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Fuel Refabrication Task

DENSITY, ACIDITY, AND CONDUCTIVITY MEASUREMENTS OF URANYL NITRATE/NITRIC ACID SOLUTIONS

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TABLE OF CONTENTS

ABSTRACT

Conductivity, density, and acidity (pH) measurements were made on a series of uranyl nitrate solutions under conditions closely simulating the process used to load weak acid resins in the preparation of the HTGR recycle fuel particle. To relate these parameters to the uranium and nitrate concentrations of the solutions, a least-squares fit of the experimental data and mathematical expressions resulting from computer curve-fitting techniques was made.

Measurements were made on solutions having concentrations of 0.05 to 1.27 M uranium, 0.1 to 2.0 M nitrate, and NO_3/U ratios from 1.56 to 2.3. These measurements were made at 25, 30, 40, 50, and 75°C.

From these experiments, the necessary data were obtained to write two computer programs which can be used to predict or calculate uranium and nitrate concentrations of the process solutions and which will allow control of the process to be exercised in the particle preparation.

INTRODUCTION

The kernel of an HTGR recycle fuel particle is prepared by loading a weak acid resin (carboxylic acid exchange groups) with uranium from an acid-deficient nitrate solution using the exchange reaction: $UO_2^{2+} + 2HR \rightleftharpoons UO_2R_2 + 2H^+$, where HR is the resin in hydrogen form. Since the degree of loading of the uranium on the resin depends primarily on the acidity of the solution, the measurement of the acidity and the uranium content of the solution are important to the process control of loading. Therefore, it is desirable to continuously monitor solution parameters to control the process and obtain maximum loading efficiency.[1]

Since the net effect of resin loading, shown in the exchange reaction above, is to exchange one uranyl ion for two hydrogen ions, it seems the loading process could be controlled by in-line measurements of the pH and density of the uranyl nitrate. However, the assumption cannot be made that this is a simple chemical reaction. The uranium in solution is hydrolyzed or otherwise complexed and is only partially present as UO_2^{2+}. As a result of these complexes, attaining an equilibrium pH requires much longer for uranyl nitrate than for simple ionic solutions. This difference has also been observed by Haas[1] and Shaffer,[2] who have studied the behavior of uranyl nitrate as related to this and other processes. The complexity of uranyl nitrate solutions due to hydrolysis has also been studied by Baes and Mesmer.[3] These same types of acid-actinide mixtures were studied by Mocken, Marchand, and de Vries,[4] who used experimentally derived equations to relate density and conductivity to uranium and acid concentration. Their work was an extension of previous publications on density and conductivity measurements of these mixtures.[5,6]

The purpose of the experiments described and discussed here was to measure the densities, pH's, and conductivities of acid-deficient uranyl nitrate solutions under process conditions and to correlate them with uranium concentration, NO_3/U ratio, and temperature as variables. Then from this correlation the feasibility of using these solution parameters as a process control could be determined. Since the loaded resin is the end product in this process, it is very important that the process control be stringent enough to make sure all the resin is loaded uniformly. This fact should dictate the type of control used.

EXPERIMENTAL

Two stock solutions of uranyl nitrate were prepared by dissolving 387.3 grams of UO_3 in 128.2 ml of concentrated nitric acid under reflux. The solutions were filtered to remove any insoluble material and diluted to 1000 ml with water. Each solution (Nos. 1 and 2) was assayed by two methods—gravimetric and volumetric. There was excellent agreement in the assays by the two methods: 1.287 M in uranium for solution No. 1 and 1.271 M for solution No. 2. Using the Kjeldahl distillation method, the nitrate concentration of each solution was determined to be 2.008 M for solution No. 1 and 1.987 M for solution No. 2. The free acid or acid deficiency of the stock solutions was determined by potentiometric titration.

The density of each solution used in the experiment was determined by pycnometric measurement to an accuracy of ±0.05%.

The conductivities, i.e., specific conductances, of the experimental solutions were measured using a Radiometer conductivity meter (type CDM3) with a dip-type conductivity cell. The cell constant for the meter was experimentally determined to be 1.00 cm within 1.34%. This meter is equipped with temperature compensation and is capable of measuring conductances from 1.5 microsiemens to 200 millisiemens.

The pH of the solutions was measured with a Beckman Research pH meter (model 101900) with a resolution of 0.0005 pH units. The instrument is calibrated against a standard cell with a precisely known potential. The electrode is standardized using a buffer solution of known pH at a given temperature.

EXPERIMENTAL DATA

Set No. 1

Six concentrated standard solutions having NO_3/U mole ratios of 1.56, 1.70, 1.90, 2.00, 2.10, and 2.30 were prepared from stock solution No. 1 by diluting a weighed aliquot of the stock solution with a weighed amount of 2.00 M nitric acid. Five additional experimental solutions were prepared from each of the six standard solutions by diluting with water. These 36 solutions were used to obtain the experimental data. Measurements of pH, conductivity, and density were made at 25, 50, and 75 ± 0.05°C.

Set No. 2

Six concentrated standard solutions having NO_3/U mole ratios of 1.56, 1.60, 1.70, 1.80, 1.90, and 2.00 were prepared from stock solution No. 2 with 2.00 M nitric acid. Five additional experimental solutions were again prepared from each of the concentrated standard solutions, and measurements of pH, conductivity, and density were made at 30, 40, and 50°C on these 36 solutions.

RESULTS AND DISCUSSION

The numerical results for each set of 36 solutions are given in Appendices I and II. Figures 1, 2, and 3 show the set No. 1 solution relationship between acidity, density, and conductivity and the uranium concentration at 25°C. Figures 4, 5, and 6 show the same relationship at 30°C and at varying uranium concentrations for the solutions from set No. 2. These experimentally derived curves show that all three parameters are quite sensitive to the NO_3/U ratio. However, the density is almost independent of the NO_3/U ratio.

If the conductivity from data set No. 1 is plotted against the NO_3/U mole ratios at a fixed uranium concentration as shown in Figure 7, a break in the curve is observed at a NO_3/U ratio of 2.00, as expected.

The relationship between pH and the NO_3/U mole ratios at a fixed uranium concentration is shown in Figure 8. A sigmoidal curve is the

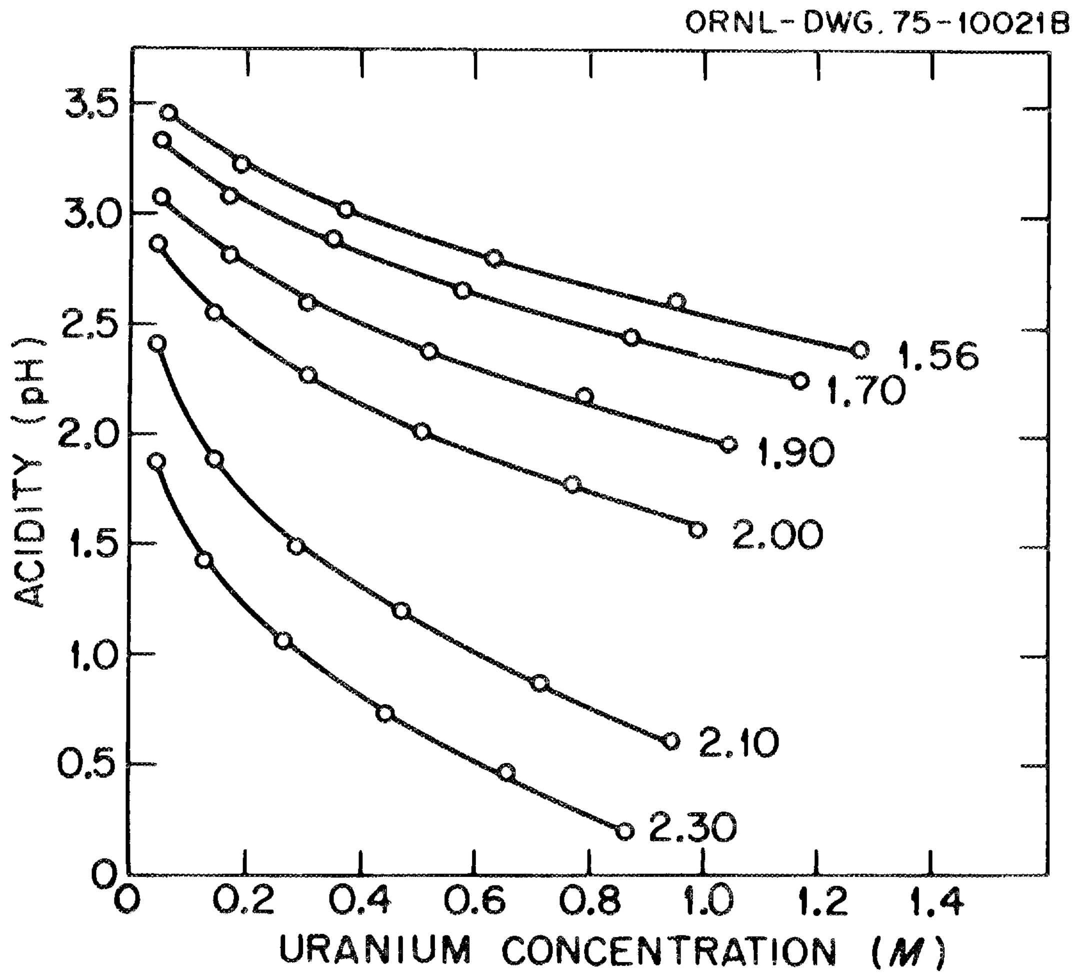

Fig. 1. Relationship between acidity and uranium concentration of uranyl nitrate solutions at 25°C and at various nitrate to uranium mole ratios.

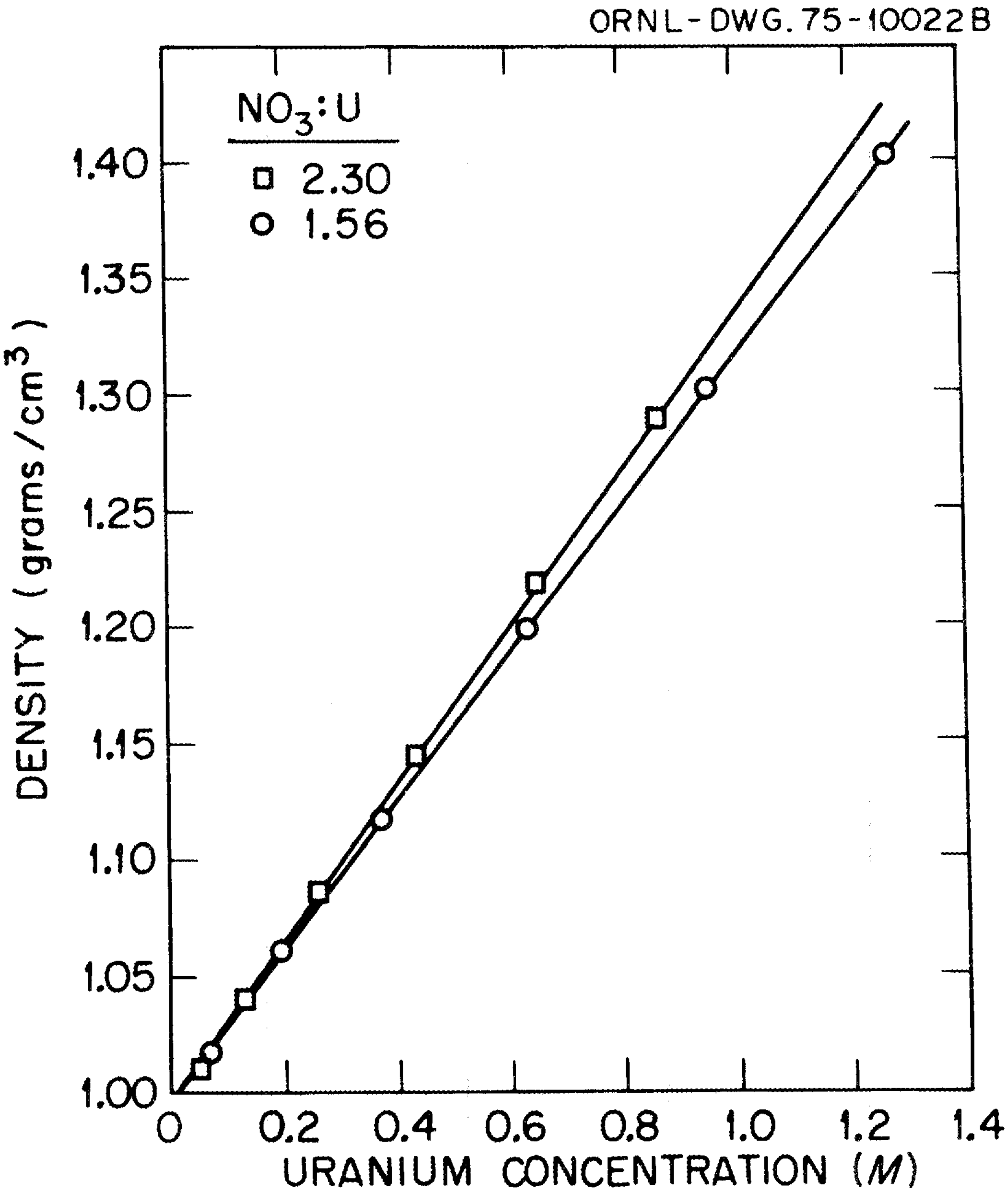

Fig. 2. Relationship between density and uranium concentration of uranyl nitrate solutions at 25°C and at extreme nitrate to uranium mole ratios.

