig. 15. Distribution of pertechnetate between aqueous HNO_3 and TBP-NDD at 60°C.

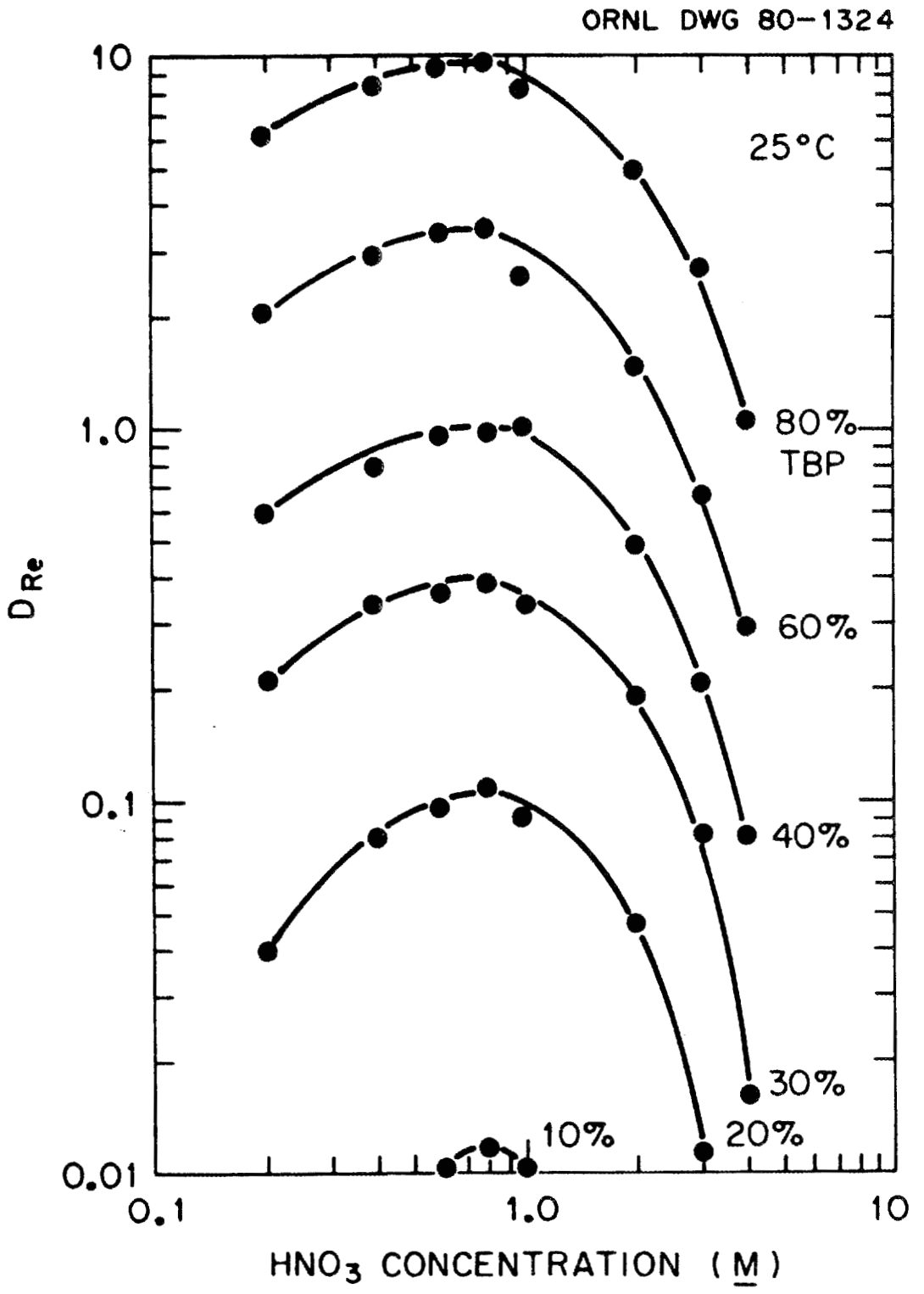

Fig. 16. Distribution of perrhenate between aqueous HNO_3 and TBP-NDD at 25°C.

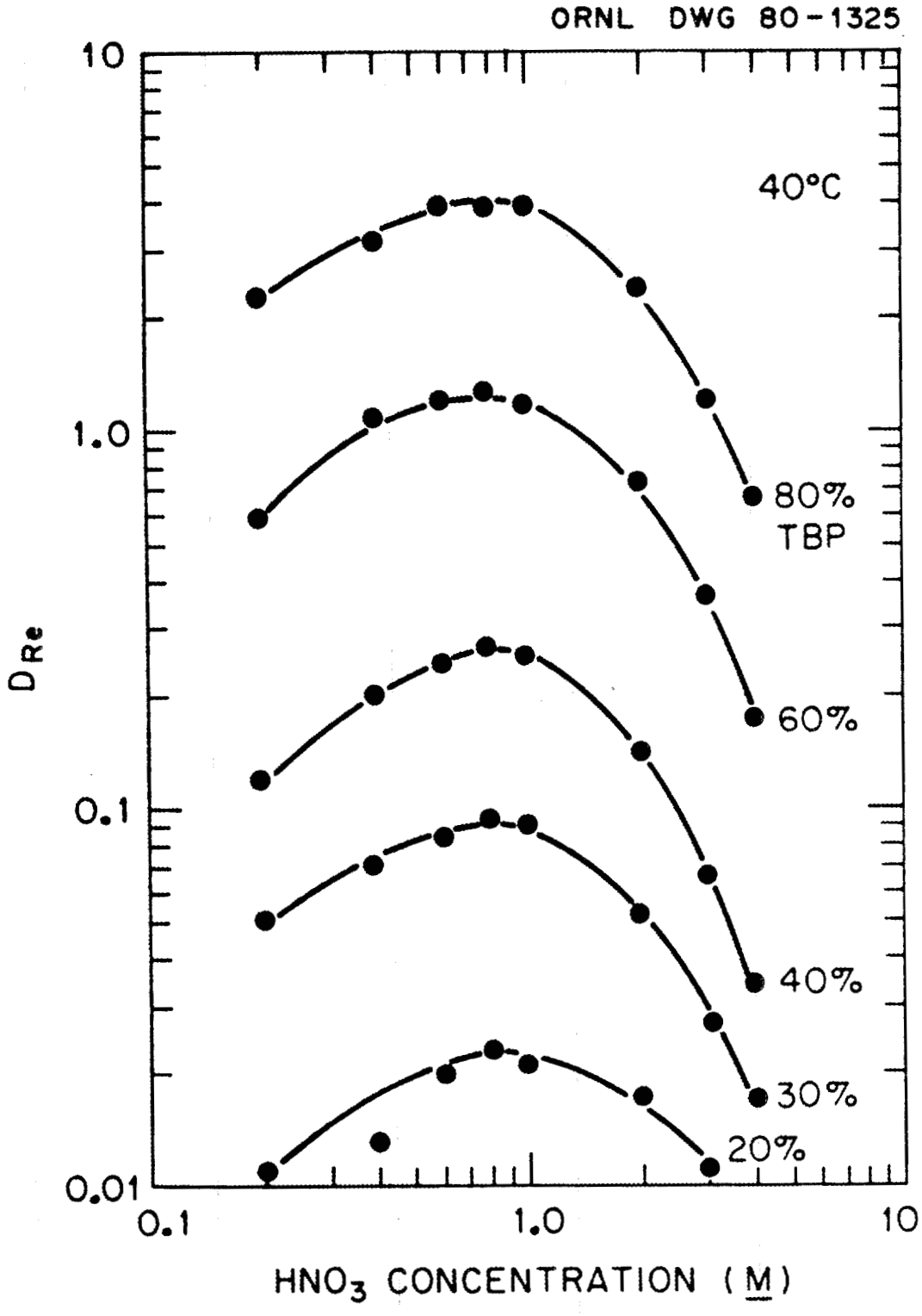

Fig. 17. Distribution of perrhenate between aqueous HNO_3 and TBP-NDD at 40°C.

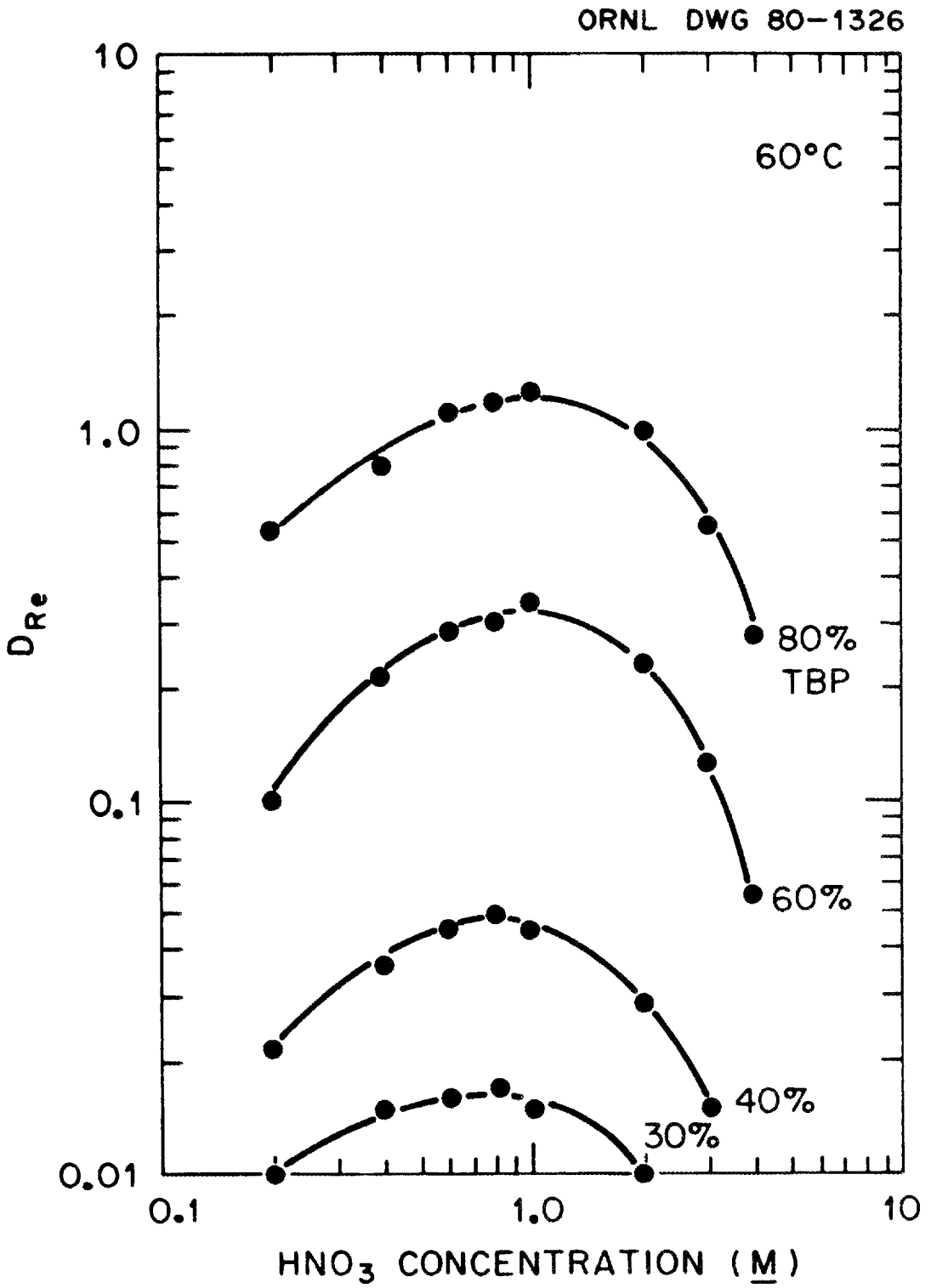

Fig. 18. Distribution of perrhenate between aqueous HNO_3 and TBP-NDD at 60°C.

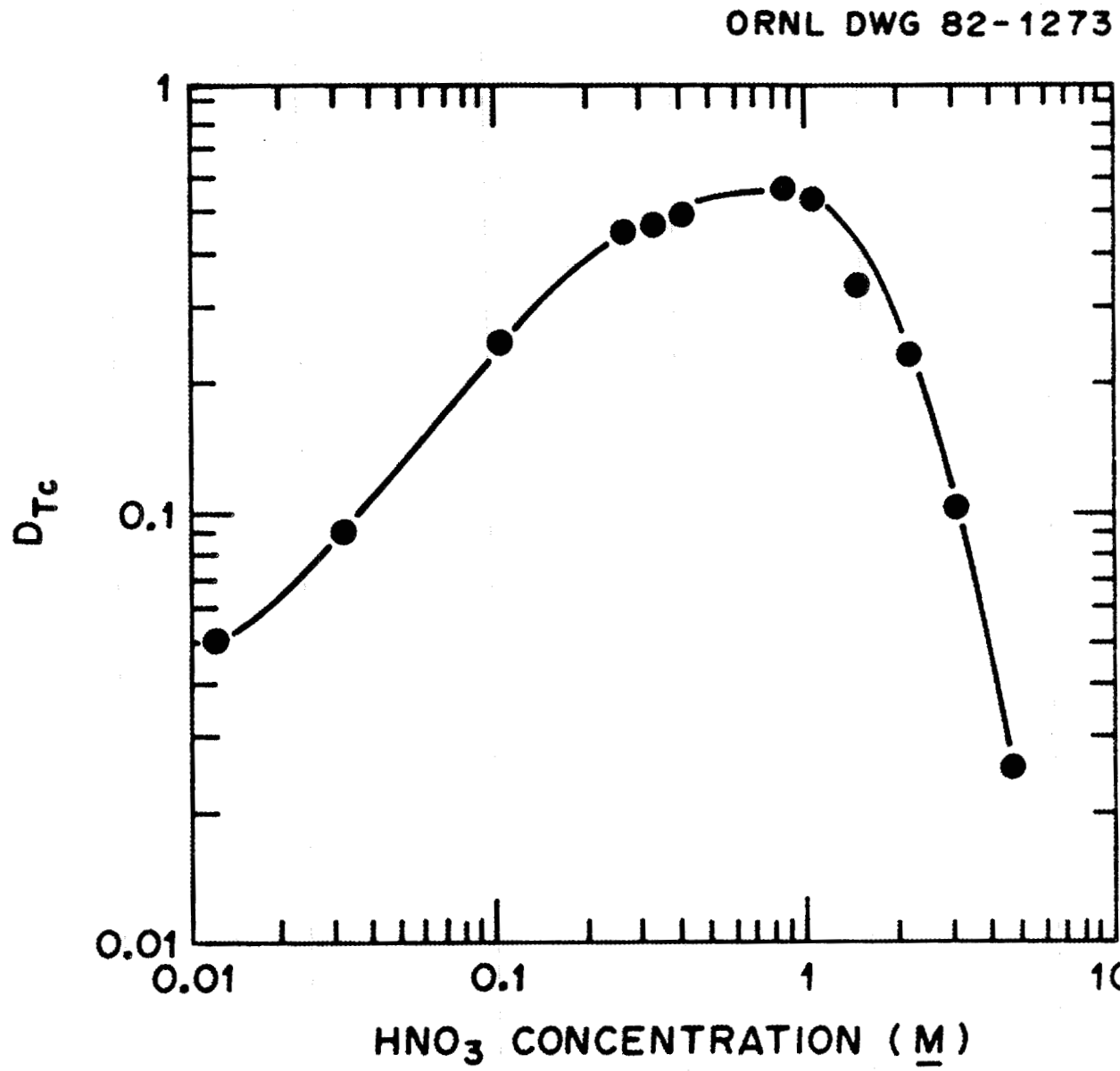

Fig. 19. Distribution of pertechnetate between aqueous HNO_3 and TBP-NDD. Data derived from ref. 6.

Previous reports on the extraction of perrhenate and pertechnetate from HNO_3 have presented conflicting conclusions regarding the dependence of the distribution coefficient on the TBP concentration. Some of the earliest work on perrhenate extraction was done by Kertes and Beck,[12] who used CCL_4 as the organic diluent and reported a TBP dependence of "~4" between 1.5 and 3.65 M (about 40 and 100 vol %) TBP (Table A.13). Their conclusion was based on a plot of log D_{Re} vs log $[TBP]_{initial}$ at a constant initial nitric acid concentration in the aqueous phase. No correction was made for the changes in effective TBP concentration and aqueous-phase HNO_3 concentration due to Eq. (13). Thus, many of the data points shown in their plot deviate significantly from linearity. Even if these points are ignored, the slopes of the lines vary from 3.9 to 5.0. A simple linear analysis of their data is not possible since both the equilibrium aqueous-phase HNO_3 concentration and the free TBP concentration are varied simultaneously. Multiple linear regression (Sects. 3.2.4 and 3.3.2) can be used successfully for this purpose, however, and it indicates a TBP dependence of slightly over 5.

A short time after the work by Kertes and Beck appeared in the literature,[12] Colton published the results of an independent study of perrhenate extraction in which he found a third-order dependence on the TBP concentration over the range 0.375 to 1.88 M (10 to 50 vol %) TBP in kerosene (Table A.14).[13] The aqueous phase was listed as 1 M HNO_3, but it is not clear whether this was an initial or equilibrium value. No temperature was reported, and, no free TBP corrections were made. Colton noted the difference between his results and those of Kertes and Beck and correctly pointed out that his results, which were obtained using lower TBP concentrations, should be more meaningful since deviations from ideality would be smaller.

For pertechnetate extraction, Siddall reported a third-order dependence on TBP concentration under conditions similar to those used in the Purex process but with no uranium present.[6] Both Spitsyn[8] and Zaitsev[9] (Table A.15), and their co-workers, reported results similar to those obtained by Siddall[6] and Pruett[1] in work done at room temperature (assumed

to be about 20°C). Several years later, Macasek suggested a third-order dependence in one paper (Table A.16)[10] and a complicated analysis involving first- to fourth-order dependences in a later paper.[20] An aqueous phase containing 4 M NH_4NO_3 or NH_4Cl and, in some experiments, dilute (0.1 M) HNO_3 was used in each of the studies; various diluents and TBP concentrations were employed as the organic phase. The unique conditions used in these experiments make it difficult to integrate Macasek's results with those of other investigators.

Most recently, Lieser and his co-workers reported a fourth-order dependence for technetium extraction by TBP.[11] Their treatment and interpretation of their solvent extraction data are unusual, and some of their assumptions are erroneous. For example, they make use of an old value for the dissociation constant of $HTcO_4$ (K_d = 0.5), which was later shown to be incorrect.[21] They calculated the solubility of TBP in the aqueous phase as a function of HNO_3 concentration from previously published data, and used the resulting values to calculate a fourth-order TBP dependence. This approach to determining solvation numbers is unique in the technetium solvent extraction literature. It is the concentration of free TBP at the interface and in the bulk organic phase that determines D_{Tc} rather than the amount of TBP dissolved in the aqueous phase. The decrease in D_{Tc} as $NaNO_3$, $NaNO_2$, or NH_4NO_3 is added to the aqueous phase is due to the shifting of the equilibrium to the left in Eq. (12), or the analogous reaction with HNO_2, thus lowering the concentration of free TBP available to complex other species. If D_{Tc} or D_{Re} is calculated at a constant free TBP concentration, it is a constant, regardless of the aqueous-phase concentration of TBP or $NaNO_3$ (Sect. 3.2.3). Despite the unusual analysis given in this report, the raw data appear to be in good agreement with those of other workers, when differences in the temperature and other parameters are considered.

In summary, while the distribution behavior of perrhenate appears to differ considerably from that of pertechnetate when log D–vs–log $[H^+]$ plots of HNO_3 systems are compared with those of HCl systems, these differences arise from the competing extraction of HNO_3 and not from

changes in the $HXO_4 \cdot 3TBP$ complex. Previous analyses of extraction data in these systems have been inconsistent, but most of the published data are in general agreement when the varying experimental conditions are taken into account.

