

OAK RIDGE NATIONAL LABORATORY

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U. S. ATOMIC ENERGY COMMISSION



DATE ISSUED:

FEB 2 1970

ORNL - TM - 2711

A MATHEMATICAL MODEL AND A COMPUTER PROGRAM FOR ESTIMATING
DISTRIBUTION COEFFICIENTS FOR PLUTONIUM, URANIUM, AND
NITRIC ACID IN EXTRACTIONS WITH TRI-n-BUTYL PHOSPHATE

D. E. Horner

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ORNL-TM-2711

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

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ABSTRACT

Based on the equilibria involved in the solvent extraction of plutonium, uranium, and nitric acid with tri-n-butyl phosphate (TBP), an empirical mathematical model and a FORTRAN program have been developed to allow computer calculation of estimated distribution coefficients. For the 15% TBP system, good agreement was shown between experimentally obtained distribution coefficients which were fitted to the model and the calculated coefficients.

1. INTRODUCTION

For development studies of the processing of plutonium (and uranium) in spent fast breeder reactor (FBR) fuels via solvent extraction by TBP, a knowledge of the distribution behavior of plutonium, uranium, and nitric acid is necessary. Because plutonium, as well as uranium and nitric acid, is present in macro concentrations in dissolver solutions of these fuels, the distribution of each of these components affects the distribution of the other two components. Most of the existing distribution data, obtained in connection with conventional, light-water reactor processing in which the plutonium is at very low concentrations, are not usable for FBR processing studies. Only a very limited amount of data for concentrated plutonium solutions is now available.

In addition to experimentally determined distribution data for this system, a method of rapidly estimating the distribution behavior over a wide range of conditions would be of value. This report describes a mathematical model and a corresponding FORTRAN computer program for rapid calculation of the estimated distribution coefficients for plutonium, uranium, and nitric acid. The model is

based on the overall equilibria involved in the extraction reactions, and uses empirical polynomial equations to fit experimental data to the model. According to the model, only the value of the equilibrium concentration, C_s , in Eq. (14) need be changed in order to permit estimated coefficients to be calculated using concentrations of TBP other than 15%. However, until this has been verified with experimental data, or, if necessary, until data obtained at different TBP concentrations are fitted to the model, estimates for coefficients using a TBP concentration other than 15% should not be made.

Many investigators have made studies of the equilibria involved in the extraction of uranium by TBP.¹⁻⁷ Some of the studies were mainly concerned with equilibrium calculations for uranium and nitric acid.¹⁻⁸ For a mathematical description in thermodynamic terms of the equilibria involved, a knowledge of the activities or activity coefficients for all the species is necessary. In addition, densities of all phases are necessary for expression of the concentrations in molality. Since not all of this information is available now, particularly the activities or activity coefficients for the organic-phase species, a thermodynamic, mathematical model cannot be fully described at this time. Extensive fundamental work to determine thermodynamic quantities such as activities and activity coefficients of uranyl nitrate, nitric acid, and TBP has been reported by W. Davis, Jr. et al.⁹⁻¹³ This work is a part of an overall program whose ultimate goal is the thermodynamic, mathematical description of the distribution behavior of all the components.¹²

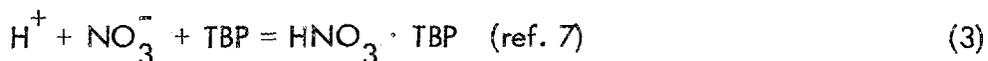
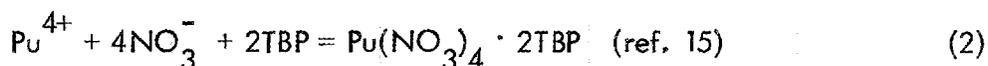
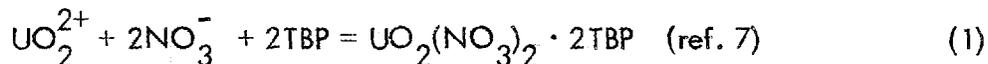
In spite of the lack of fundamental, thermodynamic data, it has been found practical to quantitatively describe the distribution behavior of the components in the system by means of expressions containing equilibrium quotients, K' , which in turn, are expressed empirically as polynomial functions of the aqueous-phase ionic strength. Use of these polynomial functions, however, is not intended to indicate that the true, mathematical relationship between the activity coefficients of species in this system and the ionic strength is a polynomial function.*

*Theoretically, according to Debye-Hückel theory, activity coefficients of ions of strong electrolytes are a function of ionic strength in dilute solutions. This suggests that the K' quantities as used in this model, being a function of the ratios of activity coefficients, are indeed a function of the ionic strength but not necessarily a polynomial function.

This model is similar to that used by Rozen and Khorkhorina,² and by Jury and Whatley¹ for the uranium--nitric acid system; and by Baumgartel, Ochsenfeld, and Schmieder¹⁴ for the plutonium--uranium--nitric acid system. The present report differs from the work described in ref. 14 in that it contains expressions for the calculation of nitric acid distributions in addition to those for plutonium and uranium. It also gives new empirical equations that were determined by fitting experimental distribution data obtained at ORNL, using 14.2% TBP, and provides a computer code for making the calculations.

