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THE DENSITIES OF URANYL SULFATE
SOLUTIONS BETWEEN 20° AND 90°C
(Information Report)

BY

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ABSTRACT

The densities of uranyl sulfate water solutions were measured for 12 concentrations between the temperatures of 20° and 90°C. The density range at 20 C is 1.0539 grams per cubic centimeter at 0.17 molal to 2.0059 grams per cubic centimeter at 3.9 molal.

INTRODUCTION

The knowledge of the densities of uranyl sulfate solutions is necessary for the calculation of viscosity and surface tension and for the conversion of weight-per cent concentration to a molar basis.

Some early work was done by de Coninck¹ on the densities of solutions between 0.03 and 0.3 molar concentration at temperatures between 10° and 16°C. In 1943, Helmholtz and Friedlander² reported the densities of solutions of three concentrations at temperatures between 0° and 90°C and of eight concentrations at 30°C.

EXPERIMENTAL

A well known method of measuring solution densities was adopted in which the change in weight of a bob was measured when immersed in the solutions. A hole was drilled through the base of an Ainsworth Type DLB Chain-o-matic balance below the left end of the beam. A Pyrex bob, one-half inch by three inches, filled with steel balls to adjust the weight, was suspended from the beam by a length of 36 gauge platinum wire so that it hung freely in the solution which was contained in a 100-milliliter vessel. This vessel was placed in a well-insulated, constant-temperature oil bath. The bob was first weighed in air and then in doubly-distilled water at each of the temperatures of the experiment. From the known densities of water,³ the volume of the bob was calculated at each temperature used.

The vessel which contained the solution originally was fitted with a reflux condenser to avoid changes of concentration due to evaporation. This vessel was discarded, first, because the holdup of the condenser was too large, secondly, because cold water was continually running back into the solution and upsetting thermal equilibrium, and thirdly, because a concentration equilibrium could not be established easily with condensate continually dripping back. The vessel used for the measurements had small openings for a stirrer and the suspension wire; thus, a small amount of evaporation occurred during the periods when temperature equilibrium was being established. However, the solution was well stirred before each measurement, and immediately afterward a sample was taken to establish the exact concentration at the time of the measurement.

A platinum resistance thermometer placed in the solution showed a temperature variation of less than $\pm 0.05^{\circ}\text{C}$ during the course of a measurement. Measurements were made at 20° , 30° , 45° , 60° , 75° and 90°C for 12 concentrations of uranyl sulfate.

The preparation of the uranyl sulfate used for this work has been described in two other reports.^{10, 11} The uranium concentration of the original material was determined by weighing a sample in a covered Vycor crucible, evaporating carefully to dryness, and then igniting to a constant weight of U_3O_8 . The sulfate was determined by using an ion exchange column filled with Dowex 50 and titrating the effluent acid for sulfuric acid. This method has been described fully in an ORNL report.¹²

After each density determination, triple determinations were made of the uranium concentration. The maximum range among triplicate analyses was within 0.06 per cent by weight of uranium, and the average range about 0.025 per cent by weight.

DISCUSSION OF RESULTS

The results of the density measurements are tabulated in Table I. As mentioned earlier in the report, samples for analysis were taken at each temperature because of the change in concentration due to evaporation. This change in concentration becomes noticeable above 60°C . A graph of the results is shown in Figure 1. So that the graph would remain legible, only the curves for 20°C and 90°C are presented in this figure. In order to present the data in more detail, the graph was divided into three sections and enlarged in Figures 2, 3, and 4.

These curves are presented as density versus molality for the reason that the molality of a solution does not change with temperature as does the molar concentration. The curve in Figure 5 may be used to convert molality to the more common molar concentration at 25°C , the temperature at which most solutions are prepared and analyzed.

Figure 6 shows a comparison of the data of various experimenters. The solid line has been drawn through the data given in this report.

