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SUMMARY

This report lists some physical property data of aqueous homogeneous reactor materials. The information has been collected primarily as an aid to those engaged in nuclear calculations. Densities of light water, heavy water, uranyl sulfate solutions, uranyl sulfate-lithium sulfate solutions, slurries of uranium oxides, and thorium oxide are presented as functions of temperature and concentration. In addition, other data useful in reactor analysis, such as viscosity, vapor pressure, etc., are listed for those materials for which data is available.

Since experimental data is frequently meager or limited in range, large extrapolations have generally been required to cover possible areas of interest. The basis and extent of such extrapolations is always indicated.

In a collection on this kind, errors and omissions are almost unavoidable. Notice of these, as well as suggestions, would be appreciated.

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I. Light Water

A. Density.

Fig. 1 shows the variation of H_2O density with pressure at several temperatures between $300^{\circ}F$ and $620^{\circ}F$. At constant temperature, the density is practically a linear function of pressure. The equations for these linear variations is marked on each isotherm, with density in grams per c.c. and pressure P in psia.

Fig. 2 is a plot of H_2O density vs. temperature for various pressures between saturation and 3500 psia for temperatures between 150 and $350^{\circ}C$.

Fig. 3 shows the ratio of the density of compressed water to that of saturated water at the same temperature as a function of pressure, for temperatures between $300^{\circ}F$. and $620^{\circ}F$. and pressures between 1000 and 4000 psia.

References: Dorsey, N. E., Properties of Ordinary Water Substance, Reinhold Publishing Company, New York (1940).

Keenan, J. H. and Keyes, Frederick G., Thermodynamic Properties of Steam, John Wiley and Sons, Inc. (1936)

B. Thermal Conductivity.

Fig. 4 shows the variation of thermal conductivity with temperature ($300 - 680^{\circ}F$) and pressure (saturation to 6000 psi.) The accuracy of the values is 5% or better over the range shown.

References: Lottes, P. A., Physical and Thermodynamic Properties of Light and Heavy Water, Chapter 1.3, Reactor Handbook, Vol. 2, (Engineering), May 1955, Technical Information Service, USAEC.

C. Viscosity.

Fig. 5 gives the viscosity of liquid water in lbs. mass/hr-ft. vs. temperature in degrees Fahrenheit (200 - 700°F.) for various pressures between saturation and 4000 psia. The accuracy varies between 1.5 and 8%, the error increasing with temperature and pressure.

References: Harrison, W. B., ORNL 156 (October 1, 1948)
Lottes, P. A. (ibid.) (Presents tables of viscosity.)

D. Heat Capacity.

Fig. 6 shows the heat capacity in Btu/lb -°F. from 30 - 690°F. for pressures between saturation and 6000 psia. The accuracy varies between 0.3 and 5%, the error increasing with temperature.

References: Harrison, W. B., (ibid.)
Lottes, P. A., (ibid.) (Tables available)

E. Vapor Pressure.

Fig. 7 shows the vapor pressure of light water as a function of temperature (150°C. - 374.2°C.) and the ratio of the vapor pressure of light water to that of heavy water at the same temperature.

Reference: Dorsey, ibid.
Oliver, G. D., Grisard, J. W., J. Am. Chem. Soc. 78 561-3 (1956)

II. Heavy Water

A. Density.

Fig. 8 shows the density of saturated D₂O as a function of temperature. The section of the curve between 260°C. and 350°C. has been interpolated by comparison with light water data, using the reasonably constant ratio of light-to-heavy water density in this range. The accuracy of this curve is believed to be 1% or better.

References: Heiks, J. R., Barnett, M. K., Jones, L. V., and Orban, E., The Density, Surface Tension, and Viscosity of Deuterium Oxide at Elevated Temperatures, J. Physical Chemistry 58, 488 (1954).
 Van Winkle, R., ORNL CF 52-1-124, Jan. 18, 1952.

