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Subject SOME PHYSICAL PROPERTIES

OF UO_2SO_4 - D_2O SOLUTIONS

By R. Van Winkle

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SOME PHYSICAL PROPERTIES OF UO_2SO_4 - D_2O SOLUTIONS

I INTRODUCTION

The physical properties of various solutions of UO_2SO_4 in D_2O must be known at certain temperatures in order to make some of the engineering calculations in designing a homogeneous reactor. As far as it is known here, experimental values of density, viscosity, heat capacity, etc. of UO_2SO_4 - D_2O solutions at high temperatures (above 100°C) have not been published. In order to provide the ISHR pre-design group with some numbers to use in the absence of experimentally-determined physical properties, this memo presents some calculated values of a few properties, using known physical properties of pure H_2O , UO_2SO_4 , and D_2O , with the assumption that the ideal solution laws hold for these mixtures (plus a few other simplifying assumptions), in making the calculations.

The individual physical properties are treated each in its own section of this memo.

The surface tension, viscosity, density and specific heat of three different concentrations of UO_2SO_4 in D_2O have been measured at several temperatures between 28° and 100°C by Roarty, Kaplan, Powers and Redmond of this laboratory. Although their work has not been published yet, some of their results are listed in this article to show a comparison between observed and calculated values; also, their viscosity determinations were used to obtain the extrapolations shown since ideal solution laws provide no basis for calculation of solution viscosities from physical properties of the pure constituents of the solution when the solute is a solid.

II DENSITY OF UO_2SO_4 IN D_2O SOLUTIONS

The project handbook lists the densities of UO_2SO_4 and $\text{UO}_2\text{SO}_4 \cdot 3\text{H}_2\text{O}$ as follows:

$$\rho_{\text{UO}_2\text{SO}_4} = 5.24 \text{ g/cc at } 18^\circ\text{C}$$

$$\rho_{\text{UO}_2\text{SO}_4 \cdot 3\text{H}_2\text{O}} = 3.28 \text{ g/cc at } 16.5^\circ\text{C}$$

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Molecular weights of some uranyl sulfates, H_2O and D_2O , are listed as follows:

$$\text{Mol. wt. of } UO_2SO_4 = 366.13$$

$$\text{Mol. wt. of } UO_2SO_4 \cdot 3H_2O = 420.18$$

$$\text{Mol. wt. of } UO_2SO_4 \cdot 3D_2O = 426.19$$

$$H_2O = 18.016$$

$$D_2O = 20.0284$$

$$\text{Calculated density of } UO_2SO_4 \cdot 3D_2O = 3.31 \text{ g/cc at } 30^\circ C$$

An expression for the density of a mixture of UO_2SO_4 and D_2O may be obtained if it is assumed that the unit volume of UO_2SO_4 in solution remains constant over the entire range of temperature considered, while the unit volume of D_2O in the solution varies with temperature exactly the same amount as pure D_2O .

Using the density of $UO_2SO_4 \cdot 3H_2O$ given above, the apparent volume of one gram mole of UO_2SO_4 in H_2O solution may be calculated

$$\text{Volume of one gram mole } UO_2SO_4 \cdot 3H_2O \text{ at } 16.5^\circ C = \frac{420.18}{3.28} = 128.10 \text{ cc}$$

$$\text{Volume of } H_2O \text{ in one gram mole } UO_2SO_4 \cdot 3H_2O \text{ at } 16.5^\circ C =$$

$$\frac{3 \times 18.016}{0.9989} = 54.11 \text{ cc}$$

$$\text{Apparent volume of } UO_2SO_4 \text{ in one gram mole } UO_2SO_4 \cdot 3H_2O = 128.10 - 54.11$$

$$= 73.99 \text{ cc/mole}$$

This value for the gram-molar volume of UO_2SO_4 in water solutions will be assumed to be the same for D_2O solutions.