2. MODEL DEFINING DISTRIBUTION COEFFICIENTS OF PLUTONIUM, URANIUM, AND NITRIC ACID

The model consists of equations derived to define the distribution coefficients of plutonium, uranium, and nitric acid in extractions by TBP as a function of the equilibrium aqueous-phase concentrations of these components and equilibrium quotients. The equations are based on the equilibrium expressions obtained from a consideration of the overall chemical reaction equations for formation of the extracted complexes:



Equilibrium expressions for these reactions are, respectively:

$$K_U = \frac{[\text{UO}_2(\text{NO}_3)_2 \cdot 2\text{TBP}]}{[\text{UO}_2^{2+}][\text{NO}_3^-]^2[\text{TBP}]^2} \times \frac{y_{\text{UO}_2(\text{NO}_3)_2 \cdot 2\text{TBP}}}{y_{\text{UO}_2^{2+}} y_{\text{NO}_3^-}^2 y_{\text{TBP}}^2} \quad (4)$$

$$K_{Pu} = \frac{[Pu(NO_3)_4 \cdot 2TBP]}{[Pu^{4+}][NO_3^-]^4 [TBP]^2} \times \frac{y_{Pu(NO_3)_4 \cdot 2TBP}}{y_{Pu^{4+}}^4 y_{NO_3^-}^4 y_{TBP}^2} \quad (5)$$

$$K_H = \frac{[HNO_3 \cdot TBP]}{[H^+][NO_3^-][TBP]} \times \frac{y_{HNO_3 \cdot TBP}}{y_{H^+} y_{NO_3^-} y_{TBP}} \quad (6)$$

The quantities in brackets represent concentrations of the reacting species expressed in molarity, and the y's represent the activity coefficients of these species.

The product of each equilibrium constant, K , and the ratio of the activity coefficients can be expressed as an equilibrium quotient, K' ; thus,

$$K'_U = K_U \times \frac{y_{UO_2^{2+}} y_{NO_3^-}^2 y_{TBP}^2}{y_{UO_2(NO_3)_2 \cdot 2TBP}} = \frac{[UO_2(NO_3)_2 \cdot 2TBP]}{[UO_2^{2+}][NO_3^-]^2 [TBP]^2} \quad (7)$$

$$K'_{Pu} = K_{Pu} \times \frac{y_{Pu^{4+}} y_{NO_3^-}^4 y_{TBP}^2}{y_{Pu(NO_3)_4 \cdot 2TBP}} = \frac{[Pu(NO_3)_4 \cdot 2TBP]}{[Pu^{4+}][NO_3^-]^4 [TBP]^2} \quad (8)$$

$$K'_H = K_H \times \frac{y_{H^+} y_{NO_3^-} y_{TBP}}{y_{HNO_3 \cdot TBP}} = \frac{[HNO_3 \cdot TBP]}{[H^+][NO_3^-][TBP]} \quad (9)$$

The K' quantities, reflecting changes in values of the activity coefficients with changes in concentrations of the reacting species, are not constant and are defined as empirical polynomial functions of the ionic strength of the aqueous phase. The K' quantities thus take on different values with each value of total ionic strength, μ , defined for this system as:

$$\mu = [H^+] + 3[UO_2^{2+}] + 10[Pu^{4+}] . \quad (10)$$

The distribution coefficient, D , for each component is defined as follows:

$$D_U = \frac{[UO_2(NO_3)_2 \cdot 2TBP]}{[UO_2^{2+}]} = K'_U [NO_3^-]^2 [TBP]^2 \quad (11)$$

$$D_{Pu} = \frac{[Pu(NO_3)_4 \cdot 2TBP]}{[Pu^{4+}]} = K'_{Pu} [NO_3^-]^4 [TBP]^2 \quad (12)$$

$$D_{HNO_3} = \frac{[HNO_3 \cdot TBP]}{[H^+]} = K'_H [NO_3^-][TBP] \quad (13)$$

The quantity $[TBP]$ in the above equations is the concentration of uncombined ("free") TBP, calculated as:

$$[TBP] = C_s - [HNO_3 \cdot TBP] - 2[UO_2(NO_3)_2 \cdot 2TBP] - 2[Pu(NO_3)_4 \cdot 2TBP] . \quad (14)$$

Here C_s is the equilibrium concentration of the total amount of TBP in the organic phase. As a simplifying assumption, the initial concentration, C , which differs from C_s due to volume changes during extraction of the various components, was used in place of C_s in the calculations. The initial concentration, C , of TBP in the water-saturated organic phase was calculated from the volume percent TBP (determined by chemical analysis) by the relation:

$$C = \frac{\text{vol \% TBP} \times 0.973 \times 1000}{266.3}$$

The error introduced by use of C instead of C_s is significant only when the ratio $[TBP]/C_s$ is less than about 0.1.

