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MASTER

THE MOLAL FREEZING POINT DEPRESSION CURVE OF URANIUM HEXAFLUORIDE AND ITS APPLICATION TO A METHOD OF ANALYSIS

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UNION CARBIDE CORPORATION
NUCLEAR DIVISION

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A B S T R A C T

The freezing point curves for the binary systems uranium hexafluoride-trichloroheptafluorobutane ($C_4Cl_3F_7$), uranium hexafluoride-perfluorodimethylcyclohexane (C_8F_{16}), and uranium hexafluoride-dichlorotetrafluoroethane ($C_2Cl_2F_4$) have been determined over the ranges of 0 to 2.5 mole percent of the individual solutes in uranium hexafluoride. In addition, the uranium hexafluoride-hydrogen fluoride system as determined by Wertz and Hedge (3) has been included on a molal basis. The observed molal freezing point depressions produced constants of 19.4, 19.6, 19.2, and 18.8°C per mole of solute added to 1000 grams of uranium hexafluoride, respectively. The average of these constants, 19.2 ± 0.7, is the molal freezing point constant.

The deviations resulting from non-ideality are discussed. Corrections for these deviations are applied by extension of observed freezing point depressions and ratio of liquid volume to total volume.

A method is introduced whereby the individual gross, liquid, and vapor compositions of the binary systems at their triple points may be estimated.

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THE MOLAL FREEZING POINT DEPRESSION CURVE OF URANIUM HEXAFLUORIDE AND ITS APPLICATION TO A METHOD OF ANALYSIS

Freezing point depression measurements offer a precise method for the determination of molal quantities of solutes in solutions. Many such molal freezing point constants have been established for various solvents as water, benzene, etc. The classical method for determining such constants may be found in the standard physical chemistry text books. In general, the method involves the dilution of known amounts of a solvent with known amounts of solutes, and measuring the resulting freezing point depression. From the Clausius-Clapeyron equation, the freezing point depression has the following theoretical relationship with the solute and solvent: $M_2 = 1000\ K_f \frac{W_2}{\Delta T W_1}$; where M_2 is the gram-molecular weight of the solute, W_2 is the grams of solute dissolved in W_1 grams of solvent, ΔT is the freezing point depression in degrees centigrade, and K_f is the molal freezing point constant in degrees centigrade per mole of solute per 1000 grams of solvent. The basic assumption of the equation is ideality of the solution.

Rutledge, Jarry, and Davis (2) discussed the deviations from ideality of the binary system uranium hexafluoride-hydrogen fluoride and prepared a freezing point diagram of the system. Wertz and Hedge (3) found the diagram invalid when applied to dilute solutions, and presented a method for correlating the solution freezing point with the gross composition, thus eliminating errors from calculations involving vapor-liquid compositions. They also presented a precise method of hydrogen fluoride analysis for the binary system uranium hexafluoride-hydrogen fluoride.

This method has been applied to three other solutes in uranium hexafluoride, and by extension of the data obtained the molal freezing point depression of 19.2 ± 0.7 degrees centigrade per mole of solute per 1000 grams of uranium hexafluoride was determined.

SUMMARY

The freezing point depressions obtained by introducing progressive quantities of trichloroheptafluorobutane, perfluorodimethylcyclohexane, and dichlorotetrafluoroethane into individual containers of uranium hexafluoride were used to determine the molal freezing point constant for the solvent. The freezing point depressions obtained by Wertz and Hedge (3) using hydrogen fluoride dilutions in uranium hexafluoride, were also employed. For each of the binary systems studied, the estimated molal freezing point constant for uranium hexafluoride was 19.4, 19.6, 19.2, and 18.8, respectively; the average of these being 19.2 ± 0.7 degrees centigrade per mole of solute per 1000 grams of uranium hexafluoride.

A method for estimating the gross, vapor, and liquid compositions of binary uranium hexafluoride solutions at their triple points is presented. The method needs only the freezing point depression and the ratio of

liquid volume to total volume.