For any given mixture of UO_2SO_4 and D_2O the calculated density will be given by

$$\rho_{\text{solution}} = \frac{\text{grams } UO_2SO_4 + \text{grams } D_2O}{\text{cc } UO_2SO_4 + \text{cc } D_2O} \quad (1)$$

If the ratio of D_2O to UO_2SO_4 molecules is given by β then

$$(\rho_{\text{solution}})_T = \frac{366.13 + 20.0284 \beta}{73.99 + \frac{20.0284 \beta}{(\rho_{D_2O})_T}} \quad (2)$$

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Simplifying,

$$(\rho_{\text{solution}})_T = \frac{4.95 + 0.2707 \beta}{1 + 0.2707 \frac{\beta}{(\rho_{\text{D}_2\text{O}})_T}} \quad \text{grams/cc} \quad (3)$$

To use this equation, the density of D_2O as a function of temperature must be known. Figure 1 contains plots of $\rho_{\text{D}_2\text{O}}$ vs T , $\rho_{\text{H}_2\text{O}}$ vs T , and calc. $(\rho_{\text{solutions}}) \beta = 28, 62.9, \text{ and } 124.8$ vs T . Experimental values of these solution densities are also plotted over the temperature range reported by Roarty, et al. It is seen that the calculated densities are high by not more than 2% in two cases and low by about 1 1/2% in the third case.

Figure 2 is a plot of calculated solution density vs β at 30°C and 250°C .

Grams of uranium per liter of solution may be calculated from the following equation:

$$(\text{Grams U/liter})_T = \frac{650.237}{1 + 0.054704 \beta} (\rho_{\text{solution}})_T \quad (4)$$

Figure 3 is a plot of solution concentration, grams uranium per liter, vs β , molecules of D_2O per molecule of uranium, at 30° and 250°C .

Since there is much more density data available for $\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ solutions than for $\text{UO}_2\text{SO}_4\text{-D}_2\text{O}$ solutions, an equation for calculating densities of $\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ solutions similar to Equation (3) may be derived and used for checking calculated densities against observed densities. This is done in the Appendix.

The magnitude of error in using the Equation (3) for calculating densities of $\text{UO}_2\text{SO}_4\text{-D}_2\text{O}$ solutions will depend upon the concentration and temperature of the solution in question. Using as a basis the comparison between calculated and observed densities of $\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ solutions, it is believed that the following statements are true for $\text{UO}_2\text{SO}_4\text{-D}_2\text{O}$ solutions:

- (1) Calculated densities using Equation 3 will be low at all concentrations and temperatures.
- (2) The magnitude of deviation between calculated and observed densities appear to decrease as the value for β increases, and to increase as temperature increases.

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Drawing # 13716

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Drawing # 13717

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- (3) If $\beta < 36$, calculated solution densities may be low by 6% or more at 250°C.
- (4) If $\beta > 36$, calculated solution densities may be low by not more than 6% at 250°C, and the error will be less at lower temperatures and concentrations.
- (5) For dilute solutions, where $\beta \approx \sim 400$, the calculated densities may be low by not more than 1% at 250°C. The error will probably be even less at lower temperatures.

III VISCOSITY OF UO_2SO_4 IN D_2O SOLUTIONS

Figure 4 is a plot of viscosity vs temperature for H_2O , D_2O and the three UO_2SO_4 in D_2O solutions studied by Roarty, et al.

The values used to plot the viscosity of H_2O were from the Handbook of Chemistry and Physics and from ORNL 156-12, Physical Properties of Water and Water Vapor by W. B. Harrison.

The values used to plot the viscosity of D_2O up to 35°C were from AECD-1991, Physical Properties of Heavy Water by I. Kirshenbaum. Above 35°C the viscosity curve for D_2O is merely an extrapolation. The viscosity curve for D_2O should cross that of H_2O in the neighborhood of the critical temperature of D_2O which is 371.5°C, while the critical temperature of H_2O is 374.2°C. No attempt has been made here to guess the manner in which these curves would intersect.

The extrapolated curves of the experimental data of Roarty, et al represent a pure guess as to how the viscosity of these three solutions would vary with temperature, and the values shown for the $\beta = 28$ solution may be off (low) by as much as 50% in the neighborhood of 250°C.