By proper substitution of Eqs. (7), (9), and (12) in Eq. (14), and then solving the resulting equation simultaneously with Eq. (12) to eliminate $[TBP]$ and D_U , I obtained a quadratic equation defining D_{Pu} as a function of the aqueous-phase concentrations of the components and K'_H , K'_{Pu} , and K'_U :

$$2D_{Pu} + 2\sqrt{D_{Pu}} \sqrt{K'_{Pu}} \left[\frac{1 + K'_H [H^+][NO_3^-]}{[UO_2^{2+}] K'_U + [Pu^{4+}][NO_3^-]^2 K'_{Pu}} \right] - \frac{C_s [NO_3^-]^2 K'_{Pu}}{[UO_2^{2+}] K'_U + [Pu^{4+}][NO_3^-]^2 K'_{Pu}} = 0. \quad (15)$$

By letting

$$S = \frac{1 + K'_H [H^+][NO_3^-]}{[UO_2^{2+}] K'_U + [Pu^{4+}][NO_3^-]^2 K'_{Pu}}$$

and

$$Z = \frac{[NO_3^-]^2}{[UO_2^{2+}] K'_U + [Pu^{4+}][NO_3^-]^2 K'_{Pu}}, \quad (16)$$

Eq. (15) was simplified to:

$$D_{Pu} + \sqrt{\frac{D_{Pu}}{2}} \sqrt{K'_{Pu}} S - \frac{C_s}{2} Z K'_{Pu} = 0. \quad (17)$$

The solution of this equation (using the positive value of the square root) is the equation:

$$\sqrt{D_{Pu}} = - \frac{\sqrt{K'_{Pu}} S}{4} \left(1 - \sqrt{1 + \frac{8C_s Z}{S^2}} \right). \quad (18)$$

In a similar manner, I obtained the quadratic equation:

$$2D_U + \sqrt{D_U} \sqrt{K'_U} \left[\frac{1 + K'_H [H^+][NO_3^-]}{[Pu^{4+}] K'_{Pu} [NO_3^-]^3 + [UO_2^{2+}][NO_3^-] K'_U} \right] - \frac{C_s K'_U}{[UO_2^{2+}] K'_U + [Pu^{4+}][NO_3^-]^2 K'_{Pu}} = 0. \quad (19)$$

Solution of this equation results in the equation:

$$\sqrt{D_U} = -\frac{\sqrt{K'_U} R}{4} \left(1 - \sqrt{1 + \frac{8C_s W}{R^2}} \right), \quad (20)$$

where

$$R = \frac{1 + K'_H [H^+][NO_3^-]}{[UO_2^{2+}][NO_3^-] K'_U + K'_{Pu} [Pu^{4+}][NO_3^-]^3} \quad (21)$$

and

$$W = \frac{1}{[UO_2^{2+}] K'_U + K'_{Pu} [Pu^{4+}][NO_3^-]^2}. \quad (22)$$

Finally for D_H , I obtained the quadratic equation:

$$D_H^2 \times \frac{[UO_2^{2+}] K'_U + K'_{Pu} [NO_3^-]^2 [Pu^{4+}]}{K'_H} + \frac{D_H}{2} \left(K'_H [H^+] + \frac{1}{[NO_3^-]} \right) - \frac{C_s}{2} K'_H = 0. \quad (23)$$

Solution of this equation results in the equation:

$$D_H = - \frac{TK'_H}{4V} \left(1 - \sqrt{1 + \frac{8C_s V}{T^2}} \right), \quad (24)$$

where

$$T = K'_H [H^+] + \frac{1}{[NO_3^-]} \quad (25)$$

and

$$V = [UO_2^{2+}] K'_U + [NO_3^-]^2 [Pu^{4+}] K'_{Pu}. \quad (26)$$

For each of the three components in the system, a polynomial equation relating K' to the ionic strength, μ , was determined:

$$K' = A + B\mu + D\mu^2 + E\mu^3. \quad (27)$$

The values of the coefficients A , B , D , and E can be determined by nonlinear least-squares fitting of a set of experimental distribution data to the model.

For use as a standard of reference, the model should predict very accurate values of the distribution coefficients. This, in turn, requires that the most accurate data obtainable be used to fit the model. The distribution data now available, although adequate for its original intended use, may not be of the degree of accuracy required for the present purpose. In many instances, sets of data from different sources do not agree with each other, thereby making it impossible to know which set of data, if any, is accurate enough to use. Thus we have initiated an experimental program that is designed to give very accurate distribution data; these data will be based on the most accurate analytical determinations possible. When they become available the data will be used to fit the model.