EQUIPMENT

Apparatus

1. A 785-ml freezing point cell, 7-1/2 inches long and 3-1/2 inches in diameter with hemispherical ends, was equipped with a valve and a well for a platinum resistance thermometer (figure 1).

2. The solute container was a 3/8-inch diameter nickel tube closed at one end and equipped with a small Hoke valve at the opposite end (figure 2). An analytical balance was used to weigh the assembly to ± 0.1 milligrams.

3. A hot water bath was used to liquefy the uranium hexafluoride in the freezing point cell.

4. The constant temperature bath was a stainless steel Dewar flask, 6 inches in diameter and 14 inches deep and was surrounded by a cork block 12 inches square and 16 inches deep. The temperature of the bath, 0.1 to 0.3°C below the freezing point of the uranium hexafluoride, was maintained by a mercury thermo-regulator operating a heating element through an Emil Greiner electronic relay. A propeller driven by a 1525 rpm motor circulated the water in the bath.

5. A Leeds and Northrup calibrated platinum resistance thermometer, in conjunction with a Mueller temperature bridge and a Minneapolis-Honeywell Electronik null indicator was used to obtain the cooling curves of uranium hexafluoride. The platinum resistance thermometer was checked daily at the freezing point of distilled water.

Materials

1. Isotopically unaltered uranium hexafluoride containing less than 0.01 weight percent hydrogen fluoride was further purified by vacuum transferring the vapor portion of the material at 0 degrees centigrade. The freezing point of the purified material was within experimental error of 64.052 degrees centigrade, the triple point of pure uranium hexafluoride (1).

2. Trichloroheptafluorobutane was molecularly distilled, and the distilled material analyzed by infrared and mass spectrometer techniques. No impurities were found by the analyses. The material was vacuum transferred over "Drierite" into a two-inch nickel cylinder with a valve. A sample of the packaged material was withdrawn into a 3/8-inch fluorothene tube containing approximately 0.5 grams of uranium hexafluoride. No yellow precipitate formed, thus indicating the absence of appreciable amounts of water in the trichloroheptafluorobutane.