Figure 5 is a plot of viscosity vs β at several temperatures. These plots were made from points taken from the curves of Figure 4.

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FIGURE 4
VISCOSITY VS TEMPERATURE
FOR H_2O , D_2O AND VARIOUS SOLUTIONS
OF UO_2SO_4 IN D_2O

FIGURE 5
VISCOSITY VS β
AT VARIOUS TEMPERATURES

$\mu - 16/\text{hr-ft}$

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β - MOLECULES D_2O PER URANIUM ATOM

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IV SPECIFIC HEAT OF UO_2SO_4 IN D_2O SOLUTIONS

In attempting to estimate the heat capacities (or specific heats) of various solutions of UO_2SO_4 in D_2O , it would be of value to know the heat capacity of the hydrated (or deuterated) salt. Since these values are not in the literature, it becomes necessary to calculate the heat capacity of the salt itself.

The project handbook lists the heat capacity of a similar salt, $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, at various temperatures.

TABLE 1

Heat Capacity for $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$

<u>T</u> <u>°C</u>	<u>C_P</u> <u>Cal/mole-°C</u>	<u>Specific Heat</u> <u>cal/g °C</u>
-53.2	86.10	0.171
-33.2	92.21	0.184
-13.2	98.35	0.196
6.8	105.00	0.209
26.8	112.70	0.224

At 25° C_P of $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O} = 112.0 \text{ cal/mole-}^\circ\text{C}$.

A survey of a few metallic sulfates, both anhydrous and hydrated, revealed that at ordinary temperatures, 10-50°C, the partial molal heat capacity of SO_4 was about 20 cal/mole-°C. It was also found that $\bar{C}_P(\text{H}_2\text{O})$ in these hydrated salts was about 9.0 cal/mole-°C.

$\bar{C}_P(\text{NO}_3)$ in $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ was calculated to be 17.25 cal/mole-°C.

Using $\bar{C}_P(\text{H}_2\text{O}) = 9.0$ and $\bar{C}_P(\text{NO}_3) = 17.25$, $\bar{C}_P(\text{UO}_2)$ in $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ is 24.2. This value can be used in estimating the heat capacity of $\text{UO}_2\text{SO}_4 \cdot 3\text{H}_2\text{O}$ and $\text{UO}_2\text{SO}_4 \cdot 3\text{D}_2\text{O}$.

$$\begin{aligned} C_P (\text{UO}_2\text{SO}_4 \cdot 3\text{D}_2\text{O}) &= \bar{C}_P(\text{UO}_2) + \bar{C}_P(\text{SO}_4) + \bar{C}_P(\text{D}_2\text{O}) \\ &= 24.2 + 20.0 + 3 \times 10.0 \\ &= 74.2 \text{ cal/mole-}^\circ\text{C} \end{aligned}$$

$$\begin{aligned} C_P (\text{UO}_2\text{SO}_4 \cdot 3\text{H}_2\text{O}) &= 24.2 + 20.0 + 3 \times 9.0 \\ &= 71.2 \text{ cal/mole-}^\circ\text{C} \end{aligned}$$

The following table shows calculated values of specific heats of various solutions of UO_2SO_4 in D_2O . These values were obtained by interpolating between values for dilute solutions and for the solid tri-deuterate $\text{UO}_2\text{SO}_4 \cdot 3\text{D}_2\text{O}$ at 25°C.

45-11

TABLE 2

Weight % UO ₂ SO ₄ in D ₂ O	β Moles D ₂ O per mole UO ₂ SO ₄	Est. Specific Heat at 25°C cal/g-°C	Est. Specific Heat at 250°C cal/g-°C
0	∞	1.005	1.168
10	164.5	0.916	1.064
20	73.0	0.830	0.964
30	42.6	0.745	0.866
40	27.4	0.658	0.764
50	18.3	0.568	0.660
60	12.2	0.474	0.551
70	7.83	0.370	0.430
80	4.57	0.252	0.293
85.9	3.00	0.174	0.202

These values are plotted in Figure 6.