The data of Holmholz and Friedlander and of this report at 30°C agree reasonably well. It can be seen that the data of de Coninck appears to have a much smaller slope than that of the other experimenters.

The concentrations of uranyl sulfate solutions may be estimated from their densities provided the relationship between concentration and density is established with sufficient precision.

To obtain from these data an equation suitable for estimating molarities from measured densities, the density must be regarded as the independent variable and the "line of best fit" made to conform to the condition that the sum of squares of the deviations of the points from the line, measured in the direction parallel to the molarity axis, shall be a minimum. This equation is found to be

$$M = 3.1840 a - 3.1887$$

where M is the molarity of the solution and a is its density in grams per cubic centimeter.

The reliability of estimated molarities deduced from the above equation depends on the scatter of the observed points about the line and may be expressed in terms of the "standard error of estimate" which in this case is approximately 0.0053 mole per liter. There are 5 chances in 100 that an estimated molarity as calculated from the equation

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will deviate by as much as two standard errors or 0.0106 mole per liter from the true molarity. There is 1 chance in 100 that an estimated molarity will deviate by as much as three standard errors or 0.0159 mole per liter from the true molarity.

The applicability of the above equation is restricted to the ranges of concentration and density corresponding to the data of Table I taken at 20°C. The values for the slope of the line of best fit, its intercept, and the standard error of estimate are but estimates based on the limited data available. More numerous data should provide more reliable estimates of these parameters.

If the density of water at 20°C, 0.9982 gram per cubic centimeter, is substituted in the equation, a value of 0.0104 for M results. To pass through the point ($M = 0$, $d = 0.9970$), our line would have to depart from linearity and assume a more gentle slope at lower concentrations than at the high

