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The Mechanism of Extraction of Hexavalent

U(VI)

Uranium by Trilaurylamine Nitrate

by

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ABSTRACT

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The effects of amine, uranium and hydrogen ion concentrations on the extraction of U^{VI} from acidic nitrate solutions by tri-n-dodecylamine nitrate in toluene was studied in detail. Over a wide range of these variables, it was found that the data could be correlated quantitatively on a basis of a mechanism of extraction involving equilibria between amine nitrate monomer and dimer species, extraction of uranium by both of these species and extraction of nitric acid by the amine nitrate:

By determination of the nitric acid extracted into the organic phase by the fourth equilibrium, it was possible to calculate the concentration of amine nitrate available for participation in the other equilibria. From a reported value of $K = 125 \pm 25$ for

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the first equilibrium, values of

$$K_1 = 50 \pm 10$$

$$K_2 = 200 \pm 50$$

were estimated for the second and third equilibria, respectively.

Data is also presented on the variation of nitrate and hydrogen ions in the organic phase with uranium loading, from which it is concluded that no species other than the uranyl dinitrate complex is extracted. The difficulties caused by amine nitrate aggregation during studies on the mechanism of extraction of metals by other amines in other diluents are discussed.

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Introduction

The extraction of uranium from nitrate solutions is of interest in nuclear fuels reprocessing. The possible separation of uranium and plutonium by amine extraction was noted in 1957 (1), and several studies have been made of the extraction of both uranium and plutonium by alkylamines since that time (2) - (14).

Of some importance, since it affects the extraction of uranium, is the behavior of amines in nitrate-nitric acid systems (15). As a first step, nitric acid is extracted to form the amine salt:

The amine nitrate is capable of extracting more nitric acid

Nitric acid extracted in this fashion is referred to as "unbound" nitric acid. At very high aqueous nitric acid molarities, still more nitric acid may be extracted (10). Some water follows the nitric acid into the organic phase, but the effect is relatively slight and varies from solvent to solvent (10), (16), (17).

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Except in aromatic diluents, the range of solubility of the amine nitrate in diluents is limited. This appears to be linked to aggregation of the amine nitrate in the organic phase. Infra-red and light scattering studies (17) - (19) have shown aggregates in tri-n-octylamine nitrate-benzene, tri-iso-octylamine nitrate-toluene, and at high concentrations of trilaurylamine nitrate in benzene. A recent report (20) has shown the existence of a monomer-dimer equilibrium in tri dodecylamine nitrate in xylene. From the data given, a value of $K_{III} = 125 \pm 25$ for the equilibrium

may be derived.

The equilibrium constant for Eq. (1) is of the order of 10^5 (19) - (22). The order of magnitude of the equilibrium constant for Eq. (2) is unity (22), (23). It is of note that extraction of unbound nitric acid by Eq. (2) appears to involve solvation of the molecular nitric acid by the amine nitrate. Evidence for this is in the correlation of Skavdahl and Mason; for a wide range of nitric acid-nitrate solutions the nitric acid in the organic phase was a function of the molecular nitric acid activity only (24).

In the extraction of uranyl nitrate, two possible mechanisms may be postulated:

and

The ion exchange mechanism, Eq. (4), is sometimes quoted as implying the existence of some anionic complex (25), (26).

However, the solvation mechanism of extraction, Eq. (5), is thermodynamically equivalent to Eq. (4), so that it is only true to say that the amine to metal ratio is $(x-2)$. Studies in other systems have shown that the ion exchange mechanism is operative mainly at high salt concentrations (27).

It is usually assumed that two nitrate ions are transferred to the organic phase for each uranyl ion transferred. However, Baroncelli, (2), has postulated the extraction of some other species at high nitric acid concentrations. Keder, (6), has observed the spectrum of $\text{UO}_2(\text{NO}_3)_3^-$ in the organic phase.

Difficulty arises in the determination of the ratio, n , of the number of amine molecules per uranium molecule in the complex in the organic phase. This is normally determined from the slope of a log-log plot of the distribution coefficient versus free amine concentration, and non-integral values between 0 and 3 have been found. It may be noted, in passing, that this method of determining n will not show whether the amine is polymerized in the metal complex. Thus if polymerized amine species are involved, it becomes difficult to make a valid correction for the amine complexed to uranium in the organic phase, in determining the free amine concentration. Moreover, in determining the free amine concentration, it is necessary to correct for amine complexed with any unbound nitric acid extracted by Eq. (2), and this can only be done by simultaneous solution of Eq. (2) and Eq. (5) for the concentration of free amine nitrate.