Since the heat capacity of the salt is only an estimate and no C_p vs temperature relationship is known for the salt, the best one can do to estimate specific heats of these solutions at high temperatures is to assume that the specific heat of the solution varies with temperature in the same proportion as pure water. Table 3 lists specific heats of H₂O and D₂O at various temperatures up to 320°C.

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TABLE 3

Specific Heats of D₂O and H₂O

Temp °C	C _p D ₂ O ⁽¹⁾ Cal/Mole-°C	Specific Heat-D ₂ O Cal/g-°C	Specific Heat-H ₂ O Cal/g-°C
10	20.22	1.0095	1.0013 ⁽²⁾
15	20.18	1.0076	0.9997
20	20.15	1.0060	0.9988
25	20.13	1.0051	0.9983
30	20.12	1.0046	0.9980
35	20.10	1.0036	0.9979
40	20.10	1.0036	0.9980
45	20.10	1.0036	0.9983
50	20.11	1.0041 ⁽⁴⁾	0.9985
60		1.0049	0.9994
70		1.0062	1.0007
80		1.0078	1.0023
90		1.0099	1.0044
100		1.0125	1.0070
120		1.0186	1.013 ⁽³⁾
140		1.0286	1.023
160		1.0417	1.036
180		1.0578	1.052
200		1.0809	1.075
220		1.108	1.102
240		1.145	1.139
250		1.168	1.162
260		1.194	1.188
280		1.269	1.262
300		1.390	1.383
320		1.568	1.56

- (1) These values from AECD-1991, "Physical Properties of Heavy Water" by I. Kirshenbaum.
- (2) Specific heats of water from 10-100°C from Handbook of Chemistry and Physics.
- (3) Specific heats of water from 100-320°C from ORNL 156-12, "Physical Properties of Water and Water Vapor" by W. B. Harrison.
- (4) Specific heats of D₂O above 50°C obtained from multiplying the specific heat of H₂O at T by 1.0055.

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In using these curves, the specific heat of a given solution may be estimated at any temperature by using the following equation.

$$\text{Specific heat at } T = \text{Specific heat at } 25^{\circ}\text{C} \times \frac{\text{Specific Heat D}_2\text{O at } T}{\text{Specific Heat D}_2\text{O at } 25^{\circ}\text{C}} \quad (5)$$

Values obtained in this manner will be at best only approximate.

It might be of interest to compare these calculated values of specific heat with some that have been observed by Lyon's Group. This comparison is made in Table 4.

It can be seen that the values do not compare especially well; comparative values differ by as much as 10%. No claim can be made here in support of either the calculated or the observed values.

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Drawing # 18720

FIGURE 6
SPECIFIC HEAT VS β
 $UO_2SO_4-D_2O$ SOLUTIONS

TABLE 4

D₂O - UO₂SO₄ Solutions

β Molecules D ₂ O per Atom of Uranium	Temp. °C	Observed ΔH^T °C cal/g	Observed Mean Specific Heat 0°C - T°C cal/g-°C	Calc. Specific Heat at 25°C from Figure 6 cal/g-°C	<u>Specific Heat (Calc.)</u> <u>Specific Heat (Obs.)</u>
124.8	26	23.5	0.904		
124.8	47	39.5	0.840	0.892	1.064
124.8	103	79.4	0.771		
	C _P (Average) =		0.84		
62.9	26	20.2	0.777		
62.9	85	75.0	0.882	0.808	0.973
62.9	87	71.9	0.826		
	C _P (Average) =		0.83		
28.0	46	34.4	0.748		
28.0	67	47.4	0.707	0.662	0.907
28.0	79	58.9	0.745		
	C _P (Average) =		0.73		

V SURFACE TENSION

Dorsey lists the surface tension of pure H₂O up to 130°C. These values are plotted in Figure 7 and extrapolated to zero at the critical temperature.