Fig. 9 shows the density of compressed D_2O as a function of temperature (148 - 326°C.) for various pressures between saturation and 3500 psia. The compressed densities were computed with the aid of Fig. 3, i.e., by assuming the ratio of compressed-to-saturation pressure density ratio was the same for H_2O and D_2O at the same temperature.

B. Viscosity.

Fig. 10 shows the variation of the viscosity of 99.20% D_2O with temperature (30-250°C.) at saturation pressure. The accuracy is believed to be better than 2% and may be as low as 0.2% at the lowest temperatures.

References: Heiks, J. R., et al., *ibid.* (See also Hardy, R. C. and Cottington, R. L., Report No. A-3954, Dept. of Commerce, National Bureau of Standards (1946?).)

C. Vapor Pressure. (See Fig. 7 and Section I.E.)

III. Density of Mixtures of Light and Heavy Water

Fig. 11 gives the density of mixtures of light and heavy water at various temperatures (150 - 320°C.) vs. the weight fraction, p , of heavy water in the mixture. These curves were calculated assuming the mixtures were ideal solutions, so that the density of the mixture is given by

$$d = \frac{1}{\frac{p}{d_{D_2O}} + \frac{1-p}{d_{H_2O}}}$$

where d = density of $H_2O - D_2O$ solution, gms/cc
 d_{D_2O} = density of pure saturated D_2O at same
 temperature as solution, gms/cc
 d_{H_2O} = density of pure saturated H_2O at same
 temperature as solution, gms/cc

Pressure effects due to differences in saturation pressure have been neglected. At total pressures higher than saturation pressure, corrections to the density may be made using Fig. 3.

Fig. 12 gives the concentration in molecules per barn-cm of H_2O and D_2O vs. weight fraction D_2O in $H_2O - D_2O$ mixtures over the 150 - 320°C. temperature range. The formulas used to prepare these curves were:

$$N_{H_2O} = 0.03343 (1-p) d$$

$$N_{D_2O} = 0.03007 p d$$

where N_{H_2O} and N_{D_2O} are the molecules per barn-cm of H_2O and D_2O respectively in the mixture of density d grams per cc and p weight fraction D_2O .

References: Heiks, J. R., et al. *ibid.*
 Kirshenbaum, I., Urey, H. C., and Murphy, G. M.,
Physical Properties and Analysis of Heavy Water,
 McGraw-Hill (1951).

IV. Solutions of UO_2SO_4 in H_2O

A. Density

Figures 13 and 14 show the density of uranyl sulfate- H_2O solutions as a function of the density of water at the same temperature and pressure for uranium-water ratios between 10 and 700 grams U/kg H_2O .

Figures 15, 16, and 17 show the concentration of uranium in atoms per barn-centimeter as a function of H_2O density at the same temperature and pressure as the solution for various uranium-water weight ratios (10 - 700 grams U/kg H_2O).

Fig. 18 shows the concentration of H_2O in atoms per barn-cm vs. the concentration of uranium in grams U/kg H_2O for various values of H_2O density at the same temperature and pressure as the solution. (0-700 grams U/kg H_2O ; 0.4 - 1.0 density of H_2O in gms per cc.)

The density and atomic concentration curves of figures 13 - 18 are based on the reference-substance correlation devised by Marshall for uranyl-sulfate solutions. The density relation is:

$$d_s = \frac{1}{\frac{78.65}{U} - 1.046} + d_{\text{H}_2\text{O}}$$

where d_s is the density of the solution in grams/cc, U is the weight per cent uranium and $d_{\text{H}_2\text{O}}$ is the density of water, g/cc, at the same temperature and pressure as the solution. To find the density of a solution given the concentration of uranium in grams of U/kg H_2O , the temperature, and the pressure, Figures 1,

2, or 3 are consulted to obtain the density of H_2O , and this density and the weight ratio are used to get the solution density from Figs. 13 or 14. The atomic concentrations are obtained in similar fashion from Figs. 15 - 18. Linear interpolation between curves is acceptable.