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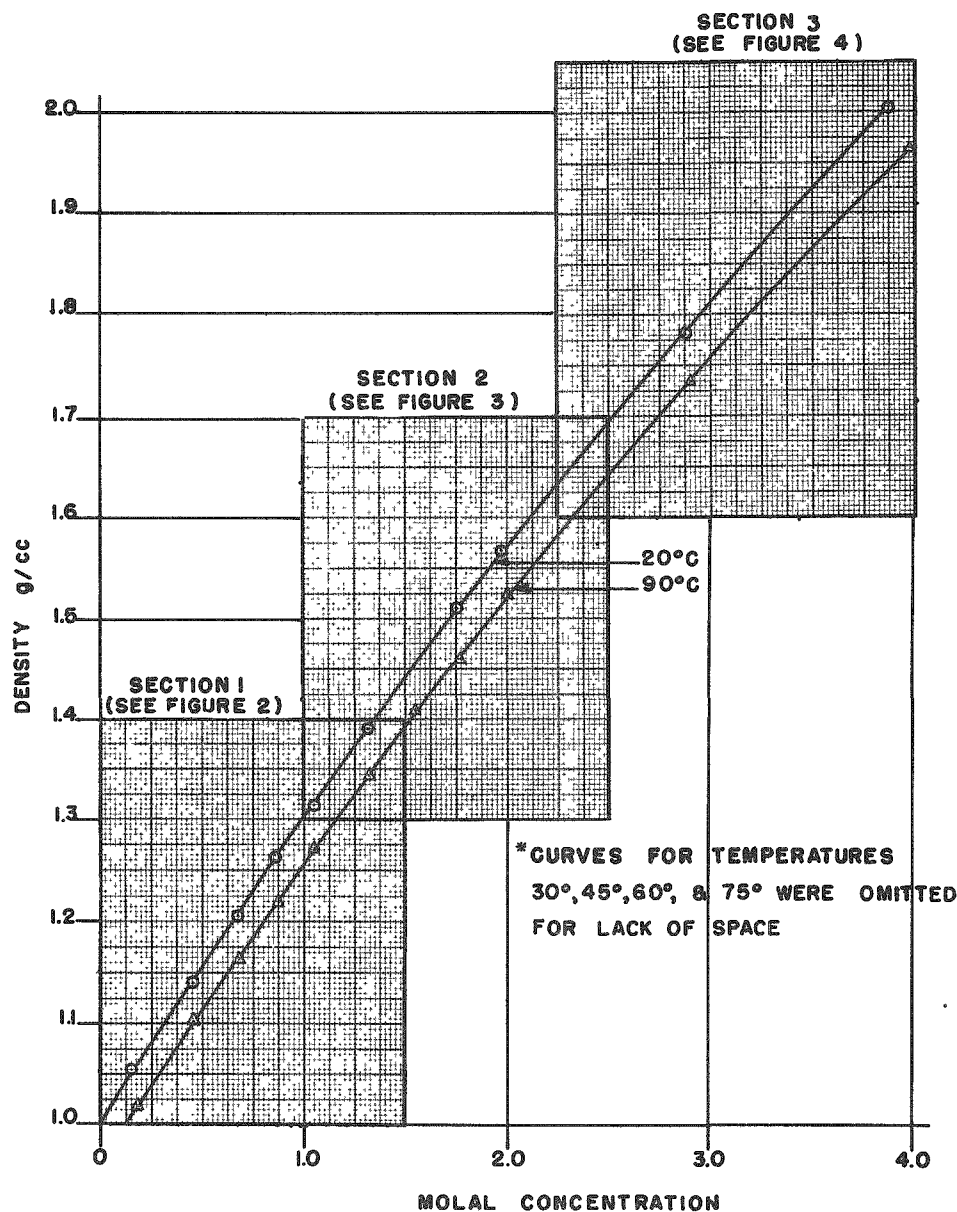
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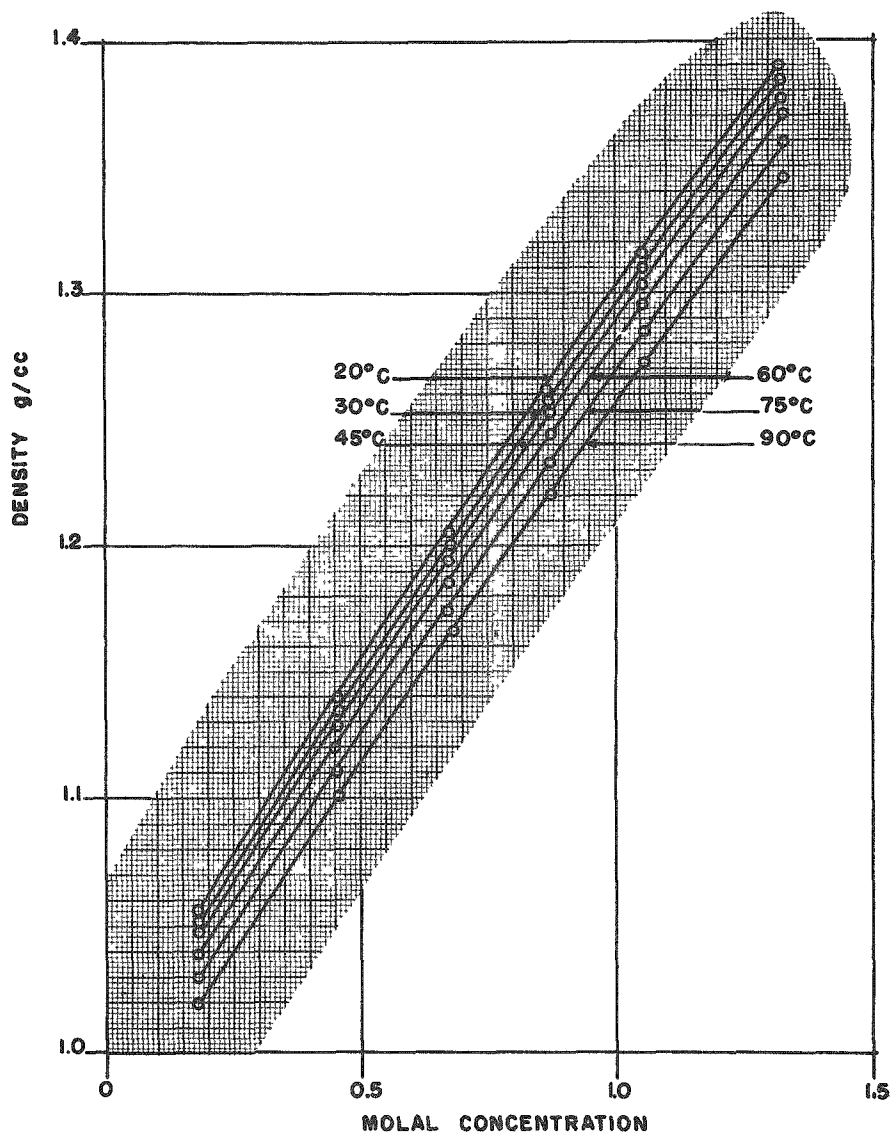
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TABLE I
THE DENSITIES OF URANYL SULFATE SOLUTIONS