Kirshenbaum states that the surface tension of pure D₂O relative to that of water is 0.99946 at 25°C, so there is essentially no difference between the surface tension of H₂O and D₂O. It can also be assumed that the surface tension of D₂O goes to zero at its critical temperature.

In Figure 7 experimental points for three different solutions of UO₂SO₄ in D₂O of Roarty, et al, are plotted and extrapolated. Plots of β vs surface tension can be made from these curves at any desired temperature.

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FIGURE 7
SURFACE TENSION VS TEMPERATURE
FOR H₂O AND VARIOUS UO₂SO₄-D₂O SOLUTIONS
O DATA OF ROARTY ET AL

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VI APPENDIX

An equation (similar to Equation 3) for calculating densities of $\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ solutions is given as follows:

$$(\rho_{\text{solution}})_T = \frac{4.95 + 0.2435 \beta}{1 + 0.2435 \frac{\beta}{(\rho_{\text{H}_2\text{O}})_T}} \quad (6)$$

Some data by Secoy in ORNL-336, p. 27 is listed below and compared with the values calculated by Equation (6) above.

TABLE 5

Comparison of Calculated Densities of $\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ Solutions
With Densities Reported by Secoy at Various High
Temperatures

<u>T</u> <u>°C</u>	<u>Wt. %</u> <u>UO_2SO_4</u>	<u>Obs. ρ_T</u>	<u>β</u>	<u>$(\rho_{\text{H}_2\text{O}})_T$</u>	<u>Calc. ρ_T</u>	<u>$\Delta \rho$</u>	<u>% Error</u>
108.8	34.93	1.361	37.86	0.952	1.326	0.035	2.64
129.2	34.97	1.341 ⁵	37.80	0.935	1.305	0.036	2.76
152.6	35.02	1.319	37.71	0.915	1.280	0.039	3.05
175.7	35.11	1.298	37.56	0.891	1.251	0.047	3.76
187.0	35.18	1.288	37.44	0.879	1.237	0.051	4.12
231.7	35.54	1.238	36.85	0.825	1.172	0.066	5.63
249.4	35.78	1.214	36.48 ⁵	0.801	1.144	0.070	6.12
274.0	36.28	1.178	35.69	0.762	1.099	0.079	7.19
303.8	37.14	1.121 ⁵	34.41	0.704	1.033	0.088 ⁵	8.57
325.8	38.26	1.066 ⁵	32.79	0.651	0.975	0.091 ⁵	9.38
345.8	39.77	1.002	30.78	0.588	0.905	0.97	10.72
356.3	40.48	0.967	29.87	0.547	0.855	0.112	13.09
366.5	41.75	0.919	28.36	0.486	0.779	0.140	17.97
371.5	42.20	0.895	27.83 ⁵	0.430	0.700	0.195	27.85

In Table 6 are listed some density data of Davenport and Powell, together with calculated densities using Equation 6 above.

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TABLE 6

Comparison of Calculated Densities of $\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ Solutions
With Densities Reported by Davenport and Powell

T °C	Molarity at 25°C	β	Obs $(\rho_{\text{soln}})_T$	Calc. $(\rho_{\text{soln}})_T$	% error
50	0.08732	631	1.0159	1.0133	0.26
50	0.125	440	1.0278	1.0242	0.35
50	0.1622	338	1.0397	1.0350	0.45
184	0.1299	423	$0.9248 \pm .0005$	0.9165	0.90
203	0.1299	423	$0.9015 \pm .0005$	0.8938	0.86
232	0.1299	423	$0.8630 \pm .0005$	0.8577	0.62

Table 7 shows a comparison between calculated values and those reported by Los Alamos at three different concentrations.

In Figure 8 are plotted a few correction factors vs β , which values were taken from Tables 5, 6 and 7. A smooth curve is drawn through six values at 50°C to show the approximate deviation of calculated densities from observed densities as a function of concentration. It can be seen that the equation appears to be much more accurate for dilute solutions.