In the meantime, to show the potential use of the model by a calculated example, batch distribution data for plutonium, uranium, and nitric acid, obtained by Moore at ORNL,¹⁶ were fitted to the model, using the Marquardt Nonlinear Least Squares computer program^{17,18} to minimize the difference in the predicted

and experimental distribution coefficients. In this fitting procedure, the experimental distribution coefficients for such components were fitted separately by means of three polynomial equations containing 11 variable coefficients.* Results of this fitting are shown by the following equations. Fit to the plutonium distribution data, the equations are:

$$K'_{Pu} = 2.415 - 0.7010\mu + 0.05271\mu^2 + 0.03330\mu^3 \quad (28)$$

$$K'_U = 12.22 + 3.810\mu - 4.798\mu^2 + 2.477\mu^3 \quad (29)$$

$$K'_H = 0.4076 - 0.1660\mu + 0.03319\mu^2. \quad (30)$$

Fit to the uranium distribution data, the equations are:

$$K'_{Pu} = 3.882 - 0.4838\mu + 0.05814\mu^2 + 0.006005\mu^3 \quad (31)$$

$$K'_U = 18.39 + 5.114\mu - 4.174\mu^2 + 1.892\mu^3 \quad (32)$$

$$K'_H = 0.4841 - 0.1445\mu + 0.02216\mu^2. \quad (33)$$

A similar attempt to fit the nitric acid distribution data to this type of equation was not successful, possibly because of inaccurate analytical determinations for the acid concentrations. In accordance with the model, the same equations should result from fitting the distributions of any of the components. Presumably, the difference in the above sets of equations fitted to each component is due to the lack of a high degree of accuracy in the distribution data.

The difference in the two equations for each component based on either plutonium or uranium distribution data is shown in Figs. 1-3. In these figures the two equations are plotted as a function of the ionic strength, ranging in values from 2 to 5, for K'_{Pu} , K'_U , and K'_H , respectively. For all calculations in this report, except the nitric acid calculations, the set of equations based on the component being calculated was used. (The nitric acid calculations were based on the equations fit to the uranium distribution data.)

*In this case, 15% was used as the value for the initial TBP concentration, C, even though the experimental data were obtained with 14.2% TBP.

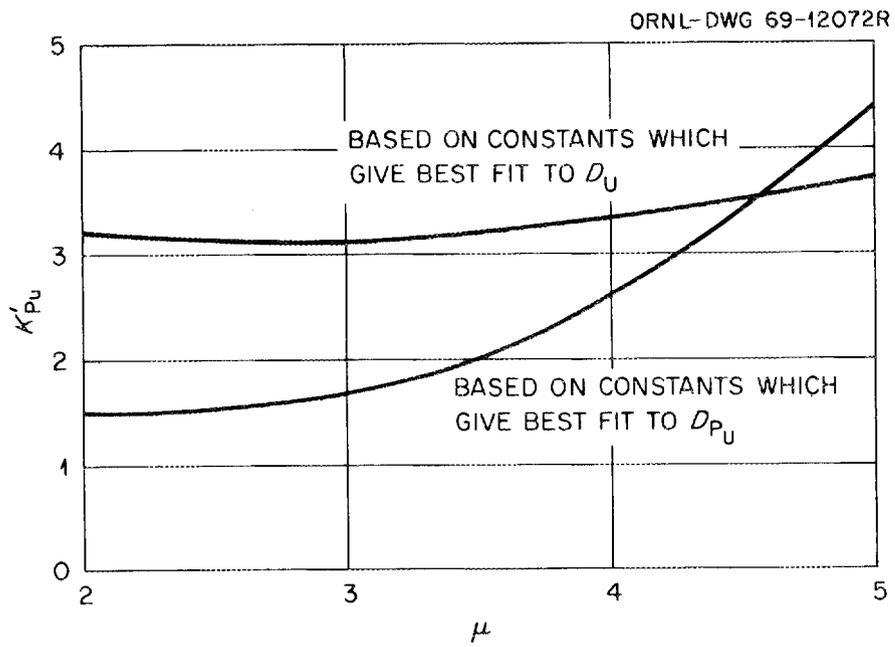


Fig. 1. Value of K'_{Pu} as a Function of the Ionic Strength, μ .