The accuracy of Marshall's density formula is believed to be $\pm 0.5\%$ from 25 - 300°C for solutions 0 - 10% uranium by weight, $\pm 1\%$ for 10 - 50% U solutions from 120 - 250°C, and $\pm 2\%$ for 10 - 50% U solutions from 25 - 120°C and 250 - 280°C. The atomic concentrations are of similar accuracy in these ranges. Note that the charts show considerable extrapolation below H_2O densities covered by experimental measurement. These extrapolated curves do not necessarily imply that the solutions to which they correspond may actually exist. They have been included only as an aid in making estimates in ranges where no data is available.

References: Lee, J. E., et al., ORNL 1332, June 18, 1952 (Secret).
Jegart, J. S., Heiks, J. R., Orban, E., MLM 728, Oct. 10, 1952.
Marshall, W. L., ORNL CF 52-1-93, Jan. 15, 1952.
Segaser, C. L., ORNL CF 54-4-203, April 29, 1954.
(Contains several references to measurements made by Orban and others at Mound Laboratory.)

B. Vapor Pressures of Uranyl Sulfate - H_2O Solutions

Fig. 19 shows the ratio of the vapor pressures of uranyl sulfate solutions to that of pure H_2O for various uranium concentrations (100 - 1000 g U/kg H_2O). This curve is based on Table I which presents data (unpublished) taken by H. O. Day, and kindly provided by Dr. C. H. Secoy. Fig. 19 was prepared on the assumption that, at a given molality, the vapor pressure ratio was temperature

independent. Actually, inspection of Table I will show that a slight temperature dependence has been observed. The average value of the ratio at each concentration has been used in Fig. 19.

Reference: Secoy, C. H., Personal Communication

C. Gas Solubilities.

For the solubility of gases in aqueous uranyl sulfate and other solvents of interest, the reader is referred to the extensive discussion by Stephan, E. F., Hatfield, N. S., Peoples, R. S., and Pray, H. A. H., "The Solubility of Gases in Water and in Aqueous Uranyl Salt Solutions at Elevated Temperatures and Pressures," BMI 1067, Jan. 23, 1956. This report presents 23 tables and 30 graphs of results obtained at Battelle covering Hydrogen, Oxygen, Helium, Deuterium and Xenon solubilities in Uranyl Sulfate, Fluoride, water and heavy water.

D. Viscosity.

The viscosity of aqueous uranyl sulfate solutions has been reported in MLM 788 by J. R. Heiks and J. S. Jegart, Dec. 19, 1952, in the range 20 - 90°C and 0.176 to 2.865 molal.

V. Density of Uranyl Sulfate - D₂O Solutions

Figure 20 gives the density of uranyl sulfate - D₂O solutions as a function of D₂O density at the same temperature and pressure as the solution for uranium - D₂O ratios between 0 and 1000 g U/kg D₂O.

Fig. 21 gives the concentration of uranium in atoms per barn-cm vs. D₂O density over the 0 - 1000 g U/kg D₂O range. Fig. 22 gives the D₂O concentration in molecules per barn-cm in the same fashion.

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Table I

Vapor Pressures of Uranyl Sulfate Solutionsin mm of Hg for Five Values of Molality*of UO_2SO_4

Temperature (°C)	<u>0</u>	<u>0.59166</u>	<u>1.14545</u>	<u>3.17433</u>	<u>4.1821</u>
25	23.749	23.516	23.254	21.637	20.491
30	31.816	31.516	31.170	29.049	27.571
35	42.167	41.775	41.331	38.468	36.672
40	55.317	54.806	54.237	50.582	48.315
45	71.869	71.194	70.493	65.979	63.032
50	92.518	91.634	90.781	85.123	81.481
60	149.406	147.963	146.712	138.640	132.532
70	233.723	231.559	229.844	217.556	208.859
80	355.215	352.108	349.560	331.953	319.638
90	525.857	521.692	517.964	493.041	-
100	760.00	754.304	-	-	-

*Molality is the average values of the high and low concentrations of a particular run.