Baroncelli and co-workers (2), (3) conclude that the effect of unbound nitric acid on the distribution of uranium is negligible; for the system tri-*n*-dodecylamine-benzene they find values of n from 1.1 to 1.8. The data of Shevchenko (9) for the system

tri-n-octylamine-xylene correspond to values of n of 0.9 and 1.1; the correction term applied by him is, however, incorrect*. For tri-n-octylamine in various diluents, Vdovenko (12) found values of n from 1.1 to 2.2. Knoch (8) found a value of n close to 1 for tri-iso-octylamine-kerosene. The lower values of n appear to correspond to diluents in which the amine nitrate is more highly aggregated. For instance, Hogfeldt (20) found monomer and dimer triisobutylamine nitrate species in xylene, but large aggregates in n-octane. These should be compared to the values for n of 1.7 in benzene (12) and approximately 1.0 for kerosene (8). If, as seems likely, large aggregates are not involved in the uranium complex, then a treatment analogous to that given by Allen for the amine sulfate system (28) shows that such a decrease may be expected. Moreover, this variation in values of n is fairly typical of the extraction of metals by different amine nitrates - for instance, Brothers and Hart (29) find values for the extraction of Pu^{IV} between 1.5 and 2.0, and Baroncelli and Scibona, (3), (30) find values between 2 and 2.9; Skavdahl (24) found a value of 1.5 for the extraction of RuNO^{III} . Thus it is with this aspect of the mechanism of extraction that this paper is particularly concerned, since the results might be expected to apply to alkylamine nitrate systems in general.

* In determining the free amine concentration, Shevchenko first corrected for the unbound nitric acid by application of a previously determined equilibrium constant for Eq. (2), and then corrected for the amine complexed with the extracted uranium. However, the extraction of uranium must reduce the available amine concentration, displacing Eq. (2) to the left, so that in effect Shevchenko neglected the equivalent equilibrium:

The values of n reported by Shevchenko are consequently too low.

Experimental

Experiments were performed by contacting an organic phase of tri dodecylamine in toluene which had been pre-equilibrated with an aqueous phase of nitric acid-ammonium nitrate, with an equivalent aqueous phase containing uranyl nitrate. Equilibration was accomplished by vigorous shaking for 1 minute, and slow shaking for one hour at 25°C to overcome any heat of reaction effects from the initial vigorous contact. After settling, the phases were rapidly separated, centrifuged and decanted, to be stored at 25°C before analysis. In this manner it was found possible to avoid change in nitric acid concentration in the organic phase which is known to occur on standing (22). The organic phase was tri-n-dodecylamine, Eastman No. 7727, of 97.5% purity, dissolved in AR toluene, standardized by potentiometric titration against perchloric acid in dioxane. Pre-equilibration was accomplished by contacting a single volume of organic phase with at least four fresh volumes of the relevant aqueous phase containing no uranium. Preliminary experiments showed that four such contacts were necessary to ensure no further extraction of nitrate or hydrogen ions. The pre-equilibrated organic phase was stored for no longer than 2 days at 25°C before use. Analysis for uranium was via the colorimetric thiocyanate method, and for nitrate by precipitation of nitron nitrate. Organic phase analysis for uranium and nitrate was by successive washings with 1M Na_2CO_3 and 0.5M HCl , as it was found that complete hydrolysis of the organic complex of uranium by 1M Na_2CO_3 alone was slow. The washings were passed through a small Whatmans No. 1 filter paper to remove amine droplets, and

combined. Analysis of a sample of the organic phase for hydrogen ions was by potentiometric titration in acetone versus standard aqueous sodium hydroxide.

Results

The results of two sets of experiments, the one on the effect of amine concentration at constant uranium concentration in the initial aqueous phase, and the other on the effect of uranium concentration at constant total amine concentration, are summarized in Tables 1 and 2. The nitrate concentration was held approximately constant, and the hydrogen ion varied from .01M to 1M.