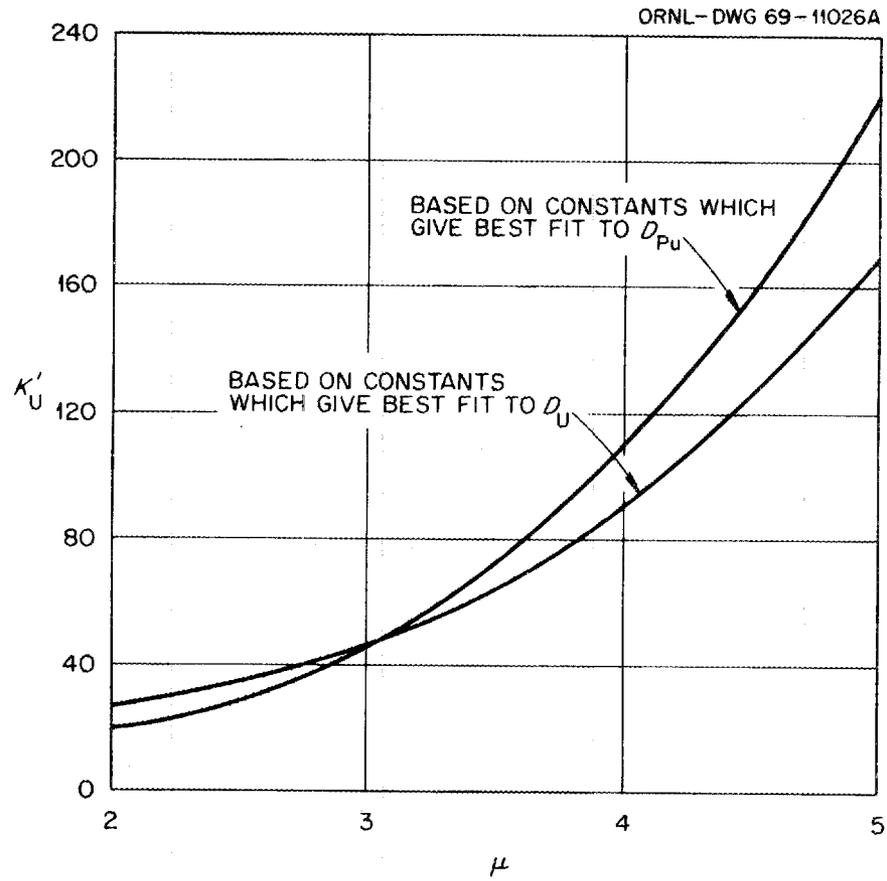


Fig. 2. Value of K'_U as a Function of the Ionic Strength, μ .

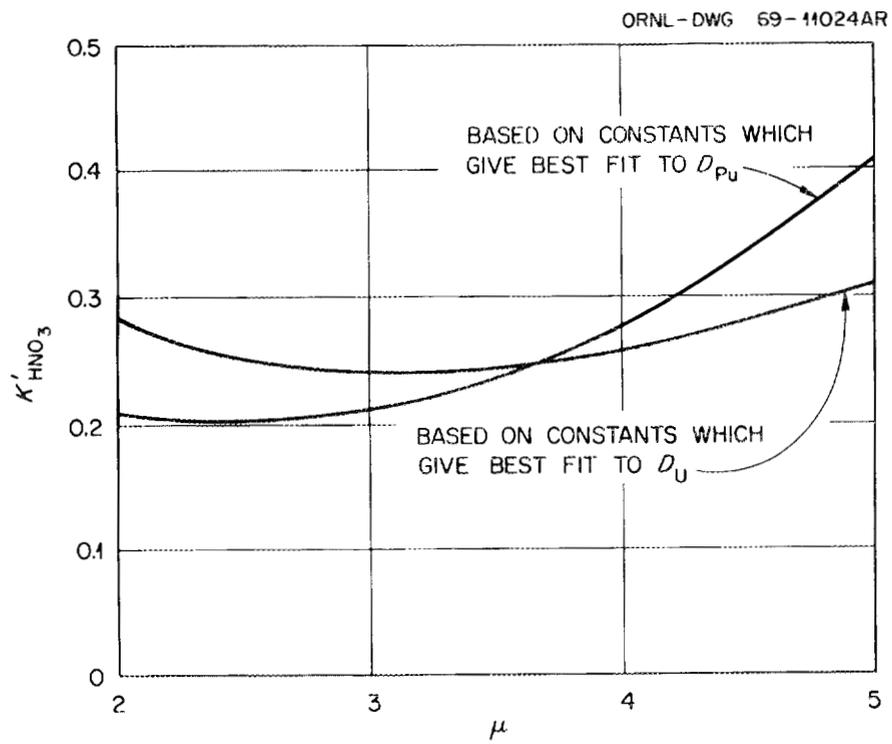


Fig. 3. Value of K'_{HNO_3} as a Function of the Ionic Strength, μ .

The FORTRAN program corresponding to the complete mathematical model is shown in Fig. 4 for the calculation of D_{Pu} , and, with five statement changes, for the calculation of D_U in Fig. 5 and for D_{HNO_3} in Fig. 6.

In these programs, the input data are:

$A(I)$ = equilibrium molar concentration of UO_2^{2+}

$B(I)$ = equilibrium molar concentration of Pu^{4+}

$C(I)$ = equilibrium molar concentration of H^+

The total molar concentration is calculated as:

$$\Sigma [NO_3^-] = 2[UO_2^{2+}] + 4[Pu^{4+}] + [H^+]. \quad (34)$$

Stepwise statements are executed in a loop to obtain values of the distribution coefficients (X), which are printed out along with the values of the calculated aqueous-phase concentration of plutonium in g/liter (AX) and the organic-phase concentration of plutonium in g/liter (YC).

3. TEST CALCULATIONS

The first type of test calculation shows results of the fitting of experimental distribution coefficients for both plutonium and uranium to the model (Figs. 7 and 8). For these figures, the experimentally obtained distribution coefficients are plotted versus the resulting predicted distribution coefficients calculated from the model. The good agreement between experimental and calculated values is shown by the close scatter of the points around the 45° line. The standard error of fit calculated by the Marquardt program was about 10% for plutonium and 6% for uranium.