URANIUM (Wt - %)	MOLALITY	DENSITY BY DISPLACEMENT (g/cc)	URANIUM (Wt - %)	MOLALITY	DENSITY BY DISPLACEMENT (g/cc)
20.0°C			59.8°C		
3.94	0.176	1.0539	3.96	0.177	1.0343
9.23	0.452	1.1384	9.20	0.450	1.1208
12.83	0.671	1.2050	12.83	0.671	1.1862
15.62	0.864	1.2630	15.72	0.870	1.2432
18.04	1.049	1.3141	18.04	1.049	1.2948
21.20	1.321	1.3911	21.22	1.323	1.3692
25.42	1.753	1.5089	25.46	1.756	1.4867
27.31	1.978	1.5672	27.38	1.986	1.5430
33.29	2.865	1.7797	33.32	2.870	1.7533
38.12	3.872	2.0059	38.16	3.880	1.9752
25.3°C			75.0°C		
38.12	3.872	2.0022	3.98	0.178	1.0293
30.0°C			9.27	0.454	1.1113
3.94	0.176	1.0510	12.80	0.669	1.1761
9.23	0.452	1.1349	15.69	0.868	1.2334
12.83	0.671	1.2012	18.10	1.053	1.2845
15.62	0.864	1.2588	21.28	1.329	1.3579
16.58	0.935	1.2770	25.48	1.761	1.4735
18.04	1.049	1.3098	27.50	2.002	1.5328
21.20	1.321	1.3865	38.48	3.959	1.9708
23.17	1.512	1.4409	90.0°C		
25.42	1.753	1.5032	4.03	0.180	1.0193
27.38	1.987	1.5613	9.33	0.458	1.1008
33.27	2.862	1.7736	12.98	0.681	1.1654
38.12	3.872	1.9983	15.81	0.877	1.2219
44.8°C			18.12	1.055	1.2720
3.94	0.176	1.0445	21.22	1.323	1.3458
9.23	0.452	1.1282	23.62	1.557	1.4113
12.83	0.671	1.1939	25.57	1.770	1.4620
15.63	0.864	1.2513	27.56	2.009	1.5236
16.62	0.938	1.2702	38.64	3.999	1.9646
18.04	1.049	1.3023			
21.20	1.321	1.3779			
23.13	1.507	1.4329			
25.42	1.753	1.4956			
27.35	1.983	1.5516			
33.25	2.858	1.7633			
38.17	3.872	1.9856			