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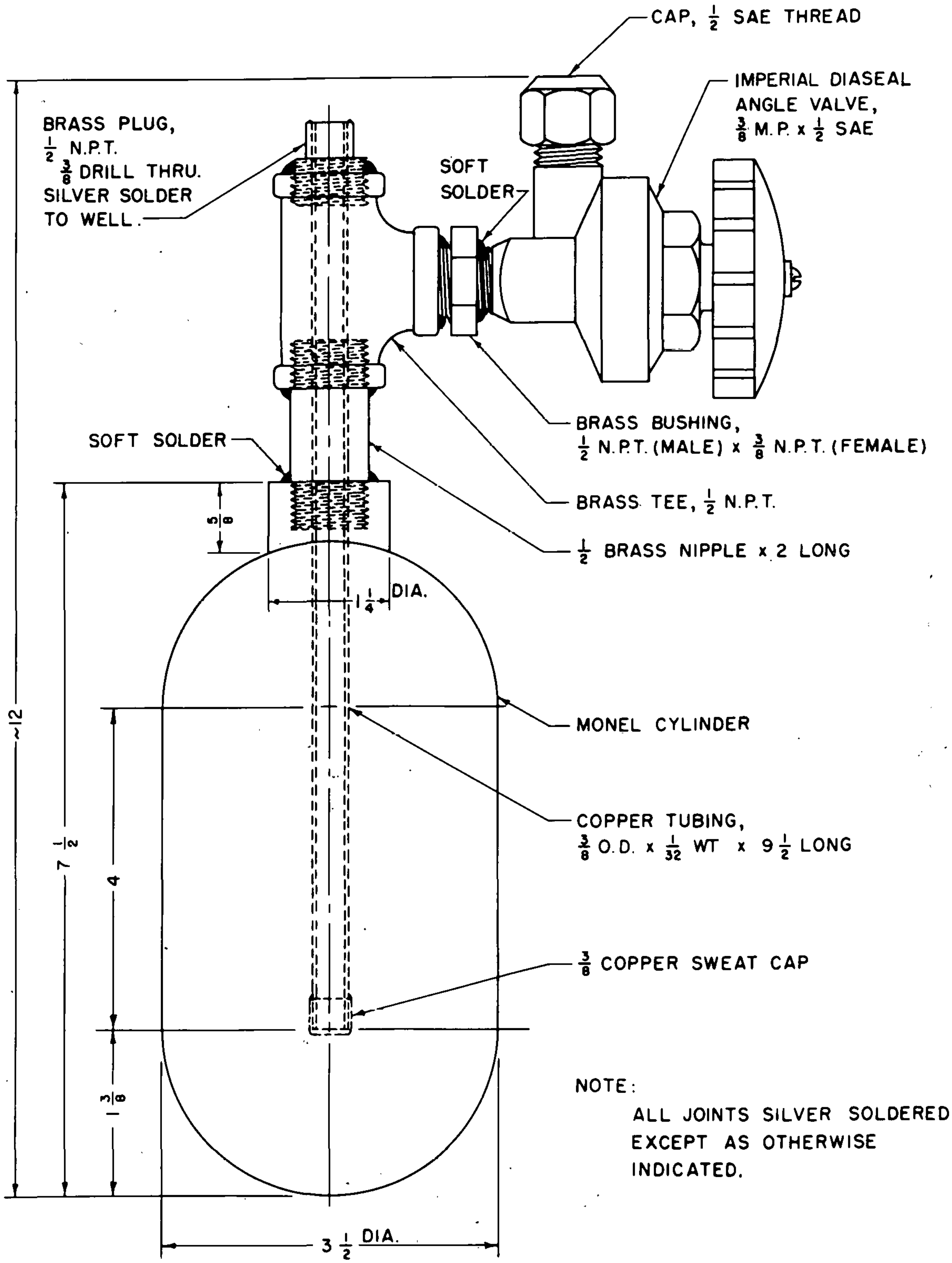