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These figures are based on Marshall's reference-substance formula for $\text{UO}_2\text{SO}_4 - \text{D}_2\text{O}$ solutions:

$$d_s = \frac{1}{\frac{71.0}{U} - 0.944} + d_{\text{D}_2\text{O}}$$

where d_s and $d_{\text{D}_2\text{O}}$ are the densities of the solution and of D_2O respectively and U is the weight per cent uranium.

The amount of experimental justification for these curves is much less than that for the corresponding light water curves discussed previously. Measurements were made on three solutions (20, 40, 60 g U/liter) between 30 and 250°C. at Mound Laboratories. The Marshall formula agrees with these measurements to within 3%.

References: Marshall, *ibid.*

Orban, E., Mound Laboratories CF No. 53-12-96 (December 28, 1953).

VI. Density of Equimolar Solutions of Uranyl Sulfate

with Li_2SO_4 or BeSO_4 in H_2O

Fig. 23 compares some preliminary density data furnished by R. S. Greeley with a reference substance correlation devised by the writer. Data was taken at room temperature for five concentrations ranging between 0 and 800 g U/kg H_2O , and for a 3.2 Molal $\text{UO}_2\text{SO}_4 - 3.4$ Molal Li_2SO_4 solution between 25 and 310°C. The room temperature data agrees within 2% with the correlation, while the higher temperature data are at most 5% lower than the values predicted by the formula.

Fig. 24 shows the density of equimolal $\text{UO}_2\text{SO}_4 - \text{Li}_2\text{SO}_4$ solutions in H_2O vs. the density of H_2O at the same temperature and pressure as the solution. The H_2O density ranges between 0.7 and 1.0 g/cc and the U concentration range is 0 - 1000 g/kg H_2O . Figures 25 and 26 show the uranium concentration in atoms per barn-cm and the H_2O concentration in molecules per barn-cm for H_2O and uranium to H_2O weight ratios approximately the same as those in Fig. 24.

The reference substance correlation devised to fit the data available assumed that the density of an equimolal solution of UO_2SO_4 and $\text{Li}_2\text{SO}_4 - \text{H}_2\text{O}$ could be estimated as the average of the densities of a $\text{UO}_2\text{SO}_4 - \text{H}_2\text{O}$ and a Li_2SO_4 solution each of double the molality of the equimolal solution. It was found that the formula

$$\rho_{\text{Li}} = \frac{1}{\frac{116.226}{p} - 0.28094} + \rho_{\text{H}_2\text{O}}$$

(where ρ_{Li} = density in g/cc of a $\text{Li}_2\text{SO}_4 - \text{H}_2\text{O}$ solution of p weight per cent Li_2SO_4 at a temperature and pressure where the density of pure H_2O would be $\rho_{\text{H}_2\text{O}}$) accurately (better than 1%) correlated density data for Li_2SO_4 solutions. Using this equation and the Marshall formula for $\text{UO}_2\text{SO}_4 - \text{H}_2\text{O}$ solutions gave the result

$$\rho = \frac{1}{2} \left[\frac{1}{\frac{116.226}{p} - 0.28094} + \frac{1}{\frac{78.65}{U} - 1.046} \right] + \rho_{\text{H}_2\text{O}}$$

where ρ is the density of an equimolal $\text{UO}_2\text{SO}_4 - \text{Li}_2\text{SO}_4$ solution in H_2O in g/cc and p and U are the weight per cents of Li_2SO_4 and uranium, respectively, in H_2O solutions containing only one of the solutes which correspond to twice the stated molality of the

equimolal solution. (The charts themselves are in terms of the actual ratio of uranium to water in g U/kg H₂O.)