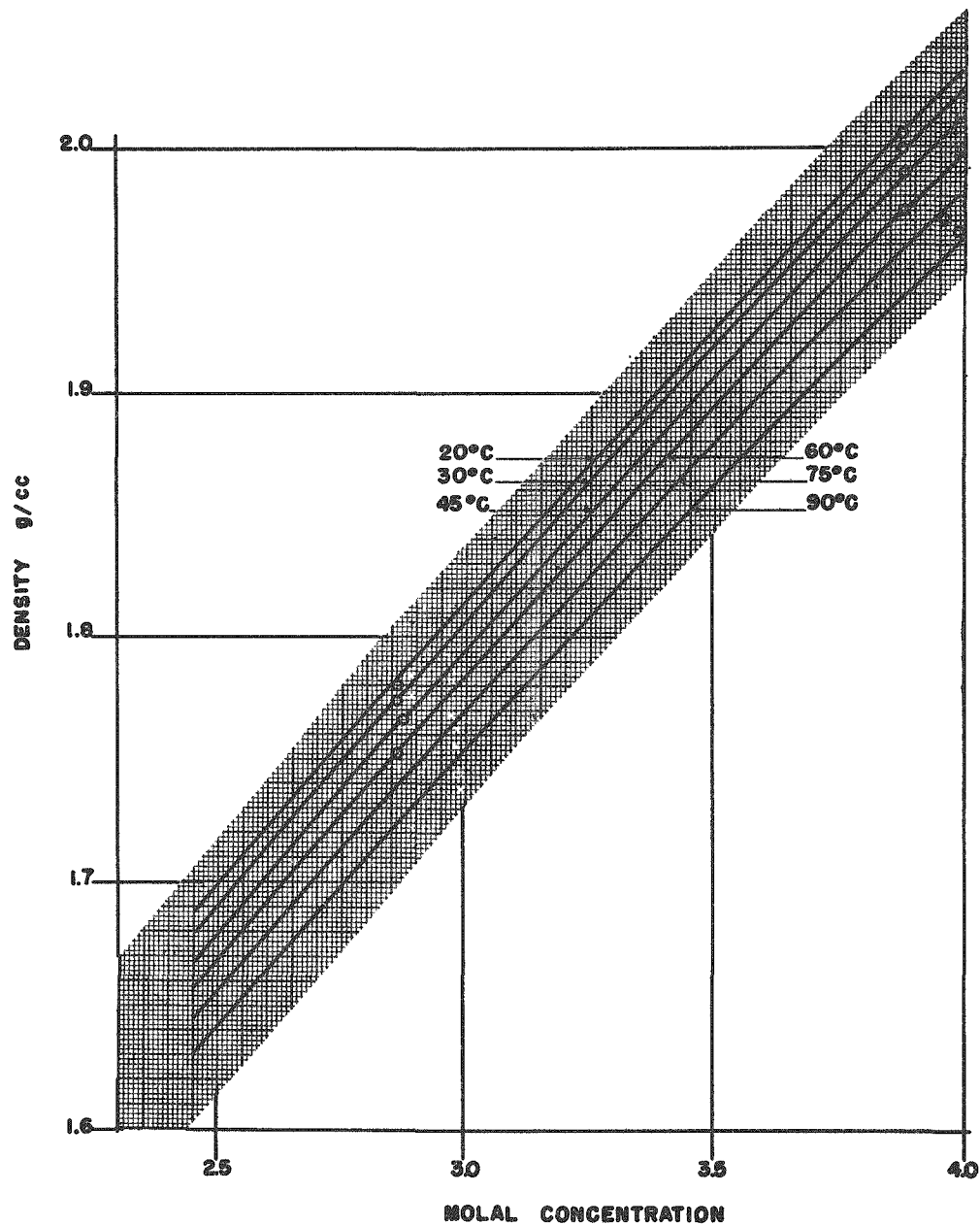


DENSITY CHANGE WITH CONCENTRATION
AT 20°C AND 90°C *



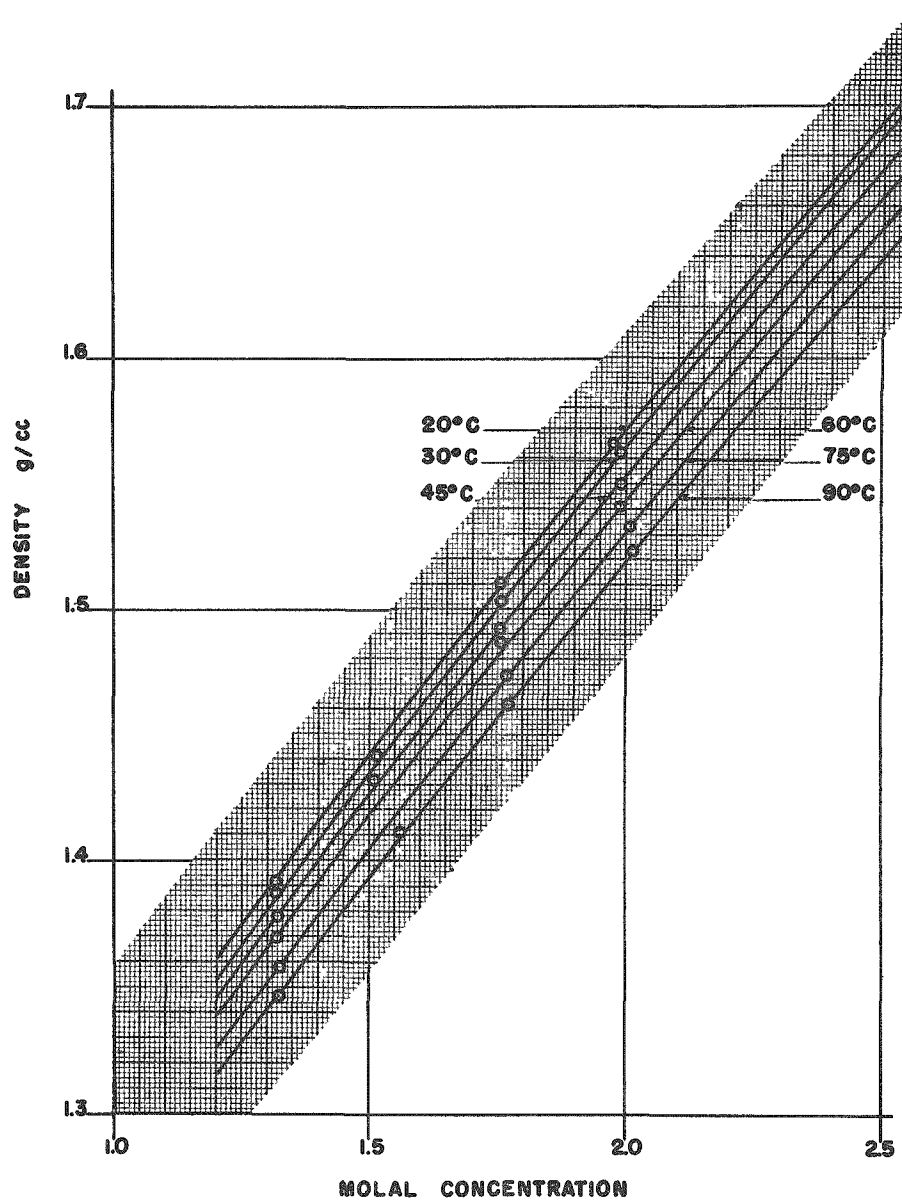
DENSITY VERSUS CONCENTRATION
(ENLARGEMENT OF SECTION I FIGURE 1)

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DENSITY VERSUS CONCENTRATION
(ENLARGEMENT OF SECTION 3 FIGURE 1)

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DENSITY VERSUS CONCENTRATION