Discussion

In determining the number of nitrate ions transferred with each uranyl ion in the organic phase, it was assumed that the total nitrate in the organic phase was derived from the amine nitrate, any unbound nitric acid present, and the nitrate transferred with the uranium, only, i.e.

$$\underline{C_{NO_3}} = C_{R_3N} + C_{unbound} H^+ + \underline{AC_U} \quad (6)$$

where the organic phase concentrations of species which may be present in both the aqueous and organic phases are indicated by underlining. The unbound nitric acid was determined by titration. Typical titration curves are shown in Figure 1. The first end point for phases in equilibrium with 1M H^+ solutions corresponds to the unbound nitric acid. The second end point, where uranium is present, corresponds to the completion of the uranium precipitation reaction, and the final end point to total hydrolysis of the

Table 1

Effect of Amine Concentration of Uranium Distribution Coefficient

Aqueous				Organic							Phase Ratio	Mass Bal.	D _U
Amine Concn.	Initial Uranium	Initial Nitrate	Initial H ⁺	Equil. Uranium	Equil. Nitrate	Equil. H ⁺	Equil. Uran.	Equil. Nitrate	Equil. Total H ⁺	Equil. Unbound H ⁺			
M	M	M	M	M	M	M	M	M	M	M	A/O	%	
.346 ^a	.0170	2.72	.010	.00537	2.72	.010	.0158	.372	.393	M11	4/3	101	2.91
.210 ^a	"	"	"	.00941	2.72	.010	.00983	.229	.239	M11	4/3	99	1.04
.109 ^a	"	"	"	.0141	-	-	.00357	.116	-	-	4/3	99	.254
.0501 ^a	"	"	"	.0162	-	-	.00102	-	-	-	4/3	100	.0624
.0201 ^a	"	"	"	.0168	-	-	.00024	-	-	-	4/3	100	.0141
.0101 ^a	"	"	"	.0169	-	-	.00005	-	-	-	1/1	100	.0030
										Initial Unbound H ⁺	Final Unbound H ⁺		
.339 ^b	.0152	2.94	.945	.00609	2.90	.929	.0089	.446	.0984	.0933	1/1	98	1.45
.204 ^b	"	"	"	.0083	2.93	.947	.0059	.269	.0531	.0512	1/1	94	.706
.103 ^b	"	"	"	.0128	2.95	.945	.00210	.129	.0240	.0239	1/1	96	.169
.0515 ^b	"	"	"	.0140	-	-	.00084	.0661	.0118	.0118	1/1	98	.060
.0205 ^b	"	"	"	.0150	-	-	.000126	-	.0049	.0049	1/1	100	.0084
.0105 ^b	"	"	"	.0152	-	-	.000084	-	.0025	.0024	1/1	100	.0006

a) Amine in equilibrium with 2.75 M NO₃⁻, 0.010 M H⁺b) Amine in equilibrium with 2.95 M NO₃⁻, 0.93 M H⁺

Table 2

Effect of Uranium Concentration on Distribution Coefficient

Aqueous							Organic							Phase Ratio	Mass Bal.	D ₀
Amine Concen.	Initial Uranium	Initial Nitrate	Initial H ⁺	Equil. Uranium	Equil. Nitrate	Equil. H ⁺	Equil. Uranium	Equil. Nitrate	Equil. H ⁺	Initial Unbound H ⁺	Final Unbound H ⁺					
<u>M</u>	<u>M</u>	<u>M</u>	<u>M</u>	<u>M</u>	<u>M</u>	<u>M</u>	<u>M</u>	<u>M</u>	<u>M</u>	<u>M</u>	<u>M</u>	A/O	%			
.346 ^a	.0153	2.72	.0048	.00350	2.71	.0048	.0114	.363	N11	N11	N11	1/1	98	3.27		
"	.0374	2.74	.0048	.0108	2.76	.0045	.0256	.387	-	-	-	1/1	97	2.37		
"	.0910	2.80	.0046	.0415	2.82	.0046	.0486	.428	-	-	-	1/1	99	1.18		
"	.187	2.86	.0042	.115	2.86	.0041	.0687	.474	-	-	-	1/1	99	.597		
"	.384	2.80	.0043	.287	2.80	.0041	.0921	.511	N11	N11	N11	1/1	98	.321		
.339 ^b	.0383	2.90	.945	.0172	2.91	.946	.0203	.468	.105	.093	.093	1/1	98	1.18		
"	.0936	2.92	.938	.0530	2.84	.940	.0410	.490	.105	.072	.072	1/1	100	.775		
"	.189	2.92	.940	.123	2.85	.950	.0616	.518	"	.058	.058	1/1	98	.503		
"	.405	3.22	.941	.312	3.16	.956	.0950	.578	"	.046	.046	1/1	101	.305		
"	.405	3.22	.941	.355	3.07	.962	.0996	.582	"	.042	.042	2/1	100	.281		

a) In equilibrium with 2.72 M NO₃, .0047 M H⁺

b) In equilibrium with 2.90 M NO₃, .940 M H⁺

FIG. I TITRATION OF ORGANIC EXTRACT PHASES.