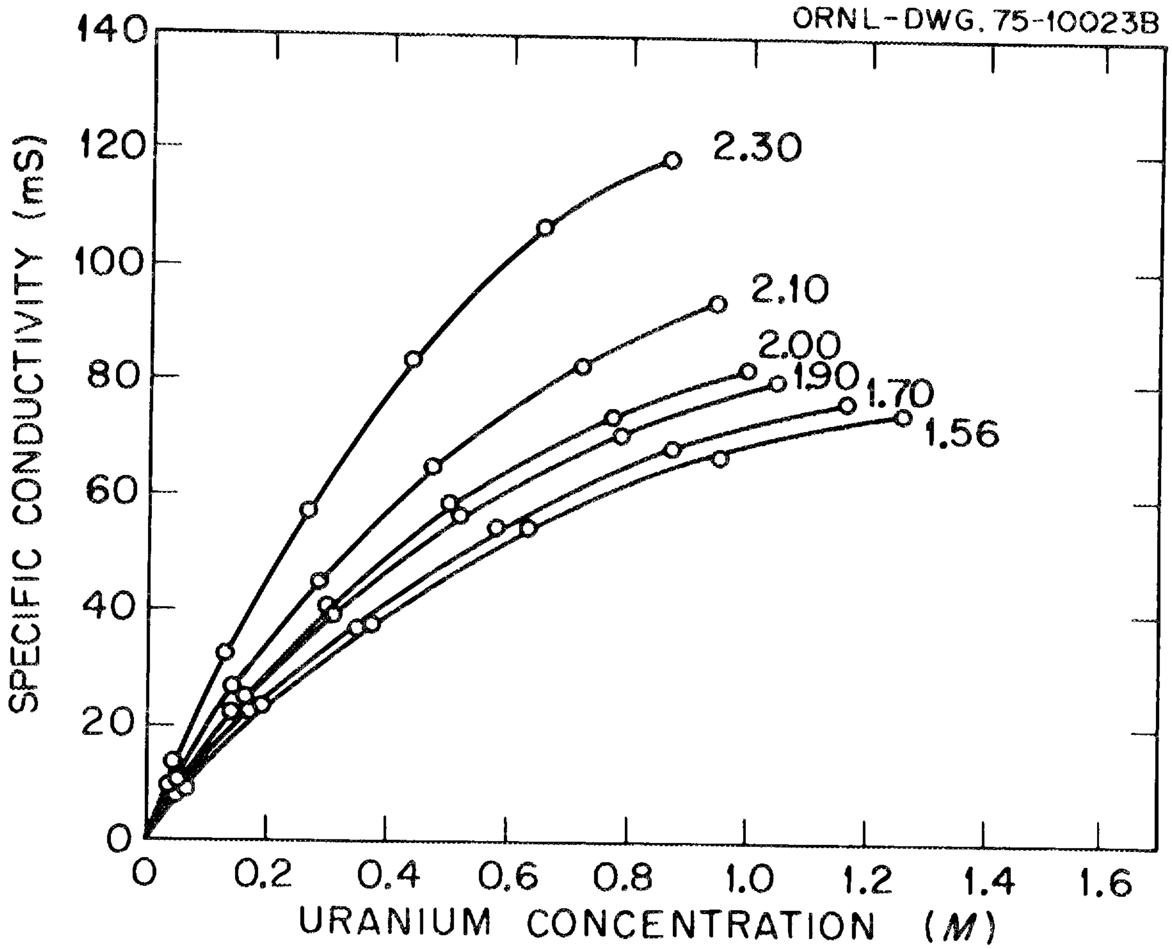

Fig. 3. Relationship between conductivity and uranium concentration of uranyl nitrate solutions at 25°C and at various nitrate to uranium mole ratios.

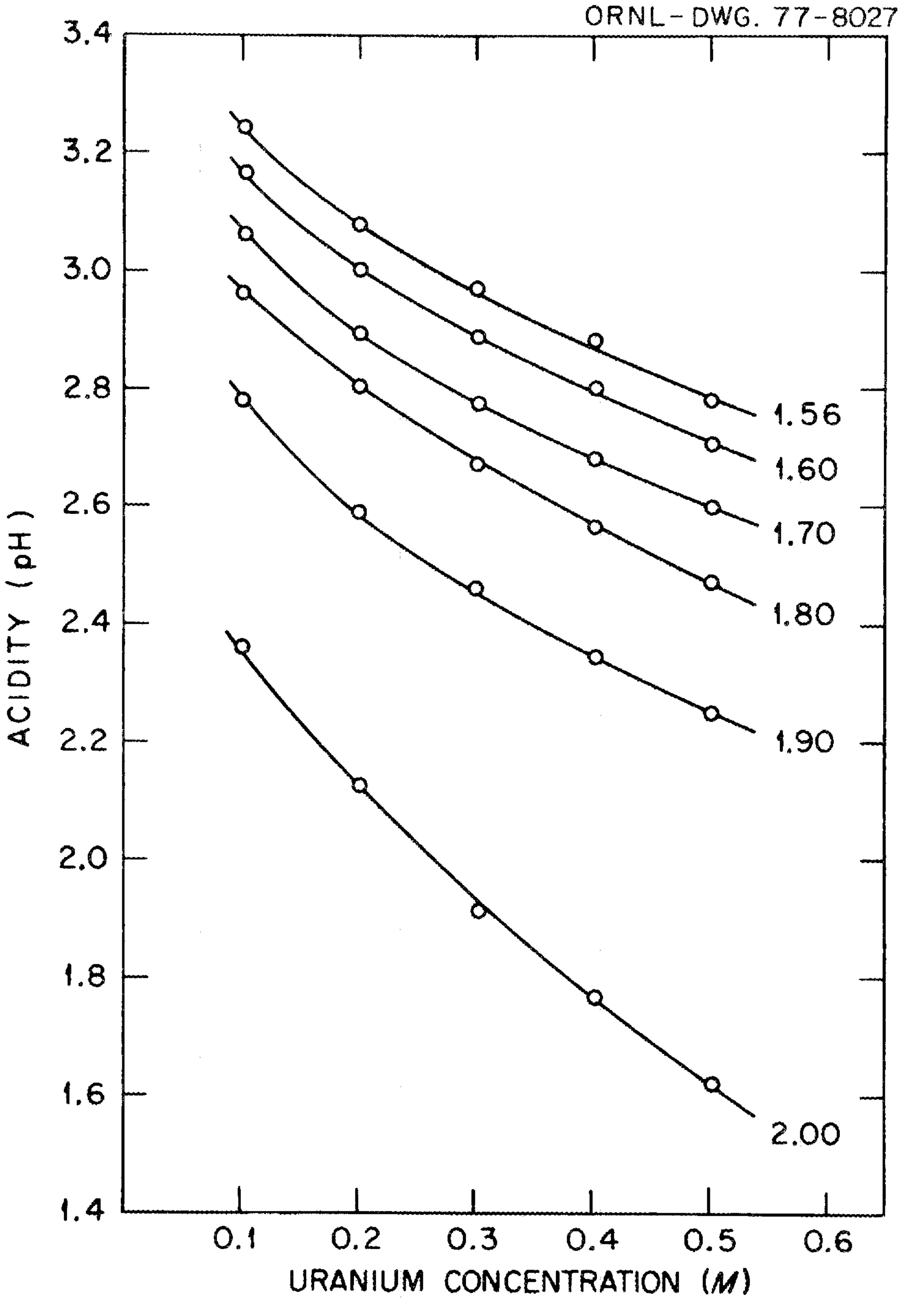

Fig. 4. Relationship between the acidity and uranium concentration of uranyl nitrate solutions at 30°C and various nitrate to uranium mole ratios.

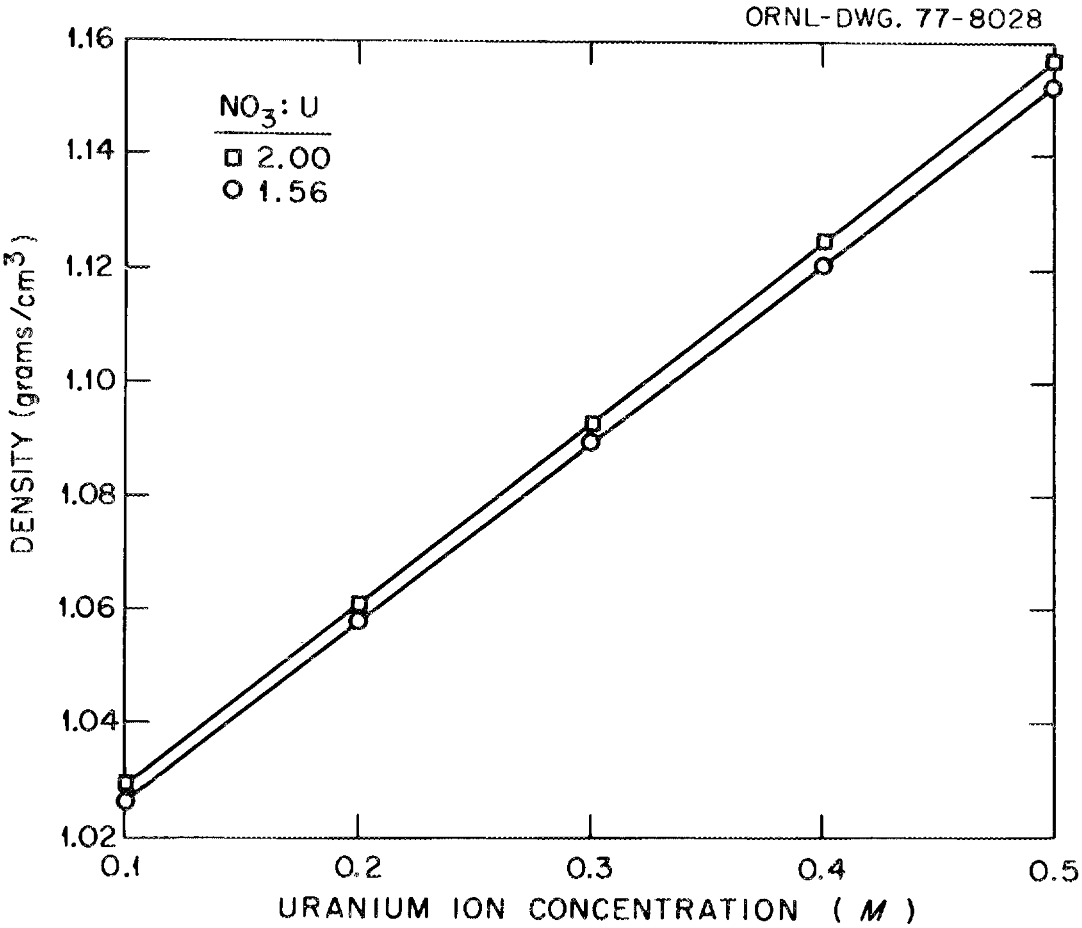

Fig. 5. Relationship between density and uranium concentration of uranyl nitrate solutions at 30°C and at extreme nitrate to uranium mole ratios.

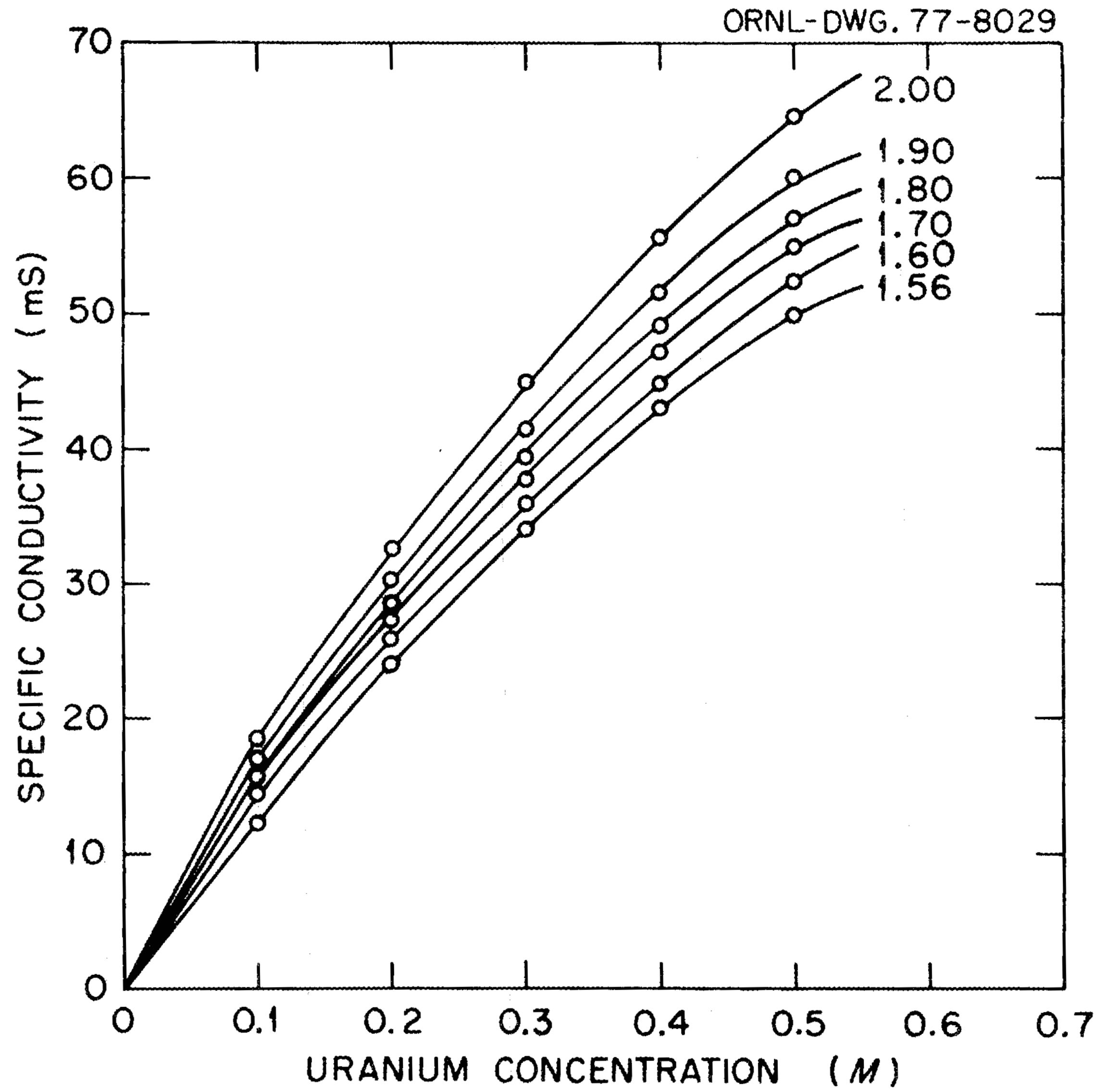

Fig. 6. Relationship between conductivity and uranium concentration of uranyl nitrate solutions at 30°C and at various nitrate to uranium mole ratios.