For a second, more difficult, type of test, another computer program, SEPHIS,¹⁹ developed by Groenier of ORNL, was used to compare equilibrium concentrations of plutonium and nitric acid from an experimental batch countercurrent extraction cascade with those predicted by this program. The SEPHIS program, which uses the

```

PROGRAM PU U
DIMENSION A(200), B(200), C(200), X(200), AX(200), YC(200)
I = 1
9 READ 10, A(I), B(I), C(I), K
10 FORMAT (3F8.0, I1)
I = I + 1
IF (K.EQ.0)9,8
8 NO = I - 2
DO 24 I = 1, NO
11 D = 2.0*A(I) + 4.0*B(I) + C(I)
12 E = C(I) + 3.0*A(I) + 10.0*B(I)
13 F = 0.4076 - 0.1660*E + 0.03319*E**2
14 G = 12.22 + 3.810*E - 4.798*E**2 + 2.477*E**3
15 H = 2.415 - 0.7010*E + 0.05271*E**2 + 0.03330*E**3
16 T = A(I)*G + B(I)*D**2*H
17 S = (1.0 + F*C(I)*D)/T
18 Z = D**2/T
19 Y = (-SQRT(H)*S/4.0)*(1.0-SQRT(1.0 + 4.385*Z/S**2))
20 X(I) = Y**2
23 AX(I) = B(I)*239.0
24 YC(I) = X(I)*AX(I)
DO 21 I = 1, NO
21 PRINT 22, A(I), B(I), C(I), X(I), YC(I), AX(I)
22 FORMAT (6E20.4)
RETURN
END

```

Fig. 4. FORTRAN Program for Calculation of Plutonium Distribution Coefficients.

```

PROGRAM PU U
DIMENSION A(50), B(50), C(50), X(50)
I = 1
9 READ 10, A(I), B(I), C(I), K
10 FORMAT (3F8.0, I1)
I = I + 1
IF(K.EQ.0)9,8
8 NO = I - 2
DO 20 I = 1, NO
11 D = 2.0*A(I) + 4.0*B(I) + C(I)
12 E = C(I) + 3.0*A(I) + 10.0*B(I)
13 F = 0.4841 - 0.1445*E + .02216*E**2
14 G = 18.39 + 5.114*E - 4.174*E**2 + 1.892*E**3
15 H = 3.882 - 0.4838*E + 0.05814*E**2 + 0.006005*E**3
16 T = A(I)*G*D + H*B(I)*D**3
17 S = (1.0 + F*C(I)*D)/T
18 Z = D/T
19 Y = (-SQRT(G)*S/4.0)*(1.0-SQRT(1.0 + 4.385*Z/S**2))
20 X(I) = Y**2
DO 21 I = 1, NO
21 PRINT 22, A(I), B(I), C(I), X(I)
22 FORMAT (4E20.4)
RETURN
END

```

Fig. 5. FORTRAN Program for Calculation of Uranium Distribution Coefficients.

```
PROGRAM PU U
DIMENSION A(200), B(200), C(200), X(200)
I = 1
9 READ 10, A(I), B(I), C(I), K
10 FORMAT (3F8.0, I1)
I = I + 1
IF(K.EQ.0)9,8
8 NO = I - 2
DO 18 I = 1, NO
11 D = 2.0*A(I) + 4.0*B(I) + C(I)
12 E = C(I) + 3.0*A(I) + 10.0*B(I)
13 F = 0.4841 - 0.1445*E + .02216*E**2
14 G = 18.39 + 5.114*E - 4.174*E**2 + 1.892*E**3
15 H = 3.882 - 0.4838*E + 0.05814*E**2 + 0.006005*E**3
16 T = A(I)*G*D + H*B(I)*D**3
S = F*C(I) + 1.0/D
18 X(I) = -S/2.0*(1.0-SQRT(1.0 + 1.096*T/S**2))
DO 21 I = 1, NO
21 PRINT 22, A(I), B(I), C(I), X(I)
22 FORMAT (4E20.4)
RETURN
END
```

Fig. 6. FORTRAN Program for Calculation of HNO_3 Distribution Coefficients.