amine nitrate. The precipitation of uranium as sodium diuranate requires three OH^- equivalents per uranium, so it is noteworthy that the total OH^- consumed in the titrations did correspond to the sum of the free nitric acid, three times the uranium concentration, and the amine concentration. This is evidence that no uranyl nitrate acid, $\text{H}_{x-2}\text{UO}_2(\text{NO}_3)_x$, is extracted. In the presence of uranium it is necessary to extrapolate the curve for the nitric acid end point to the half-wave potential for the reaction in order to obtain the true end point.

A plot of (6) is given in Figure 2. The line through the experimental points is of slope 2.0, so that two nitrate ions must be transferred with each uranyl ion during the extraction. Thus neither acidic nor hydrolyzed complex species of uranium seem to be present in the organic phase under these extraction conditions.

In attempting to find the amine to uranium ratio in the organic phase, log-log plots of distribution coefficient versus free amine concentration were made for the data of Tables 1 and 2 as shown in Figures 3 and 4. If it is assumed that the extraction of uranium is by

$$K_C = \frac{C_{US}^n}{C_U \cdot C_{T_1}^n}$$

$$\text{and } D_U = \frac{C_{US}^n}{C_U} = K_C C_{T_1}^n \quad (8)$$

C_{T_1} is the free amine concentration, which for Eq. (8) is calculated as the total amine concentration less the amine complexed

FIG.2 PLOT OF EQUATION (6)

FIG. 3 VARIATION OF URANIUM DISTRIBUTION COEFFICIENT WITH CALCULATED FREE AMINE CONCENTRATION, 0.01 M H^+

FIG. 4 VARIATION OF URANIUM DISTRIBUTION COEFFICIENT WITH AMINE CONCENTRATION, 1 $\underline{M}$ H^+

to nitric acid and to uranium, i.e.

$$C_{T_1} = C_F - nC_U \quad (9)$$

where $C_F = C_{R_3N} - C_{\text{unbound } H^+}$ (10)

If Eq. (8) applies, the log-log plots in Figures 3 and 4 should be linear and of slope n . In each figure, the data of Table 1 representing low and uniform uranium concentrations are consistent with a value of $n \approx 2$. However, the data of Table 2 representing various uranium concentrations follow quite a different trend. In order to show this difference more clearly, the isotherm for the extraction of uranium from $.01M H^+$ by $.346M$ amine is shown in Figure 5, together with the theoretical isotherm calculated by combining Eq. (8) and (9), assuming $n = 1, 2, 3$ and 4 , and using the values of D_U and C_{R_3N} at point A to determine K_C .

As Eq. (7) did not provide an explanation to the experimental data, it was necessary to postulate a different mechanism of extraction. Similar difficulties have been encountered in previous studies (30) - (34), and a semi-empirical equation of the form $D_U = K(C_F - nC_U)^n$ has been proposed for the isotherm. This could occur if the amine is polymerized, since

$$K'_C = \frac{C_U(S_x)_n}{C_U \cdot C_{T_{11}}^n}$$

and $D_U = \frac{C_U(S_x)_n}{C_U} = K'_C C_{T_{11}}^n$ (12)

where $C_{T_{11}}$ is given by

$$C_{T_{11}} = C_F - nC_U \quad (13)$$

FIG. 5 COMPARISON OF EXPERIMENTAL WITH THEORETICAL EXTRACTION ISOTHERMS.

In this case, combining Eq. (12) and (13), will give an equation of the same form of the semi-empirical one mentioned above. Then a plot of $D_U^{1/n}/C_F$ versus C_U/C_F should yield a straight line, the intercept of which at $D_U^{1/n}/C_F = 0$ should be $1/xn$. Plots of this type are given in Figures 6 and 7.

The best straight lines through the experimental points lead to values of xn of 3 and 2.5. The scatter in the points at low values of C_U may be ascribed to the necessity of using small differences between large numbers in the calculation. However, the points at high values of C_U deviate from the straight lines drawn by more than could be expected from the slight increases in aqueous nitrate concentration associated with these points. Moreover, by the above mechanism which assumes a single polymeric amine species only, it is difficult to explain the range in the values of xn .