3.3.2 Reaction Stoichiometry and Mathematical Modeling

As mentioned in Sect. 3.3.1, the extraction reaction for perrhenate or pertechnetate is expected to be the same from aqueous HNO_3 solution as it would from HCl solution because only the proton (and not the anion) of the acid is involved. However, the stoichiometric concentrations of HNO_3 and TBP cannot be used for $[H^+]$ and [TBP] in Eq. (5) as a basis for modeling the data since these species are involved in reactions (12) and (13) as well. Rather, these stoichiometric concentrations must be corrected to reflect the amount of TBP complexed by HNO_3 in the organic phase and the amount of undissociated HNO_3 in the aqueous phase. The concentration of uncomplexed, or "free," TBP can be calculated from Eq. (14),

$$[\overline{TBP}]_f = [\overline{TBP}]_0 - [\overline{HNO_3}] - 3[\overline{X}] \ , \qquad (14)$$

where $[\overline{TBP}]_0$ is the initial, stoichiometric TBP concentration and $[\overline{X}]$ is the concentration of rhenium or technetium in the organic phase. Under the conditions of the experiments reviewed here, the third term of Eq. (14) is negligible; only the HNO_3 term is ever large enough to affect $[TBP]_f$.

The data obtained by Davis in his study of the distribution of HNO_3 between TBP—Amsco 125-82 and water at 25°C[26] were used to derive a correlation between $[HNO_3]_{aq}^{equilibrium}$ and the $[TBP]_f$. It is assumed that substituting TBP—NDD for TBP—Amsco makes a negligible change in the HNO_3 distribution. Although these data can be analyzed in a variety of theoretical and semiempirical ways, the simple expression of Eq. (15) was found to provide reasonable agreement between calculated and experimental values of $[\overline{TBP}]_f$ over the range of conditions for which technetium and rhenium extraction data are available:

$$[\overline{TBP}]_f = 1.0026\ [\overline{TBP}]_0 - 0.2054\ [HNO_3]_{aq}^{equilibrium}\ [\overline{TBP}]_0\ . \qquad (15)$$

Values of the stoichiometric activity of HNO_3, $a_s = [HNO_3]\gamma_s = [HNO_3]\gamma\pm\alpha$, were calculated using Eq. (16),

$$\ln \gamma_s = 0.0082 - 1.0515\ [HNO_3]^{1/2} + 0.907\ [HNO_3] - 0.1967\ [HNO_3]^{3/2} + 0.966\ [HNO_3]\ , \qquad (16)$$

which was obtained by fitting literature values of γ_s to a fourth-order polynomial in $[HNO_3]^{1/2}$ from 0.001 to 21.0 M HNO_3. Agreement between Eq. (16) and the literature values was excellent, with a correlation coefficient (R^2) of 0.9997 and differences of less than ±3% between calculated and measured values of γ_s.

Although data for the activity of HNO_3 and its distribution between TBP–NDD and water at temperatures other than 25°C are not readily available, Adamskii et al. have reported that the distribution does not change significantly between 25 and 70°C.[27] While this would indicate that Eq. (15) is probably valid for calculating $[\overline{TBP}]_f$ at 40 and 60°C, it is likely that γ_s will change substantially over this temperature range. Therefore, Eqs. (15) and (16) were used to calculate $[\overline{TBP}]_f$ and γ_s at 25°C only, and the experimental data obtained at that temperature were fit to an equation similar to Eq. (6) to yield Eqs. (17) and (18):

$$\ln D_{Tc} = 0.0933 + 0.634\ \ln a_s^{HNO_3} + 2.94\ \ln [\overline{TBP}]_f\ , \qquad (17)$$

$$\ln D_{Re} = -0.519 + 0.642\ \ln a_s^{HNO_3} + 3.19\ \ln [\overline{TBP}]_f\ . \qquad (18)$$

The agreement between the D_{Tc} and D_{Re} values calculated using these equations is reasonably good, especially when the errors and assumptions in determining the values of D, $a_s^{HNO_3}$, and $[\overline{TBP}]_f$ are considered.

While activity data for HNO_3 at 40 and 60°C are not available, it is possible to fit all of the experimental results in Tables A.10-A.12 to an expression of the form of Eq. (7), using $[\overline{TBP}]_f$ and the stoichiometric HNO_3 concentration for $[H^+]$. The resulting correlations, Eqs. (19) and (20),

$$\ln D_{Tc} = -20.13 + 6013/T + 0.865 \ln [H^+] + 2.91 \ln [\overline{TBP}]_f , \quad (19)$$

$$\ln D_{Re} = -25.88 + 7534/T + 0.944 \ln [H^+] + 3.19 \ln [\overline{TBP}]_f , \quad (20)$$

predict D_{Tc} and D_{Re} values that are usually within about 30% of the measured values, and always within a factor of 2. By comparison, distribution coefficients measured by different workers, and often even by the same worker at different times, frequently vary by factors at least as large as these. It should also be noted that the correlations show an approximately first-order dependence on acidity and an approximately third-order dependence on $[\overline{TBP}]_f$, as expected from Eq. (4). Further, the values of "($\Delta H/R$)" and "($\Delta S/R$)" are comparable to those in Eqs. (8) and (9), which would be expected if the same extraction reactions are involved.

3.4 EXTRACTION FROM OTHER MEDIA

3.4.1 Extraction from Other Acids

Table A.5 includes essentially all of the published information on perrhenate extraction from acids other than HCl and HNO_3, while Tables A.6, A.7, and A.20 show similar information for pertechnetate extraction. These limited data show no unusual behavior for the extraction of either anion. As in the HCl and HNO_3 systems, the effect of the acid concentration in the aqueous phase on D_{Re} and D_{Tc} appears to be solely a function of the strength of the acid (i.e., the activity of the hydrogen ion) and the tendency of the mineral acid to compete with perrhenate and pertechnetate for extraction by TBP. Acids that drive Eq. (4) to the

right increase the distribution coefficient, while those that react with TBP as HNO_3 does in Eq. (13) decrease the distribution coefficient at higher acidities. For example, $HClO_4$ is a very strong acid, but it is readily extracted and competes very strongly for TBP; thus, D_{Re} and D_{Tc} from this medium are very low. On the other hand, H_2SO_4 is a strong acid that is very poorly extracted by TBP; therefore, distribution coefficients from this medium are quite high.

One other effect must be considered when interpreting these data. Some acids, such as concentrated solutions of HCl (about 6 M and above) will quickly reduce TcO_4^- or ReO_4^- and thereby change the chemistry of the system entirely. Even relatively dilute solutions of HI produce the same effect.

3.4.2 Extraction as Neutral Salts

Boyd and Larson[5] were the first to report data on the extraction of pertechnetate as a neutral alkali metal or ammonium salt (Table A.8 and Fig. 20). Dakar et al.[19] studied the extraction of perrhenate salts from sodium carbonate, sodium nitrate, and each of the alkali metal hydroxides (Tables A.21 and A.22, Fig. 21). There are few other references to the extraction of MXO_4 (X = Tc or Re, M = Li, Na, K, Rb, Cs, or NH_4), except for those noting that the anions extract from acidic solutions of their potassium salts only as acids and not as potassium salts.[1-4,12] These data show that the distribution coefficient of either anion decreases as M is varied in the following order: Li > NH_4 > Na > K > Rb > Cs. Further, under otherwise identical conditions, $D_{MTcO_4} > D_{MTcO_4}$ and $D_{HXO_4} > D_{MXO_4}$.

Table A.23 and Fig. 22 and 23 show the results of a new study of the extraction of pertechnetate from various ammonium and alkali metal chloride solutions. As a first approximation, pertechnetate might be expected to extract from these systems as $MTcO_4 \cdot 3TBP$, by analogy to the acid systems. If so, the plots of D_{Tc} as a function of metal chloride activity shown in Fig. 22 should be linear with slopes near 1. The slope is almost exactly 1.0 for $HTcO_4$ and $LiTcO_4$ extraction, and it is

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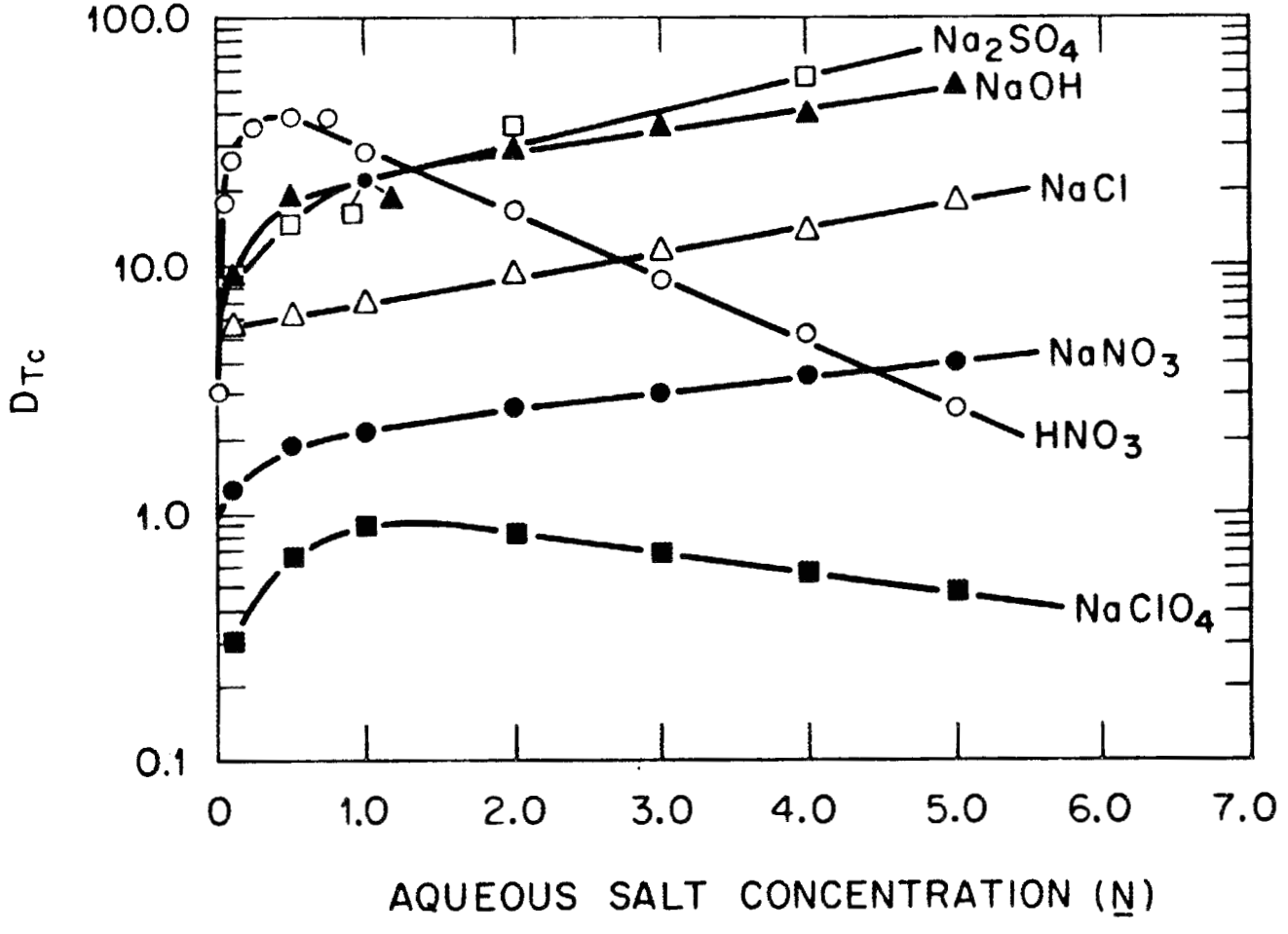

Fig. 20. Extraction of pertechnetate from aqueous salt solutions by pure TBP. Data derived from ref. 5.

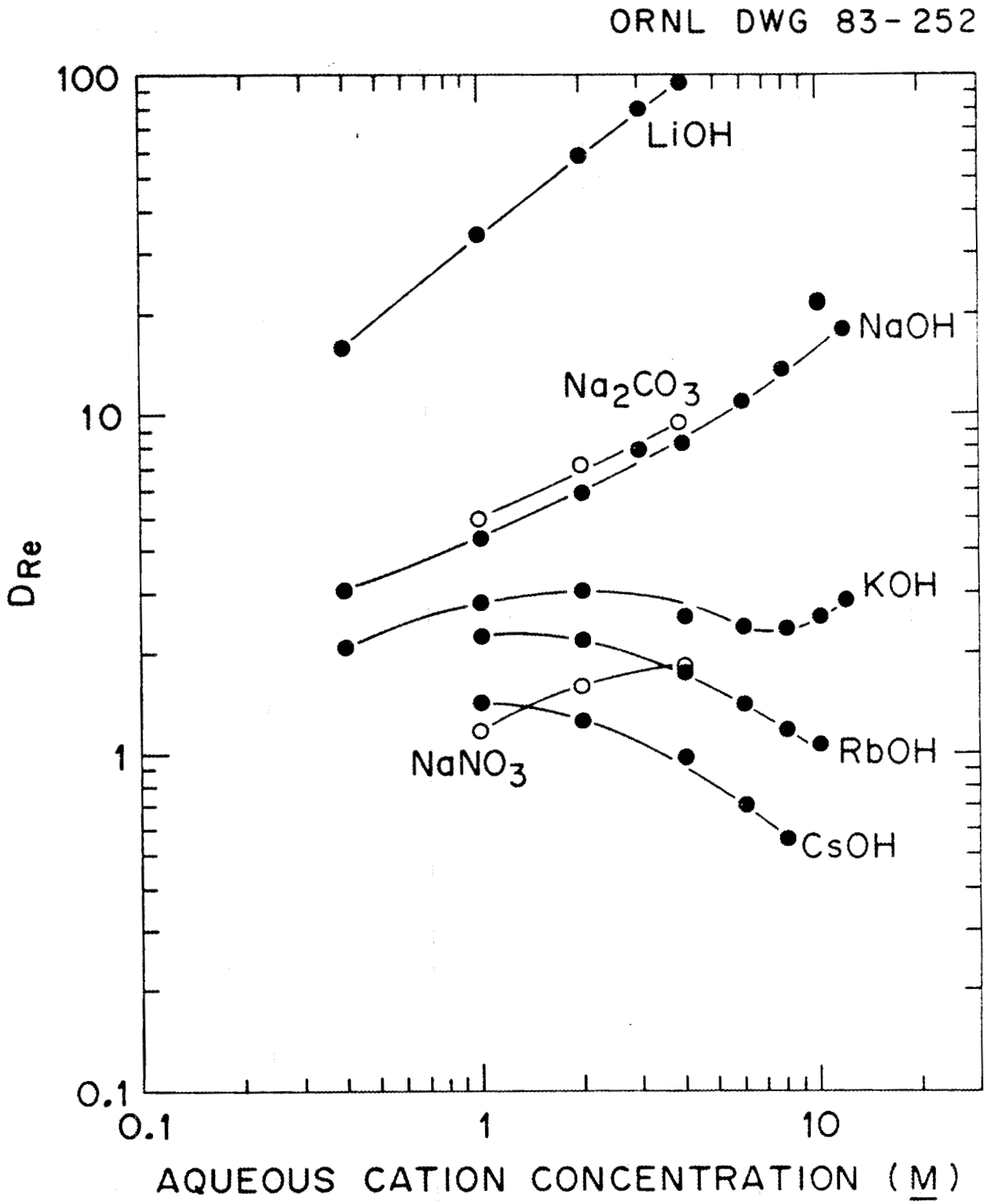

Fig. 21. Extraction of perrhenate from various aqueous alkali metal hydroxide and salt solutions by pure TBP. Data derived from ref. 19.

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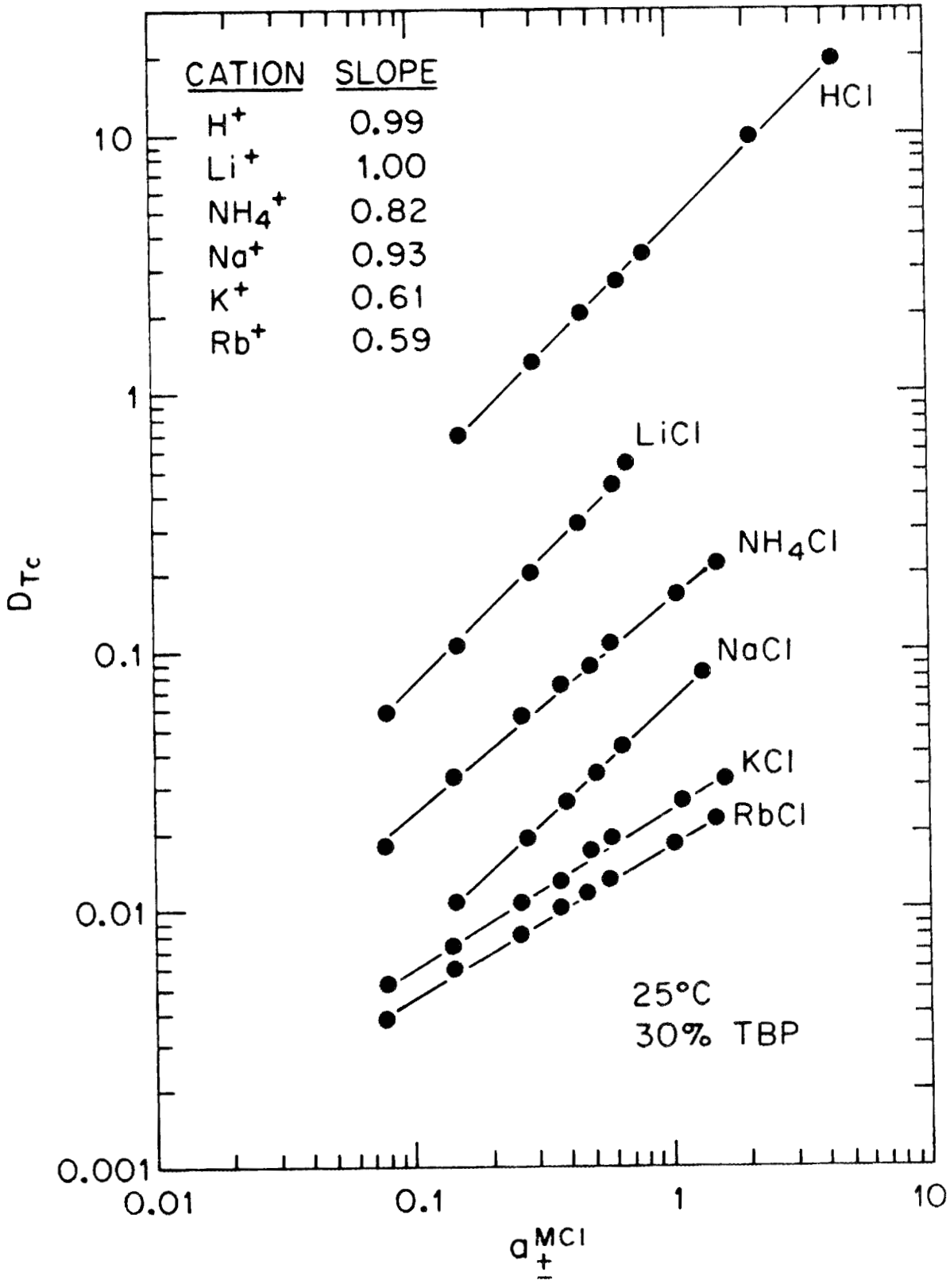

Fig. 22. Extraction of pertechnetate from various neutral chloride salt solutions with 30% TBP-NDD as a function of the aqueous cation concentration.