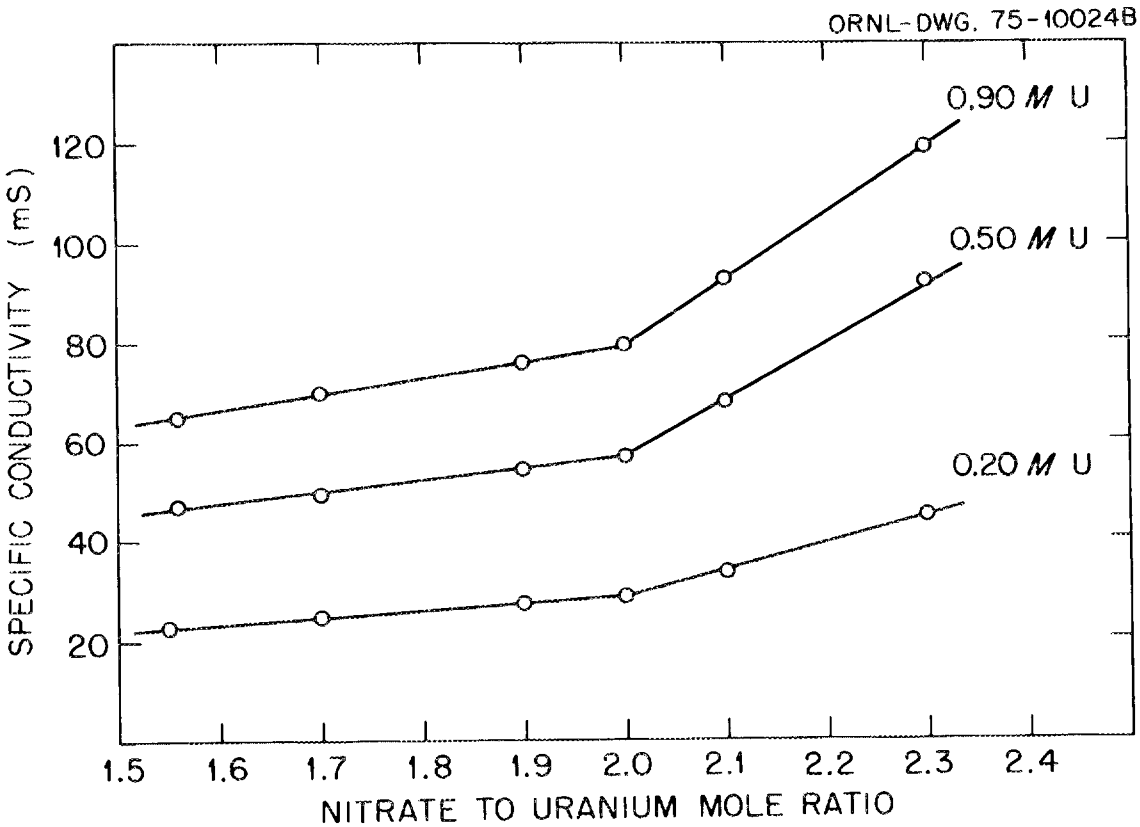

Fig. 7. Relationship between conductivity and the nitrate to uranium mole ratios of uranyl nitrate solutions at 25°C and at various fixed uranium concentrations.

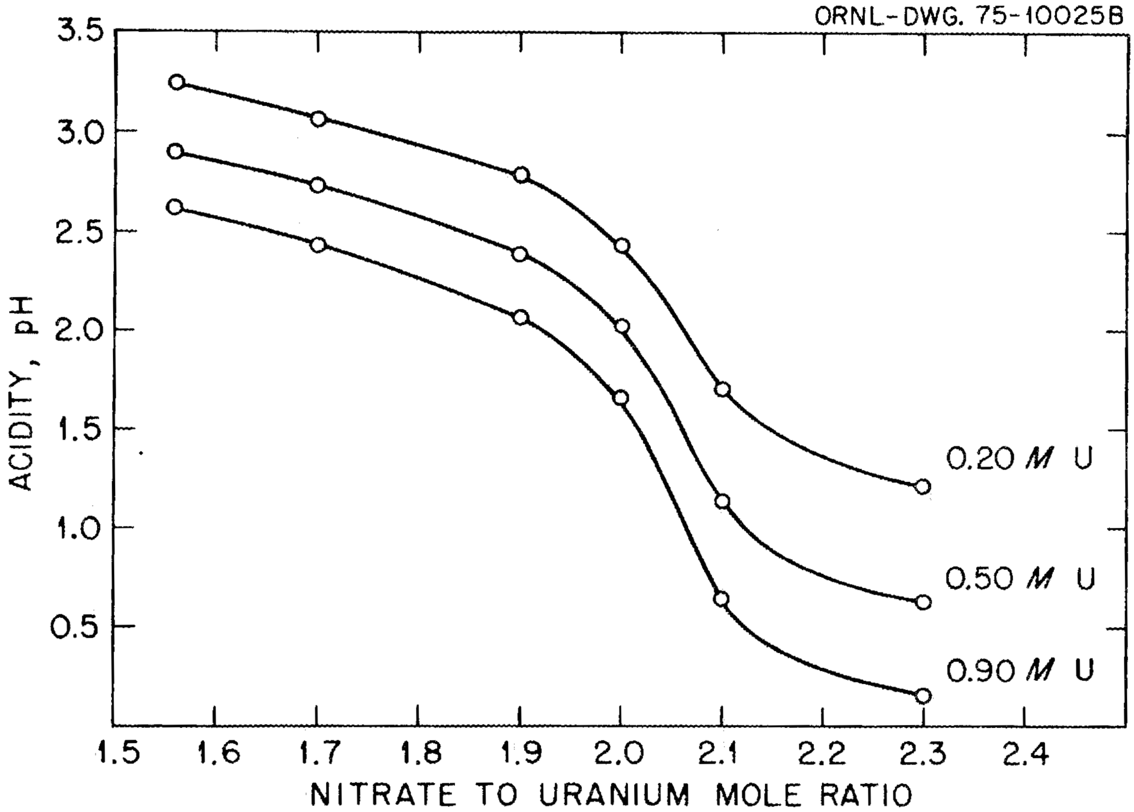

Fig. 8. Relationship between acidity and the nitrate to uranium mole ratios of uranyl nitrate solutions at 25°C and at various fixed uranium concentrations.

result, having an inflection point at a NO_3/U mole ratio of about 2.05 rather than 2.00 as expected. The deviation from stoichiometry is probably due to the uranium complexes present in these acid-deficient uranyl nitrate solutions. The attainment of an equilibrium pH requires a longer time compared to that of simple ionic solution because of these complexes.[1]

In Figures 9, 10, 11, and 12, the relationships among pH, conductivity, density, uranium concentration, and temperature are shown. Figure 9 illustrates pH values measured at both extremes of free acid and acid deficiency. This figure also indicates a slight disagreement found in the pH measured on the solutions made from stock solution No. 1 and those made from stock solution No. 2. The comparison between the two pH measurements can be made at 50°C since this was the only temperature common to both sets of data used in the experiment.

Figures 10, 11, and 12 show excellent agreement between the two data sets with respect to density and conductivity at 50°C. The precision of the density measurements for the two data sets when compared with the theoretical density of water is given in Tables I and II. Also shown in Figures 10 and 11 is the effect of temperature on conductivity and density, respectively. The conductivity of a given solution will increase while the density will decrease with temperature.

Since differences have been noted in the pH measured on the two stock solutions, a study was made to determine the reliability of the measurement. The acidity (pH) of a number of acid-deficienct uranyl nitrate preparations was measured. The dilutions on which the measurements were made were prepared from the original stock uranyl nitrate solution No. 2. The pH of each dilution was measured immediately after preparation at 30, 40, and 50°C. These measurements are shown in Table III.

A new stock uranyl nitrate solution (No. 3) was prepared in the same manner as were stock Nos. 1 and 2. The pH of dilutions made from the new stock solution was measured at 30°C and is shown in Table IV.

A comparison of the pH measurements of these stock solutions, shown in Figure 13, indicates that when pH measurements are made on two solutions having the same acid concentrations, the results will vary by as much as ±0.2 pH units. This would account for the deviation indicated in Figures 9 and 13.

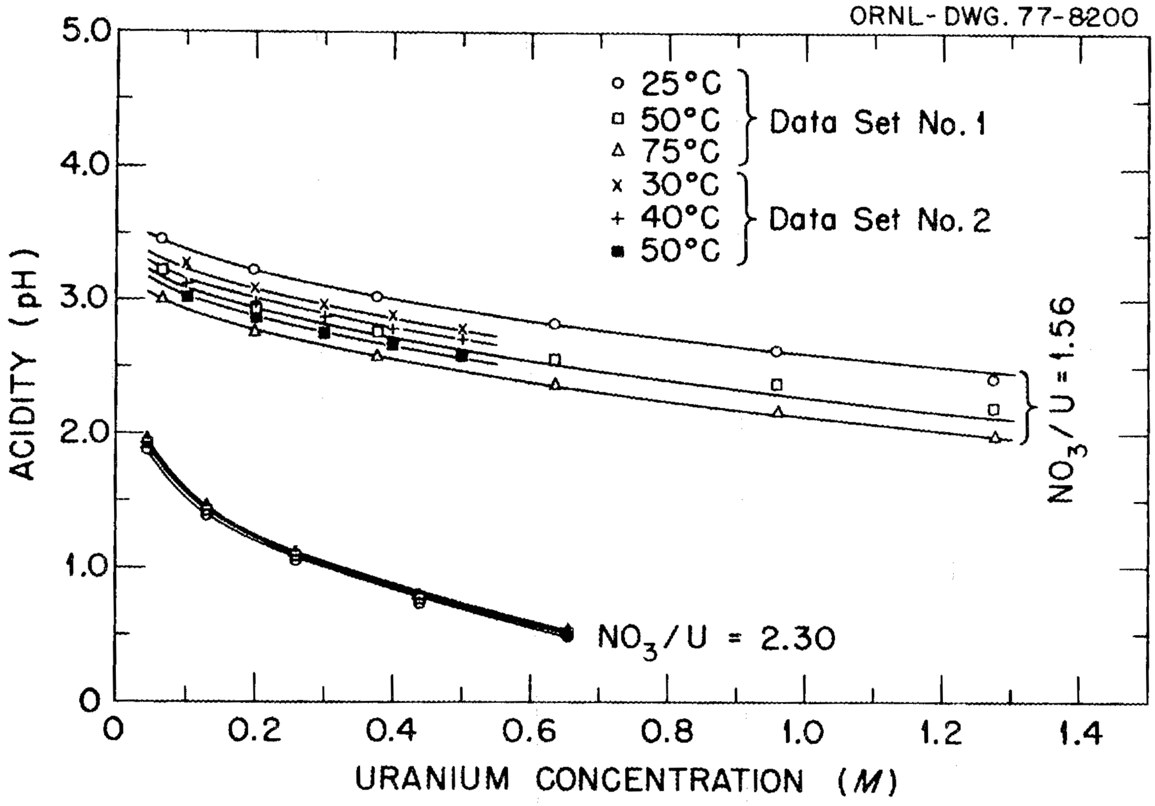

Fig. 9. Relationship between acidity and uranium concentration of uranyl nitrate solutions at various temperatures and extreme nitrate to uranium ratios.