Figure 1
FREEZING POINT CYLINDER

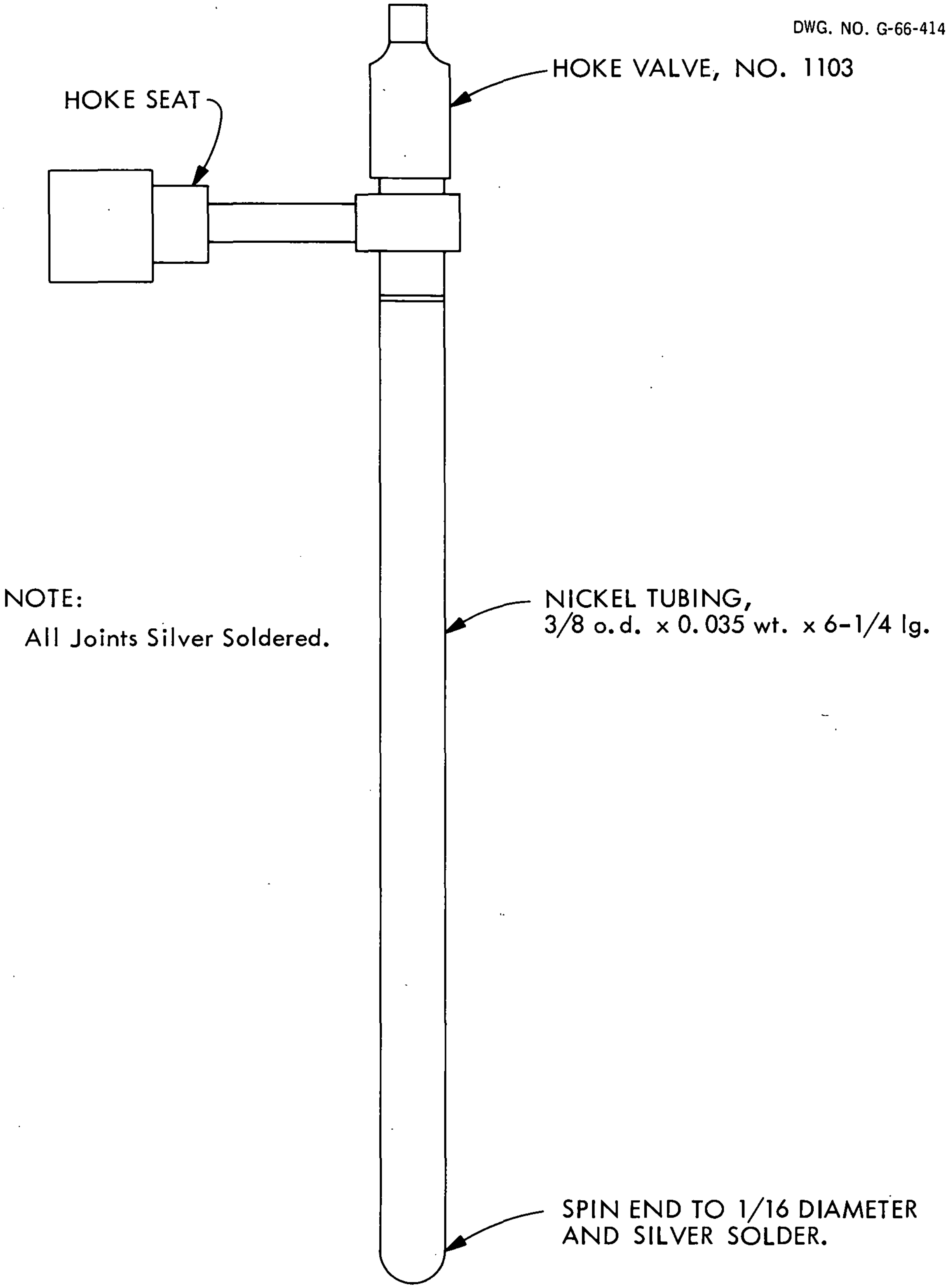

Figure 2
SOLUTE CONTAINER

3. The perfluorodimethylcyclohexane analyses by infrared and mass spectrometer techniques indicated no impurities. The material was packaged and tested for moisture as above.

4. The dichlorotetrafluoroethane analyses by infrared and mass spectrometer techniques indicated no impurities. The material was packaged and tested for moisture as above.

EXPERIMENTAL AND PROCEDURE

Preparing the Solutions

The purified uranium hexafluoride in each of the freezing point cells was measured for its freezing point to confirm its purity. Incremental quantities of the desired solute were packaged in the solute container, and vapor transferred to the freezing point cells by liquid nitrogen condensation. The weight of solute transferred was determined by the difference of weight before and after transfer as weighed to ± 0.1 mg on an analytical balance.

Measuring the Freezing Point

Two ml of mineral oil was placed in the thermowell of the freezing point cell to act as the heat transfer agent between the well and the platinum resistance thermometer. The freezing point cell was placed in the hot water bath to liquefy its contents. Once liquefied the cell was placed in the constant temperature bath which was adjusted to a temperature slightly below the expected freezing point. The cooling curve was followed by the Mueller bridge reading from the platinum resistance thermometer. When the solution supercooled, the cell was tapped to start crystallization.

Data

The freezing point depressions as $\Delta T(64.052^{\circ}C$ - freezing point), mole fractions, weight percents, and ratios of liquid volume to total volume are shown in table I for the uranium hexafluoride-trichloroheptafluorobutane synthetic binary mixtures. Figure 3 is a plot of the data from table I. The data for the system uranium hexafluoride-perfluorodimethylcyclohexane is shown in table II. Figure 4 is a plot of the data from table II. Table III is the data obtained for the binary mixtures of uranium hexafluoride-dichlorotetrafluoroethane. Figure 5 is a plot of the data contained in table III. Table IV contains the equations for the systems observed, including that of uranium hexafluoride-hydrogen fluoride as determined by Wertz and Hedge (3).