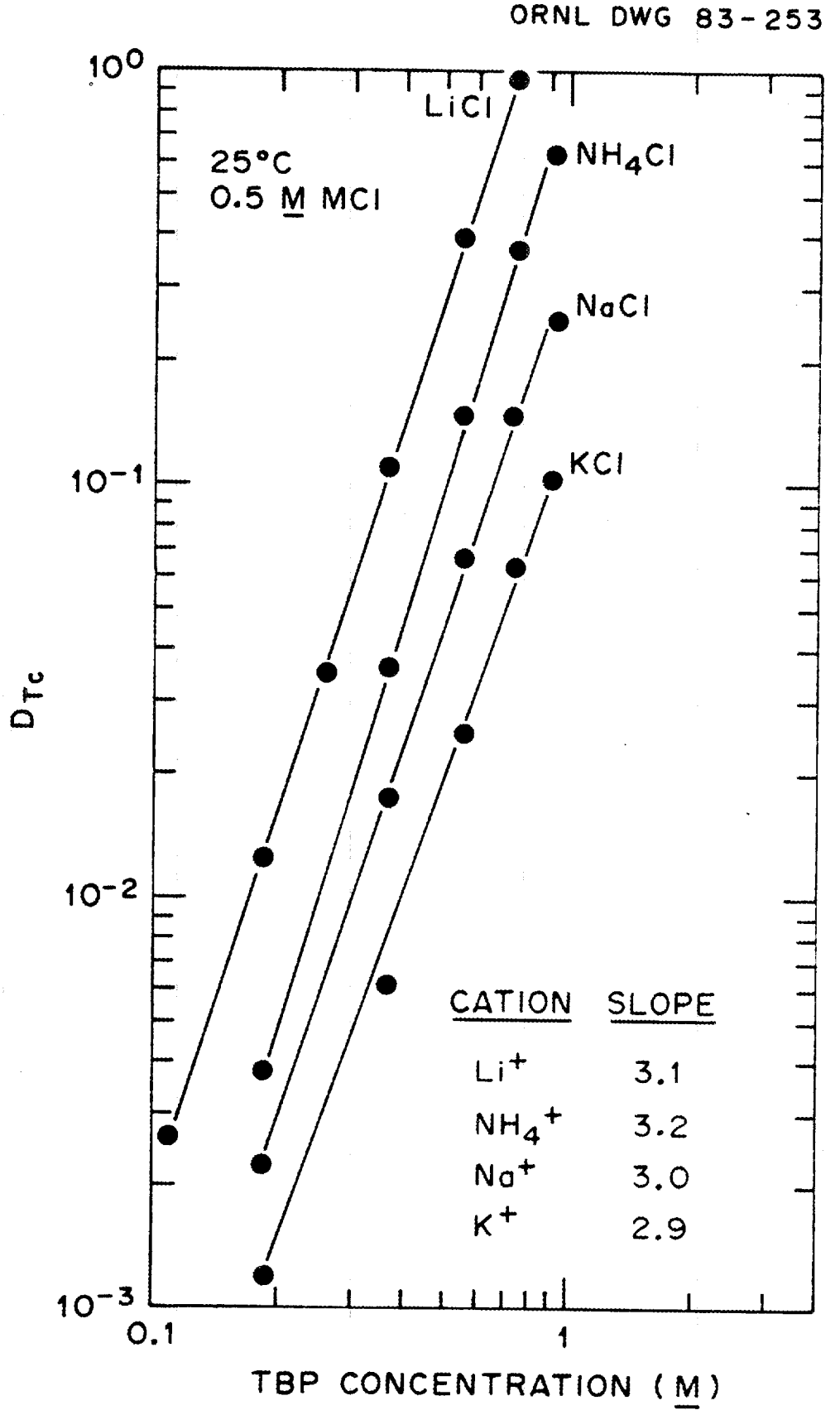

Fig. 23. Extraction of pertechnetate from 0.5 M neutral chloride salt solutions as a function of TBP concentration.

nearly so for $NaTcO_4$. However, there is a systematic decrease in the slope as the cation is varied in the order Li > Na >NH_4 > K > Rb, which is the same order in which the ionic radii and calculated single ion activities increase. Furthermore, while the slopes of these plots are constant between 25 and 60°C for M = H, Li, or Na, they increase with temperature for M = NH_4, K, and Rb. For example, the slope of a plot of log D_{Tc} vs log $a_{\pm}^{KCl}$ is 0.61 at 25°C but increases to 0.74 at 60°C.

On the other hand, all of the plots of log D_{Tc} vs TBP concentration show the expected third-order dependence (Fig. 23). Therefore, it is likely that each of the organic-phase species has the stoichiometry $MTcO_4 \cdot 3TBP$. The apparent deviation from this behavior is probably due to a temperature-dependent phenomenon (such as ion pairing) that is not corrected for in this simple, indirect method of determining the stoichiometry of the organic-phase species.

These data may be modeled using the same techniques that were described earlier for the acid systems (see Sects. 3.2.4 and 3.3.2). The equilibrium constant expression for the extraction reaction can be written as

$$K = \frac{[MTcO_4 \cdot 3TBP]\gamma_{MTcO_4 \cdot 3TBP}}{[M^+]\gamma_m + [TcO_4^-]\gamma_{TcO_4} - [TBP]^3\gamma^3{}_{TBP}} . \qquad (21)$$

Substituting $D_{Tc} = [MTcO_4 \cdot 3TBP]/[TcO_4^-]$ and taking logarithms results in the simple linear equation

$$\ln D_{Tc} = \ln K + \ln \frac{\gamma_{TcO_4^-}}{\gamma_{MTcO_4 \cdot 3TBP}} + \ln [M^+]\gamma_{M^+} + 3 \ln [TBP]\gamma_{TBP} . \qquad (22)$$

Finally, the temperature dependence of D_{Tc} can be expressed explicitly by substituting $\ln K = \Delta S/R - \Delta H/RT$ to obtain

$$\ln D_{Tc} = \Delta S/R - \Delta H/RT + \ln \frac{\gamma_{TcO_4^-}}{\gamma_{MTcO_4 \cdot 3TBP}} + \ln [M^+]\gamma_{M^+} + 3 \ln [TBP]\gamma_{TBP} \ . \quad (23)$$

Given all of the activity coefficients in Eq. (23), ΔS and ΔH could be calculated from the data in Table A.23 and D_{Tc} could be calculated under any conditions. Unfortunately, the salts are the only species in these systems for which appreciable activity data are available. Thus, since Eq. (23) cannot be solved rigorously, we make use of the functional form of the equation and the available mean molar activities of the chloride salts to derive the more empirical equation

$$\ln D_{Tc} = a - b/T + c \ln a_{\pm}^{MCl} , \quad (24)$$

where $a = \Delta S/R + 3 \ln [TBP]$, $b = \Delta H$, and the remaining activity coefficients are ignored. A simple least-squares fit of the distribution data to this equation yields the values of a, b, and c for each cation, as shown in Table 3. The variation in the value of c is consistent with the slopes observed in Fig. 22. It is tempting to try to derive estimates of ΔS and ΔH from the values of a and b. However, the variation in the value of c from its "theoretical" value of 1 and the lack of an obvious pattern in the variation of a and b (except when c is very close to 1, such as for the H, Li, and Na systems) suggest that any attempt to relate the statistical parameters to the thermodynamic parameters for these systems would be of questionable value.

Dakar et al.[19] have reported the most complete study of perrhenate extraction from an alkali metal hydroxide solution (Table A.21, Fig. 21). They observed that, as the hydroxide concentration is increased, D_{Re} passes through a "maximum" that occurs at lower concentrations for the heavier alkali metals. The LiOH curve never reaches this maximum because of the relatively low solubility limit for LiOH. They also noted that the D_{Re} from Na_2CO_3 was approximately equal to that from NaOH

Table 3. Values of the parameters a, b, and c from Eq. (24)[a] for the various MCl systems

M	Value of parameter		
	a	b	c
H	-24.7	7798	1.02
Li	-22.9	6645	1.00
Na	-20.5	5279	0.93
K	-21.7	5374	0.66
Rb	-25.2	6313	0.65
NH_4	-22.5	6153	0.83

[a] $\ln D_{Tc} = a - b/T + c \ln a_{\pm}^{MCl}$.

of the same normality but was much higher than that from $NaNO_3$. They attribute this behavior to ionization of the salts in the organic phase and the suppression of that ionization by extractable salts such as $NaNO_3$. However, the maximum in the NaOH curve at 10 M NaOH more closely resembles an erroneous data point (Fig. 21). The suppression of D_{Re} by $NaNO_3$ is due to a decrease in the free TBP concentration, analogous to the one that occurs when HNO_3 is extracted by TBP (Sect. 3.3). The same effect is undoubtedly responsible for the observed maximum in D_{Re} for each alkali hydroxide as well. Even though D_{MOH} is generally quite small, the concentration of alkali in the system (about 10 M) is considerably larger than that of rhenium (about 10^{-6} M). Only relatively small amounts of the bases need to be extracted in order to have a significant effect on D_{Re}, because of its third-order dependence on the free TBP concentration.

In summary, the distribution coefficients for perrhenate and pertechnetate from neutral or basic solution as alkali metal or ammonium salts are substantially lower than those from acidic solutions. Salts

that are easily extracted by TBP, such as $NaClO_4$, compete effectively for the solvent and suppress the distribution of Tc(VII) or Re(VII) to the organic phase. Salts that are very poorly extracted, such as Na_2SO_4, increase the distribution of Tc(VII) or Re(VII) to the organic phase by increasing the activity of the cation in the aqueous phase [Eq. (21)]. Small cations that are strongly solvated by TBP (e.g., H^+, Li^+) produce the most extractable XO_4^- compounds, while large, poorly solvated cations produce the less extractable compounds. Finally, a poorly extractable salt with a cation that can be fairly well solvated by TBP (e.g., NaOH, Na_2SP_4) acts as a much more effective salting-out agent than one that is more extractable.

3.5 EXTRACTION FROM URANYL NITRATE–NITRIC ACID SOLUTION

Siddall was the first to report that the addition of uranyl nitrate to these systems greatly enhances the distribution of technetium into the organic phase.[6] Shortly thereafter, this work was extended by Campbell (Table A.18).[7] Several years later, Macasek observed a similar effect using dilute solutions of uranium and technetium in 4.0 M ammonium nitrate. (Table A.24).[10,20] Very recently, Lieser and co-workers published a study of this effect using solutions containing uranium, technetium, HNO_3, and TBP concentrations comparable to those found in some stages of the Purex process.[11] The new work reported here (Tables A.25-A.27) was undertaken to supplement the limited data available on the coextraction of technetium in the Purex process and to develop equations to correlate its distribution coefficient with other, more readily measurable process parameters over a wide range of conditions. The behavior of rhenium under these conditions was also examined in order to evaluate its utility as a nonradioactive model for technetium in Purex-like solvent extraction systems (Table A.28-A.30).

When solutions of TBP in an inert diluent are contacted with aqueous solutions containing uranyl nitrate, HNO_3, and potassium pertechnetate, technetium can be extracted in at least two different ways:[1,6]

$$H^+ + TcO_4^- + 3\overline{TBP} \rightleftarrows [\overline{HTcO_4 \cdot 3TBP}] \ , \qquad (25)$$

$$UO_2^{2+} + NO_3^- + TcO_4^- + 2\overline{TBP} \rightleftarrows [\overline{UO_2(NO_3)(TcO_4) \cdot 2TBP}] \ . \qquad (26)$$

In principle, a uranyl dipertechnetate complex might also extract; however, since the concentration of technetium in these experiments and in systems of practical interest is much smaller than the uranium concentration, this reaction is not significant. This also means that the bulk of the uranium will extract as the familiar dinitrate-bis(tributyl phosphate) complex according to the following equation:

$$UO_2^{2+} + 2NO_3^- + 2\overline{TBP} \rightleftarrows [\overline{UO_2(NO_3)_2 \cdot 2TBP}] \ . \qquad (27)$$

Further, the distribution coefficient of pertechnetic acid declines rapidly as the concentration of uncomplexed or "free" TBP in the organic phase decreases (Sect. 3.3.1). Hence, under conditions where the uranium/technetium ratio is very large and much of the TBP in the organic phase has been complexed by reaction (27), reaction (25) is negligible and reaction (26) becomes the dominant one for extraction of technetium.

Historically, the coextraction of technetium by uranium has been viewed in two conceptually different, but thermodynamically equivalent, ways. Siddall analyzed his data by considering the effect of reactions (25) and (26) occurring simultaneously.[6] Masacek[10] viewed the system as having already attained equilibrium (27), after which technetium extracts via an interphase anion exchange reaction such as

$$[\overline{UO_2(NO_3)_2 \cdot 2TBP}] + TcO_4^- \rightleftarrows [\overline{UO_2(NO_3)TcO_4 \cdot 2TBP}] + NO_3^- \ . \qquad (28)$$

In the following discussion, Macasek's formalism will be followed and the extraction of technetium will be considered on the basis of Eq. (28). It should be noted that no data are available to determine the actual mechanism by which technetium extracts in the presence of uranium and that this formalism is chosen merely for convenience. The thermodynamic relationship between the two treatments becomes obvious when it is recognized that the equilibrium constant for Eq. (28) is the quotient of the equilibrium constants for Eqs. (26) and (27):

$$K = \frac{[\overline{UO_2(NO_3)(TcO_4)\cdot 2TBP}][NO_3^-]}{[\overline{UO_2(NO_3)_2\cdot 2TBP}][TcO_4^-]} \cdot Q \ , \qquad (29)$$

where Q is the quotient of the appropriate molar activity coefficients. Each of the concentration terms in Eq. (28) can be readily determined or calculated from concentrations that are routinely measurable. For the reasons discussed above, the concentration of the organic-phase uranyl pertechnetate complex can be equated with the measured total organic-phase technetium concentration. In all of the results reported below, the amount of technetium in the organic phase is almost negligible in comparison with the amount of uranium present in that phase; thus, the concentration of the organic-phase uranyl dinitrate complex is essentially equal to the measured total organic-phase uranium concentration. The aqueous nitrate concentration is the sum of the equilibrium HNO_3 concentration and twice the measured aqueous-phase uranium concentration. Because $HTcO_4$ is a very strong acid, the measured aqueous concentration of technetium can be equated with $[TcO_4^-]$ in Eq. (29).

These approximations may be summarized as follows:

$$D_{Tc} = \frac{[\overline{Tc}]}{[Tc]} = \frac{[\overline{UO_2(NO_3)(TcO_4)\cdot 2TBP}]}{[TcO_4^-]} \ . \qquad (30)$$

Combining Eqs. (25) and (26) and taking logarithms yields:

$$\ln D_{Tc} = \ln(K/Q) + \ln [\overline{U}] - \ln [NO_3^-] \ . \qquad (31)$$

where $[NO_3^-]$ is the total, stoichiometric concentration of nitrate in the aqueous phase at equilibrium (i.e., $[NO_3^-] = [HNO_3] + 2[UO_2(NO_3)_2]$). Equation (30) might be used to calculate values for D_{Tc} under conditions similar to those used in the Purex process, in terms of two process variables that are routinely monitored and controlled. However, this would require knowledge of how K and Q vary as functions of temperature and concentration and, as in the simpler HCl and HNO_3 systems, use of a less rigorous method. Hence, the same statistical methods described in Sect. 3.2.4 and 3.3.2 were used to fit the distribution data from three different temperatures to an equation of the form of Eq. (31) and to obtain functions to predict values of D_{Tc} at 25, 40, and 60°C. These functions are shown as Eqs. (31)-(33), respectively:

$$\ln D_{Tc} = 0.147 + 0.370 \ln [\overline{U}] - 0.897 \ln [NO_3^-] \text{ at } 25°C , \quad (32)$$

$$\ln D_{Tc} = 0.341 + 0.573 \ln [\overline{U}] - 0.832 \ln [NO_3^-] \text{ at } 40°C , \quad (33)$$

$$\ln D_{Tc} = 0.394 + 0.656 \ln [\overline{U}] - 0.750 \ln [NO_3^-] \text{ at } 60°C . \quad (34)$$

Unfortunately, this treatment completely ignores the changes in Q that occur as the concentrations of the various reagents change, making it impossible to assign any physical meaning to the coefficients in Eqs. (32)-(34). Nonetheless, Figs. 24-26 show the success of this approach by comparing the values of D_{Tc} calculated using Eqs. (32)-(34) with the actual experimental values listed in Tables A.24-A.26. Only one calculated value of D_{Tc} in each of the 40 and 60°C data sets differ from the measured value by more than 10%, while there are four such values for the 25°C data set. The difference between the measured and experimental values of D_{Tc} never exceeds 21%.

One further modification of Eq. (31) can be made. As in the derivation of Eq. (7), the relationship $\Delta G = -RT \ln K = \Delta H - T\Delta S$ is used to introduce a temperature-dependent term in Eq. (31) and allow all of the data in Tables A.25-A.27 to be fit to a single equation. The result of this procedure is Eq. (35):

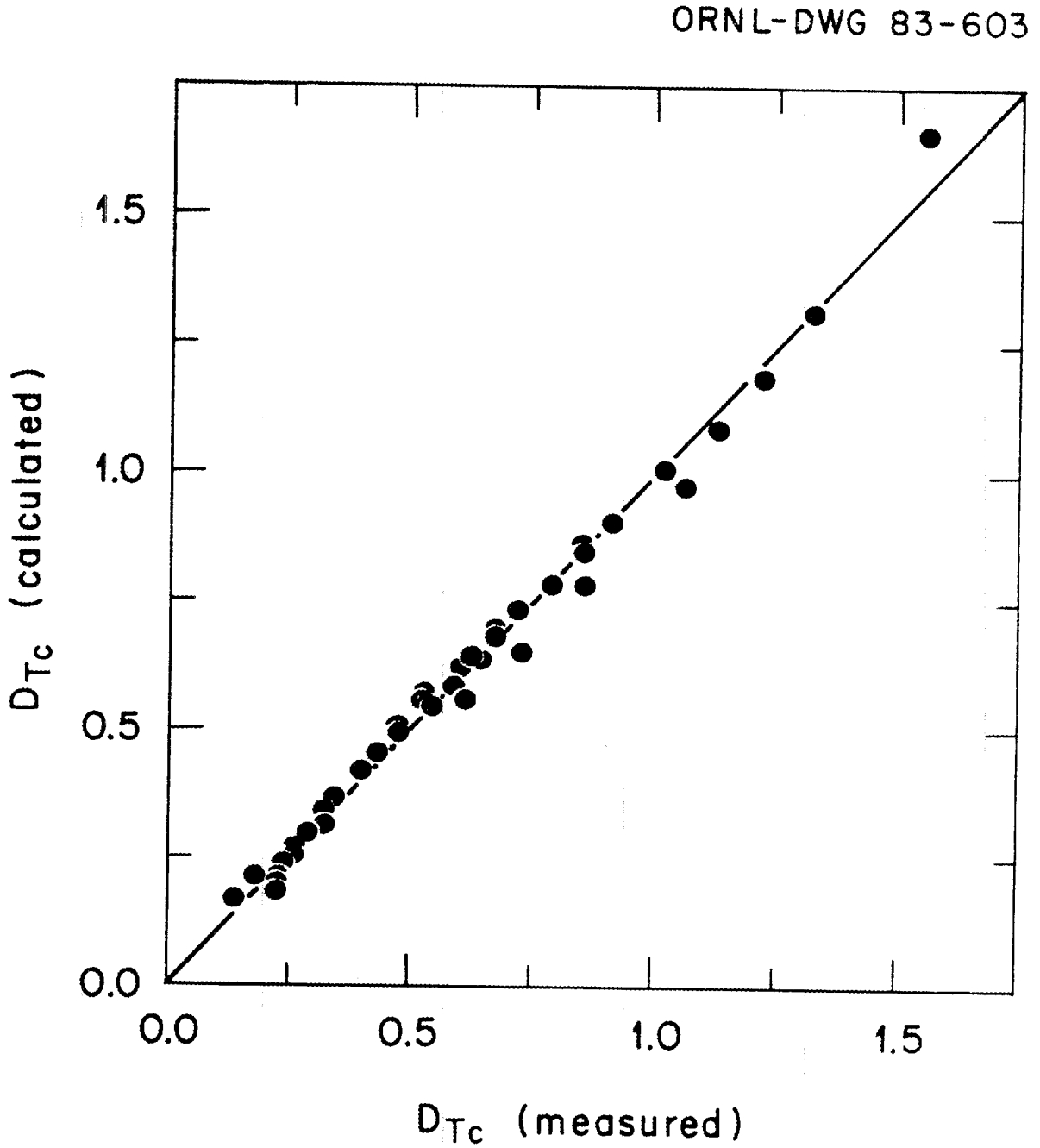

Fig. 24. Comparison of D_{Tc} in the presence of uranium at 25°C as calculated from Eq. (31) with experimental values of D_{Tc}.