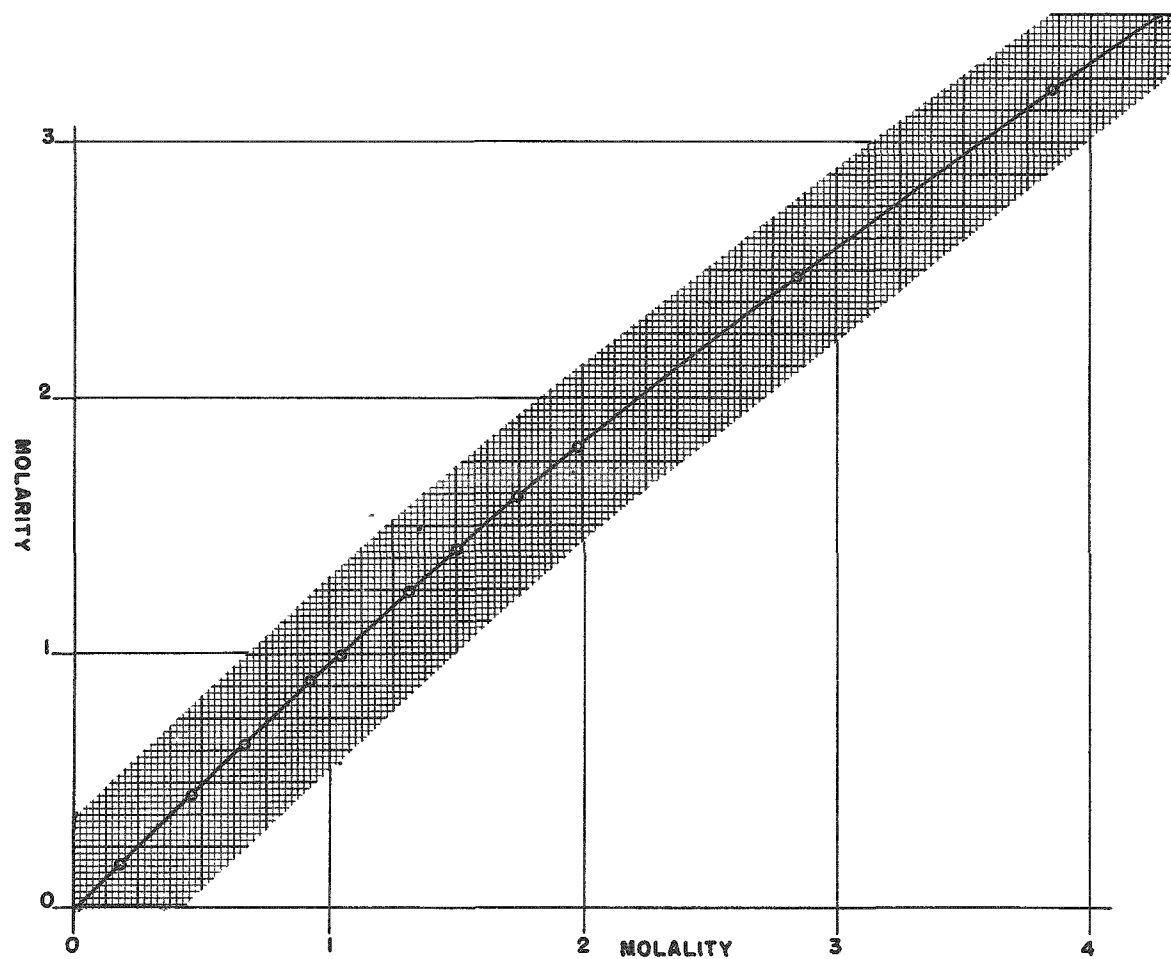
(ENLARGEMENT OF SECTION 2 FIGURE 1)