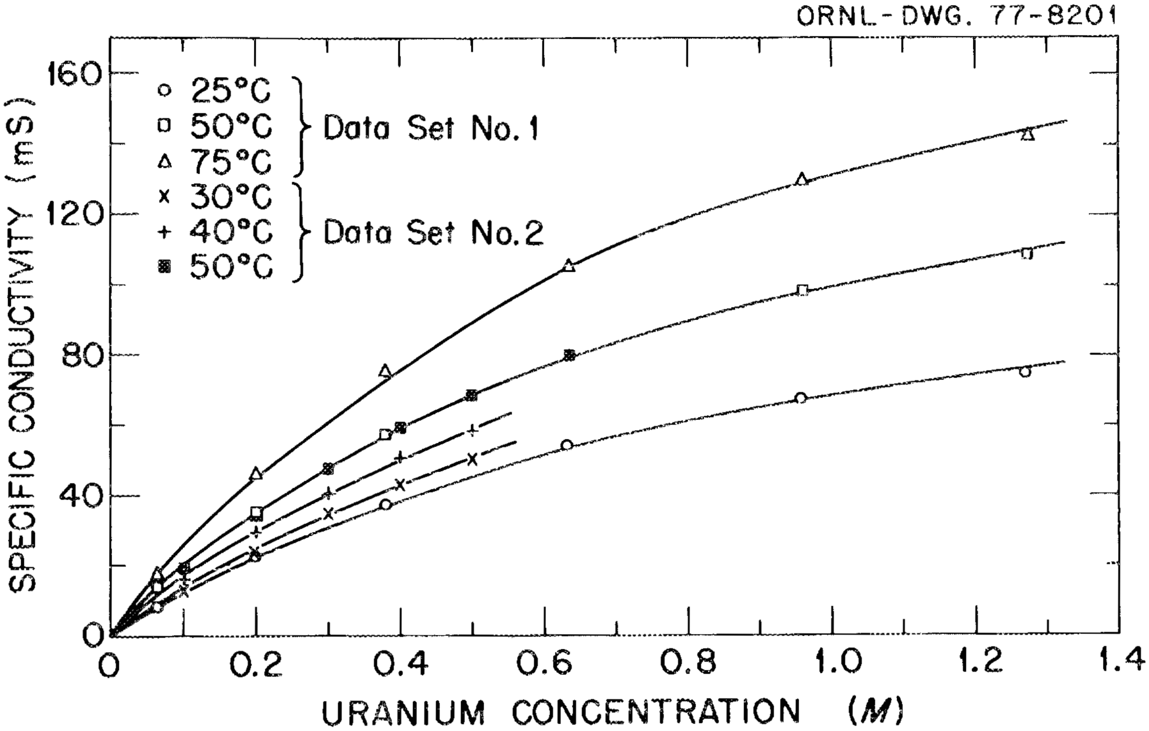

Fig. 10. Relationship between conductivity and uranium concentration at various temperatures (NO_3/U mole ratio = 1.56).

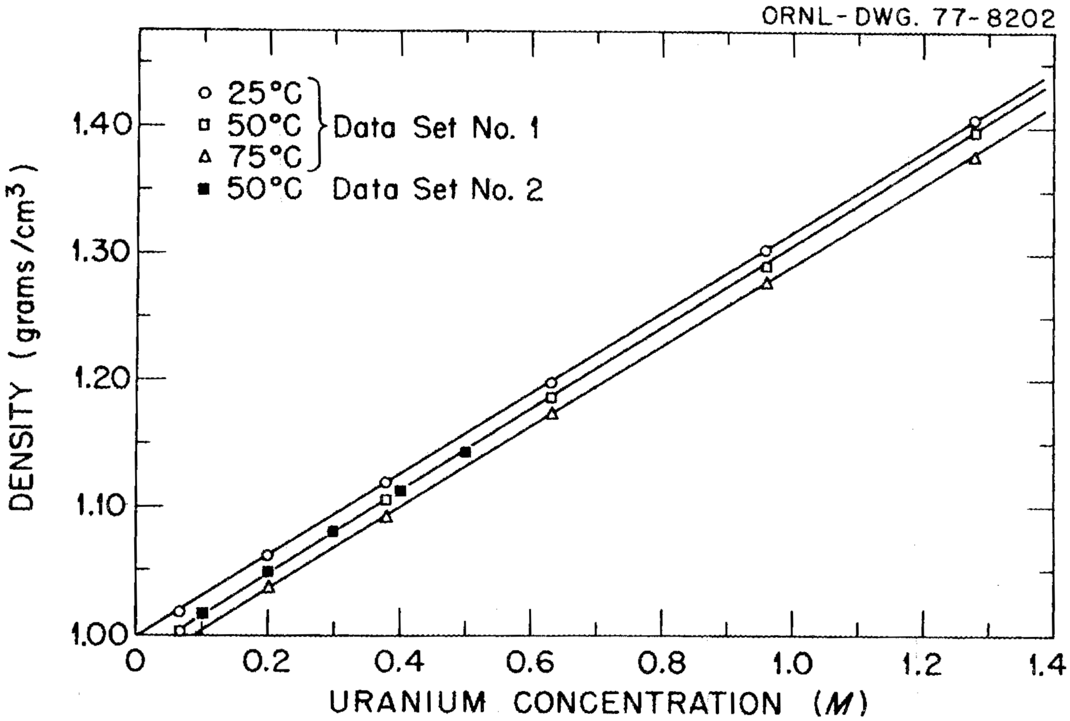

Fig. 11. Relationship between density and uranium concentration at various temperatures (NO_3/U mole ratio = 1.56).

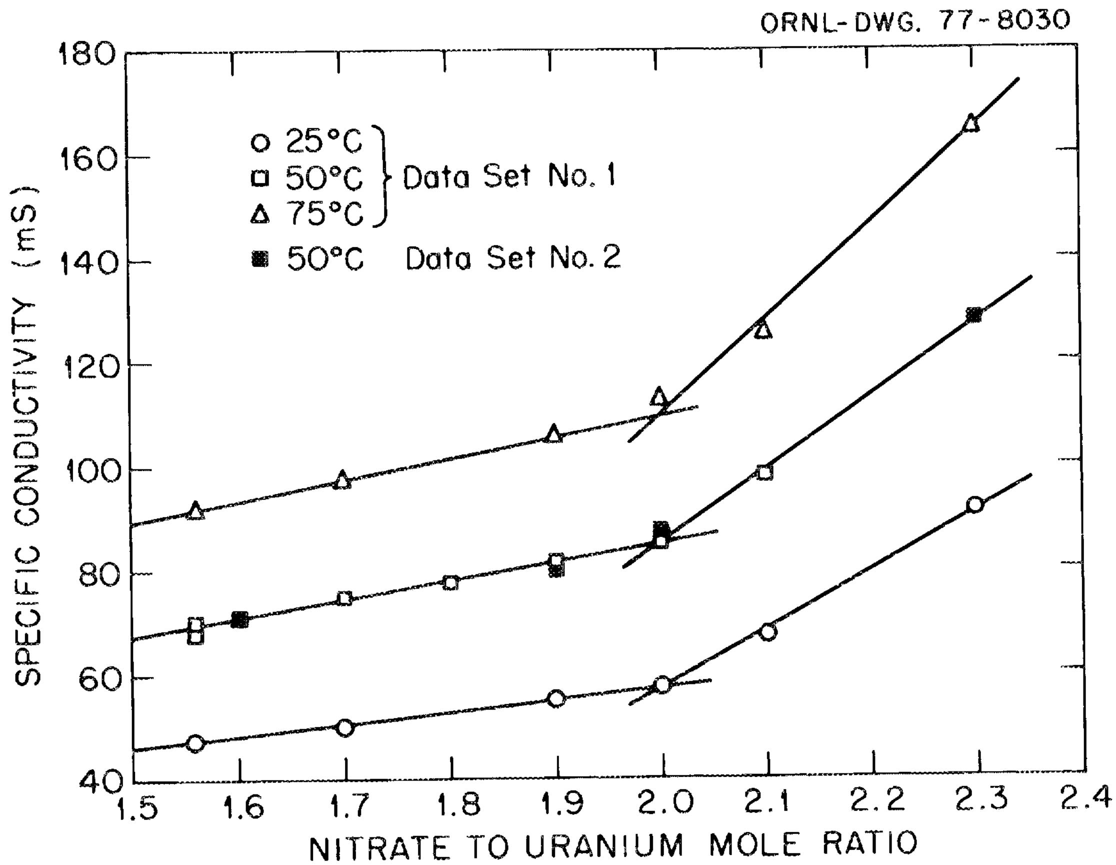

Fig. 12. Relationship between conductivity and the nitrate to uranium mole ratios of uranyl nitrate solutions at various temperatures (uranium concentration: 0.5*M*).

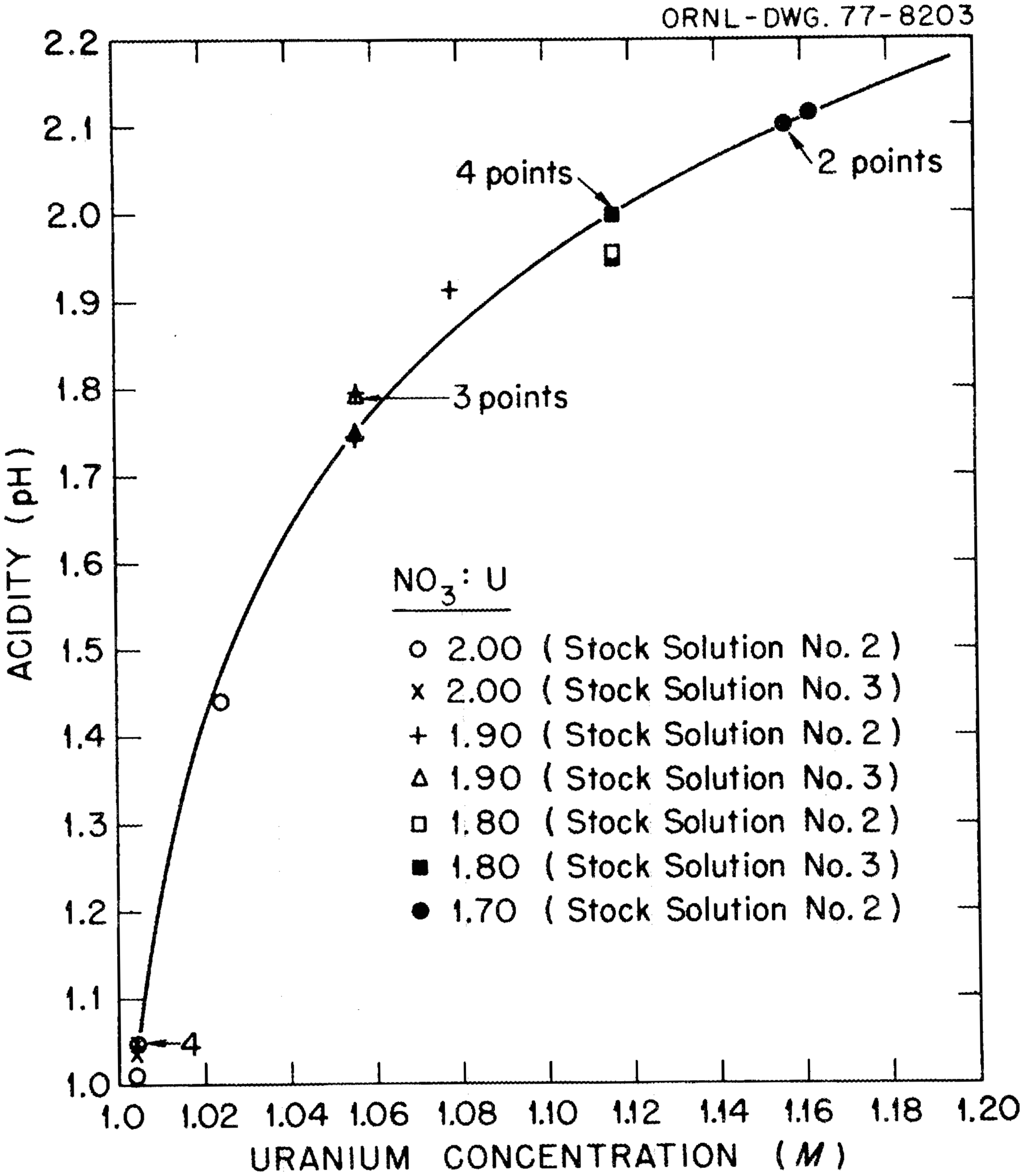

Fig. 13. Acidity of replicate acid deficient uranyl nitrate preparations at 30°C.

Table I. Density of water by extrapolation of uranyl nitrate density data at 25°C, 50°C, and 75°C (data set No. 1)*

Mole ratio NO_3: U	25°C		50°C		75°C	
	Slope	Intercept	Slope	Intercept	Slope	Intercept
1.56	0.3197	0.99651	0.3145	0.98921	0.3115	0.97528
1.70	0.3248	0.99873	0.3229	0.99584	0.3242	0.97587
1.90	0.3282	0.99714	0.3223	0.98962	0.3228	0.97486
2.00	0.3298	0.99875	0.3288	0.98834	0.3241	0.97547
2.10	0.3341	0.99664	0.3146	0.99074	0.3121	0.97699
Mean		0.9978		0.9908		0.9757
Standard deviation		±0.0014		±0.0030		±0.0008

*Theoretical density of water at 25°C = 0.9971; 50°C = 0.9881; 75°C = 0.9749.

Table II. Density of water by extrapolation of uranyl nitrate density data at 30°C, 40°C, and 50°C (data set No. 2)*

Mole ratio NO_3: U	30°C		40°C		50°C	
	Slope	Intercept	Slope	Intercept	Slope	Intercept
1.56	0.3140	0.99524	0.3128	0.99300	0.3109	0.98783
1.60	0.3085	0.99575	0.3053	0.99281	0.3040	0.98840
1.70	0.3137	0.99613	0.3126	0.99302	0.3119	0.98823
1.80	0.3176	0.99596	0.3169	0.99285	0.3155	0.98797
1.90	0.3197	0.99587	0.3193	0.99225	0.3177	0.98781
2.00	0.3222	0.99590	0.3229	0.99211	0.3215	0.98791
Mean		0.9958		0.9927		0.9880
Standard deviation		±0.0003		±0.0004		±0.0002

*Theoretical density of water at 30°C = 0.9957; 40°C = 0.9922; 50°C = 0.9881.