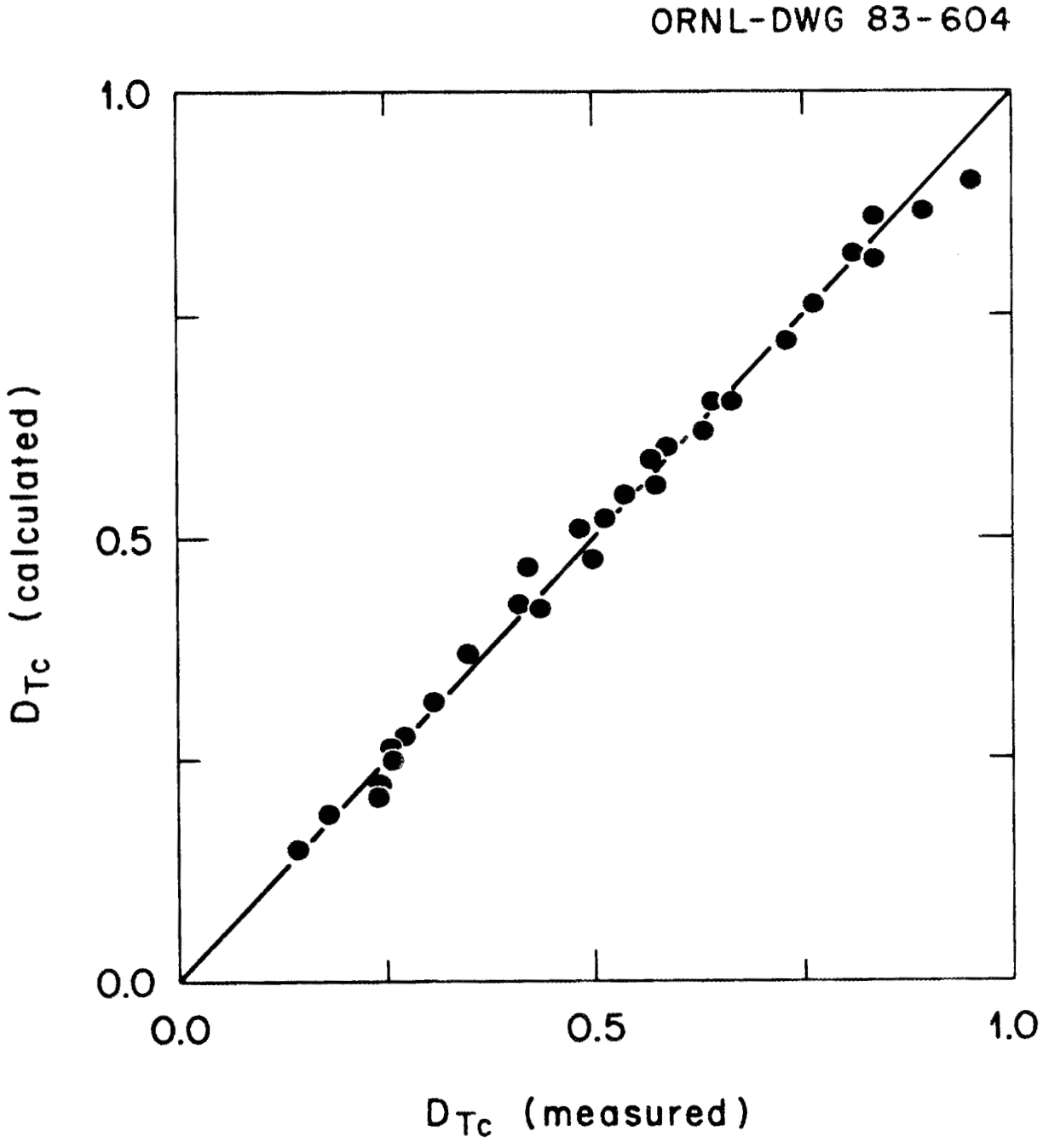

Fig. 25. Comparison of D_{Tc} in the presence of uranium at 40°C as calculated from Eq. (32) with experimental values of D_{Tc}.

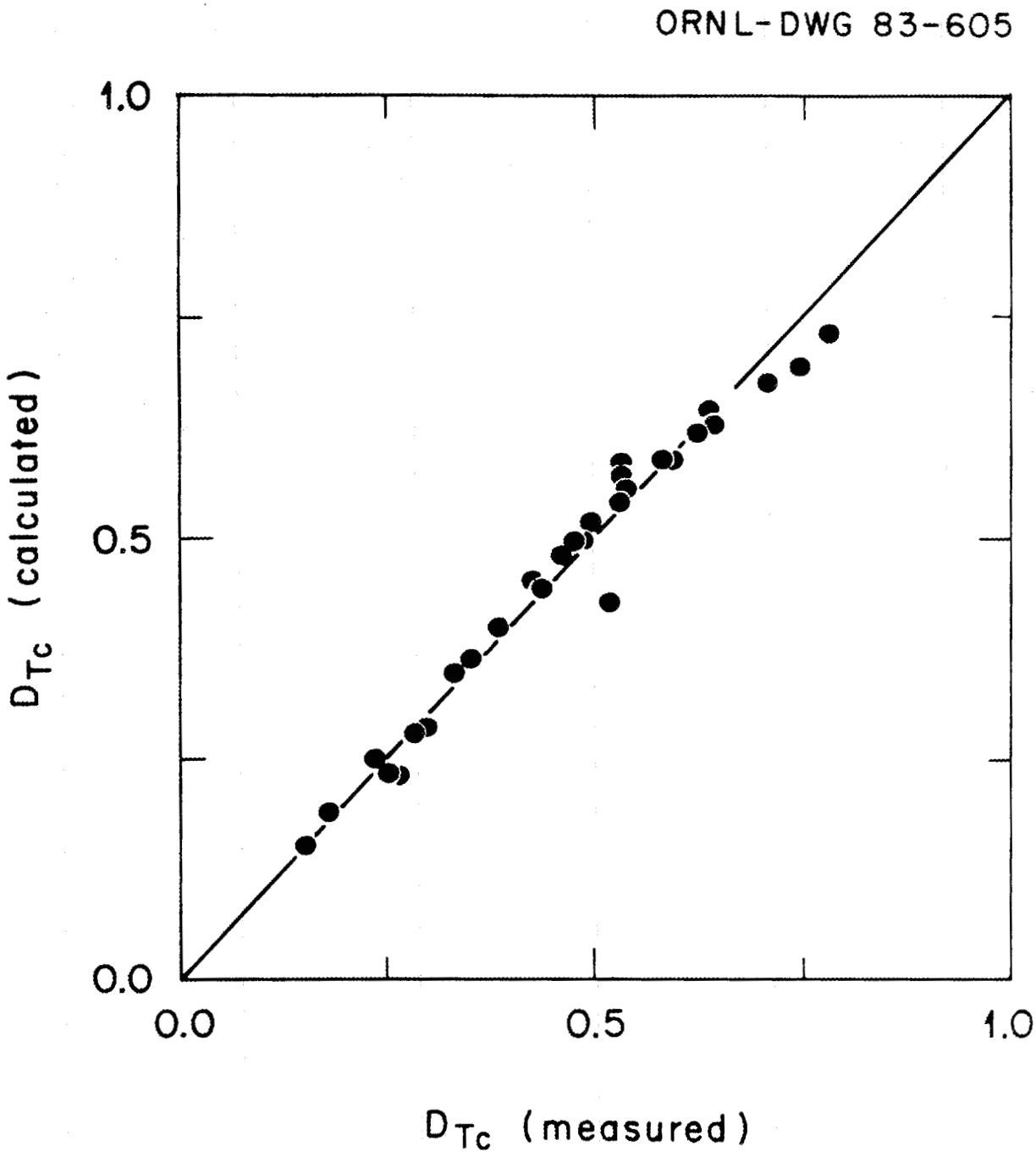

Fig. 26. Comparison of D_{Tc} in the presence of uranium at 60°C as calculated from Eq. (33) with experimental values of D_{Tc}.

$$\ln D_{Tc} = -0.548 - 54.4/T + 0.641 \ln [\overline{U}] - 0.879 \ln [NO_3^-] \ . \quad (35)$$

Figure 27 shows that, with the exception of the six points connected with a dashed line, Eq. (24) correlates a broad range of data quite successfully. The six outlying points indicated in the figure are the first six points listed in Table A.25. These points could probably be ignored since they were obtained under conditions where the assumptions used to justify exclusion of the effects of equilibrium (25) are the poorest [i.e., the $[U]_{org}$ is low and the $[HTcO_4]_{org}$ is relatively high]. The fact that these six calculated values of D_{Tc} are all lower than the measured values suggests that some technetium is being extracted as $HTcO_4$. However, attempts to correct these values using the known behavior of $HTcO_4$ resulted in only a modest improvement, probably because of the uncertainty in calculating the free TBP concentrations in these solutions.

These equations (and the data from which they are derived) indicate that only a relatively small amount, certainly less than 5%, of the technetium that enters the solvent extraction portion of a fuel reprocessing plant will remain with the product uranium stream if a high-acid Purex flowsheet is used. Using similar data, Siddall[6] calculated that between 4 and 40% of this technetium would be recovered with the uranium product, depending on the flowsheet considered. Using a low-acid flowsheet and a laboratory-scale, multiple-batch, countercurrent mockup of a Purex solvent extraction process, Campbell[7] found that almost 80% of the technetium that entered the process followed the uranium stream. The author was unable to obtain data on the amount of technetium that has actually been found in the product of operating fuel reprocessing plants. While it is well established that sufficient technetium is present in reprocessed uranium to cause operational problems in gaseous diffusion plans,[28] the fraction of technetium carried with the product uranium has apparently not been reported. In order to validate the results of calculations based on Eqs. (32)-(35) and the results of both Siddall and Campbell (all of which are in good agreement), it will be necessary to obtain data from an operating Purex plant. Other factors,

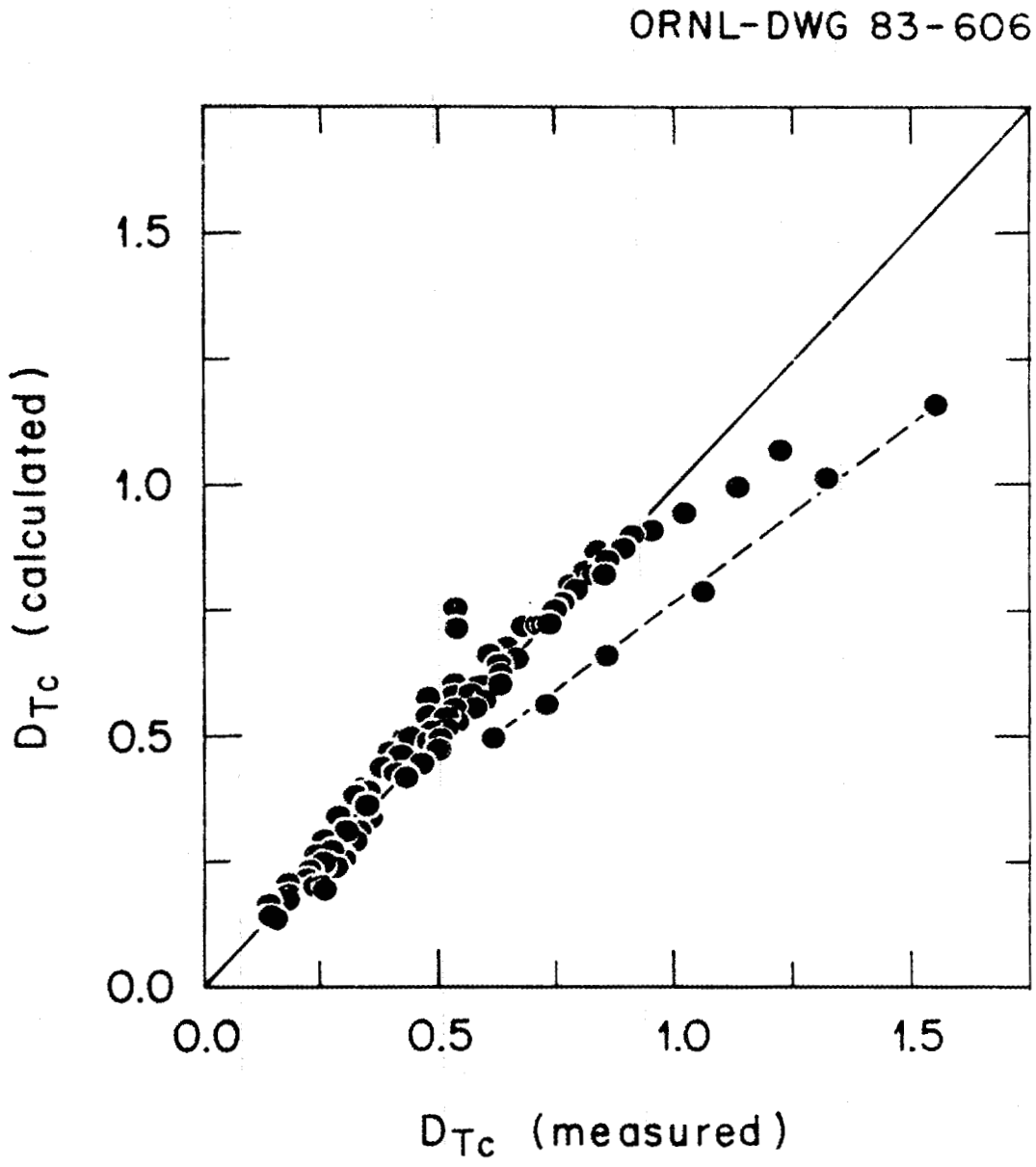

Fig. 27. Comparison of D_{Tc} in the presence of uranium at 25 to 60°C as calculated from Eq. (34) with experimental values of D_{Tc}.

such as the presence of the other actinide ions (discussed below), may have significant effects on technetium decontamination.

As expected from earlier work showing the similarity of pertechnetate and perrhenate in solvent extraction systems,[1-4] it was found that the addition of uranyl nitrate to HNO_3–TBP–NDD solvent extraction systems greatly enhances the distribution of perrhenate into the organic phase (Table A.28-A.30). Analysis of these data using the methods described above for the technetium data shows that the distribution coefficient for rhenium can be calculated from the following equations:

$$\ln D_{Re} = 1.58 + 0.216 \ln [\overline{U}] - 0.938 \ln [NO_3^-] \text{ at } 25°C , \quad (36)$$

$$\ln D_{Re} = -1.12 + 0.593 \ln [\overline{U}] - 0.940 \ln [NO_3^-] \text{ at } 40°C , \quad (37)$$

$$\ln D_{Re} = -0.963 + 0.678 \ln [\overline{U}] - 0.907 \ln [NO_3^-] \text{ at } 60°C , \quad (38)$$

$$\ln D_{Re} = -0.980 - 66.4/T + 0.523 \ln [\overline{U}] - 0.907 \ln [NO_3^-] \text{ at all } T . \quad (39)$$

3.6 EXTRACTION FROM THORIUM NITRATE–NITRIC ACID SOLUTION

A survey of the literature reveals that the only published data for the mixed thorium-technetium system are those reported by Macasek and Kadrabova.[20] In a brief study at 25°C, they demonstrated that technetium could be extracted as $Th(TcO_4)(NO_3)_3 \cdot 2TBP$ from 4 M NH_4NO_3, and that a given concentration of thorium in the organic phase extracted on the order of ten times more technetium than did the same concentration of uranium.

Tables A.31-A.36 summarize the results of a series of experiments performed by the author to determine the effect of thorium on the extraction of technetium and rhenium by 30.1 vol % (1.10 M) from HNO_3 at 25, 40, and 60°C. These data were analyzed by the same method used for the uranium nitrate–nitric acid system (Sect. 3.5), except that Eq. (40) was assumed to be the extraction reaction for pertechnetate and perrhenate,

$$\overline{Th(NO_3)_4 \cdot 2TBP} + MO_4 \quad \overline{Th(MO_4)(NO_3)_3 \cdot 2TBP} + NO_3^- \ . \qquad (40)$$

Fitting the data to an equation of the form of Eq. (31), but using $[\overline{Th}]$ instead of $[\overline{U}]$, yields Eqs. (41) and (42) for technetium and rhenium, respectively:

$$\ln D_{Tc} = -0.560 + 1398/T + 0.921 \ln [\overline{Th}] - 0.780 \ln [NO_3^-] \ , \qquad (40)$$

$$\ln D_{Re} = -2.03 + 1408/T + 0.814 \ln [\overline{Th}] - 0.806 \ln [NO_3^-] \ . \qquad (41)$$

As expected, D_{Tc} and D_{Re} show roughly a first-order dependence on $[\overline{Th}]$ and an inverse first-order dependence on $[NO_3^-]$. The greatest difference between the uranium and thorium systems is expressed in the "$\Delta H/R$" term, which indicates a ΔH value of about 0.03 kcal/mol for the uranium nitrate—XO_4^- exchange reaction (i.e., the reaction is only weakly temperature dependent) and a value of -2.8 kcal/mol for the thorium nitrate—XO_4^- exchange reaction. Apparently, the relatively "hard" TcO_4^- and ReO_4^- ions are more strongly bonded to the hard Th(IV) cation than to the relatively softer UO_2^{2+} cation. This observation has important consequences in the LMFBR fuel cycle, where Pu(IV) can be expected to show an affinity for TcO_4^- very similar to that observed for Th(IV), This expectation is confirmed by the data discussed in the following section.

3.7 EXTRACTION FROM PLUTONIUM NITRATE—NITRIC ACID SOLUTION

The limited amount of data obtained on the simultaneous extraction of Tc(VII) and Pu(IV) is included in Table A.37. These data are somewhat less reliable than those for the uranium-technetium and thorium-technetium systems, for several reasons. Only the aqueous phase was analyzed for plutonium and technetium. The initial aqueous phases were made up from stock solutions of known Pu(IV), Tc(VII), and HNO_3 concentrations. Aliquots of these initial solutions were kept for analysis, while the bulk of the solution was equilibrated with an equal volume of 30.1% TBP. However, there was no constant-temperature bath

available in the plutonium glove box, and the ambient temperature may have varied by as much as ±2°C from the nominal value of 25°C. After equilibration, only the aqueous phase was analyzed for plutonium, technetium, and HNO_3; the organic-phase concentrations were determined by material balance.