In the case of BeSO₄ - UO₂SO₄ solutions, the little data available indicates that the density of an equimolal Li₂SO₄ - UO₂SO₄ solution is the same as that of an equimolal BeSO₄ - UO₂SO₄ solution. The molecular weights of BeSO₄ and Li₂SO₄ are close enough to allow the approximation that solutions of the same molality in these salts have the same weight per cent of these salts. For estimation purposes, it seems reasonable, therefore, to recommend the use of Figs. 24, 25, and 26 unaltered for equimolar uranyl sulfate - beryllium sulfate solutions until specific data for these are available.

References: Marchall, *ibid.*

R. S. Greeley, Personal Communication of Preliminary Data
International Critical Tables, p. 78, V. III, McGraw-Hill
Book Company, Inc., 1928.

VII. Density of Slurries

A. Thorium Oxide Slurries in Light and Heavy Water.

Fig. 27 gives the concentration of thorium in atoms per barn-cm in aqueous slurries of ThO₂ vs. the density of H₂O or D₂O at the same temperature and pressure as the slurry for various concentrations of Th in g/kg of water. The Thorium-water ratio varies between 100 and 2000 g Th/kg water, while the water density is between 0.6 and 1.1.

Fig. 28 gives the molecules of H₂O/barn-cm. These concentrations were calculated assuming ThO₂ to be completely insoluble in water, and using a density of ThO₂ of 9.69 g/cc. (While recent diffraction data indicate a theoretical density of ThO₂ of 10.03, the required

changes in the curves using the higher value are slight. Moreover, the minor ThO_2 density variation with temperature has also been neglected.)

References: Haubenreich, P. N., "Some Properties of ThO_2 - D_2O Slurries," TBR Memo No. 10, ORNL CF 55-2-51 (Feb. 10, 1955).

Crowley, P. R., and Kitzes, A. S., "Physical Properties of Thorium Oxide Slurries," ORNL CF 54-5-59 (May 13, 1954).

Handbook of Chemistry and Physics, 36th Edition Chemical Rubber Publishing Co. (1954).

B. Uranium Dioxide Slurries in Light and Heavy Water

Fig. 30 gives the concentration of uranium in atoms/barn-cm, and Fig. 31 gives the concentration of H_2O and D_2O in molecules/barn-cm. in UO_2 - heavy or light water slurries as a function of water density at the same temperature and pressure. These graphs were prepared assuming that UO_2 is completely insoluble in water and has a density of 10.9 g/cm^3 .

References: Handbook of Chemistry and Physics, 36th Edition, Chemical Rubber Publishing Co. 1954.

Shapiro, Z. M., WAPD-PMM-167, June 30, 1955 (Confidential).

C. Uranium Trioxide Slurries in Light Water.

Fig. 32 gives the concentration of uranium atoms/barn-cm and Fig. 33 gives the concentration of H_2O in molecules/barn-cm in uranium trioxide-light water slurries as a function the density of H_2O at the same temperature and pressure as the slurry. Uranium trioxide was assumed to be in the form $\text{UO}_3 \cdot \text{H}_2\text{O}$ which, in turn, was assumed water insoluble. The density of the monohydrate was taken as 6 g/cm^3 . Actually, the monohydrate may exist in more than one crystalline form having densities somewhat different from 6, but this value is considered adequate for the purposes of the present collection.

References: Katz, Joseph J. and Rabinowitch, "The Chemistry of Uranium," (National Nuclear Energy Series, Vol. VIII-5), 1st Edition, McGraw-Hill Book Co., Inc., (1951).