If, however, a smooth curve is drawn through the experimental points in Figures 6 and 7, at high uranium concentrations xn intercept tends to a value of 2 and at low concentrations to a value of 4, in both cases. This suggests that there is some form of amine monomer-dimer equilibrium, with both species capable of extraction.

At this stage in the investigation, attention was drawn to the results of Hogfeldt (20) on the monomer-dimer equilibrium in the system tridodecylamine nitrate-xylene. It seemed likely his results would hold in the present system, with toluene replacing xylene as diluent.

FIG. 6 PLOT OF EQUATION (12) FOR 0.01 M H.⁺

FIG. 7 PLOT OF EQUATION (12) FOR 1M H^+

A mechanism of extraction consistent with the results obtained above is

The equilibrium constants of equilibria (14) and (15) are given by

$$K_I = \frac{c_{\text{US}_2}}{c_{\text{U}} \cdot c_{\text{S}}^2} \quad (16)$$

$$K_{II} = \frac{c_{\text{U}(\text{S}_2)_2}}{c_{\text{U}} \cdot c_{\text{S}_2}^2} \quad (17)$$

Moreover

$$c_{\text{T}} = c_{\text{S}} + 2c_{\text{S}_2} = c_{\text{F}} - 2c_{\text{US}_2} - 4c_{\text{U}(\text{S}_2)_2} \quad (18)$$

$$K_{III} = \frac{c_{\text{S}_2}}{c_{\text{S}}} \quad (19)$$

$$\text{and } D_{\text{U}} = \frac{c_{\text{US}_2} + c_{\text{U}(\text{S}_2)_2}}{c_{\text{U}}} = K_I c_{\text{S}}^2 + K_{II} c_{\text{S}_2}^2 \quad (20)$$

It should be possible to solve these five equations for the unknowns K_I , K_{II} , K_{III} , c_{S} , c_{S_2} , c_{US_2} and $c_{\text{U}(\text{S}_2)_2}$ by simultaneous solution using various sets of values of D_{U} , c_{U} and c_{F} obtained experimentally. In order to simplify the solution however, the data of Hogfeldt (20) were used to give $K_{III} = 125 \pm 25$. Using the data of Table 1 at 0.01M H^+ where, as there is no extraction of excess nitric acid, $c_{\text{F}} = c_{\text{R}_3\text{N}}$, values of K_I and K_{II} were

obtained assuming values for K_{III} of 100, 125 and 150. The best fit to this data was for $K_{III} = 125$, for which

$$K_I = 50 \pm 10$$

$$K_{II} = 200 \pm 50$$

To test the other data using these values of K_I , K_{II} and K_{III} , and thereby to test the proposed mechanism of extraction, C_T was calculated for the experimental values of D_U , C_U and C_P . It was necessary to assume firstly that the amine nitrate-nitric acid complex is present only as a monomer (Eq. (2)), and secondly that this complex is incapable as such of extracting uranium. The first assumption has an experimental basis - Baroncelli (17) has observed that the concentration of triaurylamine nitrate aggregates decreases during the course of the introduction of unbound nitric acid into the organic phase. The second assumption seems reasonable both from the fact that uranium displaces unbound nitric acid from the organic phase, and by analogy with the amine sulfate system, where the amine bisulfate was shown to be unable to extract uranium (28).

The experimental values of D_U and the values of C_T calculated from the data are shown in Figure 8, together with the D_U versus C_T relationship defined implicitly by Eq. (16) - (20). All the experimental points and the analytical curve based on the data taken only at low uranium and acid concentrations show very close agreement. It is of interest that the analytical curve is nearly linear, and that its apparent slope is non-integral.

FIG. 8 CORRELATION OF URANIUM DISTRIBUTION COEFFICIENT
 WITH FREE AMINE NITRATE CONCENTRATION CALCULATED
 FOR AMINE MONOMERS AND DIMERS.

The variation in the average number of amines associated with each uranium in the organic phase may be estimated qualitatively by considering Figure 9, a plot of C_S and C_{S_2} versus C_T from Eq. (18) and (19) for a value of $K_{III} = 125$. At a constant and relatively high total amine concentration and low uranium loading, most of the amine will be dimerized, and roughly four amines will be associated with each uranium. Increasing the uranium concentration will decrease the available amine concentration, and thereby increase the proportion of monomer to dimer, so in the limit two amines should be associated with each uranium, at the highest uranium concentrations. This trend is illustrated in Figure 10, where the amine to uranium ratio, $(C_F - C_T)/C_U$, is plotted against the percentage saturation, $2(C_U/C_F) \times 100$, for the experimental values of C_U and C_F and the values of C_T calculated from them.