Nonetheless, even with these limitations in mind, the data clearly show the very strong effect of the presence of plutonium on D_{Tc}. It is clear that the interaction between pertechnetate and Pu(IV) in the organic phase has a substantial influence on the overall decontamination of uranium and plutonium from technetium in Purex systems, especially when fuels containing a large amount of plutonium (i.e., breeder fuels) are reprocessed.

4. CONCLUSIONS

The following conclusions can be drawn from the results of the study presented here:

1. The fate of technetium entering a nuclear fuel reprocessing plant has not been determined quantitatively in the past.
2. Previous studies of the solvent extraction behavior of technetium in the Purex process, while in some cases well done for a limited range of conditions,[7] were generally too limited in scope to allow reliable calculation of the flow of technetium through a reprocessing plant, especially in cases where fuels with high plutonium contents were involved.
3. The results presented here, along with the data obtained by other workers on the behavior of technetium during the dissolution of a variety of nuclear fuels, make it possible to calculate, for the first time, reasonable estimates for the amount of technetium that will flow through each portion of a given reprocessing flowsheet.

However, actual data from an operating reprocessing plant must be obtained in order to validate the calculated estimates.

4. Because of the limited quantity and relatively poor quality of the mixed plutonium-technetium extraction data, additional studies will be required before these systems can be accurately modeled. Specifically, laboratory-scale or pilot-plant-scale studies of solvent extraction systems containing U(VI), Pu(IV or III), and Tc(VII), similar to those for the U(VI)-Tc(VII) system,[7] are needed.

5. Perrhenate can serve as an excellent, nonradioactive model for technetium in any Purex-like solvent extraction system under nonreducing conditions.

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APPENDIX

Table A.1. Distribution of perrhenate or pertechnetate between aqueous hydrochloric acid and tributyl phosphate in n-dodecane at 25°C

$[HCl]_{aq}$ (M)	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}	D_{Tc}/D_{Re}
[TBP] = 0.339 M (9.28 vol %)							
0.20	—	--	--	0.0788	3.88	0.0203	—
0.40	0.043	2.78	0.015	0.151	3.89	0.0388	2.6
0.60	0.091	2.80	0.033	0.206	3.75	0.0549	1.7
0.80	0.102	2.80	0.036	0.291	3.64	0.0799	2.2
1.00	0.129	2.88	0.045	0.292	3.70	0.0789	1.8
2.00	0.360	2.64	0.136	0.748	3.25	0.230	1.7
3.00	0.778	2.23	0.349	1.92	3.25	0.591	1.7
4.00	1.18	1.90	0.621	1.87	2.09	0.895	1.4
[TBP] = 0.738 M (20.2 vol %)							
0.20	0.204	2.59	0.079	0.559	3.31	0.169	2.1
0.40	0.381	2.43	0.157	0.980	3.04	0.322	2.1
0.60	0.569	2.21	0.257	1.40	2.69	0.520	2.0
0.80	0.612	2.14	0.286	1.66	2.36	0.703	2.5
1.00	0.821	1.98	0.415	1.99	2.19	0.909	2.2
2.00	1.44	1.29	1.12	2.90	1.17	2.48	2.2
3.00	1.99	0.805	2.47	3.51	0.658	5.33	2.2
4.00	2.21	0.553	4.00	4.00	0.373	10.7	2.7
[TBP] = 1.11 M (30.5 vol %)							
0.20	0.644	2.19	0.294	0.984	1.42	0.693	2.4
0.40	1.06	1.77	0.599	1.38	1.06	1.30	2.2
0.60	1.48	1.44	1.03	1.64	0.817	2.01	1.9
0.80	1.65	1.20	1.38	1.72	0.654	2.63	1.9
1.00	1.87	1.01	1.85	1.80	0.532	3.38	1.8
2.00	2.35	0.489	4.81	2.16	0.224	9.64	2.0
3.00	2.56	0.263	9.73	2.30	0.118	19.5	2.0
4.00	2.69	0.150	17.9	2.32	0.0685	34.1	1.9
[TBP] = 1.45 M (39.6 vol %)							
0.20	1.20	1.81	0.663	2.53	1.66	1.52	2.3
0.40	1.83	1.14	1.61	3.02	1.00	3.02	1.9
0.60	2.21	0.843	2.62	3.33	0.722	4.61	1.8
0.80	2.29	0.671	3.41	3.51	0.562	6.25	1.8
1.00	2.46	0.537	4.58	3.67	0.428	8.57	1.9
2.00	2.83	0.242	11.7	3.96	0.166	23.9	2.0
3.00	3.03	0.102	29.7	4.10	0.0918	44.6	1.5
4.00	3.09	0.059	52.4	4.20	0.0605	68.9	1.3
[TBP] = 2.18 M (59.7 vol %)							
0.20	2.24	0.752	2.98	4.14	0.780	5.31	1.8
0.40	2.53	0.419	6.04	4.43	0.407	10.9	1.8
0.60	2.56	0.306	8.37	4.63	0.283	16.4	2.0
0.80	2.49	0.231	10.8	4.74	0.211	22.5	2.1
1.00	2.75	0.166	16.6	4.68	0.147	31.8	1.9
2.00	2.85	0.075	38.0	4.80	0.0573	83.8	2.2
3.00	2.82	0.043	65.6	4.78	0.0278	172	2.6
4.00	2.78	0.027	103	4.51	0.0141	320	3.1
[TBP] = 2.90 M (79.3 vol %)							
0.20	2.49	0.268	9.29	3.65	0.244	15.0	1.6
0.40	2.61	0.156	16.7	3.83	0.116	33.0	2.0
0.60	2.61	0.123	21.2	3.89	0.0799	48.7	2.3
0.80	2.68	0.097	27.6	3.84	0.0693	55.4	2.0
1.00	2.69	0.059	45.6	3.85	0.0383	101	2.2
2.00	2.71	0.027	100	3.83	0.0160	239	2.4
3.00	2.72	0.014	194	3.87	0.0085	455	2.3
4.00	2.74	0.009	304	3.80	0.0043	884	2.9

Table A.2. Distribution of perrhenate or pertechnetate between aqueous hydrochloric acid and tributyl phosphate in n-dodecane at 40°C

$[HCl]_{aq}$ (M)	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}	D_{Tc}/D_{Re}
[TBP] = 0.339 M (9.28 vol %)							
0.20	--	--	--	0.0126	3.91	0.00322	--
0.40	--	--	--	0.0257	3.96	0.00649	--
0.60	--	--	--	0.0389	3.93	0.00990	--
0.79	--	--	--	0.0558	3.94	0.0141	--
0.99	--	--	--	0.0718	3.94	0.0182	--
1.98	0.070	2.82	0.025	0.203	3.86	0.0526	2.1
2.96	0.171	2.70	0.063	0.413	3.56	0.116	1.8
3.92	0.295	2.68	0.110	0.804	3.19	0.252	2.3
[TBP] = 0.738 M (20.0 vol %)							
0.20	0.056	2.80	0.020	0.156	3.73	0.0418	2.1
0.40	0.064	2.70	0.024	0.297	3.67	0.0809	3.4
0.60	0.102	2.70	0.0378	0.423	3.61	0.117	3.1
0.79	0.183	2.63	0.0696	0.579	3.34	0.173	2.5
0.99	0.274	2.55	0.107	0.716	3.21	0.223	2.1
1.98	0.569	2.21	0.258	1.40	2.06	0.680	2.6
2.96	1.01	1.74	0.581	2.42	1.56	1.55	2.7
3.92	1.41	1.31	1.08	3.26	1.03	3.17	2.9
[TBP] = 1.11 M (30.5 vol %)							
0.20	0.199	2.37	0.0840	0.495	3.06	0.162	1.9
0.40	0.483	2.36	0.205	0.831	2.59	0.321	1.6
0.60	0.553	2.22	0.249	1.10	2.34	0.470	1.9
0.79	0.591	2.14	0.276	1.45	2.09	0.694	2.5
0.99	0.741	2.00	0.371	1.58	1.90	0.832	2.2
1.98	1.47	1.28	1.15	2.12	1.01	2.10	1.8
2.96	1.88	0.891	2.11	2.88	0.561	5.13	2.4
3.92	2.14	0.580	3.69	2.19	0.299	7.32	2.0
[TBP] = 1.45 M (39.6 vol %)							
0.20	0.553	2.36	0.234	0.973	2.56	0.380	1.6
0.40	0.875	1.99	0.440	1.57	2.01	0.781	1.8
0.60	1.12	1.72	0.651	1.91	1.67	1.14	1.8
0.79	1.45	1.40	1.04	2.07	1.40	1.48	1.4
0.99	1.51	1.29	1.17	2.35	1.10	2.14	1.8
1.98	2.22	0.650	3.42	2.86	0.560	5.11	1.5
2.96	2.42	0.365	6.63	3.22	0.228	14.1	2.1
3.92	2.67	0.166	16.1	3.35	0.142	23.6	1.5
[TBP] = 2.18 M (59.7 vol %)							
0.20	1.11	1.61	0.689	2.35	1.47	1.60	2.3
0.40	1.40	1.28	1.09	2.86	0.972	2.94	2.7
0.60	2.01	0.789	2.55	3.10	0.693	4.47	1.8
0.79	2.19	0.585	3.74	3.28	0.519	6.32	1.7
0.99	2.22	0.478	4.64	3.44	0.407	8.45	1.8
1.98	2.55	0.188	13.6	3.66	0.148	24.7	1.8
2.96	2.67	0.113	23.6	3.73	0.0728	51.2	2.2
3.92	2.68	0.0483	55.5	3.79	0.0452	83.8	1.5
[TBP] = 2.90 M (79.3 vol %)							
0.20	2.10	0.762	2.76	3.21	0.676	4.75	1.7
0.40	2.39	0.462	5.17	3.49	0.361	9.67	1.9
0.60	2.73	0.322	8.48	3.71	0.236	15.7	1.9
0.79	2.64	0.242	10.9	3.32	0.125	26.6	2.4
0.99	2.74	0.193	14.2	3.84	0.131	29.3	2.1
1.98	2.90	0.0859	33.8	3.82	0.0428	89.3	2.6
2.96	2.79	0.0537	52.0	3.81	0.0201	190	3.6
3.92	2.73	0.0268	102	3.74	0.0179	209	2.0

Table A.3. Distribution of perrhenate or pertechnetate between aqueous hydrochloric acid and tributyl phosphate in n-dodecane at 60°C

$[HCl]_{aq}$ (M)	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}	D_{Tc}/D_{Re}
[TBP] = 0.339 M (9.28 vol %)							
0.20	—	—	—	0.0035	3.44	0.0010	—
0.40	—	—	—	0.0066	3.47	0.0019	—
0.60	—	—	—	0.0115	3.45	0.0033	—
0.78	—	—	—	0.0155	3.49	0.0044	—
0.98	—	—	—	0.0215	3.51	0.0061	—
1.96	—	—	—	0.0618	3.41	0.0181	—
2.93	—	—	—	0.139	3.34	0.0416	—
3.88	—	—	—	0.293	3.18	0.0921	—
[TBP] = 0.738 M (20.2 vol %)							
0.20	—	—	—	0.0370	3.33	0.0111	—
0.40	0.037	2.83	0.013	0.0685	3.47	0.0197	1.5
0.60	0.070	2.76	0.025	0.0994	3.37	0.0295	1.2
0.78	0.107	2.81	0.0381	0.132	3.31	0.0399	1.0
0.98	0.135	2.75	0.0491	0.185	3.29	0.0562	1.1
1.96	0.276	2.65	0.104	0.473	3.07	0.154	1.5
2.93	0.472	2.37	0.199	0.893	2.64	0.338	1.7
3.88	0.805	2.08	0.387	1.46	2.12	0.689	1.8
[TBP] = 1.11 M (30.5 vol %)							
0.20	0.0537	2.73	0.0197	0.144	3.34	0.0431	2.2
0.40	0.118	2.79	0.0423	0.255	3.29	0.0775	1.8
0.60	0.204	2.63	0.0776	0.378	3.14	0.120	1.5
0.78	0.328	2.56	0.128	0.489	3.04	0.161	1.3
0.98	0.376	2.46	0.153	0.605	2.96	0.204	1.3
1.96	0.719	2.17	0.331	1.29	2.20	0.586	1.8
2.93	1.27	1.58	0.804	2.03	1.39	1.46	1.8
3.88	1.56	1.20	1.30	2.55	0.907	2.81	2.2
[TBP] = 1.45 M (39.6 vol %)							
0.20	0.123	2.72	0.0452	0.318	3.28	0.0970	2.1
0.40	0.225	2.58	0.0872	0.549	3.06	0.179	2.1
0.60	0.478	2.35	0.203	0.761	2.85	0.267	1.3
0.78	0.644	2.22	0.290	0.959	2.56	0.375	1.3
0.98	0.660	2.14	0.308	1.24	2.38	0.521	1.7
1.96	1.43	1.39	1.03	2.04	1.32	1.55	1.5
2.93	1.92	0.907	2.12	2.67	0.763	3.50	1.7
3.88	2.24	0.558	4.01	3.04	0.457	6.65	1.7
[TBP] = 2.18 M (59.7 vol %)							
0.20	0.526	2.36	0.223	1.08	2.41	0.448	2.0
0.40	0.929	1.96	0.474	1.56	1.99	0.784	1.7
0.60	1.11	1.68	0.661	1.93	1.71	1.13	1.7
0.78	1.55	1.35	1.15	2.14	1.36	1.57	1.4
0.98	1.60	1.24	1.29	2.44	1.21	2.02	1.6
1.96	1.15	0.403	2.85	3.12	0.540	5.78	2.0
2.93	2.58	0.242	10.7	3.41	0.251	13.6	1.3
3.88	2.76	0.124	22.3	3.56	0.148	24.1	1.1
[TBP] = 2.90 M (79.3 vol %)							
0.20	1.33	1.57	0.847	2.14	1.64	1.30	1.5
0.40	1.73	1.11	1.56	2.47	1.06	2.33	1.5
0.60	2.08	0.811	2.56	2.75	0.815	3.37	1.3
0.78	2.38	0.569	4.18	2.71	0.630	4.30	1.0
0.98	2.52	0.408	6.18	2.99	0.431	6.94	1.1
1.96	2.79	0.183	15.2	3.33	0.190	17.5	1.2
2.93	3.04	0.113	26.9	3.29	0.0859	38.3	1.4
3.88	2.85	0.054	52.8	3.53	0.0465	75.9	1.4

Table A.4. Mean molar activity coefficients for hydrochloric acid

25°C			40°C			60°C		
[HCl]	$\gamma_{\pm}$	$a_{\pm}$	[HCl]	$\gamma_{\pm}$	$a_{\pm}$	[HCl]	$\gamma_{\pm}$	$a_{\pm}$
0.20	0.768	0.154	0.20	0.754	0.151	0.20	0.735	0.147
0.40	0.758	0.303	0.40	0.739	0.296	0.40	0.714	0.286
0.60	0.779	0.467	0.60	0.756	0.454	0.60	0.713	0.428
0.80	0.795	0.636	0.79	0.767	0.606	0.78	0.723	0.564
1.00	0.827	0.827	0.99	0.795	0.787	0.98	0.756	0.741
2.00	1.07	2.14	1.98	1.02	2.02	1.96	0.953	1.87
3.00	1.48	4.44	2.96	--	--	2.93	--	--
4.00	2.15	8.60	3.92	--	--	3.88	--	--

Table A.5. Distribution of perrhenate[a] between aqueous acid solutions and 20 vol % tributyl phosphate[b,c]

Normality	HNO_3	$HClO_4$	HCl	H_2SO_4
0	0.015	0.015	0.015	0.015
1	0.14	0.031	0.54	0.39
2	0.087	0.007	1.27	0.91
3	0.032	0.001	2.77	1.42
4	0.015	--	5.99	--
5.6	--	--	--	5.65
6	0.0018	--	--	--
7.2	--	--	--	11.5
9	<0.0001	--	--	18.44
12	<0.0001	--	--	--

[a]Initial $[HReO_4]$ = 14.2 mM in the aqueous phase.
[b]Data adapted from ref. 13.
[c]No temperature reported for this study.

Table A.6. Distribution coefficient of pertechnetate between various 1 N aqueous solutions and tributyl phosphate in cyclohexane at 25°C[a]

[TBP] (M)	H_2SO_4	Na_2SO_4	NaOH
(neat)	44	8.3	7.5
0.1	0.27	0.002	0.001

[a]Data adapted from ref. 5.

Table A.7. Distribution coefficient of pertechnetate between 1 N solutions of various strong acids and pure TBP at room temperature[a]

H_2SO_4	HCl	HBr	HNO_3	HI	$HClO_4$
44	120	91	29	79[b]	5.6

[a]Data adapted from ref. 5.
[b]The authors note that Tc(VII) is thermodynamically unstable toward reduction in 1 N HI and that the valence state extracted may have been Tc(V).

Table A.8. Distribution coefficient of pertechnetate between various 1 M nitrate solutions and pure TBP at room temperature[a]

HNO_3	$LiNO_3$	NH_4NO_3	$NaNO_3$	KNO_3	$Ca(NO_3)_2$	$Cu(NO_3)_2$	$Al(NO_3)_2$
28.3	14.7	4.6	2.2	1.1	15.6	5.4	4.4

[a]Data adapted from ref. 5.