The data of Secoy appears to indicate that the deviation of calculated densities from observed densities increases with temperature above 100°C and at 250°C for $\beta = \sim 36$ the error is over 6%.

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TABLE 7

Comparison of Calculated Densities of $\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ Solutions

With Densities Reported by Los Alamos

T °C	$(\rho_{\text{H}_2\text{O}})_T$	C = 51%; $\beta = 19.52$			C = 39.9%; $\beta = 30.61$			C = 29.9%; $\beta = 47.64$		
		Obs. $(\rho_{\text{soln}})_T$	Calc. $(\rho_{\text{soln}})_T$	% Error	Obs. $(\rho_{\text{soln}})_T$	Calc. $(\rho_{\text{soln}})_T$	% Error	Obs. $(\rho_{\text{soln}})_T$	Calc. $(\rho_{\text{soln}})_T$	% Error
0	0.99987	--	--	-	1.5350	1.4671	4.63	1.3547	1.3134	3.14
15	0.99823	1.7959	1.6854	6.56	1.5283	1.4662	4.24	1.3499	1.3125	2.86
20	0.99823	1.7926	1.6843	6.43	1.5257	1.4650	4.14	1.3479	1.3114	2.78
25	0.99567	1.7891	1.6825	6.34	1.5228	1.4635	4.08	1.3455	1.3100	2.71
30	0.99567	1.7854	1.6805	6.24	1.5199	1.4616	3.99	1.3430	1.3083	2.65
35	0.99406	1.7816	1.6781	6.17	1.5166	1.4596	3.90	1.3403	1.3063	2.60
40	0.99224	1.7777	1.6758	6.08	1.5133	1.4572	3.85	1.3376	1.3041	2.57
45	0.99025	1.7736	1.6729	6.02	1.5099	1.4546	3.80	-	-	-
50	0.98807	1.7696	1.6699	5.97	1.5064	1.4518	3.76	1.3314	1.2990	2.49
55	0.98573	1.7655	1.6666	5.94	1.5028	1.4488	3.73	-	-	-
60	0.98324	1.7612	1.6632	5.89	1.4992	1.4455	3.71	1.3249	1.2932	2.45
65	0.98059	1.7569	1.6594	5.87	-	-	-	-	-	-
70	0.97781	1.7526	1.6555	5.86	1.4913	1.4385	3.67	1.3178 ^{70.20}	1.2865	2.43
75	0.97489	1.7472	1.6514	5.86	-	-	-	-	-	-
80	0.97183	1.7427	1.6471	5.80	1.4829 ^{79.90}	1.4308	3.64	1.3106 ^{79.40}	1.2797	2.41
93.6	0.96294	1.7300	1.6345	5.84	1.4713 ^{93.40}	1.4192	3.67	1.2998 ^{93.20}	1.2688	2.44

$$\text{Calc. } (\rho_{\text{soln}})_T = \frac{4.95 + 0.2435 \beta}{1 + 0.2435 \beta} (\rho_{\text{H}_2\text{O}})_T$$

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-20-

Thus, from the above comparisons the derived equation for calculating densities of $\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ solutions appears to have the following limitations:

- (1) If $\beta \leq 36$, calculated solution densities may be low by more than 6% at 250°C.
- (2) If $\beta > 36$ calculated solution densities may be expected to be low by not more than 6% at 250°C.
- (3) If $\beta > 400$ calculated solution densities may be low by not more than 1% at 250°C.
- (4) The curve in Figure 8 appears to describe fairly accurately the error in calculated densities for all solutions at 50°C where $\beta > 20$.
- (5) A family of correction factor vs β curves, with temperature as the parameter, similar to the one plotted in Figure 8 may exist. There is not enough published data to attempt such a correlation.

R. VanWinkle

/wj

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FIGURE 8
CORRECTION FACTOR VS β
TO CORRECT CALCULATED DENSITIES
OF $UO_2SO_4-H_2O$ SOLUTIONS TO THE
VALUES REPORTED BY VARIOUS OBSERVERS

KS-32

Drawing # 13722