Table III. Acidity of replicate acid-deficient uranyl nitrate preparations at various temperatures (stock solution No. 2)

		Acidity, pH		
NO_3/U	(U),M	30°C	40°C	50°C
2.00	1.004	1.012	1.004	1.026
	1.004	1.042	1.043	1.080
	1.024	1.441	1.361	1.300
1.90	1.056	1.740	1.682	1.631
	1.056	1.784	1.710	1.697
	1.078	1.913	1.808	1.705
1.80	1.116	1.950	1.880	1.830
	1.116	2.003	1.925	1.865
	1.114	2.000	1.882	1.832
1.70	1.162	2.116	2.020	1.960
	1.156	2.105	2.022	1.966
	1.156	2.100	2.030	1.969

Table IV. Acidity of replicate acid-deficient uranyl nitrate preparations at 30°C (stock solution No. 3)

NO_3/U	(U),M	Acidity, pH
2.00	1.004	1.039
	1.004	1.040
	1.004	1.049
	1.004	1.049
	1.004	1.050
		1.045 ± 0.005
1.90	1.056	1.750
	1.056	1.790
	1.056	1.788
	1.056	1.793
		1.780 ± 0.018
1.80	1.116	1.955
	1.116	1.957
	1.116	2.000
	1.116	2.000
		1.978 ± 0.022

Additional tests were made on the stability of dilute uranyl nitrate solutions by measuring the pH of dilutions made from stock solution No. 2 on successive days at 30°C. The stability of these dilute solutions is quite good, varying about 0.02 pH units in 24 hours, as shown in Table V.

Table V. Acid stability of dilute acid-deficient uranyl nitrate solutions at 30°C

		Acidity, pH		
NO_3/U	(U),M	a*	b*	Deviation, pH units
2.00	0.100	2.343	2.276	0.067
	0.300	1.889	1.868	0.021
	0.500	1.616	1.590	0.026
1.90	0.100	2.585	2.611	0.026
	0.300	2.272	2.292	0.020
	0.500	2.090	2.103	0.013
1.70	0.100	2.860	2.870	0.010
	0.300	2.600	2.591	0.009
	0.500	2.404	2.416	0.012
1.56	0.100	2.978	2.956	0.022
	0.300	2.716	2.700	0.016
	0.500	2.543	2.532	0.011

*a = initial pH measurement; b = pH measurement made 24 hours later.

To further establish the reliability in preparing these solutions, the deviation of the pH measurement between two uranyl nitrate stock solutions and between dilutions made from a given stock solution is shown in Tables VI and VII.

USE OF EXPERIMENTAL DATA

The experimental data given in Appendices I and II can be used to derive an expression to compute the uranium and nitrate concentrations of uranyl nitrate solutions, given the density, conductivity, pH, and temperature. Since resin loading is most efficient in the acid-deficient region, only data in the range of 1.56 to 2.00 NO_3/U ratio will be considered. This limited range with one set of constants gives the best fit of the

Table VI. Acidity of two different uranyl nitrate stock solutions at 0.2 M uranium and 50°C

Ratio NO_3/U	Acidity, pH Stock I	Stock II	Deviation, pH units
2.00	2.260	2.066	0.194
1.90	2.520	2.410	0.110
1.70	2.810	2.700	0.110
1.56	2.940	2.810	0.130

Table VII. Acidity of two different uranyl nitrate dilutions from stock solution II at 0.2 M uranium and 30°C

Ratio NO_3/U	Acidity, pH Stock II Dilution I	Stock II Dilution II	Deviation, pH units
2.00	2.124	2.104	0.020
1.90	2.596	2.440	0.156
1.80	2.803	-	-
1.70	2.895	2.720	0.175
1.60	3.004	-	-
1.56	3.008	2.82	0.188

data. A second set of constants could be obtained for the region of 2.00 to 2.30 NO_3/U if desired.

The density and conductivity data set No. 2 were fitted to the following equations. The fit of the data is given in Table VIII using the values shown.

$$\text{Density} = \text{DENW} + A(U) + B(NO_3), \text{ and} \tag{1}$$

$$\text{Conductivity} = \frac{A_3 + B_3(NO_3)}{C_3 + U}, \tag{2}$$

where

$\text{DENW} = 0.992247 - 0.0003806\ \text{DT} - 0.00000375(\text{DT})^2$ (density of pure water),

DT = temperature -40,

Temp = temperature of the test solution, °C,

$A = 2.65684E-01$,

U = uranium concentration of test solution, molar,

$B = 2.82071E-02$,

NO_3 = nitrate concentration of test solution, molar,

$A_3 = A_1 + (A_2)(DT)$,

$A_1 = 7.56708E-01$,

$A_2 = 3.52224E-02$,

$B_3 = B_1 + (B_2)(DT)$,

$B_1 = 1.13629E+02$,

$B_2 = 1.50419$,

$C_3 = C_1 + (C_2)(DT)$,

$C_1 = 1.02522$,

$C_2 = -2.21409E-03$.

Equations 1 and 2 were then solved for the uranium and nitrate concentrations, and the following expressions were obtained:

$$U = B_3(D-B)(C_3)(\text{Prod})(\text{Divd}), \text{ and} \tag{3}$$

$$NO_3 = (A)(C_3 + D)(\text{Prod})(\text{Divd}), \tag{4}$$

where

D = DENS - DENW,

Dens = density of test solution, grams,

Prod = cond - A_3,

Table VIII. Prediction of the density and conductivity of uranyl nitrate solutions from known uranium and nitrate values at 30°C, 40°C, and 50°C

Mole ratio NO_3/U	Values at 30°C							
	Experimental conc, molar		Density, grams/cc			Conductivity, millisiemens		
	Uranium	Nitrate	Observed	Calc	Error, %	Observed	Calc	Error, %
1.56	0.100	0.156	1.0266	1.0266	0.0	12.2	13.8	13.2
	0.200	0.312	1.0579	1.0576	0.0	24.0	25.1	4.4
	0.300	0.468	1.0898	1.0886	-0.1	34.0	34.6	1.9
	0.400	0.624	1.1207	1.1196	-0.1	43.0	42.9	-0.2
	0.500	0.780	1.1522	1.1505	-0.1	50.0	50.1	0.2
1.60	0.100	0.160	1.0266	1.0268	0.0	14.4	14.2	-1.7
	0.200	0.320	1.0575	1.0578	0.0	25.8	25.7	-0.4
	0.301	0.480	1.0885	1.0892	0.1	36.0	35.5	-1.4
	0.400	0.640	1.1192	1.1200	0.1	44.7	44.0	-1.6
	0.500	0.800	1.1493	1.1511	0.2	52.5	51.4	-2.1
1.70	0.100	0.170	1.0275	1.0270	0.0	15.4	15.0	-2.5
	0.200	0.340	1.0588	1.0584	0.0	27.4	27.3	-0.4
	0.300	0.510	1.0902	1.0898	0.0	37.9	37.7	-0.5
	0.400	0.680	1.1219	1.1211	-0.1	47.1	46.7	-0.8
	0.500	0.850	1.1528	1.1525	0.0	55.0	54.6	-0.8
1.80	0.100	0.180	1.0278	1.0273	0.0	15.9	15.9	-0.2
	0.200	0.360	1.0594	1.0590	0.0	28.5	28.9	1.3
	0.300	0.540	1.0912	1.0906	-0.1	39.4	39.9	1.3
	0.400	0.720	1.1230	1.1223	-0.1	49.0	49.4	0.9
	0.500	0.900	1.1548	1.1539	-0.1	57.0	57.7	1.3
1.90	0.100	0.190	1.0279	1.0276	0.0	16.8	16.7	-0.4
	0.200	0.380	1.0599	1.0595	0.0	30.2	30.4	0.8
	0.300	0.570	1.0915	1.0915	0.0	41.5	42.1	1.5
	0.400	0.760	1.1238	1.1234	0.0	51.5	52.2	1.3
	0.500	0.950	1.1558	1.1553	0.0	60.0	60.9	1.6
2.00	0.101	0.202	1.0283	1.0282	0.0	18.5	17.7	-4.1
	0.200	0.400	1.0605	1.0601	0.0	32.7	32.0	-2.1
	0.301	0.602	1.0930	1.0926	0.0	45.0	44.4	-1.3
	0.401	0.802	1.1250	1.1249	0.0	55.7	55.0	-1.3
	0.500	1.000	1.1570	1.1567	0.0	64.5	64.1	-0.6

Table VIII (continued)

Mole ratio NO_3/U	Values at 40°C							
	Experimental conc, molar		Density, grams/cc			Conductivity, millisiemens		
	Uranium	Nitrate	Observed	Calc	Error, %	Observed	Calc	Error, %
1.56	0.100	0.156	1.0242	1.0232	-0.1	16.2	16.5	1.9
	0.200	0.312	1.0554	1.0542	-0.1	29.0	29.7	2.4
	0.300	0.468	1.0873	1.0852	-0.2	40.5	40.9	0.9
	0.400	0.624	1.1180	1.1161	-0.2	50.7	50.5	-0.4
	0.500	0.780	1.1493	1.1471	-0.2	58.5	58.9	0.6
1.60	0.100	0.160	1.0233	1.0233	0.0	17.2	16.9	-1.7
	0.200	0.320	1.0542	1.0544	0.0	30.7	30.4	-0.9
	0.301	0.480	1.0849	1.0853	0.1	42.5	41.9	-1.5
	0.400	0.640	1.1157	1.1166	0.1	52.5	51.8	-1.4
	0.500	0.800	1.1457	1.1477	0.2	61.5	60.4	-1.9
1.70	0.100	0.170	1.0242	1.0236	-0.1	18.2	17.9	-1.5
	0.200	0.340	1.0557	1.0550	-0.1	32.5	32.3	-1.0
	0.300	0.510	1.0867	1.0863	0.0	44.8	44.5	-0.7
	0.400	0.680	1.1181	1.1177	0.0	55.5	55.0	-1.0
	0.500	0.850	1.1493	1.1491	0.0	64.5	64.1	-0.6
1.80	0.100	0.180	1.0244	1.0239	0.0	19.1	18.9	-0.9
	0.200	0.360	1.0564	1.0555	-0.1	33.8	34.1	1.0
	0.300	0.540	1.0879	1.0872	-0.1	46.7	47.1	0.8
	0.400	0.720	1.1197	1.1188	-0.1	57.5	58.2	1.1
	0.500	0.900	1.1512	1.1505	-0.1	67.0	67.8	1.2
1.90	0.100	0.190	1.0246	1.0242	0.0	20.2	19.9	-1.3
	0.200	0.380	1.0556	1.0561	0.0	35.4	36.0	1.7
	0.300	0.570	1.0879	1.0880	0.0	48.8	49.6	1.7
	0.400	0.760	1.1201	1.1200	0.0	60.5	61.3	1.4
	0.500	0.950	1.1520	1.1519	0.0	70.0	71.5	2.2
2.00	0.101	0.202	1.0244	1.0248	0.0	21.8	21.1	-3.0
	0.200	0.400	1.0570	1.0567	0.0	38.5	37.9	-1.9
	0.301	0.602	1.0894	1.0892	0.0	53.0	52.3	-1.3
	0.401	0.802	1.1218	1.1214	0.0	65.0	64.7	-0.5
	0.500	1.000	1.1533	1.1533	0.0	76.0	75.3	-1.0

Table VIII (continued)

Mole ratio NO_3/U	Values at 50°C							
	Experimental conc, molar		Density, grams/cc			Conductivity, millisiemens		
	Uranium	Nitrate	Observed	Calc	Error, %	Observed	Calc	Error, %
1.56	0.100	0.156	1.0188	1.0190	0.0	18.0	19.3	7.3
	0.200	0.312	1.0499	1.0500	0.0	33.8	34.5	2.0
	0.300	0.468	1.0815	1.0810	0.0	47.3	47.3	0.0
	0.400	0.624	1.1122	1.1119	0.0	58.0	58.3	0.6
	0.500	0.780	1.1431	1.1429	0.0	68.0	67.9	-0.2
1.60	0.100	0.160	1.0186	1.0191	0.1	19.8	19.8	-0.1
	0.200	0.320	1.0493	1.0502	0.1	35.2	35.3	0.4
	0.301	0.480	1.0800	1.0816	0.1	49.3	48.5	-1.7
	0.400	0.640	1.1103	1.1124	0.2	60.5	59.8	-1.2
	0.500	0.800	1.1401	1.1435	0.3	71.0	69.6	-2.0
1.70	0.100	0.170	1.0195	1.0194	0.0	21.3	20.9	-1.7
	0.200	0.340	1.0506	1.0508	0.0	37.7	37.5	-0.6
	0.300	0.510	1.0817	1.0822	0.0	51.0	51.5	0.9
	0.400	0.680	1.1129	1.1135	0.1	64.0	63.5	-0.8
	0.500	0.850	1.1443	1.1449	0.1	74.5	73.9	-0.8
1.80	0.100	0.180	1.0196	1.0197	0.0	22.2	22.1	-0.4
	0.200	0.360	1.0511	1.0514	0.0	39.2	39.6	1.0
	0.300	0.540	1.0826	1.0830	0.0	54.0	54.4	0.8
	0.400	0.720	1.1138	1.1146	0.1	66.5	67.1	1.0
	0.500	0.900	1.1460	1.1463	0.0	77.0	78.2	1.5
1.90	0.100	0.190	1.0199	1.0200	0.0	22.7	23.3	2.5
	0.200	0.380	1.0509	1.0519	0.1	41.4	41.8	0.8
	0.300	0.570	1.0831	1.0838	0.1	56.5	57.4	1.6
	0.400	0.760	1.1150	1.1158	0.1	70.0	70.8	1.2
	0.500	0.950	1.1467	1.1477	0.1	81.0	82.4	1.8
2.00	0.101	0.202	1.0199	1.0206	0.1	25.5	24.7	-3.3
	0.200	0.400	1.0528	1.0525	0.0	44.8	43.9	-2.0
	0.301	0.601	1.0845	1.0850	0.0	61.5	60.4	-1.8
	0.401	0.801	1.1174	1.1172	0.0	75.3	74.5	-1.0
	0.500	1.001	1.1482	1.1491	0.1	87.5	86.8	-0.8

Cond = specific conductance of test solution, millisiemens,

Divd = $[(B)(Prod) + (A)(B_3)]^{-1}$.