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- Fig. 23 Density Data for $\text{UO}_2\text{SO}_4\text{-Li}_2\text{SO}_4\text{-H}_2\text{O}$ solutions.
- Fig. 24 The Density of Equimolal $\text{UO}_2\text{SO}_4\text{-Li}_2\text{SO}_4$ solutions in H_2O vs. the Density of H_2O at the Same Pressure and Temperature as the Solutions for Various Concentrations of Uranium in Grams U/kg H_2O .
- Fig. 25 Concentration of Uranium in Atoms per barn-cm in Equimolal $\text{UO}_2\text{SO}_4\text{-Li}_2\text{SO}_4$ solutions in H_2O vs. Density of H_2O at the Same Temperature and Pressure as the Solutions for Various Concentrations of Uranium in Grams U/kg H_2O .
- Fig. 26 Concentration of H_2O in Molecules per barn-cm vs. Uranium Concentration in Grams U/kg H_2O in Equimolal Solutions of UO_2SO_4 and Li_2SO_4 in H_2O for Various Values of H_2O Density at the Same Pressure and Temperature as the Solutions.
- Fig. 27 Concentration of Thorium in Atoms per Barn-cm in Aqueous Slurries of ThO_2 vs. the Density of H_2O or D_2O at the Same Temperature and Pressure as the Slurry for Various Concentrations of Th in Grams of Thorium/kg of Water.
- Fig. 28 H_2O Molecules per Barn-cm in $\text{H}_2\text{O}\text{-ThO}_2$ slurries vs. Density of H_2O at Same Temperature and Pressure as Slurry for Various Values of Th Concentration in Grams Th/kg H_2O .

- Fig. 29 Concentration of Heavy Water in Molecules per Barn-cm in D_2O - ThO_2 slurries vs. Density of D_2O at Same Temperature and Pressure as Slurry for Various Concentrations of Th in Grams Th/kg D_2O .
- Fig. 30 Concentration of Uranium in Atoms per Barn-cm in UO_2 - Water (H_2O or D_2O) Slurries vs. Water Density at Same Temperature and Pressure as Slurry for Various Uranium Concentrations in Grams U/kg Water.
- Fig. 31 Molecules of Water per Barn-cm in Uranium Dioxide - Water Slurries vs. Density of Water at Same Temperature and Pressure as Slurry for Various Uranium Concentrations in Grams U/kg Water. (Solid Curves Refer to Light Water Slurries; Dashed Curves to D_2O Slurries.)
- Fig. 32 Concentration of Uranium Atoms in Atoms per Barn-cm in Uranium Trioxide - H_2O Slurries vs. Density of H_2O at Same Temperature and Pressure as Slurry for Various Uranium Concentrations in Grams U/kg H_2O .
- Fig. 33 Concentration of H_2O in Molecules per barn-cm in Uranium Trioxide - Water Slurries vs. Density of H_2O at Same Temperature and Pressure as Slurry for Various Uranium Concentrations in Grams U/kg H_2O .

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Fig. 6

Heat Capacity of Liquid H_2O at Constant Pressure
(ORNL-156 Part 2, p. 11)

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Fig. 10 Viscosity of Heavy Water (99.20% D₂O) vs. Temperature

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Fig. 22 Concentration of D_2O in $UD_{2804}-D_2O$ Solutions in Molecules per barn-cm. vs. Uranium Concentration for Various Values of Density of D_2O at Same Pressure and Temperature as Solution.

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Fig. 24
The Density of Equimolar $\text{CO}_2\text{SO}_3 - \text{Li}_2\text{SO}_3$ Solutions in H_2O vs. the Density of H_2O at the Same Pressure and Temperature as the Solutions for Various Concentrations of Uranium in g. U per kg H_2O .

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Fig. 26 Concentration of H_2O in molecules per barn-cm. vs. Uranium Concentration in Grams U per kg. H_2O in Equimolar Solutions of UO_2SCl_4 and Li_2SCl_4 in H_2O for various values of H_2O Density at the same Pressure and Temperature as the Solutions.

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Fig. 33 Concentration of H_2O in Molecules per barn-cm in Uranium Trioxide-Water Slurries vs. Density of H_2O at Same Temperature and Pressure as Slurry for Various Uranium Concentrations in Grams U per kg H_2O .

Molecules of H_2O per barn-cm