Table A.9. Effect of adding LiCl, NaCl, or KCl to 1 M HCl on the extraction of 0.002 M $KTcO_4$ by 30 vol % tributyl phosphate in n-dodecane

[MCl] (M)	Temperature (°C)	D_{Tc} LiCl	NaCl	KCl
0.1	25	3.86	3.54	2.43
0.2	25	4.25	3.40	2.35
0.3	25	4.48	--	--
0.4	25	4.69	3.68	2.46
0.5	25	5.05	--	--
0.6	25	5.29	--	--
0.78	25	5.90	--	--
0.8	25	--	3.62	2.34
1.0	25	--	3.98	2.43
2.0	25	--	5.41	2.15
3.0	25	--	6.74	2.33
4.0	25	--	9.28	--
0.1	40	1.11	0.825	0.820
0.2	40	--	0.830	0.791
0.3	40	1.22	--	--
0.4	40	1.25	0.918	0.847
0.5	40	1.34	--	--
0.6	40	1.51	--	--
0.78	40	1.61	--	--
0.8	40	--	0.966	0.730
1.0	40	--	1.05	0.740
2.0	40	--	1.42	0.734
3.0	40	--	1.77	0.786
4.0	40	--	2.52	--
0.1	60	0.259	0.219	0.202
0.2	60	--	0.228	0.205
0.3	60	0.299	--	--
0.4	60	0.312	0.241	0.214
0.5	60	0.347	--	--
0.6	60	0.375	--	--
0.78	60	0.406	--	--
0.8	60	--	0.265	0.210
1.0	60	--	0.272	0.191
2.0	60	--	0.395	0.214
3.0	60	--	0.513	0.225
4.0	60	--	0.687	--

Table A.10. Distribution of perrhenate and pertechnetate between aqueous nitric acid and tributyl phosphate in n-dodecane at 25°C

$[HNO_3]_{aq}$ (M)	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}	D_{Tc}/D_{Re}
[TBP] = 0.365 M (9.99 vol %)							
0.10	—	—	—	0.0311	4.71	0.00660	—
0.20	—	—	—	0.0554	4.61	0.0120	—
0.40	—	—	—	0.0913	4.61	0.0198	—
0.50	—	—	—	0.104	4.58	0.0227	—
0.60	0.030	3.00	0.010	0.116	4.61	0.0252	2.5
0.70	—	—	—	0.121	4.60	0.0263	—
0.80	0.035	2.92	0.012	0.125	4.56	0.0274	2.3
0.90	—	—	—	0.126	4.55	0.0277	—
1.00	0.032	3.00	0.011	0.126	4.63	0.0272	2.5
2.00	—	—	—	0.0791	4.63	0.0171	—
3.00	—	—	—	0.0348	4.68	0.00744	—
4.00	—	—	—	0.0145	4.60	0.00315	—
[TBP] = 0.732 M (19.8 vol %)							
0.10	—	—	—	0.230	4.63	0.0497	—
0.20	0.11	2.67	0.041	0.402	4.57	0.0880	2.1
0.40	0.20	2.54	0.079	0.636	4.14	0.154	1.9
0.50	—	—	—	0.743	4.00	0.186	—
0.60	0.25	2.61	0.096	0.824	3.92	0.210	2.2
0.70	—	—	—	0.906	3.97	0.228	—
0.80	0.26	2.49	0.10	0.876	3.89	0.225	2.2
0.90	—	—	—	0.881	3.99	0.221	—
1.00	0.23	2.58	0.089	0.826	3.87	0.213	2.4
2.00	0.13	2.72	0.048	0.443	4.29	0.103	2.1
3.00	0.032	2.90	0.011	0.202	4.69	0.0431	3.9
4.00	—	—	—	0.0858	4.72	0.0182	—
[TBP] = 1.10 M (30.0 vol %)							
0.10	—	—	—	0.616	2.02	0.305	—
0.20	0.48	2.31	0.21	0.896	1.61	0.557	2.7
0.40	0.72	2.08	0.35	1.10	1.35	0.815	2.3
0.60	0.74	2.03	0.36	1.17	1.33	0.880	2.4
0.80	0.79	2.11	0.37	1.18	1.30	0.908	2.5
1.00	0.70	2.08	0.34	1.05	1.33	0.789	2.3
1.50	—	—	—	0.841	1.47	0.572	—
2.00	0.44	2.35	0.19	0.619	1.80	0.344	1.8
2.50	—	—	—	0.428	1.87	0.229	—
3.00	0.21	2.70	0.078	0.281	1.96	0.143	1.8
3.50	—	—	—	0.187	2.02	0.0926	—
4.00	0.048	2.97	0.016	0.122	2.11	0.0578	3.6
5.00	—	—	—	0.0640	2.40	0.0267	—
6.00	—	—	—	0.0318	2.31	0.0138	—
[TBP] = 1.46 M (40.1 vol %)							
0.10	—	—	—	—	—	—	—
0.20	1.12	1.86	0.602	—	—	—	—
0.40	1.27	1.65	0.770	—	—	—	—
0.50	—	—	—	—	—	—	—
0.60	1.44	1.50	0.960	—	—	—	—
0.70	—	—	—	—	—	—	—
0.80	1.49	1.54	0.968	—	—	—	—
0.90	—	—	—	—	—	—	—
1.00	1.47	1.45	1.01	—	—	—	—
2.00	0.913	1.87	0.488	—	—	—	—
3.00	0.467	2.27	0.206	—	—	—	—
4.00	0.209	2.68	0.0780	—	—	—	—

Table A.10 (continued)

$[HNO_3]_{aq}$ (M)	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}	D_{Tc}/D_{Re}
[TBP] = 2.18 M (59.8 vol %)							
0.10	—	—	—	3.28	1.43	2.29	—
0.20	1.86	0.907	2.05	3.61	0.965	3.74	1.8
0.40	2.14	0.736	2.91	3.72	0.714	5.21	1.8
0.50	—	—	—	3.87	0.696	5.56	—
0.60	2.18	0.660	3.30	4.05	0.704	5.75	1.7
0.70	—	—	—	3.76	0.639	5.88	—
0.80	2.23	0.639	3.49	3.63	0.648	5.60	1.6
0.90	—	—	—	3.65	0.702	5.20	—
1.00	2.10	0.816	2.57	3.82	0.802	4.76	1.9
2.00	1.78	1.19	1.50	2.99	1.51	1.98	1.3
3.00	1.16	1.76	0.659	2.06	2.53	0.814	1.2
4.00	0.687	2.39	0.287	1.19	3.46	0.344	1.2
[TBP] = 2.93 M (80.2 vol %)							
0.10	—	—	—	4.12	0.676	6.10	—
0.20	2.48	0.408	6.08	4.69	0.355	13.2	2.2
0.40	2.46	0.301	8.17	4.77	0.263	18.1	2.2
0.50	—	—	—	4.70	0.259	18.1	—
0.60	2.40	0.258	9.30	4.68	0.259	18.1	1.9
0.70	—	—	—	4.66	0.274	17.0	—
0.80	2.57	0.263	9.77	4.30	0.261	16.5	1.7
0.90	—	—	—	4.46	0.297	15.0	—
1.00	2.47	0.306	8.07	4.32	0.323	13.4	1.7
2.00	2.37	0.483	4.91	3.92	0.776	5.05	1.0
3.00	2.12	0.795	2.67	2.94	1.53	1.92	0.72
4.00	1.37	1.33	1.03	2.08	2.47	0.842	0.82

Table A.11. Distribution of perrhenate and pertechnetate between aqueous nitric acid and tributyl phosphate in n-dodecane at 40°C

$[HNO_3]_{aq}$ (M)	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}	D_{Tc}/D_{Re}
[TBP] = 0.365 M (9.99 vol %)							
0.10	—	—	—	0.0091	4.85	0.0019	—
0.20	—	—	—	0.0168	4.78	0.0035	—
0.40	—	—	—	0.0314	4.76	0.0066	—
0.50	—	—	—	0.0352	4.86	0.0072	—
0.60	—	—	—	0.0390	4.75	0.0082	—
0.70	—	—	—	0.0416	4.83	0.0086	—
0.80	—	—	—	0.0456	4.77	0.0096	—
0.90	—	—	—	0.0478	4.82	0.0099	—
1.00	—	—	—	0.0469	4.78	0.0098	—
2.00	—	—	—	0.0339	4.80	0.0071	—
3.00	—	—	—	0.0182	4.86	0.0037	—
4.00	—	—	—	0.0077	4.86	0.0016	—
[TBP] = 0.732 M (19.8 vol %)							
0.10	—	—	—	0.114	4.78	0.0238	—
0.20	0.032	2.89	0.011	0.209	4.66	0.0448	4.1
0.40	0.038	2.79	0.014	0.307	4.48	0.0685	4.9
0.50	—	—	—	0.377	4.38	0.0861	—
0.60	0.054	2.74	0.020	0.375	4.45	0.0843	4.2
0.70	—	—	—	0.378	4.41	0.0857	—
0.80	0.064	2.86	0.022	0.382	4.51	0.0847	3.8
0.90	—	—	—	0.382	4.43	0.0862	—
1.00	0.059	2.82	0.021	0.388	4.48	0.0866	4.1
2.00	0.048	3.07	0.016	0.226	4.62	0.0489	3.1
3.00	0.032	2.98	0.011	0.103	4.69	0.0220	2.0
4.00	—	—	—	0.0465	4.82	0.0096	—
[TBP] = 1.10 M (30.0 vol %)							
0.10	—	—	—	0.448	4.43	0.101	—
0.20	0.140	2.74	0.0511	0.742	4.21	0.176	3.4
0.40	0.188	2.60	0.0723	1.01	3.83	0.264	3.6
0.50	—	—	—	1.14	3.86	0.295	—
0.60	0.204	2.49	0.0819	1.12	3.72	0.301	3.7
0.70	—	—	—	1.15	3.74	0.307	—
0.80	0.242	2.54	0.0953	1.15	3.66	0.314	3.3
0.90	—	—	—	1.12	3.64	0.308	—
1.00	0.231	2.53	0.0913	1.11	3.83	0.290	3.2
2.00	0.134	2.60	0.0515	0.732	3.94	0.186	3.6
3.00	0.0752	2.79	0.0270	0.315	5.04	0.0625	2.3
4.00	0.0483	2.83	0.0171	0.159	4.78	0.0333	1.9
[TBP] = 1.46 M (40.1 vol %)							
0.10	—	—	—	0.912	4.11	0.222	—
0.20	0.290	2.39	0.121	1.34	3.54	0.379	3.1
0.40	0.440	2.19	0.201	1.76	3.14	0.561	2.8
0.50	—	—	—	1.86	3.07	0.606	—
0.60	0.505	2.10	0.240	1.90	3.07	0.619	2.6
0.70	—	—	—	1.94	3.08	0.630	—
0.80	0.553	2.07	0.267	1.87	3.06	0.611	2.3
0.90	—	—	—	1.85	3.03	0.611	—
1.00	0.531	2.12	0.250	1.90	3.64	0.522	2.1
2.00	0.322	2.34	0.138	1.11	3.97	0.280	2.0
3.00	0.172	2.59	0.0664	0.577	4.08	0.141	2.1
4.00	0.0913	2.68	0.0341	0.282	4.93	0.0572	1.7

Table A.11 (continued)

$[HNO_3]_{aq}$ (M)	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}	D_{Tc}/D_{Re}
[TBP] = 2.18 M (59.8 vol %)							
0.10	—	—	—	2.21	2.55	0.867	—
0.20	1.02	1.77	0.576	2.85	1.94	1.47	2.6
0.40	1.52	1.40	1.09	3.28	1.54	2.13	2.0
0.50	—	—	—	3.46	1.43	2.42	—
0.60	1.55	1.30	1.19	3.29	1.39	2.37	2.0
0.70	—	—	—	3.43	1.41	2.43	—
0.80	1.61	1.25	1.29	3.45	1.42	2.43	1.9
0.90	—	—	—	3.39	1.43	2.37	—
1.00	1.52	1.29	1.18	3.37	1.47	2.29	1.9
2.00	1.19	1.63	0.730	2.52	2.24	1.13	1.5
3.00	0.768	2.12	0.362	1.52	3.27	0.465	1.3
4.00	0.403	2.43	0.166	0.838	3.99	0.210	1.3
[TBP] = 2.93 M (80.2 vol %)							
0.10	—	—	—	3.47	1.40	2.48	—
0.20	1.90	0.832	2.28	4.07	1.01	4.03	1.8
0.40	2.00	0.633	3.16	4.27	0.625	6.83	2.2
0.50	—	—	—	4.33	0.632	6.85	—
0.60	4.64	1.22	3.80	4.26	0.614	6.94	1.8
0.70	—	—	—	4.05	0.579	7.00	—
0.80	2.16	0.564	3.83	4.37	0.645	6.78	1.8
0.90	—	—	—	4.22	0.629	6.71	—
1.00	2.15	0.564	3.81	4.04	0.625	6.46	1.7
2.00	1.99	0.816	2.44	3.50	1.27	2.76	1.1
3.00	1.44	1.20	1.20	2.56	2.17	1.18	1.0
4.00	1.08	1.63	0.663	1.80	3.03	0.594	0.9

Table A.12. Distribution of perrhenate and pertechnetate between aqueous nitric acid and tributyl phosphate in n-dodecane at 60°C

$[HNO_3]_{aq}$ (M)	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}	D_{Tc}/D_{Re}
[TBP] = 0.365 M (9.99 vol %)							
0.10	—	—	—	0.0024	4.99	0.00048	—
0.20	—	—	—	0.0042	4.97	0.00084	—
0.40	—	—	—	0.0082	4.88	0.0017	—
0.50	—	—	—	0.0093	5.00	0.0019	—
0.60	—	—	—	0.0105	4.93	0.00213	—
0.70	—	—	—	0.0157	4.93	0.00318	—
0.80	—	—	—	0.0118	4.89	0.00242	—
0.90	—	—	—	0.0134	4.98	0.00269	—
1.00	—	—	—	0.0139	5.19	0.00269	—
2.00	—	—	—	0.0121	4.90	0.00268	—
3.00	—	—	—	0.0086	4.97	0.0017	—
4.00	—	—	—	0.0041	4.91	0.00084	—
[TBP] = 0.732 M (19.8 vol %)							
0.10	—	—	—	0.0246	4.69	0.00525	—
0.20	—	—	—	0.0429	4.70	0.00913	—
0.40	—	—	—	0.0734	4.82	0.0152	—
0.50	—	—	—	0.0860	4.78	0.0180	—
0.60	—	—	—	0.0954	4.74	0.0201	—
0.70	—	—	—	0.100	4.71	0.0212	—
0.80	—	—	—	0.104	4.72	0.0220	—
0.90	—	—	—	0.108	4.68	0.0231	—
1.00	—	—	—	0.111	4.28	0.0259	—
2.00	—	—	—	0.0800	4.73	0.0169	—
3.00	—	—	—	0.0419	4.81	0.00871	—
4.00	—	—	—	0.0229	4.80	0.00477	—
[TBP] = 1.10 M (30.0 vol %)							
0.10	—	—	—	0.104	4.81	0.0216	—
0.20	0.027	2.69	0.010	0.167	4.70	0.0355	3.6
0.40	0.043	2.86	0.015	0.278	4.71	0.0590	3.9
0.50	—	—	—	0.312	4.58	0.0681	—
0.60	0.043	2.69	0.016	0.337	4.48	0.0752	4.7
0.70	—	—	—	0.344	4.40	0.0782	—
0.80	0.047	2.75	0.017	0.362	4.44	0.0815	4.8
0.90	—	—	—	0.366	4.52	0.0810	—
1.00	0.043	2.80	0.015	0.355	4.34	0.0818	5.5
2.00	0.028	2.82	0.010	0.259	4.67	0.0555	5.5
3.00	—	—	—	0.178	4.67	0.0381	—
4.00	—	—	—	0.090	4.85	0.0186	—
[TBP] = 1.46 M (40.1 vol %)							
0.10	—	—	—	0.224	4.61	0.0486	—
0.20	0.070	2.79	0.025	0.357	4.34	0.0823	3.3
0.40	0.102	2.80	0.0364	0.541	4.23	0.128	3.5
0.50	—	—	—	0.608	4.26	0.143	—
0.60	0.118	2.74	0.0431	0.652	4.16	0.157	3.6
0.70	—	—	—	0.640	4.02	0.159	—
0.80	0.118	2.40	0.0491	0.661	4.02	0.164	3.3
0.90	—	—	—	0.667	4.04	0.165	—
1.00	0.118	2.63	0.0449	0.678	4.05	0.167	3.7
2.00	0.081	2.82	0.029	0.457	4.32	0.106	3.7
3.00	0.043	2.80	0.015	0.237	4.39	0.054	3.6
4.00	—	—	—	0.131	4.73	0.028	—

Table A.12 (continued)

$[HNO_3]_{aq}$ (M)	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}	D_{Tc}/D_{Re}
[TBP] = 2.18 M (59.8 vol %)							
0.10	—	—	—	0.740	3.45	0.214	—
0.20	0.242	2.37	0.102	1.17	3.42	0.342	3.3
0.40	0.440	2.04	0.216	1.57	3.06	0.513	2.4
0.50	—	—	—	1.69	2.83	0.597	—
0.60	0.580	2.07	0.280	1.85	2.78	0.665	2.4
0.70	—	—	—	1.76	2.70	0.652	—
0.80	0.633	2.05	0.309	1.78	2.71	0.657	2.1
0.90	—	—	—	1.74	2.70	0.644	—
1.00	0.687	2.02	0.340	1.88	2.74	0.686	2.0
2.00	0.489	2.09	0.234	1.35	3.19	0.423	1.8
3.00	0.290	2.33	0.124	1.09	3.84	0.284	2.3
4.00	0.150	2.71	0.055	0.455	4.32	0.105	1.9
[TBP] = 2.93 M (80.2 vol %)							
0.10	—	—	—	2.13	2.71	0.786	—
0.20	0.929	1.78	0.522	2.72	2.19	1.24	2.4
0.40	1.19	1.50	0.793	3.24	1.73	1.87	2.4
0.50	—	—	—	3.35	1.67	2.01	—
0.60	1.39	1.27	1.09	3.35	1.48	2.26	2.1
0.70	—	—	—	3.39	1.38	2.46	—
0.80	1.42	1.24	1.15	3.47	1.51	2.30	2.0
0.90	—	—	—	3.45	1.48	2.33	—
1.00	1.42	1.18	1.20	3.51	1.49	2.36	2.0
2.00	1.34	1.41	0.950	2.75	2.18	1.26	1.3
3.00	0.982	1.78	0.552	1.75	2.78	0.629	1.1
4.00	0.607	2.17	0.280	1.17	3.65	0.321	1.1

Table A.13. Distribution of nitric acid and perrhenate between aqueous nitric acid and tributyl phosphate in carbon tetrachloride at 20°C[a]

$[HNO_3]_{aq}$ (M)	$[HNO_3]_{org}$ (M)	$[TBP]_f$ (M)	$[Re]_{org}$ (μM)	$[Re]_{aq}$ (μM)	D_{Re}
$[TBP]_o$ = 3.65 M (100 vol %)					
0.083	0.024	3.63	905	61.7	14.7
0.143	0.066	3.58	926	37.6	24.6
0.308	0.209	3.44	925	20.2	25.8
0.570	0.464	3.19	932	12.8	73.0
0.726	0.623	3.03	922	18.8	49.0
1.035	0.901	2.74	851	70.3	12.1
2.245	1.620	2.03	748	20.7	3.60
3.290	1.905	1.74	535	442	1.21
5.225	2.718	0.92	236	820	0.290
$[TBP]_o$ = 3.00 M (82.2 vol %)					
0.088	0.018	2.98	613	378	1.62
0.162	0.048	2.95	666	320	2.08
0.347	0.170	2.83	790	187	4.23
0.646	0.398	2.60	875	93.0	9.40
0.838	0.521	2.48	866	103	8.40
1.170	0.730	2.27	802	166	4.83
2.476	1.402	1.60	552	442	1.25
3.487	1.750	1.25	338	682	0.495
5.630	2.321	0.68	126	936	0.135
$[TBP]_o$ = 2.00 M (54.8 vol %)					
0.097	0.009	1.94	206	807	0.260
0.185	0.023	1.98	366	635	0.576
0.426	0.092	1.91	460	543	0.848
0.778	0.261	1.74	652	343	1.90
1.008	0.378	1.62	599	393	1.52
1.415	0.498	1.50	425	572	0.741
2.780	1.118	0.882	186	844	0.220
$[TBP]_o$ = 1.50 M (41.1 vol %)					
0.101	0.004	1.50	63.1	906	0.071
0.198	0.009	1.49	114	891	0.127
0.461	0.058	1.44	193	819	0.236
0.866	0.171	1.33	201	811	0.248
1.099	0.281	1.22	243	767	0.317
1.535	0.377	1.12	116	884	0.131

[a]Data adapted from ref. 12.

Table A.14. Distribution of perrhenic acid between nitric acid and tributyl phosphate in odorless kerosene[a,b]

[TBP] (vol %)	[TBP] (M)	[HNO_3] (M)	[$HReO_4$] (M)	D_{Re}
20	0.75	1.0	0.0377	0.14
20	0.750	1.0	0.0269	0.14
20	0.75	1.0	0.0142	0.13
20	0.75	1.0	0.0054	0.14
10	0.375	1	0.0107	0.023
20	0.750	1	0.0107	0.14
30	4.13	1	0.0107	0.50
40	1.50	1	0.0107	1.02
50	1.88	1	0.0107	2.10
20	0.75	0	0.0142	0.015
20	0.75	1	0.0142	0.14
20	0.75	2	0.0142	0.087
20	0.75	3	0.0142	0.032
20	0.75	4	0.0142	0.015
20	0.75	6	0.0142	0.0018
20	0.75	9	0.0142	$<10^{-4}$
20	0.75	12	0.0142	$<10^{-4}$

[a]Data adapted from ref. 13.
[b]No temperature reported for this study.