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FIGURE 5

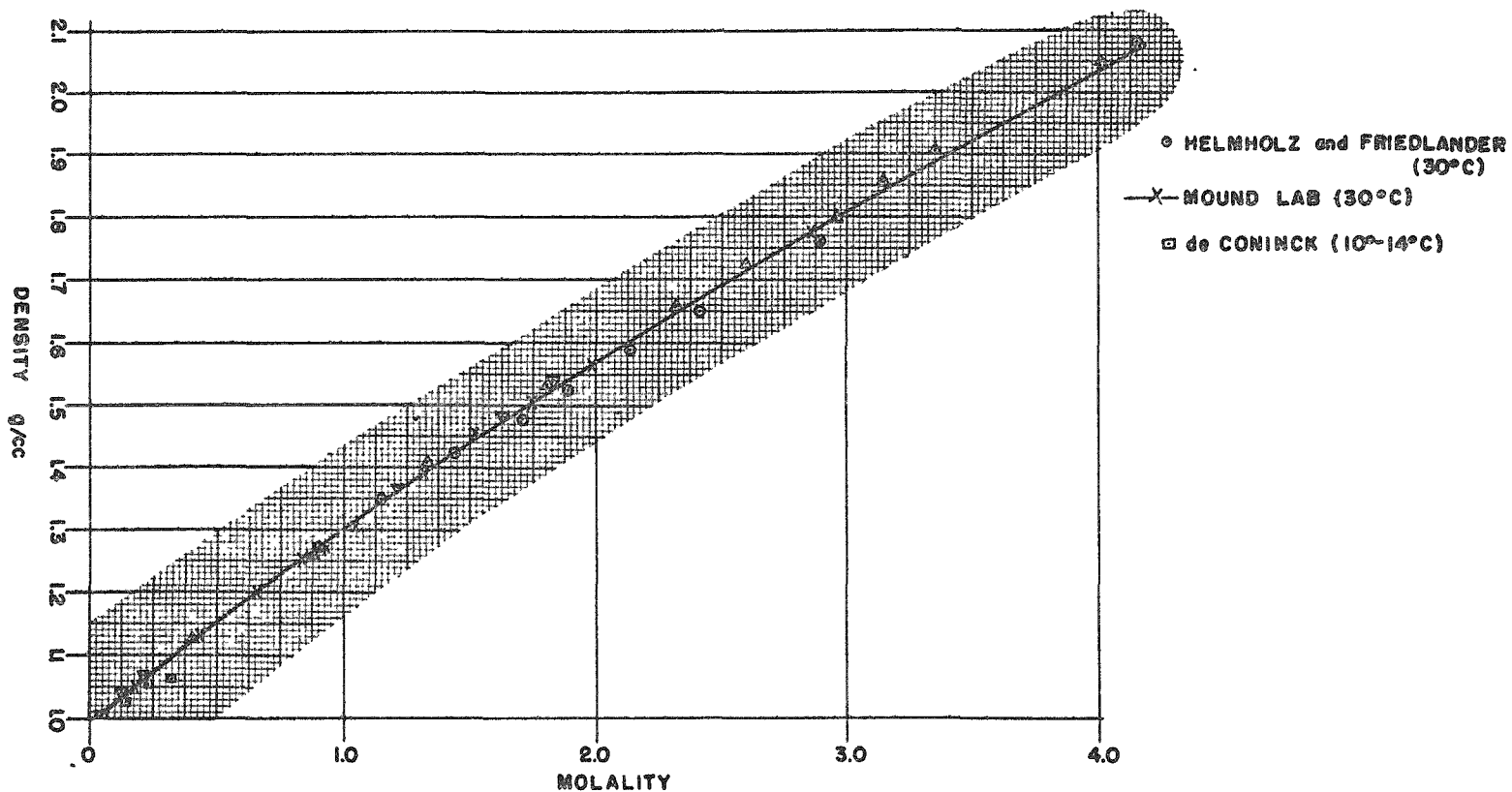
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MOLALITY VERSUS MOLALITY OF URANYL SULFATE SOLUTIONS AT 25°C

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DENSITY OF URANYL SULFATE SOLUTIONS
DATA OF VARIOUS EXPERIMENTERS

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FIGURE 6

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