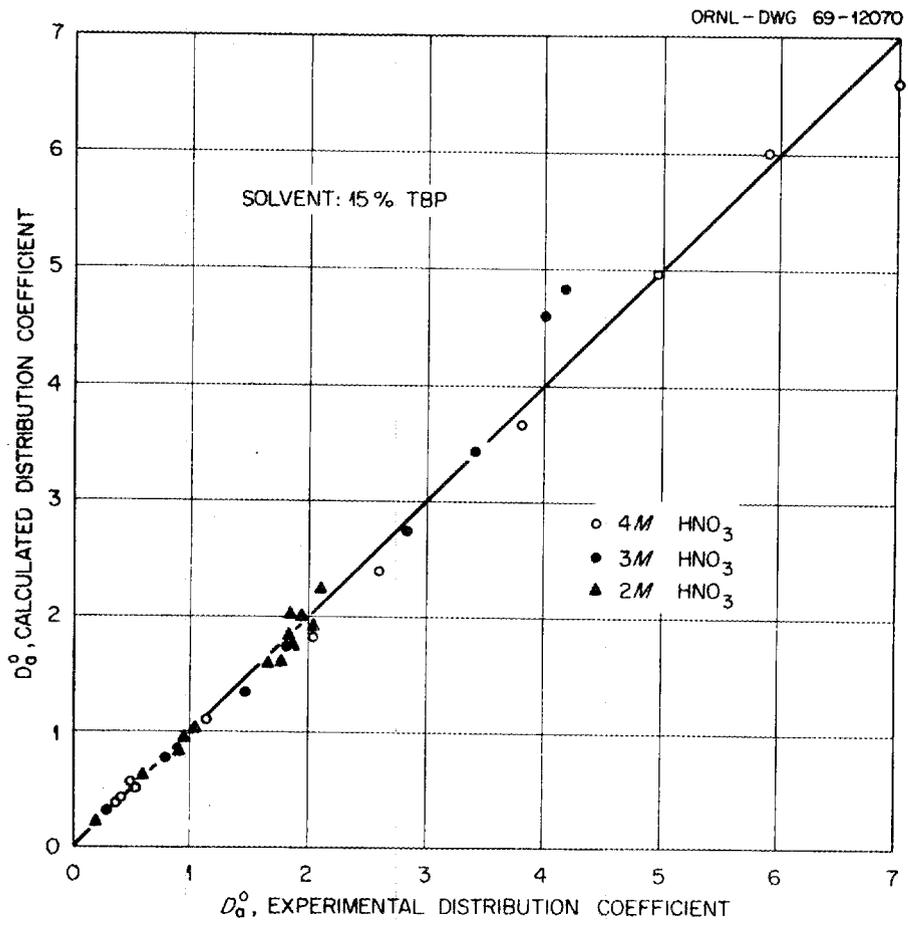


Fig. 7. Calculated vs Experimental Plutonium Distribution Coefficients.

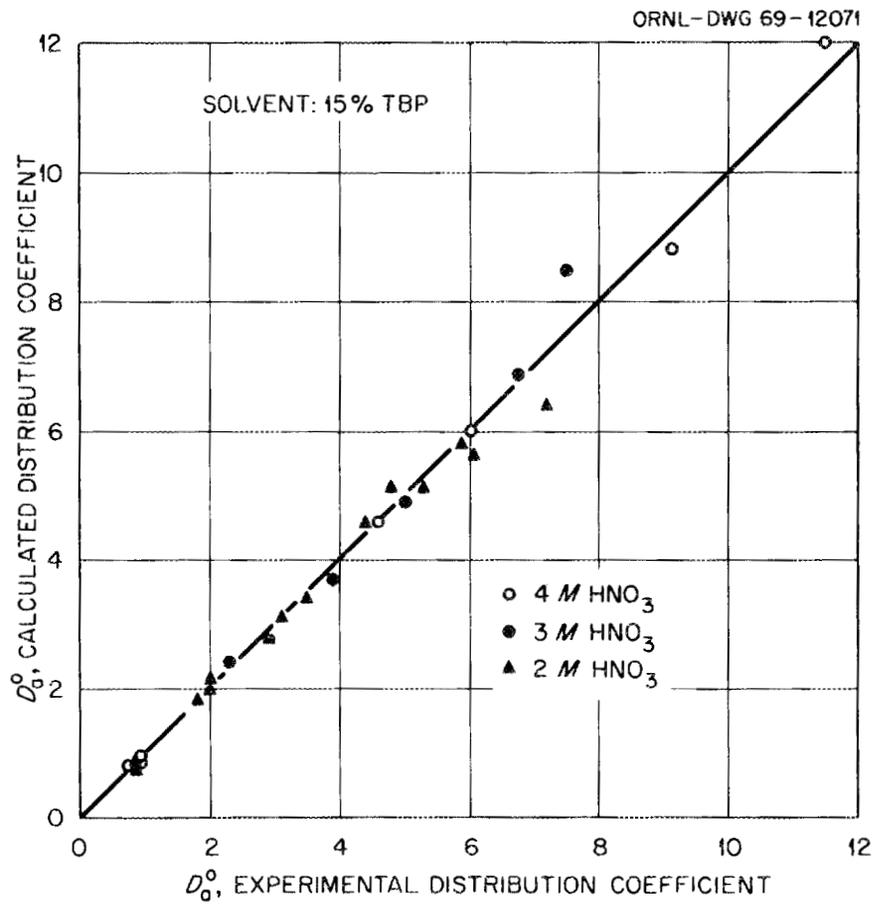


Fig. 8. Calculated vs Experimental Uranium Distribution Coefficients.

program of the equilibrium extraction model as a subprogram, is based on a model of a theoretical extraction stage used to simulate a countercurrent extraction cascade. Results of this test for extraction from a system containing plutonium and nitric acid (but no uranium) are shown in Table 1 as predicted and experimental stage concentration profiles. The generally good agreement between experimental and calculated profiles reflects agreement between the batch data used to fit the extraction model and the batch countercurrent data. In addition, the results show the potential capability of the models and the methods for practical predictions of an extraction system. Similar tests with a system containing plutonium, uranium, and nitric acid have not yet resulted in as good agreement. Reasons for the lack of close agreement are currently being sought.