Table A.15. Distribution of pertechnetate between aqueous nitric acid and tributyl phosphate in synthine[a,b]

[HNO_3] (M)	D_{Tc}	
	100% TBP	20% TBP
0.01	1.8	0.00045
0.1	26	0.052
0.5	46	0.16
1.0	65	0.16
2.0	22	0.087
4.0	13	0.0024
6.0	1.3	0.0055
8.0	0.32	0.0026
12.0	0.13	0.0046
18.4	0.30	c

[a]Data adapted from ref. 9.
[b]No temperature reported for this study.
[c]Third phase formed.

Table A.16. Distribution of pertechnetate between 4 M NH_4NO_3 in aqueous nitric acid and tributyl phosphate in carbon tetrachloride at 25°C[a]

pH	[TBP] (m)	D_{Tc}
1.95	0.00	5.72×10^{-5}
1.96	0.075	9.42×10^{-5}
1.98(5)	0.150	0.000150
1.99	0.225	0.000257
1.99(5)	0.301	0.000455
2.01(5)	0.451	0.00140
2.04	0.753	0.00367
2.08	0.930	0.00938
2.14(5)	1.13	0.0167
2.23(5)	1.50	0.0365
2.33	1.88	0.0687
2.51(5)	2.63	0.177

[a]Data adapted from ref. 10.

Table A.17. Distribution of pertechnetate between aqueous nitric acid and 30 vol % tributyl phosphate in n-dodecane at 20°C[a]

$[HNO_3]^{equil}_{aq}$ (M)	D_{Tc}
0	0.00162
0.1	0.437
0.3	0.908
0.5	1.138
0.7	1.168
1.0	0.890
2.0	0.370
3.0	0.141

[a]Data derived from ref. 11.

Table A.18. Distribution of pertechnetate between 0.75 M nitric acid and 30 vol % tributyl phosphate in kerosene as a function of temperature and organic-phase uranyl nitrate concentration[a]

Temperature (°C)	$[U]_{organic}$ 0.0 M	$[U]_{organic}$ 0.441 M
11	2.28	—
18	1.37	—
24	0.75	1.33
30	0.56	1.35
40	0.22	1.34
50	0.17	1.35
60	0.13	—

[a]Data derived from ref. 7.

Table A.19. Distribution of pertechnetate between nitric acid and tributyl phosphate as a function of nitric acid concentration at various tributyl phosphate concentrations[a,b]

$[HNO_3]$ (M)	D_{Tc} 15% TBP (0.548 M)	D_{Tc} 30% TBP (1.1 M)	D_{Tc} 45% TBP (1.64 M)
0 (pH 6.7)	0.0	0.0	0.0
0.2	0.045	0.385	1.19
0.4	0.060	0.68	1.34
0.75	0.065	0.75	1.42
1.0	0.065	0.65	1.16
2.0	0.04	0.22	0.65
3.0	0.03	0.145	0.30
4.0	0.01	0.045	0.09

[a]Data derived from ref. 7.
[b]Conditions: diluent, kerosene; temperature, 24 ± 2°C; initial aqueous technetium concentration, $\sim 10^{-6}$ M.

Table A.20. Distribution of technetium between aqueous sulfuric or phosphoric acid and 30 vol % tributyl phosphate in n-dodecane at 25°C

Acid concentration (M)	D_{Tc}	
	H_2SO_4	H_3PO_4
0.1	0.322	—
0.5	1.50	—
1.0	3.18	—
2.0	12.0	2.27
3.0	31.4	3.28
4.0	71.2	5.90
5.0	—	9.17
6.0	—	13.5

Table A.21. Distribution coefficient of perrhenate[a] between aqueous alkali metal hydroxides and pure tributyl phosphate[b,c]

$[OH^-]_{aq}^{init}$	LiOH	NaOH	KOH	RbOH	CsOH
0.40	15.9	3.03	2.07	—	—
1	34	4.30	2.81	2.27	1.41
2	58	5.41	3.04	2.17	1.25
3	80	7.91	—	—	—
4	95	8.10	2.52	1.74	0.97
6	—	11.0	2.37	1.40	0.70
8	—	14.0	2.35	1.15	0.56
10	—	21.8	2.50	1.00	—
12	—	18.1	2.80	—	—

[a]Initial rhenium concentration, 6×10^{-5} M.
[b]Data adapted from ref. 19.
[c]No temperature reported for this study.

Table A.22. Distribution coefficient of perrhenate[a] between aqueous NaOH, Na_2CO_3, or $NaNO_3$ and pure tributyl phosphate[b,c]

$[Na^+]_{aq}^{init}$	NaOH	Na_2CO_3	$NaNO_3$
1	4.30	5.00	1.16
2	5.41	7.03	1.60
4	8.10	9.43	1.80

[a]Initial rhenium concentration, 6×10^{-5} M.
[b]Data adapted from ref. 17.
[c]No temperature reported for this study.

Table A.23. Distribution coefficient of technetium from various salt solutions containing an initial $KTcO_4$ concentration of 2.0 mM into 30 vol % tributyl phosphate in n-dodecane

$[Cl^-]$ (M)	T (K)	D_{Tc} HCl	LiCl	NaCl	KCl	RbCl	NH_4Cl
0.100	298	--	0.0574	0.00545	0.00518	0.00387	0.0171
0.200	298	0.693	0.106	0.0106	0.00729	0.00601	0.0329
0.400	298	1.30	0.201	0.0185	0.0111	0.00805	0.0554
0.600	298	2.01	0.310	0.0261	0.0130	0.0103	0.0729
0.800	298	2.63	0.441	0.0333	0.0171	0.0117	0.0872
0.900	298	--	0.507	--	--	--	--
1.00	298	3.38	--	0.0411	0.0190	0.0131	0.103
2.00	298	9.64	--	0.0818	0.0264	0.0185	0.166
3.00	298	19.5	--	0.134	0.0309	0.0228	0.213
0.0995	313	--	0.0181	0.00203	0.00186	0.00123	0.00632
0.199	313	0.162	0.0356	0.00420	0.00301	0.00168	0.0112
0.398	313	0.321	0.0655	0.00739	0.00458	0.00261	0.0189
0.597	313	0.470	0.102	0.0100	0.00533	0.00349	0.0246
0.796	313	0.694	0.146	0.0116	0.00644	0.00387	0.0310
0.896	313	--	0.178	--	--	--	--
0.993	313	0.832	--	0.0146	0.00706	0.00450	0.0364
1.99	313	2.10	--	0.0342	0.0106	0.00649	0.0627
2.98	313	5.13	--	0.0506	0.0140	0.00806	0.0852
0.0985	333	--	0.00536	0.000810	0.000542	0.000345	0.00194
0.197	333	0.0431	0.00978	0.00144	0.000993	0.000538	0.00340
0.394	333	0.0775	0.0182	0.00263	0.00173	0.000860	0.00589
0.591	333	0.120	0.0312	0.00375	0.00214	0.00108	0.00759
0.789	333	0.161	0.0398	0.00476	0.00225	0.00125	0.00950
0.888	333	--	0.0509	--	--	--	--
0.984	333	0.204	--	0.00576	0.00295	0.00152	0.0110
1.97	333	0.586	--	0.0127	0.00402	0.00240	0.0188
2.96	333	1.46	--	0.0218	0.00582	0.00279	0.0278

Table A.24. Distribution of uranium and technetium between aqueous 4 M NH_4NO_3 and 1.0 M tributyl phosphate in carbon tetrachloride at 25°C[a]

pH	$[U]_{aq}$ (M × 10^3)	$[U]_{org}$ (M × 10^3)	D_U	$[TBP]_{free}$ (M)	D_{Tc}
2.10	0.17	6.36	0.027	0.987	0.0147
2.08	0.32	16.7	0.019	0.967	0.0201
2.02	0.53	33.5	0.016	0.933	0.0278
2.00	1.7	49.5	0.034	0.901	0.0343
1.96	--	62.7	--	0.874	0.0430
1.91	5.8	96.8	0.060	0.806	0.0557
1.89	--	139	--	0.738	0.0696
1.87	10	161	0.062	0.678	0.0836
1.86	14	192	0.073	0.616	0.0916
1.81	29	243	0.119	0.507	0.111

[a]Data adapted from ref. 10.

Table A.25. Distribution of uranium and technetium between aqueous nitric acid and 30 vol % tributyl phosphate in n-dodecane

$[HNO_3]_{aq}$ (M)	$[U]_{org}$ (M)	$[U]_{aq}$ (M)	D_U	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}
Initial aqueous uranium concentration = 0.2 M (nominal)						
0.01	0.0819	0.114	0.718	2.47	1.60	1.54
0.21	0.121	0.0298	1.52	2.32	1.76	1.32
0.43	0.139	0.0618	2.25	2.10	1.99	1.06
0.66	0.160	0.0500	3.20	1.91	2.23	0.857
0.88	0.171	0.0361	4.74	1.71	2.34	0.731
1.02	0.184	0.0290	6.35	1.53	2.49	0.614
2.11	0.200	0.0143	14.0	0.993	3.07	0.324
3.16	0.198	0.00966	20.5	0.620	3.43	0.181
4.30	0.216	0.00756	28.6	0.491	3.54	0.139
Initial aqueous uranium concentration = 0.50 M (nominal)						
0.01	0.234	0.259	0.904	2.25	1.83	1.23
0.10	0.251	0.249	1.01	2.20	1.94	1.13
0.21	0.271	0.234	1.16	2.14	2.10	1.02
0.45	0.292	0.210	1.39	1.91	2.24	0.853
0.67	0.303	0.190	1.59	1.75	2.43	0.720
0.90	0.318	0.175	1.82	1.62	2.59	0.626
1.12	0.328	0.157	2.09	1.45	2.73	0.531
2.27	0.371	0.114	3.25	1.07	3.16	0.339
3.43	0.396	0.101	3.92	0.889	3.38	0.263
4.58	0.397	0.0906	4.38	0.787	3.44	0.229
Initial aqueous uranium concentration = 0.75 M (nominal)						
0.01	0.339	0.407	0.833	1.96	2.15	0.912
0.10	0.350	0.397	0.882	1.91	2.23	0.857
0.21	0.362	0.393	0.921	1.81	2.29	0.790
0.43	0.382	0.372	1.03	1.68	2.47	0.680
0.66	0.394	0.362	1.09	1.56	2.65	6.589
0.88	0.421	0.348	1.21	1.45	2.74	0.529
1.11	0.416	0.332	1.25	1.36	2.84	0.479
2.29	0.451	0.302	1.49	1.03	3.18	0.324
3.43	0.458	0.294	1.56	0.873	3.38	0.258
4.57	0.465	0.296	1.57	0.797	3.41	0.234
Initial aqueous uranium concentration = 1.0 M (nominal)						
0.01	0.406	0.594	0.684	1.67	2.48	0.673
0.10	0.412	0.595	0.692	1.65	2.56	0.644
0.21	0.427	0.578	0.739	1.57	2.61	0.602
0.45	0.426	0.568	0.750	1.47	2.72	0.540
0.69	0.440	0.567	0.776	1.37	2.89	0.474
0.93	0.440	0.552	0.797	1.28	2.98	0.430
1.16	0.445	0.541	0.823	1.21	3.05	0.397
2.31	0.464	0.527	0.880	0.956	3.31	0.289
3.46	0.466	0.526	0.886	0.825	3.47	0.238
4.60	0.467	0.527	0.886	0.774	3.44	0.225

Table A.26. Distribution of uranium and technetium between aqueous nitric acid and 30 vol % tributyl phosphate in n-dodecane at 40°C

$[HNO_3]_{aq}$ (M)	$[U]_{org}$ (M)	$[U]_{aq}$ (M)	D_U	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}
Initial aqueous uranium concentration = 0.2 M (nominal)						
0.01	0.0672	0.129	0.521	1.87	2.24	0.835
0.10	0.0791	0.116	0.682	1.85	2.28	0.811
0.21	0.0950	0.104	0.913	1.80	2.36	0.763
0.43	0.117	0.0811	1.44	1.66	2.50	0.664
0.66	0.132	0.0639	2.97	1.53	2.66	0.575
0.88	0.139	0.0528	2.63	1.38	2.77	0.498
1.02	0.147	0.0411	3.58	1.26	2.90	0.434
2.11	0.169	0.0232	7.28	0.854	3.35	0.255
3.16	0.174	0.0161	10.8	0.641	3.60	0.178
4.30	0.179	0.0136	13.2	0.529	3.70	0.143
Initial aqueous uranium concentration = 0.50 M (nominal)						
0.01	0.209	0.281	0.744	1.99	2.09	0.952
0.10	0.229	0.268	0.854	1.95	2.18	0.984
0.21	0.238	0.251	0.948	1.87	2.24	0.835
0.45	0.264	0.232	1.13	1.75	2.40	0.729
0.67	0.283	0.209	1.35	1.63	2.54	0.642
0.90	0.297	0.192	1.55	1.50	2.65	0.566
1.12	0.294	0.176	1.67	1.41	2.76	0.511
2.27	0.352	0.135	2.61	1.09	3.13	0.348
3.43	0.365	0.117	3.12	0.911	3.35	0.272
4.58	0.373	0.114	3.27	0.816	3.38	0.241
Initial aqueous uranium concentration = 1.0 M (nominal)						
0.01	0.370	0.604	0.613	1.56	2.35	0.664
0.10	0.376	0.603	0.624	1.52	2.42	0.628
0.21	0.386	0.590	0.654	1.47	2.50	0.588
0.45	0.398	0.574	0.693	1.39	2.58	0.529
0.69	0.407	0.563	0.723	1.30	2.70	0.481
0.93	0.411	0.560	0.734	1.18	2.80	0.421
1.16	0.410	0.566	0.724	1.17	2.86	0.409
2.31	0.424	0.540	0.785	0.946	3.11	0.304
3.46	0.438	0.554	0.791	0.838	3.25	0.258
4.60	0.436	0.534	0.816	0.781	3.25	0.240

Table A.27. Distribution of uranium and technetium between aqueous nitric acid and 30 vol % tributyl phosphate in n-dodecane at 60°C

$[HNO_3]_{aq}$ (M)	$[U]_{org}$ (M)	$[U]_{aq}$ (M)	D_U	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}
Initial aqueous uranium concentration = 0.2 M (nominal)						
0.01	0.0583	0.140	0.416	1.43	2.69	0.532
0.10	0.0714	0.127	0.562	1.46	2.72	0.537
0.21	0.0852	0.115	0.741	1.41	2.71	0.520
0.43	0.105	0.0922	1.14	1.34	2.80	0.479
0.66	0.119	0.0758	1.57	1.27	2.90	0.438
0.88	0.131	0.0639	2.05	1.17	3.04	0.385
1.02	0.140	0.0555	2.52	1.09	3.10	0.352
2.11	0.165	0.0319	5.17	0.797	3.38	0.236
3.16	0.174	0.0232	7.50	0.644	3.60	0.179
4.30	0.175	0.0209	8.37	0.551	3.63	0.152
Initial aqueous uranium concentration = 0.50 M (nominal)						
0.01	0.197	0.301	0.654	1.77	2.27	0.780
0.10	0.204	0.287	0.711	1.77	2.36	0.750
0.21	0.223	0.276	0.808	1.69	2.38	0.710
0.45	0.249	0.253	0.984	1.61	2.50	0.644
0.67	0.271	0.229	1.18	1.52	2.61	0.582
0.90	0.285	0.214	1.33	1.44	2.71	0.531
1.12	0.294	0.195	1.51	1.36	2.80	0.486
2.27	0.335	0.157	2.13	1.10	3.09	0.356
3.43	0.349	0.140	2.49	0.940	3.22	0.292
4.58	0.356	0.142	2.51	0.844	3.35	0.252
Initial aqueous uranium concentration = 1.0 M (nominal)						
0.01	0.372	0.610	0.610	1.58	2.47	0.640
0.10	0.375	0.603	0.622	1.56	2.50	0.624
0.21	0.377	0.600	0.628	1.53	2.58	0.593
0.45	0.400	0.590	0.678	1.45	2.69	0.539
0.69	0.405	0.573	0.707	1.37	2.75	0.498
0.93	0.412	0.573	0.719	1.31	2.83	0.463
1.16	0.419	0.566	0.740	1.24	2.89	0.429
2.31	0.439	0.548	0.801	1.03	3.12	0.330
3.46	0.442	0.552	0.801	0.921	3.25	0.283
4.60	0.435	0.556	0.782	0.857	3.31	0.259

Table A.28. Distribution of uranium and rhenium between aqueous nitric acid and 30.7 vol % (1.2 M) tributyl phosphate in n-dodecane at 25°C

$[HNO_3]_{aq}$ (M)	$[U]_{org}$ (M)	$[U]_{aq}$ (M)	D_U	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}
Initial aqueous uranium concentration = 0.20 M (nominal)						
0.01	0.0909	0.117	0.777	0.70	2.42	0.29
0.10	0.0965	0.0984	0.981	0.75	2.36	0.32
0.20	0.120	0.0854	1.41	0.70	2.36	0.30
0.40	0.138	0.0634	2.18	0.64	2.42	0.26
0.60	0.151	0.0438	3.45	0.59	2.42	0.24
0.80	0.163	0.0383	4.26	0.54	2.47	0.22
1.00	0.170	0.0305	5.57	0.48	2.58	0.19
2.00	0.176	0.0142	12.4	0.21	2.79	0.075
3.00	0.186	0.0089	20.9	0.16	2.90	0.055
4.00	0.192	0.0066	28.9	0.08	2.95	0.027
Initial aqueous uranium concentration = 0.50 M (nominal)						
0.01	0.251	0.248	1.01	1.40	4.67	0.30
0.10	0.267	0.233	1.15	1.40	4.14	0.34
0.20	0.281	0.226	1.24	1.29	5.37	0.24
0.40	0.309	0.195	1.58	1.02	5.48	0.19
0.60	0.324	0.174	1.87	0.70	5.69	0.12
0.80	0.349	0.157	2.22	0.64	5.32	0.12
1.00	0.358	0.140	2.55	0.54	5.53	0.098
2.00	0.400	0.0983	4.07	0.32	5.32	0.060
3.00	0.421	0.0826	5.10	0.21	6.07	0.035
4.00	0.439	0.0794	5.53	0.26	6.12	0.042
Initial aqueous uranium concentration = 1.0 M (nominal)						
0.01	0.458	0.741	0.618[a]	0.43	4.62	0.093[a]
0.10	0.412	0.536	0.769	0.70	4.30	0.16
0.20	0.424	0.546	0.777	0.59	4.14	0.14
0.40	0.438	0.530	0.826	0.54	4.30	0.13
0.60	0.447	0.522	0.856	0.48	4.35	0.11
0.80	0.447	0.516	0.866	0.51	4.67	0.11
1.00	0.471	0.515	0.915	0.43	4.24	0.10
2.00	0.477	0.484	0.986	0.27	4.78	0.056
3.00	0.479	0.497	0.964	0.21	4.62	0.045
4.00	0.478	0.486	0.984	0.16	4.62	0.035

[a]Initial aqueous uranium concentration = 1.22 M.