TABLE I

FREEZING POINT OF URANIUM HEXAFLUORIDE-TRICHLOROHEPTAFLUOROBUTANE AT DIFFERENT RATIOS OF LIQUID VOLUME TO TOTAL VOLUME

Cylinder	Ratio	ΔT	95% C. L.	Mole Fractions	Weight Percent
MD 2121	0.862	0.000	0.008	0.00000	0.000
(2483 g UF_6)		0.089	0.008	0.00134	0.110
		0.180	0.017	0.00258	0.211
		0.239	0.013	0.00371	0.304
		0.311	0.014	0.00500	0.409
		0.508	0.015	0.00802	0.656
		0.615	0.012	0.00992	0.812
		0.801	0.011	0.01222	1.000
		0.843	0.021	0.01390	1.138
		0.971	0.016	0.01575	1.272
		1.094	0.153	0.01811	1.460
		1.144	0.048	0.01916	1.543
		1.273	0.071	0.02187	1.758
MD 2108	0.707	0.000	0.010	0.00000	0.000
(2038 g UF_6)		0.081	0.005	0.00116	0.095
		0.127	0.004	0.00216	0.176
		0.225	0.016	0.00365	0.298
		0.310	0.012	0.00502	0.411
		0.463	0.040	0.00709	0.580
		0.539	0.022	0.00861	0.704
		0.707	0.020	0.01080	0.884
		0.741	0.019	0.01213	0.993
		0.895	0.020	0.01392	1.126
		0.961	0.023	0.01545	1.248
		1.048	0.041	0.01664	1.342
		1.163	0.051	0.01738	1.402
MD 2133	0.396	0.000	0.004	0.00000	0.000
(1141 g UF_6)		0.109	0.016	0.00121	0.099
		0.209	0.009	0.00254	0.208
		0.330	0.024	0.00396	0.323
		0.402	0.011	0.00507	0.414
		0.445	0.014	0.00598	0.490
		0.566	0.018	0.00778	0.636
		0.670	0.008	0.00933	0.763
		0.775	0.009	0.01048	0.858
		0.985	0.080	0.01309	1.060
		1.086	0.041	0.01546	1.251
		1.193	0.002	0.01712	1.381
		1.423	0.018	0.01866	1.504