Uranium and nitrate concentrations were computed from measured values of conductivity and density and are compared in Table IX. In the case of acidity (pH) and density, the least-squares fit of the experimental data from set No. 2 gave the following expressions for uranium and nitrate:

$$U = \frac{(A + B)(DEN + C)}{(pH - D)^2} \text{, and} \tag{5}$$

$$NO_3 = P + (Q)(DENW) + (R)pH + (5)(pH)^2 \text{,} \tag{6}$$

where

$A = A_1 + (A_2)(DT)$,

A_1 = 1.7587E - 02,

A_2 = 1.1642E - 04,

DT = temperature -40,

Temp = temperature of test solution, °C,

$B = B_1 + (B_2)(DT)$,

B_1 = 3.2219,

B_2 = 1.6471E - 03,

DEN = DENS - DENW,

DENS = density of test solution, g/cc,

DENW = $0.992247 - 0.0003806(DT) - 0.00000375(DT)^2$,

$C = C_1 + (C_2)(DT)$,

C_1 = -4.0335E - 01,

C_2 = 1.7881E - 02,

pH = acidity of test solution,

$D = D_1 + (D_2)(DT)$,

D_1 = -1.6537,

D_2 = 7.878E - 02,

$P = P_1 + (P_2)(DT)$,

P_1 = 4.868E - 01,

P_2 = 5.8927E - 03,

$Q = Q_1 + (Q_2)(DT)$,

Q_1 = 5.1134,

Q_2 = -2.4523E - 03,

Table IX. Predictions of uranium and nitrate concentrations in uranyl nitrate solutions from measured densities and conductivities at 30°C, 40°C, and 50°C

			Values at 30°C					
			Nitrate conc, molar			Uranium conc, molar		
Mole ratio NO_3/U	Meas cond (mS)	Meas density (gm/cc)	Observed	Calc	Error (%)	Observed	Calc	Error (%)
1.56	12.2	1.0266	0.156	0.137	-11.9	0.100	0.102	1.8
	24.0	1.0579	0.312	0.299	-4.1	0.200	0.202	1.2
	34.0	1.0898	0.468	0.461	-1.5	0.300	0.305	1.8
	43.0	1.1207	0.624	0.627	0.5	0.400	0.404	1.0
	50.0	1.1522	0.780	0.782	0.2	0.500	0.506	1.2
1.60	14.4	1.0266	0.160	0.163	1.7	0.100	0.099	-0.9
	25.8	1.0575	0.320	0.321	0.3	0.200	0.199	-0.7
	36.0	1.0685	0.480	0.486	1.2	0.301	0.298	-1.1
	44.7	1.1192	0.640	0.649	1.3	0.400	0.396	-1.0
	52.5	1.1493	0.800	0.813	1.7	0.500	0.492	-1.6
1.70	15.4	1.0275	0.170	0.175	2.8	0.100	0.101	1.2
	27.4	1.0588	0.340	0.342	0.6	0.200	0.201	0.6
	37.9	1.0902	0.510	0.513	0.6	0.300	0.301	0.4
	47.1	1.1219	0.680	0.687	1.0	0.400	0.402	0.5
	55.0	1.1528	0.850	0.857	0.8	0.500	0.500	0.1
1.80	15.9	1.0276	0.180	0.181	0.3	0.100	0.102	1.7
	28.5	1.0594	0.360	0.356	-1.1	0.200	0.202	1.0
	39.4	1.0912	0.540	0.534	-1.1	0.300	0.303	0.9
	49.0	1.1230	0.720	0.715	-0.7	0.400	0.403	0.8
	57.0	1.1548	0.900	0.891	-1.0	0.500	0.504	0.9
1.90	16.8	1.0279	0.190	0.191	0.5	0.100	0.101	1.0
	30.2	1.0599	0.380	0.377	-0.7	0.200	0.202	0.8
	41.5	1.0915	0.570	0.562	-1.4	0.300	0.301	0.3
	51.5	1.1238	0.760	0.751	-1.1	0.400	0.402	0.6
	60.0	1.1558	0.950	0.937	-1.3	0.500	0.503	0.6
2.00	18.5	1.0283	0.202	0.211	4.3	0.101	0.100	-0.6
	32.7	1.0605	0.400	0.409	2.2	0.200	0.201	0.3
	45.0	1.0930	0.602	0.610	1.4	0.301	0.302	0.2
	55.7	1.1250	0.802	0.812	1.3	0.401	0.401	-0.1
	64.5	1.1570	1.000	1.006	0.6	0.500	0.500	0.1

Table IX (continued)

Mole ratio NO_3/U	Meas cond (mS)	Meas density (gm/cc)	Values at 40°C — Nitrate conc, molar: Observed	Calc	Error (%)	Uranium conc, molar: Observed	Calc	Error (%)
1.56	16.2	1.0242	0.156	0.153	-1.6	0.100	0.104	4.0
	29.0	1.0554	0.312	0.306	-2.0	0.200	0.205	2.6
	40.5	1.0873	0.468	0.466	-0.3	0.300	0.308	2.8
	50.7	1.1180	0.624	0.629	0.8	0.400	0.407	1.6
	58.5	1.1493	0.780	0.779	-0.1	0.500	0.508	1.7
1.60	17.2	1.0233	0.160	0.163	1.7	0.100	0.100	-0.4
	30.7	1.0542	0.320	0.323	0.8	0.200	0.199	-0.5
	42.5	1.0849	0.480	0.486	1.2	0.301	0.297	-1.3
	52.5	1.1157	0.640	0.647	1.1	0.400	0.396	-1.0
	61.5	1.1457	0.800	0.811	1.3	0.500	0.491	-1.7
1.70	18.2	1.0242	0.170	0.173	1.8	0.100	0.102	1.9
	32.6	1.0557	0.340	0.344	1.2	0.200	0.202	1.2
	44.8	1.0867	0.510	0.514	0.8	0.300	0.301	0.3
	55.5	1.1181	0.680	0.687	1.0	0.400	0.401	0.2
	64.5	1.1493	0.850	0.856	0.7	0.500	0.500	0.1
1.80	19.1	1.0244	0.180	0.182	1.1	0.100	0.102	1.7
	33.8	1.0564	0.360	0.357	-0.7	0.200	0.204	1.8
	46.7	1.0879	0.540	0.537	-0.5	0.300	0.303	1.0
	57.5	1.1197	0.720	0.714	-0.9	0.400	0.404	1.0
	67.0	1.1512	0.900	0.891	-1.0	0.500	0.504	0.7
1.90	20.2	1.0246	0.190	0.193	1.5	0.100	0.101	1.3
	35.4	1.0556	0.380	0.373	-1.8	0.200	0.199	-0.6
	48.8	1.0879	0.570	0.561	-1.7	0.300	0.301	0.2
	60.5	1.1201	0.760	0.750	-1.3	0.400	0.402	0.4
	70.0	1.1520	0.950	0.931	-2.0	0.500	0.502	0.5
2.00	21.8	1.0244	0.202	0.208	3.1	0.101	0.099	-2.1
	38.6	1.0570	0.400	0.408	2.0	0.200	0.200	0.2
	53.0	1.0894	0.602	0.610	1.3	0.301	0.301	-0.0
	65.0	1.1218	0.802	0.807	0.6	0.401	0.402	0.2
	76.0	1.1533	1.000	1.009	0.9	0.500	0.499	-0.2

Table IX (continued)

Mole ratio NO_3/U	Meas cond (mS)	Meas density (gm/cc)	Values at 50°C Nitrate conc, molar Observed	Calc	Error (%)	Uranium conc, molar Observed	Calc	Error (%)
1.56	18.0	1.0188	0.156	0.145	-7.2	0.100	0.100	0.3
	33.8	1.0499	0.312	0.306	-2.0	0.200	0.200	0.1
	47.3	1.0815	0.468	0.468	0.1	0.300	0.302	0.6
	58.0	1.1122	0.624	0.621	-0.5	0.400	0.401	0.3
	68.0	1.1431	0.780	0.782	0.2	0.500	0.501	0.1
1.60	19.8	1.0166	0.160	0.160	-0.0	0.100	0.098	-2.1
	35.2	1.0493	0.320	0.318	-0.7	0.200	0.197	-1.6
	49.3	1.0800	0.480	0.486	1.2	0.301	0.294	-2.2
	60.5	1.1103	0.640	0.644	0.6	0.400	0.392	-2.1
	71.0	1.1401	0.800	0.809	1.1	0.500	0.486	-2.7
1.70	21.3	1.0195	0.170	0.173	1.8	0.100	0.100	-0.1
	37.7	1.0506	0.340	0.342	0.5	0.200	0.199	-0.5
	51.0	1.0817	0.510	0.505	-1.0	0.300	0.299	-0.4
	64.0	1.1129	0.680	0.684	0.6	0.400	0.397	-0.7
	74.5	1.1443	0.850	0.856	0.7	0.500	0.497	-0.6
1.80	22.2	1.0196	0.180	0.181	0.4	0.100	0.100	-0.5
	39.2	1.0511	0.360	0.356	-1.1	0.200	0.199	-0.3
	54.0	1.0826	0.540	0.535	-0.9	0.300	0.299	-0.3
	66.5	1.1138	0.720	0.712	-1.1	0.400	0.398	-0.6
	77.0	1.1460	0.900	0.887	-1.5	0.500	0.500	0.1
1.90	22.7	1.0199	0.190	0.185	-2.6	0.100	0.100	0.2
	41.4	1.0509	0.380	0.376	-1.1	0.200	0.197	-1.7
	56.5	1.0831	0.570	0.560	-1.7	0.300	0.298	-0.6
	70.0	1.1150	0.760	0.750	-1.3	0.400	0.398	-0.5
	81.0	1.1467	0.950	0.932	-1.9	0.500	0.498	-0.4
2.00	25.5	1.0199	0.202	0.209	3.3	0.101	0.098	-3.3
	44.8	1.0528	0.400	0.409	2.2	0.200	0.200	0.1
	61.5	1.0845	0.601	0.611	1.6	0.301	0.298	-1.0
	75.3	1.1174	0.801	0.810	1.1	0.401	0.401	-0.0
	87.5	1.1482	1.001	1.006	0.5	0.500	0.496	-0.8

$$R = R_1 + (R_2)(DT),$$
$$R_1 = -2.3192E-01,$$
$$R_2 = -4.0891E-03,$$
$$S = S_1 + (S_2)(DT),$$
$$S_1 = 2.2317E-02,$$
$$S_2 = 6.0217E-04.$$

The same types of equations were derived from experimental data set No. 1. These equations are not shown because, due to the availability of more data points, a better fit of the data was obtained using data set No. 2. Also, the second data set is more useful because it more closely simulates or approximates process conditions.

The use of the above equations is cumbersome and time-consuming without access to a computer. For that reason, two short programs for use in the PDP-10 computer have been written to calculate the uranium and nitrate concentrations of uranyl nitrate solutions given solution parameters of density, conductivity, acidity (pH), and temperature. The program given in Appendix III is for the case of conductivity and density. It can be used for any temperature in the range of 25°C to 75°C, for any density up to 1.4 grams/cc, and for specific conductances up to 160 millisiemens. Appendix IV gives the program for acidity, pH, and density. It can be used for any temperature in the range of 25°C to 75°C, for any density up to 1.4 grams/cc, and for acidities ranging from 0 to 3.5.

In most cases the errors between the calculated and observed concentrations of uranium and nitrate are less than 2% in Table IX, where the concentrations were calculated from known densities and conductivities. The error shown in Table X, where the concentrations are calculated from the measured values for density and pH, is in general less than 2% for uranium and 5% for the nitrate.

A computer calculation was made using the density and pH values given in Table X at 30°C to determine the effects of errors in the pH measurement on the computed uranium and nitrate concentration. In this calculation, the pH was allowed to vary ±0.5 pH units. The data from this calculation, shown in Table XI, indicate that a 1.0% error in pH measurement will cause a 0.2% error in the calculated uranium concentration and a 1.8% error in the nitrate concentration.