4. POTENTIAL USES OF THE MODEL AND THE PROGRAM

With this equilibrium model and the corresponding computer program, estimates of the distribution behavior of any one, two, or three of the extractable components — plutonium, uranium, or nitric acid — under a wide variety of conditions can be made. The effect of solvent loading on the distribution coefficients can be calculated or automatically plotted as an isotherm by addition of the auxiliary program, subroutine ISOTHERM (Fig. 9), and the "Intrigue" plotting package.²⁰

Use of this program in conjunction with SEPHIS to calculate performance of a cascade has many potential applications. These applications include calculations of concentration profiles of an extraction, scrub, or stripping cascade during transient or at steady-state conditions. Such calculations should be particularly useful for studying maximum concentrations of plutonium that can be expected under a given set of conditions. This information may be very important, for example, for criticality considerations in a flowsheet. The effect of any operating parameter on the performance of the cascade, including the effect of a maloperation, can be calculated. The validity of the calculations should be checked experimentally, particularly until the accuracy of the data which are fitted to the model are improved.

Table 1. Estimated and Experimental Countercurrent Plutonium and Nitric Acid Concentration Profiles

Solvent: 15% TBP--85% NDD

Aqueous Feed: 4.1 M HNO_3 containing 19.2 g of Pu/liter

Scrub: 1 M HNO_3

Flow Ratios, feed/solvent/scrub: 1/0.9/0.11

Stage	Organic-Phase Conc. (g/liter)				Aqueous-Phase Conc. (g/liter)			
	Pu		HNO_3		Pu		HNO_3	
	Exp.	Est.	Exp.	Est.	Exp.	Est.	Exp.	Est.
Scrub-3	20.7	21.3	0.24	0.15	14.1	13.8	2.2	2.1
-2	20.9	23.0	0.24	0.16	13.9	13.0	2.5	2.3
-1	19.9	22.9	0.22	0.19	11.4	9.74	2.8	2.9
Feed	--	--	--	--	19.2		4.1	
Ext. -1	22.0	22.5	0.35	0.26	6.60	7.36	3.9	4.1
-2	7.17	9.07	0.33	0.35	1.52	1.76	3.9	4.1
-3	1.80	2.17	0.34	0.42	0.307	0.335	4.0	4.1
-4	0.35	0.413	0.36	0.43	< 0.1	0.0061	4.0	4.1
-5	0.075	0.0742	0.36	0.44	< 0.1	0.0011	4.0	4.1
-6	0.015	0.0129	0.38	0.44	< 0.1	0.0002	4.0	4.1
-7	0.0046	0.00019	0.37	0.41	< 0.1	< 0.0001	3.9	3.8

```
SUBROUTINE ISOTHERM (AX, NO, YC)
DIMENSION AX(200), A(6), YC(200)
CALL LINEAR (0, 4.0, 12, 0, 4.0, 12, 10., 1, A)
CALL LETTER (0, 19, 19HPLUTONIUM ISOTHERMS, A)
CALL LETTER (1, 24, 24HAQUEOUS PU CONCENTRATION, A)
CALL LETTER (2, 24, 24HORGANIC PU CONCENTRATION, A)
DO 10 I = 1, NO
  NAT = (9 + I)/10
  GO TO (1, 2, 3, 4, 5, 6), NAT
1  CALL POINT (I, AX(I), YC(I), 1, 0.12, 0.0, 1, A)
  GO TO 10
2  CALL POINT (I, AX(I), YC(I), 2, 0.12, 0.0, 1, A)
  GO TO 10
3  CALL POINT (I, AX(I), YC(I), 3, 0.12, 0.0, 1, A)
  GO TO 10
4  CALL POINT (I, AX(I), YC(I), 4, 0.12, 0.0, 1, A)
  GO TO 10
5  CALL POINT (I, AX(I), YC(I), 5, 0.12, 0.0, 1, A)
  GO TO 10
6  CALL POINT (I, AX(I), YC(I), 6, 0.12, 0.0, 1, A)
  GO TO 10
10 CONTINUE
  CALL ADVANCE (A)
  RETURN
END
```

Fig. 9. An Example of a FORTRAN Subroutine Program Used to Plot Calculated Plutonium Isotherms.

Generally, these types of calculated estimates should be of value in pointing out areas in which further experimental work should be done and, in conjunction with the experimental program, should be of value in general process design studies and flowsheet optimization studies.

5. ACKNOWLEDGMENTS

The author gratefully acknowledges encouragement in this work offered by J. W. Roddy, W. Davis, Jr., and R. H. Rainey. J. W. Roddy and W. Davis, Jr., were particularly helpful in their advice concerning use of the computer codes.

The cascade calculations using the SEPHIS Code, contributed by W. S. Groenier, also are gratefully acknowledged.

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