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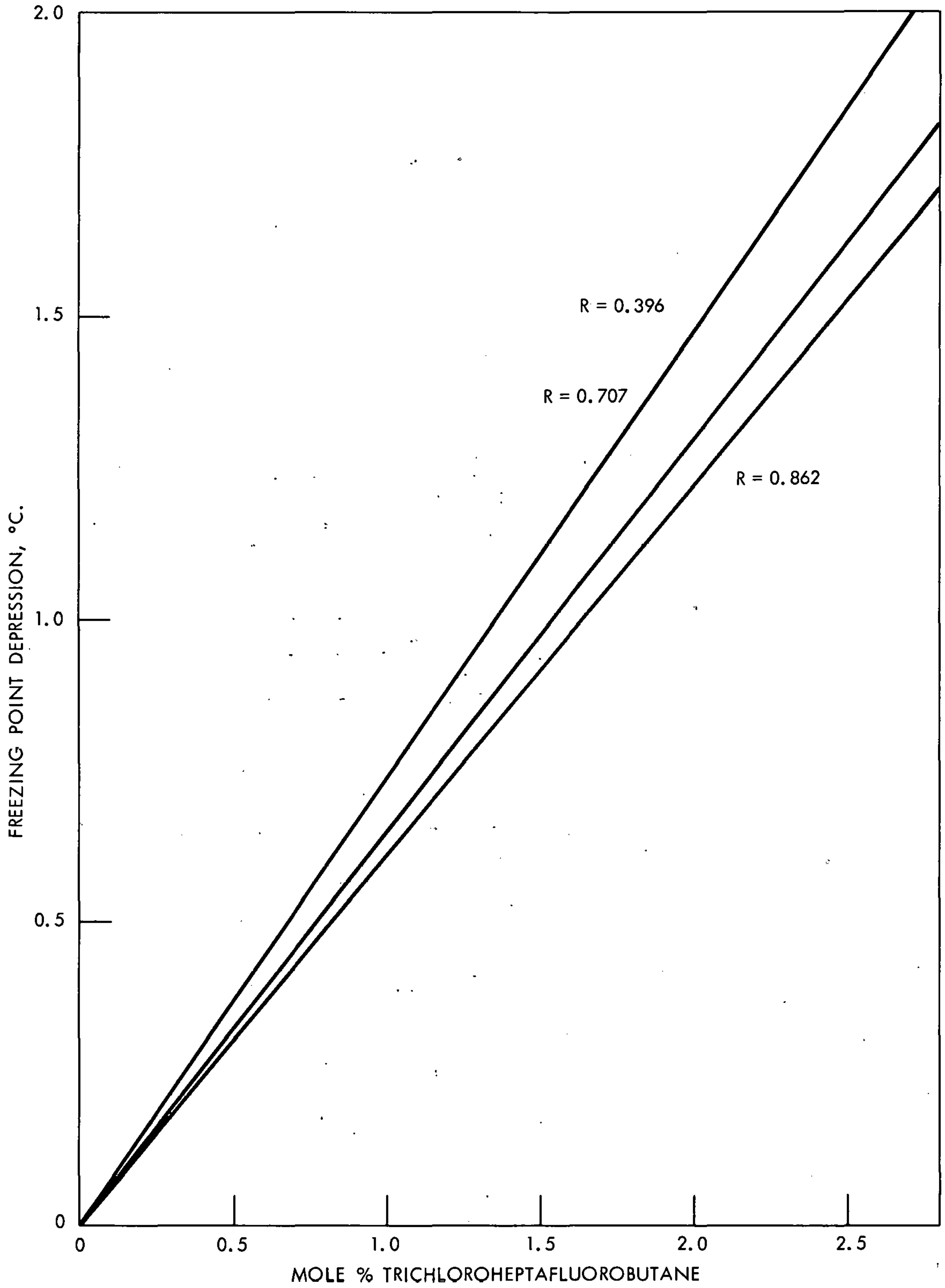

Figure 3

FREEZING POINT CURVES OF THE UF_6-$C_4Cl_3F_7$ IN FREEZING POINT CELLS WITH DIFFERENT LIQUID VOLUME TO TOTAL VOLUME RATIOS

TABLE II

FREEZING POINT OF URANIUM HEXAFLUORIDE-PERFLUORODIMETHYLCYCLOHEXANE AT DIFFERENT RATIOS OF LIQUID VOLUME TO TOTAL VOLUME

Cylinder	Ratio	ΔT	95% C. L.	Mole Fractions	Weight Percent
ORGDP 13					
(2496 g UF_6)	0.889	0.0000	0.011	0.00000	0.000
		0.248	0.023	0.00410	0.248
		0.519	0.009	0.00855	0.970
		0.742	0.047	0.01287	1.460
		0.927	0.075	0.01768	2.004
		1.280	0.052	0.02182	2.472
MD 2214					
(2040 g UF_6)	0.708	0.000	0.018	0.00000	0.000
		0.061	0.011	0.00063	0.071
		0.155	0.055	0.00160	0.182
		0.230	0.052	0.00298	0.339
		0.373	0.021	0.00498	0.566
		0.433	0.026	0.00682	0.774
		0.571	0.017	0.00881	1.000
		0.747	0.012	0.01248	1.415
		0.858	0.013	0.01421	1.612
		1.097	0.040	0.01758	1.993
		1.393	0.021	0.02254	2.553
MD 2133					
(1200 g UF_6)	0.416	0.000	0.009	0.00000	0.000
		0.129	0.029	0.00102	0.116
		0.202	0.016	0.00181	0.206
		0.277	0.019	0.00344	0.391
		0.405	0.026	0.00530	0.599
		0.469	0.055	0.00704	0.799
		0.599	0.013	0.00812	0.921
		0.813	0.060	0.01079	1.155
		0.880	0.008	0.01185	1.345
		1.140	0.012	0.01451	1.645
		1.587	0.026	0.01871	2.121