Table X. Prediction of uranium and nitrate concentration in uranyl nitrate solutions from measured densities and pH at 30°C, 40°C, and 50°C

	Values at 30°C							
			Uranium conc, molar			Nitrate conc, molar		
Mole ratio NO_3/U	Density (gm/cc)	pH	Observed	Calc	Error (%)	Observed	Calc	Error (%)
1.56	1.0266	3.240	0.100	0.100	-0.2	0.156	0.139	-11.0
	1.0579	3.088	0.200	0.199	-0.4	0.312	0.313	0.3
	1.0898	2.970	0.300	0.301	0.2	0.468	0.488	4.2
	1.1207	2.880	0.400	0.399	-0.3	0.624	0.655	5.0
	1.1522	2.780	0.500	0.499	-0.2	0.780	0.827	6.0
1.60	1.0266	3.163	0.100	0.099	-0.7	0.160	0.146	-9.0
	1.0575	3.004	0.200	0.197	-1.4	0.320	0.319	-0.4
	1.0885	2.888	0.301	0.296	-1.7	0.480	0.489	1.9
	1.1192	2.800	0.400	0.394	-1.6	0.640	0.655	2.4
	1.1493	2.707	0.500	0.489	-2.2	0.800	0.819	2.4
1.70	1.0275	3.060	0.100	0.102	1.5	0.170	0.159	-6.2
	1.0588	2.895	0.200	0.201	0.3	0.340	0.336	-1.2
	1.0902	2.770	0.300	0.300	0.1	0.510	0.509	-0.1
	1.1219	2.680	0.400	0.401	0.3	0.680	0.682	0.2
	1.1528	2.600	0.500	0.499	-0.1	0.850	0.849	-0.2
1.80	1.0278	2.964	0.100	0.102	1.8	0.180	0.170	-5.6
	1.0594	2.803	0.200	0.202	0.9	0.360	0.348	-3.4
	1.0912	2.668	0.300	0.303	0.9	0.540	0.525	-2.8
	1.1230	2.562	0.400	0.404	0.9	0.720	0.700	-2.8
	1.1548	2.474	0.500	0.505	0.9	0.900	0.873	-3.0
1.90	1.0279	2.780	0.100	0.101	0.7	0.190	0.188	-0.9
	1.0599	2.596	0.200	0.202	0.8	0.380	0.372	-2.2
	1.0915	2.460	0.300	0.302	0.6	0.570	0.549	-3.7
	1.1238	2.341	0.400	0.404	1.0	0.760	0.728	-4.2
	1.1558	2.250	0.500	0.506	1.1	0.950	0.903	-4.9
2.00	1.0283	2.381	0.101	0.098	-2.7	0.202	0.233	15.4
	1.0605	2.124	0.200	0.199	-0.7	0.400	0.429	7.2
	1.0930	1.917	0.301	0.300	-0.3	0.602	0.622	3.3
	1.1250	1.766	0.401	0.400	-0.1	0.802	0.806	0.5
	1.1570	1.620	0.500	0.501	0.1	1.000	0.990	-1.0

Table X (continued)

Mole ratio NO_3/U	Density (gm/cc)	pH	Values at 40°C: Uranium conc, molar: Observed	Calc	Error (%)	Nitrate conc, molar: Observed	Calc	Error (%)
1.56	1.0242	3.120	0.100	0.103	2.8	0.156	0.144	-7.8
	1.0554	2.980	0.200	0.202	1.1	0.312	0.317	1.5
	1.0873	2.860	0.300	0.304	1.3	0.468	0.492	5.2
	1.1180	2.780	0.400	0.402	0.6	0.624	0.658	5.4
	1.1493	2.700	0.500	0.502	0.5	0.780	0.826	6.0
1.60	1.0233	3.065	0.100	0.100	-0.5	0.160	0.144	-9.7
	1.0542	2.897	0.200	0.198	-1.1	0.320	0.319	-0.3
	1.0849	2.783	0.301	0.296	-1.8	0.480	0.488	1.7
	1.1157	2.694	0.400	0.394	-1.5	0.640	0.655	2.4
	1.1457	2.604	0.500	0.490	-2.1	0.800	0.819	2.4
1.70	1.0242	2.950	0.100	0.102	1.5	0.170	0.160	-5.7
	1.0557	2.785	0.200	0.202	0.8	0.340	0.338	-0.4
	1.0867	2.656	0.300	0.300	0.1	0.510	0.511	0.2
	1.1181	2.567	0.400	0.400	0.1	0.680	0.682	0.3
	1.1493	2.483	0.500	0.500	0.0	0.850	0.852	0.2
1.80	1.0244	2.857	0.100	0.101	1.4	0.180	0.171	-5.1
	1.0564	2.689	0.200	0.203	1.4	0.360	0.353	-2.1
	1.0879	2.552	0.300	0.303	1.0	0.540	0.529	-2.0
	1.1197	2.447	0.400	0.404	1.1	0.720	0.705	-2.1
	1.1512	2.363	0.500	0.505	0.9	0.900	0.876	-2.6
1.90	1.0246	2.677	0.100	0.100	0.3	0.190	0.191	0.7
	1.0556	2.491	0.200	0.198	-0.9	0.380	0.372	-2.2
	1.0879	2.362	0.300	0.301	0.3	0.570	0.553	-3.0
	1.1201	2.255	0.400	0.403	0.8	0.760	0.731	-3.8
	1.1520	2.157	0.500	0.505	0.9	0.950	0.907	-4.5
2.00	1.0244	2.335	0.101	0.096	-5.1	0.202	0.231	14.5
	1.0570	2.071	0.200	0.197	-1.4	0.400	0.433	8.3
	1.0894	1.875	0.301	0.298	-0.9	0.602	0.627	4.2
	1.1218	1.725	0.401	0.400	-0.3	0.802	0.816	1.7
	1.1533	1.600	0.500	0.498	-0.3	1.000	0.996	-0.4

Table X (continued)

Mole ratio NO_3/U	Density (gm/cc)	pH	Values at 50°C: Uranium conc, molar: Observed	Calc	Error (%)	Nitrate conc, molar: Observed	Calc	Error (%)
1.56	1.0188	2.956	0.100	0.101	0.6	0.156	0.143	-8.1
	1.0499	2.810	0.200	0.200	0.0	0.312	0.318	1.8
	1.0815	2.700	0.300	0.301	0.4	0.468	0.491	5.0
	1.1122	2.620	0.400	0.400	-0.0	0.624	0.657	5.2
	1.1431	2.543	0.500	0.499	-0.2	0.780	0.824	5.7
1.60	1.0186	2.929	0.100	0.100	-0.3	0.160	0.145	-9.3
	1.0493	2.767	0.200	0.198	-1.1	0.320	0.319	-0.2
	1.0800	2.660	0.301	0.296	-1.6	0.480	0.488	1.9
	1.1103	2.568	0.400	0.393	-1.7	0.640	0.654	2.2
	1.1401	2.494	0.500	0.489	-2.2	0.800	0.815	1.9
1.70	1.0195	2.870	0.100	0.102	2.1	0.170	0.156	-8.1
	1.0506	2.700	0.200	0.201	0.6	0.340	0.334	-1.8
	1.0817	2.591	0.300	0.301	0.3	0.510	0.506	-0.9
	1.1129	2.497	0.400	0.401	0.2	0.680	0.677	-0.5
	1.1443	2.416	0.500	0.502	0.3	0.850	0.847	-0.3
1.80	1.0196	2.777	0.100	0.102	1.6	0.180	0.167	-7.1
	1.0511	2.582	0.200	0.202	0.8	0.360	0.351	-2.5
	1.0826	2.475	0.300	0.302	0.8	0.540	0.525	-2.7
	1.1138	2.390	0.400	0.402	0.6	0.720	0.695	-3.4
	1.1460	2.311	0.500	0.506	1.1	0.900	0.870	-3.3
1.90	1.0199	2.611	0.100	0.101	0.9	0.190	0.189	-0.7
	1.0509	2.410	0.200	0.199	-0.5	0.380	0.373	-1.9
	1.0831	2.272	0.300	0.301	0.5	0.570	0.556	-2.5
	1.1150	2.177	0.400	0.403	0.8	0.760	0.732	-3.7
	1.1467	2.103	0.500	0.505	0.9	0.950	0.905	-4.8
2.00	1.0199	2.343	0.101	0.098	-3.3	0.202	0.224	11.0
	1.0528	2.066	0.200	0.200	-0.0	0.400	0.433	8.1
	1.0845	1.889	0.301	0.299	-0.6	0.601	0.622	3.5
	1.1174	1.777	0.401	0.403	0.5	0.801	0.809	1.0
	1.1482	1.616	0.500	0.499	-0.3	1.001	0.994	-0.7

Table XI. The effect of pH error on computer uranium and nitrate concentrations in uranyl nitrate solutions from measured densities and pH at 30°C

Mole ratio NO_3/U	Density (gm/cc)	pH	Uranium conc (M) Exp value	Uranium conc (M) Calc	Calculated error, % Obs pH -0.5	Calculated error, % Observed pH	Calculated error, % Obs pH +0.5	Nitrate conc (M) Exp value	Nitrate conc (M) Calc	Calculated error, % Obs pH -0.5	Calculated error, % Observed pH	Calculated error, % Obs pH +0.5
1.56	1.0266	3.240	0.100	0.100	-3.8	-0.2	2.6	0.156	0.139	19.0	-11.0	-35.7
	1.0579	3.088	0.200	0.199	-2.4	-0.4	1.1	0.312	0.313	16.1	0.3	-12.8
	1.0898	2.970	0.300	0.301	-1.2	0.2	1.3	0.468	0.488	15.2	4.2	-4.9
	1.1207	2.880	0.400	0.399	-1.4	-0.3	0.6	0.624	0.655	13.4	5.0	-2.1
	1.1522	2.780	0.500	0.499	-1.1	-0.2	0.5	0.780	0.827	13.0	6.0	0.1
1.60	1.0266	3.163	0.100	0.099	-4.5	-0.7	2.2	0.160	0.146	21.0	-9.0	-33.9
	1.0575	3.004	0.200	0.197	-3.4	-1.4	0.2	0.320	0.319	15.4	-0.4	-13.7
	1.0885	2.888	0.301	0.296	-3.2	-1.7	-0.6	0.480	0.489	12.8	1.9	-7.4
	1.1192	2.800	0.400	0.394	-2.8	-1.5	-0.7	0.640	0.655	10.8	2.4	-4.7
	1.1493	2.707	0.500	0.489	-3.2	-2.2	-1.4	0.800	0.819	9.4	2.4	-3.5
1.70	1.0275	3.060	0.100	0.101	-2.5	1.5	4.6	0.170	0.159	23.0	-5.2	-30.7
	1.0588	2.895	0.200	0.201	-1.9	0.3	2.0	0.340	0.336	14.2	-1.2	-14.3
	1.0902	2.770	0.300	0.300	-1.5	0.1	1.3	0.510	0.509	10.6	-0.1	-9.2
	1.1219	2.680	0.400	0.401	-1.0	0.3	1.2	0.680	0.682	8.4	0.2	-6.8
	1.1528	2.600	0.500	0.499	-1.2	-0.1	0.7	0.850	0.849	6.6	-0.2	-5.9
1.80	1.0278	2.964	0.100	0.102	-2.5	1.8	5.0	0.180	0.170	22.9	-5.6	-29.6
	1.0594	2.803	0.200	0.202	-1.4	0.9	2.7	0.360	0.348	11.6	-3.4	-16.1
	1.0912	2.668	0.300	0.303	-0.8	0.9	2.1	0.540	0.525	7.6	-2.8	-11.7
	1.1230	2.562	0.400	0.404	-0.5	0.9	1.9	0.720	0.700	5.2	-2.8	-9.7
	1.1548	2.474	0.500	0.505	-0.2	0.9	1.8	0.900	0.873	3.5	-3.0	-8.7
1.90	1.0279	2.780	0.100	0.101	-4.1	0.7	4.3	0.190	0.198	27.7	-0.9	-25.2
	1.0599	2.596	0.200	0.202	-1.8	0.8	2.8	0.380	0.372	12.9	-2.2	-15.1
	1.0915	2.460	0.300	0.302	-1.4	0.6	2.0	0.570	0.549	6.7	-3.7	-12.7
	1.1238	2.341	0.400	0.404	-0.6	1.0	2.1	0.760	0.728	3.9	-4.2	-11.2
	1.1558	2.250	0.500	0.505	-0.2	1.1	2.1	0.950	0.903	1.7	-4.9	-10.7
2.00	1.0283	2.381	0.101	0.098	-8.8	-2.7	1.8	0.202	0.233	45.5	15.4	-10.7
	1.0605	2.124	0.200	0.199	-4.3	-0.7	1.9	0.400	0.429	23.4	7.2	-7.0
	1.0930	1.917	0.301	0.300	-3.1	-0.3	1.7	0.602	0.622	14.6	3.3	-6.7
	1.1250	1.766	0.401	0.400	-2.5	-0.1	1.5	0.802	0.806	9.3	0.5	-7.3
	1.1570	1.620	0.500	0.501	-2.0	0.1	1.6	1.000	0.990	6.3	-1.0	-7.5

SUMMARY

Conductivity, density, and acidity measurements were made on a series of uranyl nitrate solutions under a number of process conditions of temperature and acidity. It has been found from this study that the acidity and conductivity of the solutions were quite sensitive to the uranium and nitrate concentration, whereas the density is sensitive only to the uranium concentration.

The complex relationships among acidity, conductivity, temperature, density, and concentration were quantified in this study. Computer programs were written to quickly predict or calculate the uranium and nitrate concentrations where (a) the temperature, density, and conductivity or (b) the temperature, density, and pH are known. The use of these programs will allow precise process control to be exercised in the preparation of HTGR recycled fuel particles by the simple monitoring of the density, temperature, and either conductivity or pH of the process solution.