Conclusions

The correlation shown in Figure 8 brings together data from a wide range of hydrogen ion, amine and uranium concentrations. This supports the proposed mechanism of extraction involving the four interrelated equilibria

FIG. 9 VARIATION OF AMINE NITRATE MONOMER AND DIMER CONCENTRATION WITH FREE AMINE CONCENTRATION.

It has proved possible to use molar concentrations rather than activities throughout the development of this model, probably due partly to approximately constant ionic strength, and partly to the use of ammonium nitrate as a salting-out agent, since in the range considered, the activity of uranyl nitrate is approximately constant in this salting-out agent (35). Non-idealities in the organic phase are taken into account by the monomer-dimer equilibrium of the amine nitrate.

The effect of hydrogen ion on the uranium distribution coefficient is entirely taken into account by the extraction of unbound nitric acid and the consequent reduction in amine nitrate available for entering into a complex with uranium. This conclusion is at variance with that of Baroncelli and co-workers who found no effect from nitric acid extraction in uranium extraction (2), but is in agreement with the results in the analogous chloride system (36). The effect of uranium and amine concentration on the distribution coefficient is satisfactorily taken into account by the mass action expressions for the equilibria involving these. The final relationship between distribution coefficient and free amine nitrate concentration is complex, but the relationship is nearly linear on a log-log plot and of an apparently non-integral slope. Thus it seems reasonable to suppose that the non-integral relationships which have been found in other systems in which alkylamine nitrates are used as extractants may also involve equilibria between various amine species, and extraction of the metal by more than one of these species.

While this work has not indicated the presence of higher polymers than amine nitrate dimers, it is not impossible that such higher polymers are in fact present, but the techniques employed here were too insensitive to show their presence. Nevertheless, it seems likely that most of the amine nitrate is present as monomers and dimers in toluene diluent. The reason for the dimerization is probably quadruple interactions between the amine nitrate ion-pairs. In turn, Keder's observation of the $\text{UO}_2(\text{NO}_3)_3^-$ spectrum in the organic phase (6) does not necessarily imply anything about the bonding of the uranium to amine, and it may be that ion-pair interactions lead to a structure for the monomer complex

At higher concentrations of uranium in the organic phase, it might be possible to overcome the comparatively weak interaction between the uranium complex and amine nitrate ion pairs, in the formation of a 1:1 amine to uranium compound.

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NOMENCLATURE

A	Ratio of nitrates to uranium extracted into organic phase.
C_F	Effective amine concentration, defined by Eq. (10).
C_{NO_3}	Total nitrate concentration in organic phase.
C_{R_3N}	Total amine concentration in organic phase.
C_S	Amine nitrate monomer concentration in organic phase.
C_{S_2}	Amine nitrate dimer concentration in organic phase.
C_T	Free amine nitrate concentration, calculated by Eq. (18).
C_{T_1}	Free amine nitrate concentration, calculated by Eq. (9).
$C_{T_{11}}$	Free amine nitrate concentration, calculated by Eq. (13).
C_U	Uranium nitrate concentration in aqueous phase.
$C_{\underline{U}}$	Uranium nitrate concentration in organic phase.
$C_{unbound\ H^+}$	Unbound nitric acid concentration in organic phase.

C_{US_n}	Concentration of $(R_3NHNO_3)_n UO_2(NO_3)_2$ in organic phase.
$C_{U(S_x)_n}$	Concentration of $[(R_3NHNO_3)_x]_n UO_2(NO_3)_2$ in organic phase.
C_{US_2}	Concentration of $(R_3NHNO_3)_2 UO_2(NO_3)_2$ in organic phase.
$C_{U(S_2)_2}$	Concentration of $[(R_3NHNO_3)_2]_2 UO_2(NO_3)_2$ in organic phase.
D_U	Distribution coefficient of uranium, $= C_U/C_U$
K_I	Equilibrium constant for reaction (14).
K_{II}	Equilibrium constant for reaction (15).
K_{III}	Equilibrium constant for reaction (3).
K_C	Equilibrium constant for reaction (7).
K_U^1	Equilibrium constant for reaction (11).
n	Amine-uranium ratio determined from log-log plot of D_U versus C_{T_1} .
x	Degree of polymerization of amine nitrates.

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