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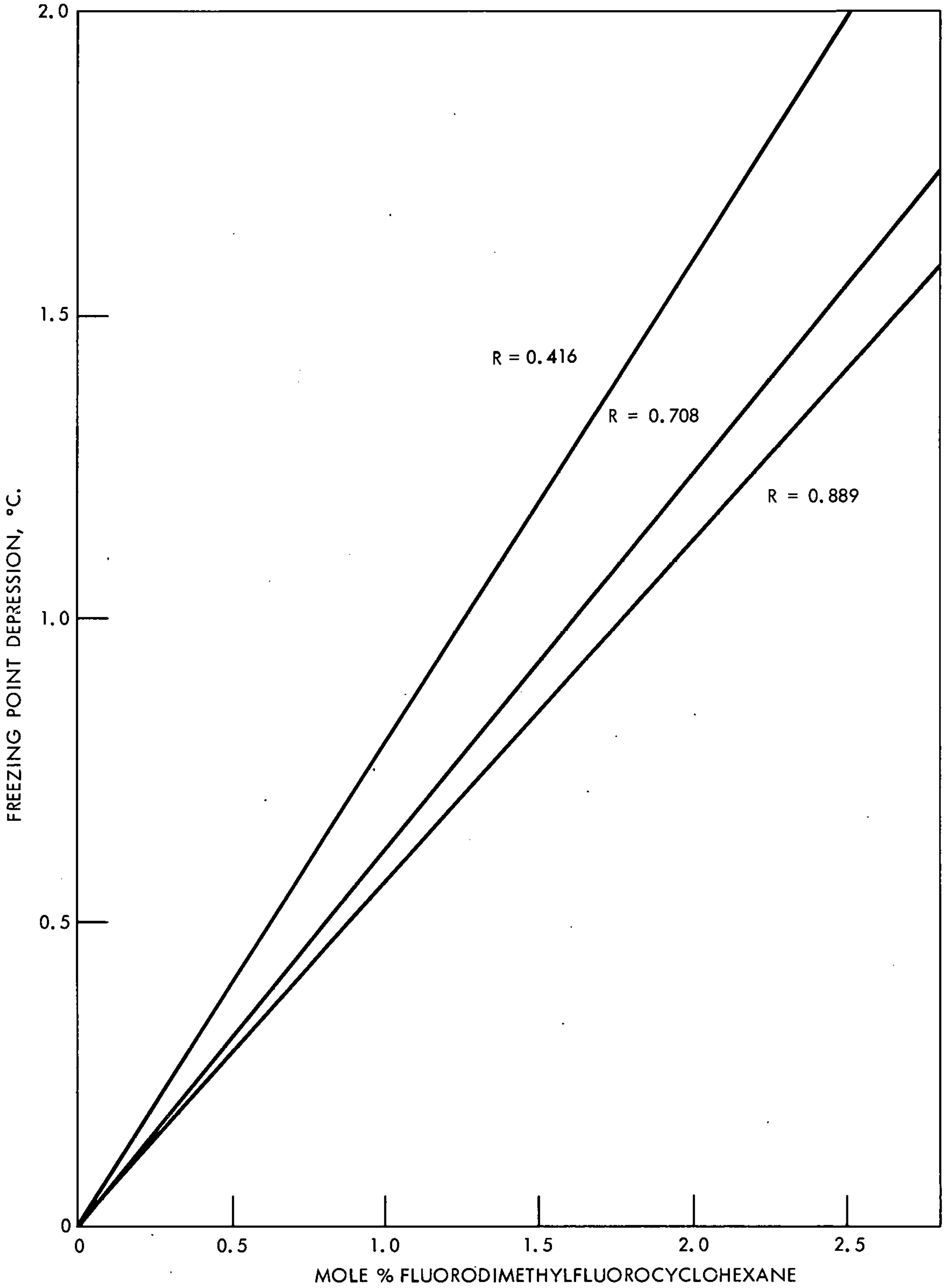