Table A.29. Distribution of uranium and rhenium between aqueous nitric acid and 30.7 vol % (1.2 M) tributyl phosphate in n-dodecane at 40°C

$[HNO_3]_{aq}$ (M)	$[U]_{org}$ (M)	$[U]_{aq}$ (M)	D_U	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}
Initial aqueous uranium concentration = 0.20 M (nominal)						
0.01	0.0720	0.130	0.554	0.48	2.63	0.18
0.10	0.0916	0.111	0.825	0.48	2.58	0.19
0.20	0.104	0.0978	1.06	0.48	2.74	0.18
0.40	0.128	0.0747	1.71	0.48	2.74	0.18
0.60	0.137	0.0579	2.37	0.43	2.63	0.16
0.80	0.146	0.0477	3.06	0.43	2.79	0.15
1.00	0.164	0.0384	4.27	0.27	2.85	0.095
2.00	0.173	0.0189	9.15	0.21	2.90	0.072
3.00	0.178	0.0126	14.1	0.11	2.95	0.037
4.00	0.189	0.0103	18.3	0.059	3.01	0.030
Initial aqueous uranium concentration = 0.50 M (nominal)						
0.01	0.220	0.272	0.808	1.29	4.67	0.276
0.10	0.253	0.258	0.981	1.18	4.73	0.249
0.20	0.253	0.236	1.07	1.02	5.05	0.202
0.40	0.289	0.216	1.34	0.81	5.58	0.15
0.60	0.310	0.192	1.61	0.70	5.05	0.14
0.80	0.331	0.175	1.89	0.81	5.58	0.15
1.00	0.340	0.157	2.16	0.59	5.48	0.11
2.00	0.405	0.117	3.47	0.32	5.75	0.055
3.00	0.407	0.0994	4.09	0.32	5.85	0.055
4.00	0.415	0.100	4.15	0.22	5.91	0.037
Initial aqueous uranium concentration = 1.0 M (nominal)						
0.01	0.441	0.763	0.578[a]	0.64	4.35	0.15[a]
0.10	0.401	0.552	0.726	0.75	4.30	0.17
0.20	0.410	0.558	0.735	0.64	4.30	0.15
0.40	0.424	0.546	0.777	0.59	4.40	0.13
0.60	0.426	0.538	0.792	0.59	4.30	0.14
0.80	0.444	0.536	0.828	0.48	4.67	0.10
1.00	0.456	0.533	0.856	0.43	4.40	0.10
2.00	0.465	0.494	0.941	0.27	4.83	0.056
3.00	0.468	0.517	0.905	0.27	4.83	0.056
4.00	0.476	0.508	0.937	0.21	4.78	0.044

[a]Initial aqueous uranium concentration = 1.22 M.

Table A.30. Distribution of uranium and rhenium between aqueous nitric acid and 30.7 vol % (1.2 M) tributyl phosphate in n-dodecane at 60°C

$[HNO_3]_{aq}$ (M)	$[U]_{org}$ (M)	$[U]_{aq}$ (M)	D_U	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}
Initial aqueous uranium concentration = 0.20 M (nominal)						
0.01	0.0641	0.141	0.455	0.55	2.74	0.20
0.10	0.0763	0.123	0.620	0.51	2.69	0.19
0.20	0.0887	0.112	0.792	0.44	2.58	0.17
0.40	0.112	0.0876	1.28	0.39	2.79	0.14
0.60	0.125	0.0703	1.78	0.32	2.85	0.11
0.80	0.135	0.0575	2.35	0.27	2.90	0.093
1.00	0.143	0.0478	2.99	0.27	2.95	0.092
2.00	0.165	0.0259	6.37	0.16	2.90	0.055
3.00	0.169	0.0188	8.99	0.11	3.01	0.037
4.00	0.172	0.0167	10.3	0.11	2.90	0.038
Initial aqueous uranium concentration = 0.50 M (nominal)						
0.01	0.217	0.291	0.746	1.02	5.10	0.200
0.10	0.233	0.277	0.841	0.97	5.21	0.19
0.20	0.254	0.267	0.951	0.91	5.90	0.15
0.40	0.274	0.242	1.13	0.91	5.37	0.17
0.60	0.296	0.217	1.36	0.70	5.80	0.12
0.80	0.310	0.198	1.57	0.64	5.64	0.11
1.00	0.325	0.179	1.82	0.59	5.80	0.10
2.00	0.370	0.139	2.66	0.59	5.69	0.10
3.00	0.391	0.120	3.26	0.22	5.26	0.042
4.00	0.392	0.125	3.14	0.22	6.01	0.037
Initial aqueous uranium concentration = 1.0 M (nominal)						
0.01	0.428	0.766	0.559[a]	0.70	4.51	0.16[a]
0.10	0.386	0.570	0.677	0.81	4.19	0.19
0.20	0.392	0.574	0.683	0.75	4.46	0.17
0.40	0.408	0.561	0.727	0.70	4.19	0.17
0.60	0.417	0.553	0.754	0.70	4.46	0.16
0.80	0.424	0.549	0.772	0.54	4.40	0.12
1.00	0.436	0.551	0.791	0.59	4.62	0.13
2.00	0.444	0.518	0.857	0.38	4.73	0.080
3.00	0.453	0.533	0.850	0.32	4.46	0.072
4.00	0.449	0.529	0.849	0.20	4.99	0.041

[a]Initial aqueous uranium concentration = 1.22 M.

Table A.31. Distribution of thorium and technetium between aqueous nitric acid and 30.1 vol % (1.10 M) tributyl phosphate in n-dodecane at 25°C

$[HNO_3]_{aq}$ (M)	$[Th]_{org}$ (M)	$[Th]_{aq}$ (M)	D_{Th}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}
Initial aqueous thorium concentration = 0.05 M (nominal)						
0.15	3.84	44.1	0.0871	2.10	1.95	1.08
0.20	4.74	43.1	0.110	2.24	1.81	1.24
0.40	9.01	38.7	0.233	2.53	1.44	1.76
0.60	14.3	34.9	0.410	2.71	1.32	2.05
0.80	18.3	30.8	0.594	2.67	1.31	2.04
1.00	21.8	27.5	0.793	2.72	1.38	1.97
2.00	33.3	17.1	1.95	2.41	1.68	1.43
3.00	38.3	12.3	3.11	2.14	1.95	1.10
4.00	40.3	9.83	4.10	1.95	2.14	0.911
Initial aqueous thorium concentration = 0.10 M (nominal)						
0.15	12.2	82.9	0.147	2.65	1.41	1.88
0.20	14.2	81.1	0.175	2.76	1.32	2.09
0.40	22.7	72.6	0.313	2.94	1.10	2.63
0.60	30.6	65.5	0.467	2.99	1.03	2.90
0.80	37.4	59.7	0.626	3.00	1.02	2.94
1.00	43.4	54.0	0.804	2.97	1.03	2.88
2.00	62.7	36.2	1.73	2.81	1.23	2.28
3.00	74.0	27.5	2.69	2.62	1.40	1.87
4.00	76.6	25.5	3.40	2.50	1.53	1.63
Initial aqueous thorium concentration = 0.20 M (nominal)						
0.15	40.3	158	0.256	3.08	0.814	3.78
0.20	42.4	161	0.264	3.08	0.780	3.95
0.40	54.9	136	0.403	3.08	0.726	4.24
0.60	65.0	133	0.488	3.14	0.704	4.46
0.80	77.1	124	0.622	3.14	0.716	4.39
1.00	83.3	115	0.723	3.17	0.741	4.28
2.00	109	87.1	1.25	3.00	0.848	3.54
3.00	132	73.1	1.81	2.95	0.951	3.10
4.00	130	64.3	2.03	2.83	1.04	2.72

Table A.32. Distribution of thorium and technetium between aqueous nitric acid and 30.1 vol % (1.10 M) tributyl phosphate in n-dodecane at 40°C

$[HNO_3]_{aq}$ (M)	$[Th]_{org}$ (M)	$[Th]_{aq}$ (M)	D_{Th}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}
Initial aqueous thorium concentration = 0.05 M (nominal)						
0.15	3.10	45.1	0.0687	1.36	2.77	0.491
0.20	3.88	44.3	0.0876	1.51	2.56	0.590
0.40	7.93	40.0	0.198	1.97	2.13	0.925
0.60	12.7	36.9	0.344	2.19	1.89	1.16
0.80	16.5	32.1	0.514	2.24	1.82	1.23
1.00	20.4	30.3	0.673	2.27	1.80	1.26
2.00	31.5	19.2	1.64	2.15	1.94	1.75
3.00	36.7	14.5	2.53	1.97	2.14	0.921
4.00	39.0	12.3	3.17	1.81	2.28	0.794
Initial aqueous thorium concentration = 0.10 M (nominal)						
0.15	9.61	85.0	0.113	2.13	2.00	1.07
0.20	11.1	83.9	0.132	2.20	1.88	1.17
0.40	18.7	75.7	0.247	2.48	1.56	1.59
0.60	26.6	68.1	0.391	2.68	1.43	1.87
0.80	34.3	63.5	0.540	2.73	1.35	2.02
1.00	39.4	62.7	0.628	2.71	1.32	2.05
2.00	60.3	39.3	1.53	2.65	1.41	1.88
3.00	67.7	31.3	2.16	2.52	1.55	1.63
4.00	70.5	27.1	2.60	2.38	1.67	1.43
Initial aqueous thorium concentration = 0.20 M (nominal)						
0.15	34.9	164	0.213	2.86	1.11	2.58
0.20	35.7	162	0.220	2.85	1.07	2.66
0.40	47.3	150	0.316	2.93	0.973	3.01
0.60	60.1	138	0.435	2.96	0.912	3.25
0.80	73.0	128	0.570	3.01	0.878	3.43
1.00	77.0	122	0.631	3.01	0.890	3.38
2.00	104	92.7	1.13	2.93	0.954	3.07
3.00	124	79.4	1.56	2.83	1.05	2.70
4.00	128	73.8	1.73	2.73	1.16	2.35

Table A.33. Distribution of thorium and technetium between aqueous nitric acid and 30.1 vol % (1.10 M) tributyl phosphate in n-dodecane at 60°C

$[HNO_3]_{aq}$ (M)	$[Th]_{org}$ (M)	$[Th]_{aq}$ (M)	D_{Th}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}
Initial aqueous thorium concentration = 0.05 M (nominal)						
0.15	2.50	48.7	0.0513	0.854	3.27	0.261
0.20	3.15	47.0	0.0670	0.976	3.11	0.314
0.40	6.64	43.6	0.152	1.41	2.63	0.536
0.60	11.0	39.9	0.276	1.70	2.43	0.700
0.80	14.7	35.6	0.413	1.78	2.28	0.781
1.00	18.4	31.5	0.584	1.86	2.25	0.827
2.00	29.2	21.2	1.38	1.89	2.20	0.859
3.00	33.5	16.5	2.03	1.72	2.37	0.726
4.00	34.9	15.0	2.33	1.57	2.22	0.707
Initial aqueous thorium concentration = 0.10 M (nominal)						
0.15	7.80	86.5	0.0902	1.55	2.51	0.618
0.20	9.22	84.3	0.109	1.66	2.40	0.692
0.40	16.2	78.1	0.207	2.04	2.01	1.01
0.60	24.5	73.0	0.336	2.31	1.80	1.28
0.80	30.6	65.7	0.466	2.43	1.66	1.46
1.00	36.8	60.9	0.604	2.45	1.59	1.54
2.00	56.8	43.0	1.32	2.49	1.60	1.56
3.00	63.8	35.4	1.80	2.34	1.75	1.34
4.00	65.7	33.1	1.98	2.14	1.90	1.13
Initial aqueous thorium concentration = 0.20 M (nominal)						
0.15	28.3	170	0.167	2.47	1.46	1.69
0.20	29.2	168	0.174	2.50	1.42	1.76
0.40	43.4	155	0.281	2.63	1.25	2.10
0.60	57.8	145	0.399	2.76	1.16	2.38
0.80	66.8	134	0.499	2.79	1.10	2.54
1.00	71.9	125	0.573	2.80	1.08	2.59
2.00	98.3	99.8	0.985	2.70	1.10	2.45
3.00	110	88.8	1.24	2.66	1.23	2.16
4.00	111	84.9	1.31	2.51	1.37	1.83

Table A.34. Distribution of thorium and rhenium between aqueous nitric acid and 30.1 vol % (1.10 M) tributyl phosphate in n-dodecane at 25°C

$[HNO_3]_{aq}$ (M)	$[Th]_{org}$ (M)	$[Th]_{aq}$ (M)	D_{Th}	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}
Initial aqueous thorium concentration = 0.05 M (nominal)						
0.15	3.49	47.3	0.0738	1.8	3.5	0.51
0.20	4.48	45.6	0.0982	1.9	3.3	0.58
0.40	9.44	41.1	0.230	2.4	3.0	0.80
0.60	14.7	36.4	0.404	2.4	2.9	0.83
0.80	18.5	31.8	0.582	2.5	2.8	0.89
1.00	22.3	27.9	0.799	2.4	2.9	0.83
2.00	33.6	16.5	2.04	1.9	3.4	0.56
3.00	37.5	11.2	3.35	1.4	3.7	0.38
4.00	39.4	8.58	4.59	1.4	3.9	0.36
Initial aqueous thorium concentration = 0.10 M (nominal)						
0.15	12.2	91.3	0.134	2.1	3.2	0.66
0.20	14.3	88.3	0.162	2.4	2.9	0.83
0.40	22.9	79.0	0.290	2.5	2.6	0.96
0.60	31.4	70.3	0.447	2.6	2.4	1.1
0.80	38.7	64.3	0.602	2.4	2.6	0.92
1.00	44.7	57.7	0.775	2.5	2.7	0.93
2.00	63.5	37.4	1.70	2.3	3.0	0.77
3.00	71.4	27.5	2.60	2.0	3.4	0.59
4.00	78.5	22.5	3.49	1.9	3.5	0.54
Initial aqueous thorium concentration = 0.20 M (nominal)						
0.15	41.8	164	0.255	2.9	2.4	1.2
0.20	45.1	160	0.282	3.1	2.4	1.3
0.40	57.3	147	0.390	2.9	2.3	1.3
0.60	69.5	133	0.523	3.1	2.2	1.4
0.80	81.6	127	0.643	3.5	2.2	1.6
1.00	89.3	116	0.770	3.7	2.3	1.6
2.00	120	86.0	1.40	2.7	2.7	1.0
3.00	139	70.2	1.98	2.5	2.9	0.86
4.00	140	61.8	2.27	2.5	3.0	0.83

Table A.35. Distribution of thorium and rhenium between aqueous nitric acid and 30.1 vol % (1.10 M) tributyl phosphate in n-dodecane at 40°C

$[HNO_3]_{aq}$ (M)	$[Th]_{org}$ (M)	$[Th]_{aq}$ (M)	D_{Th}	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}
Initial aqueous thorium concentration = 0.05 M (nominal)						
0.15	2.72	48.0	0.0567	0.86	4.4	0.20
0.20	3.58	46.9	0.0763	0.97	4.2	0.23
0.40	7.76	42.8	0.181	1.2	3.9	0.31
0.60	12.6	37.7	0.334	1.6	3.5	0.46
0.80	17.1	33.1	0.517	1.6	3.4	0.47
1.00	20.4	29.7	0.687	1.4	3.5	0.40
2.00	32.1	17.9	1.79	1.2	3.8	0.32
3.00	35.9	12.8	2.80	1.0	3.9	0.26
4.00	38.1	10.7	3.56	1.0	4.0	0.25
Initial aqueous thorium concentration = 0.10 M (nominal)						
0.15	9.61	93.3	0.103	1.6	3.8	0.42
0.20	11.3	91.0	0.124	1.6	3.7	0.43
0.40	19.3	83.7	0.231	1.9	3.4	0.56
0.60	26.8	74.1	0.362	2.0	3.2	0.63
0.80	35.3	67.1	0.526	2.3	3.1	0.74
1.00	41.6	61.5	0.676	2.3	3.0	0.77
2.00	59.9	40.9	1.46	1.9	3.3	0.58
3.00	68.4	32.1	2.13	1.9	3.5	0.54
4.00	71.1	28.1	2.53	1.7	3.6	0.47
Initial aqueous thorium concentration = 0.20 M (nominal)						
0.15	34.0	170	0.200	2.6	3.0	0.87
0.20	37.7	167	0.226	2.8	2.9	0.97
0.40	49.2	153	0.322	2.7	2.9	0.93
0.60	61.9	142	0.436	2.6	3.1	0.84
0.80	73.6	131	0.562	3.0	2.6	1.2
8.00	81.5	121	0.674	2.8	2.6	1.1
1.00	112	91.7	1.22	2.4	3.0	0.80
3.00	130	77.4	1.68	2.4	3.2	0.75
4.00	131	73.1	1.79	2.3	3.1	0.74

Table A.36. Distribution of thorium and rhenium between aqueous nitric acid and 30.1 vol % (1.10 M) tributyl phosphate in n-dodecane at 60°C

$[HNO_3]_{aq}$ (M)	$[Th]_{org}$ (M)	$[Th]_{aq}$ (M)	D_{Th}	$[Re]_{org}$ (mM)	$[Re]_{aq}$ (mM)	D_{Re}
Initial aqueous thorium concentration = 0.05 M (nominal)						
0.15	2.11	48.8	0.0432	0.54	4.8	0.11
0.20	2.80	47.8	0.0586	0.59	4.6	0.13
0.40	6.68	43.9	0.152	0.86	4.1	0.18
0.60	11.3	39.5	0.286	0.97	4.1	0.24
0.80	14.8	35.8	0.413	1.1	4.0	0.28
1.00	18.9	32.1	0.589	1.2	3.9	0.31
2.00	29.7	20.0	1.49	1.1	3.9	0.28
3.00	33.3	15.4	2.16	1.0	4.0	0.25
4.00	35.3	14.2	2.49	0.81	4.1	0.20
Initial aqueous thorium concentration = 0.10 M (nominal)						
0.15	7.46	94.6	0.0789	1.3	4.5	0.29
0.20	9.01	93.8	0.0961	1.3	4.3	0.30
0.40	16.4	84.6	0.194	1.4	4.0	0.35
0.60	23.5	77.2	0.304	1.6	3.6	0.44
0.80	30.1	71.0	0.424	1.7	3.3	0.52
1.00	38.1	55.6	0.685	1.8	3.2	0.56
2.00	55.7	44.3	1.26	1.9	3.5	0.54
3.00	63.0	37.6	1.68	1.9	3.7	0.51
4.00	65.1	35.1	1.85	1.4	3.8	0.37
Initial aqueous thorium concentration = 0.20 M (nominal)						
0.15	28.1	173	0.162	2.0	3.5	0.57
0.20	31.8	171	0.186	2.1	3.5	0.60
0.40	42.8	156	0.309	2.3	3.1	0.74
0.60	56.2	147	0.382	2.4	3.0	0.80
0.80	67.7	136	0.498	3.0	2.7	1.1
1.00	75.1	127	0.591	2.5	3.0	0.83
2.00	106	101	1.05	2.3	3.1	0.74
3.00	115	88.4	1.30	2.4	3.1	0.77
4.00	119	86.7	1.37	2.2	3.3	0.67

Table A.37. Distribution of plutonium and technetium between aqueous nitric acid and 30.1 vol % (1.10 M) tributyl phosphate in n-dodecane at 25°C

$[HNO_3]_{aq}$ (M)	$[Pu]_{org}$ (M)	$[Pu]_{aq}$ (M)	D_{Pu}	$[Tc]_{org}$ (mM)	$[Tc]_{aq}$ (mM)	D_{Tc}
Initial aqueous plutonium concentration = 0.10 M (nominal)						
0.39	48.7	52.5	0.928	9.91	1.39	7.13
0.48	54.7	47.8	1.14	10.4	1.36	7.65
0.65	61.0	38.5	1.58	10.4	1.43	7.27
0.83	69.1	33.6	2.06	10.2	1.39	7.34
1.62	82.2	19.4	4.24	9.92	2.18	4.55
2.40	88.0	15.1	5.83	8.36	3.44	2.43
3.36	90.3	11.7	7.72	6.35	5.15	1.23
Initial aqueous plutonium concentration = 0.25 M (nominal)						
0.40	125	125	1.00	8.84	1.66	5.33
0.46	132	118	1.12	9.10	1.90	4.79
0.65	142	103	1.38	9.14	1.76	5.19
0.83	151	94.5	1.60	9.28	1.72	5.40
1.71	180	62.1	2.90	8.36	1.94	4.31
2.44	200	45.3	4.42	7.51	2.79	2.69
3.56	210	37.2	5.65	5.77	3.91	1.48

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