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APPENDIX I. Experimental data set No. 1

Mole ratio NO_3/U	Molarity		Density, grams/cc			Acidity, pH			Conductivity, millisiemens		
	Nitrate	Uranium	25°C	50°C	75°C	25°C	50°C	75°C	25°C	50°C	75°C
1.57	0.102	0.065	1.0174	1.0094	0.9954	3.474	3.203	3.000	9.7	13.2	18.0
	0.312	0.199	1.0602	1.0517	1.0371	3.220	2.953	2.752	22.8	34.4	46.2
	0.590	0.376	1.1168	1.1078	1.0930	3.024	2.760	2.582	37.9	57.0	75.0
	0.992	0.632	1.1985	1.1884	1.1722	2.810	2.576	2.380	54.3	80.0	105.5
	1.499	0.956	1.3013	1.2893	1.2725	2.614	2.374	2.170	67.5	98.7	130.0
	1.988	1.267	1.4044	1.3900	1.3725	2.413	2.187	1.988	74.5	109.0	143.0
1.70	0.091	0.053	1.0146	1.0066	0.9928	3.334	3.113	2.828	8.1	12.5	17.2
	0.299	0.176	1.0552	1.0467	1.0326	3.080	2.847	2.678	22.4	34.0	46.0
	0.590	0.347	1.1119	1.1159	1.0882	2.878	2.660	2.460	38.2	57.5	77.0
	0.989	0.582	1.1889	1.1956	1.1656	2.667	2.460	2.260	54.4	81.5	107.5
	1.484	0.872	1.2858	1.2757	1.2591	2.462	2.254	2.080	68.0	99.5	132.5
	1.988	1.169	1.3750	1.3675	1.3541	2.268	2.080	1.860	75.5	110.3	147.0
1.90	0.109	0.058	1.0157	1.0075	0.9940	3.058	2.832	2.556	9.6	14.6	20.0
	0.310	0.163	1.0505	1.0419	1.0281	2.806	2.578	2.320	23.4	35.2	47.7
	0.601	0.316	1.1011	1.0917	1.0770	2.600	2.382	2.100	39.8	59.0	79.0
	0.986	0.519	1.1680	1.1578	1.1425	2.390	2.170	1.960	56.2	83.0	109.0
	1.501	0.790	1.2568	1.2456	1.2292	2.164	1.960	1.740	71.5	105.0	137.5
	1.988	1.046	1.3398	1.3253	1.3149	1.974	1.788	1.550	80.0	117.0	153.0
2.00	0.103	0.052	1.0145	1.0048	0.9918	2.858	2.660	2.446	9.2	14.2	19.5
	0.286	0.143	1.0482	1.0359	1.0222	2.546	2.363	2.155	22.2	33.6	45.0
	0.603	0.301	1.0973	1.0875	1.0731	2.282	2.107	1.903	40.2	60.0	81.5
	1.012	0.506	1.1652	1.1547	1.1396	2.034	1.865	1.646	58.0	86.0	114.5
	1.546	0.773	1.2540	1.2426	1.2263	1.778	1.624	1.396	73.9	109.0	144.0
	1.988	0.994	1.3265	1.3150	1.2973	1.586	1.414	1.235	82.0	120.5	160.0
2.10	0.091	0.043	1.0110	1.0030	0.9893	2.412	2.394	2.260	9.0	13.6	19.0
	0.307	0.146	1.0460	1.0372	1.0227	1.872	1.880	1.803	26.3	38.5	51.5
	0.601	0.286	1.0910	1.0834	1.0690	1.496	1.529	1.450	44.5	65.5	87.0
	1.000	0.476	1.1570	1.1392	1.1249	1.172	1.240	1.167	65.5	95.0	120.5
	1.511	0.719	1.2380	1.2149	1.1988	0.862	0.940	0.855	83.5	120.0	152.5
	1.988	0.947	1.3120	1.2900	1.2741	0.624	0.518	0.633	94.2	136.0	176.0
2.30	0.099	0.043	1.0128	1.0043	0.9910	1.895	1.918	1.950	13.0	18.8	24.5
	0.300	0.130	1.0419	1.0326	1.0189	1.422	1.449	1.465	32.2	46.2	59.5
	0.602	0.262	1.0861	1.0767	1.0628	1.053	1.082	1.107	57.2	91.0	104.5
	1.009	0.439	1.1467	1.1365	1.1217	0.736	0.758	0.781	83.7	118.5	152.0
	1.504	0.654	1.2193	1.2079	1.1922	0.476	0.480	0.503	107.5	151.0	193.0
	1.988	0.864	1.2898	1.2765	1.2581	0.200	0.208	0.300	119.0	180.0	235.0

APPENDIX II. Experimental data set No. 2

Mole ratio NO_3/U	Molarity		Density, grams/cc			Acidity, pH			Conductivity, millisiemens		
	Nitrate	Uranium	30°C	40°C	50°C	30°C	40°C	50°C	30°C	40°C	50°C
1.56	0.156	0.100	1.0266	1.0242	1.0188	3.240	3.120	2.956	12.2	16.2	18.0
	0.312	0.200	1.0579	1.0554	1.0499	3.038	2.980	2.810	24.0	29.0	33.8
	0.468	0.300	1.0898	1.0873	1.0815	2.970	2.860	2.700	34.0	40.5	47.3
	0.624	0.400	1.1207	1.1180	1.1122	2.880	2.780	2.620	43.0	50.7	58.0
	0.780	0.500	1.1522	1.1493	1.1431	2.780	2.700	2.543	50.0	58.5	68.0
1.60	0.160	0.100	1.0266	1.0233	1.0186	3.163	3.065	2.929	14.4	17.2	19.8
	0.320	0.200	1.0575	1.0542	1.0493	3.004	2.897	2.767	25.8	30.7	35.2
	0.480	0.301	1.0385	1.0849	1.0800	2.888	2.783	2.660	36.0	42.5	49.3
	0.640	0.400	1.1192	1.1157	1.1103	2.800	2.694	2.568	44.7	52.5	60.5
	0.800	0.500	1.1493	1.1457	1.1401	2.707	2.604	2.494	52.5	61.5	71.0
1.70	0.170	0.100	1.0275	1.0242	1.0195	3.060	2.950	2.870	15.4	18.2	21.3
	0.340	0.200	1.0588	1.0557	1.0506	2.895	2.785	2.700	27.4	32.6	37.7
	0.510	0.300	1.0902	1.0867	1.0817	2.770	2.656	2.591	37.9	44.8	51.0
	0.680	0.400	1.1219	1.1181	1.1129	2.680	2.567	2.497	47.1	55.5	64.0
	0.850	0.500	1.1528	1.1493	1.1443	2.600	2.483	2.416	55.0	64.5	74.5
1.80	0.180	0.100	1.0278	1.0244	1.0196	2.964	2.857	2.777	15.9	19.1	22.2
	0.360	0.200	1.0594	1.0564	1.0511	2.803	2.689	2.582	28.5	33.8	39.2
	0.540	0.300	1.0912	1.0879	1.0826	2.668	2.552	2.475	39.4	46.7	54.0
	0.720	0.400	1.1230	1.1197	1.1138	2.562	2.447	2.390	49.0	57.5	66.5
	0.900	0.500	1.1548	1.1512	1.1460	2.474	2.363	2.311	57.0	67.0	77.0
1.90	0.190	0.100	1.0279	1.0246	1.0199	2.780	2.677	2.611	16.8	20.2	22.7
	0.380	0.200	1.0599	1.0556	1.0509	2.596	2.491	2.410	30.2	35.4	41.4
	0.570	0.300	1.0915	1.0879	1.0831	2.460	2.362	2.272	41.5	48.8	56.5
	0.760	0.400	1.1238	1.1201	1.1150	2.341	2.255	2.177	51.5	60.5	70.0
	0.950	0.500	1.1553	1.1520	1.1467	2.250	2.157	2.103	60.0	70.0	81.0
2.00	0.202	0.101	1.0283	1.0244	1.0199	2.381	2.335	2.343	18.5	21.8	25.5
	0.400	0.200	1.0605	1.0570	1.0528	2.124	2.071	2.066	32.7	38.6	44.8
	0.602	0.301	1.0930	1.0894	1.0845	1.917	1.875	1.889	45.0	53.0	61.5
	0.802	0.401	1.1250	1.1218	1.1174	1.765	1.725	1.777	55.7	65.0	75.3
	1.000	0.500	1.1570	1.1533	1.1482	1.620	1.600	1.616	64.5	76.0	87.5

APPENDIX III. Computer program to compute uranium and nitrate concentrations of uranyl nitrate solutions given the temperature in the range of 30-50°C, the density, and the conductivity

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      DATA A/2.65684E-01/,B/2.82071E-02/
      DATA A1/7.56706E-01/,A2/3.52224E-02/
      DATA B1/1.13629E+02/,B2/1.50419/
      DATA C1/1.02522/,C2/-2.21409E-03/
      WRITE(6,10)
10    FORMAT(2X,' PREDICTION OF URANIUM AND NITRATE CONCENTRATIONS',/,
     & 2X,' IN URANYL NITRATE SOLUTIONS FROM MEASURED',/,2X,
     & ' DENSITIES AND CONDUCTIVITIES',/)
      DO 200 K=1,3
      ITEMP=20+10*K
      WRITE(6,20) ITEMP
20    FORMAT(/,2X,'COMPARISON WITH DATA AT ',I2,' DEGREES',/)
      WRITE(6,30)
30    FORMAT(/,'  COND    DENS     NO3     NO3    ERROR     U
     *  U     ERROR')
      WRITE(6,40)
40    FORMAT(17X,'OBS     CALC',10X,'OBS      CALC',/)
      DT=ITEMP-40
      A3=A1+A2*DT
      B3=B1+B2*DT
      C3=C1+C2*DT
      DENW=0.992247-0.0003806*DT-0.00000375*DT*DT
      DO 100 I=1,6
      DO 90 J=1,5
      READ(5,50) XNO3,UM,COND,DENS
50    FORMAT(2F10.0,10X,2F10.0)
      D=DENS-DENW
      PROD=COND-A3
      DIVD=1.0/(B*PROD+A*B3)
      U=(B3*D-B*C3*PROD)*DIVD
      YNO3=((A*C3+D)*PROD)*DIVD
      W1=100.*(U/UM-1.0)
      W2=100.*(YNO3/XNO3-1.0)
      WRITE(6,60) COND,DENS,XNO3,YNO3,W2,UM,U,W1
60    FORMAT(1X,F5.1,F8.4,2F7.3,F7.1,2F8.4,F7.1)
90    CONTINUE
      WRITE(6,70)
70    FORMAT(/)
100   CONTINUE
200   CONTINUE
      STOP
      END
```

APPENDIX IV. Computer program designed to calculate the uranium and nitrate concentration of uranyl nitrate solutions, given the temperature in the range of 30-50°C, the density, and the pH

```
      DATA A1/1.7587E-02/,A2/-1.1642E-04/,B1/3.2219/,B2/1.6471E-03/
      DATA C1/-4.0335E-01/,C2/1.7881E-02/,D1/-1.6537/,D2/7.8737E-02/
      DATA P1/4.8683E-01/,P2/5.8927E-03/,Q1/5.1134/,Q2/-2.4523E-03/
      DATA R1/-2.3192E-01/,R2/-4.0891E-03/,S1/2.2317E-02/
      DATA S2/6.0217E-04/
      WRITE(6,10)
10    FORMAT(5X,'URANIUM CONC= A+B(DENS-DENW)+C/(PH-D)**2',/,5X,
     &'NITRATE CONC=P+Q(DENS-DENW)+R(PH)+S(PH)**2',/,'WHERE
     &A,B,C,D,P,Q,R,S AND DENW ARE FUNCTIONS OF TEMPERATURE',/)
      ITEMP=30
      DO 200 I=1,3
      WRITE(6,20) ITEMP
20    FORMAT(/,'TEMPERATURE= ',I2)
      WRITE(6,30)
30    FORMAT(/,5X,'DENS',5X,'PH',6X,'UM',6X,'UM',5X,'ERROR',
     &4X,'NO3     NO3      ERROR')
      WRITE(6,40)
40    FORMAT(21X,'OBS     CALC',13X,'OBS     CALC',/)
      DT=ITEMP-40
      DENW=0.992247-0.0003806*DT-0.00000375*DT*DT
      A=A1+A2*DT
      B=B1+B2*DT
      C=C1+C2*DT
      D=D1+D2*DT
      P=P1+P2*DT
      Q=Q1+Q2*DT
      R=R1+R2*DT
      S=S1+S2*DT
      DO 100 J=1,6
      DO 90 K=1,5
      READ(5,50) XNO3,UM,PH,DENS
50    FORMAT(3F10.0,10X,F10.0)
      DEN=DENS-DENW
      U=A+B*DEN+C/(PH-D)**2
      YNO3=P+Q*DEN+R*PH+S*PH*PH
      E1=100.*(U/UM-1.0)
      E2=100.*(YNO3/XNO3-1.0)
      WRITE(6,60) DENS,PH,UM,U,E1,XNO3,YNO3,E2
60    FORMAT(1X,F8.4,F8.3,2F8.4,F8.1,2F8.4,F8.1)
90    CONTINUE
      WRITE(6,70)
70    FORMAT(/)
100   CONTINUE
      ITEMP=ITEMP+10
200   CONTINUE
      STOP
      END
```

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