Figure 4

FREEZING POINT CURVES OF THE UF6-C_8F_{16} IN FREEZING POINT CELLS WITH DIFFERENT LIQUID VOLUME TO TOTAL VOLUME RATIOS

TABLE III

FREEZING POINT OF URANIUM HEXAFLUORIDE-DICHLOROTETRAFLUOROETHANE AT DIFFERENT RATIOS OF LIQUID VOLUME TO TOTAL VOLUME

Cylinder	Ratio	ΔT	± 95% C. L.	Mole Fractions	Weight Percent
MD 2108	0.897	0.000	0.010	0.00000	0.000
(2584 g UF_6)		0.097	0.006	0.00152	0.074
		0.201	0.009	0.00333	0.162
		0.313	0.023	0.00505	0.246
		0.442	0.012	0.00722	0.352
		0.611	0.010	0.01003	0.490
		0.737	0.005	0.01236	0.604
		0.919	0.014	0.01507	0.738
		1.071	0.009	0.01776	0.870
		1.246	0.011	0.02078	1.020
		1.391	0.041	0.02380	1.170
		1.583	0.024	0.02652	1.306
		1.728	0.046	0.02910	1.434
		1.881	0.022	0.03148	1.554
		2.045	0.033	0.03410	1.685
		2.200	0.032	0.03667	1.815
		2.395	0.038	0.03934	1.950
MD 2121	0.687	0.000	0.004	0.00000	0.000
(1977 g UF_6)		0.023	0.008	0.00029	0.014
		0.091	0.008	0.00149	0.072
		0.146	0.011	0.00241	0.117
		0.229	0.014	0.00369	0.180
		0.336	0.007	0.00560	0.273
		0.445	0.021	0.00713	0.348
		0.542	0.025	0.00888	0.433
		0.678	0.037	0.01084	0.529
		0.799	0.008	0.01320	0.645
		0.899	0.022	0.01434	0.702
		1.053	0.054	0.01656	0.811
		1.145	0.011	0.01811	0.888
		1.287	0.030	0.02050	1.006
		1.560	0.044	0.02420	1.190
		1.723	0.069	0.02753	1.357
		1.880	0.092	0.02969	1.464
		2.017	0.062	0.03228	1.5866
		2.164	0.061	0.03576	1.761
		2.417	0.060	0.03967	1.957
		2.629	0.054	0.04254	2.102

TABLE III

FREEZING POINT OF URANIUM HEXAFLUORIDE-DICHLOROTETRAFLUOROETHANE AT DIFFERENT RATIOS OF LIQUID VOLUME TO TOTAL VOLUME
(continued)

Cylinder	Ratio	ΔT	± 95% C. L.	Mole Fractions	Weight Percent
MD 2073	0.404	0.000	0.012	0.00000	0.000
(1164 g UF_6)		0.041	0.008	0.00053	0.026
		0.073	0.011	0.00101	0.049
		0.145	0.008	0.00206	0.101
		0.227	0.018	0.00359	0.176
		0.319	0.014	0.00514	0.252
		0.428	0.043	0.00731	0.359
		0.603	0.039	0.00957	0.470
		0.719	0.047	0.01169	0.575
		0.801	0.027	0.01304	0.642
		0.949	0.076	0.01433	0.706
		1.067	0.030	0.01656	0.817
		1.232	0.013	0.01874	0.926
		1.395	0.029	0.02145	1.060
		1.551	0.038	0.02419	1.198
		1.669	0.066	0.02526	1.252
		1.812	0.020	0.02773	1.376
		2.009	0.040	0.03195	1.581
		2.256	0.074	0.03571	1.771
		2.425	0.046	0.03983	1.979
		2.626	0.066	0.04340	2.161

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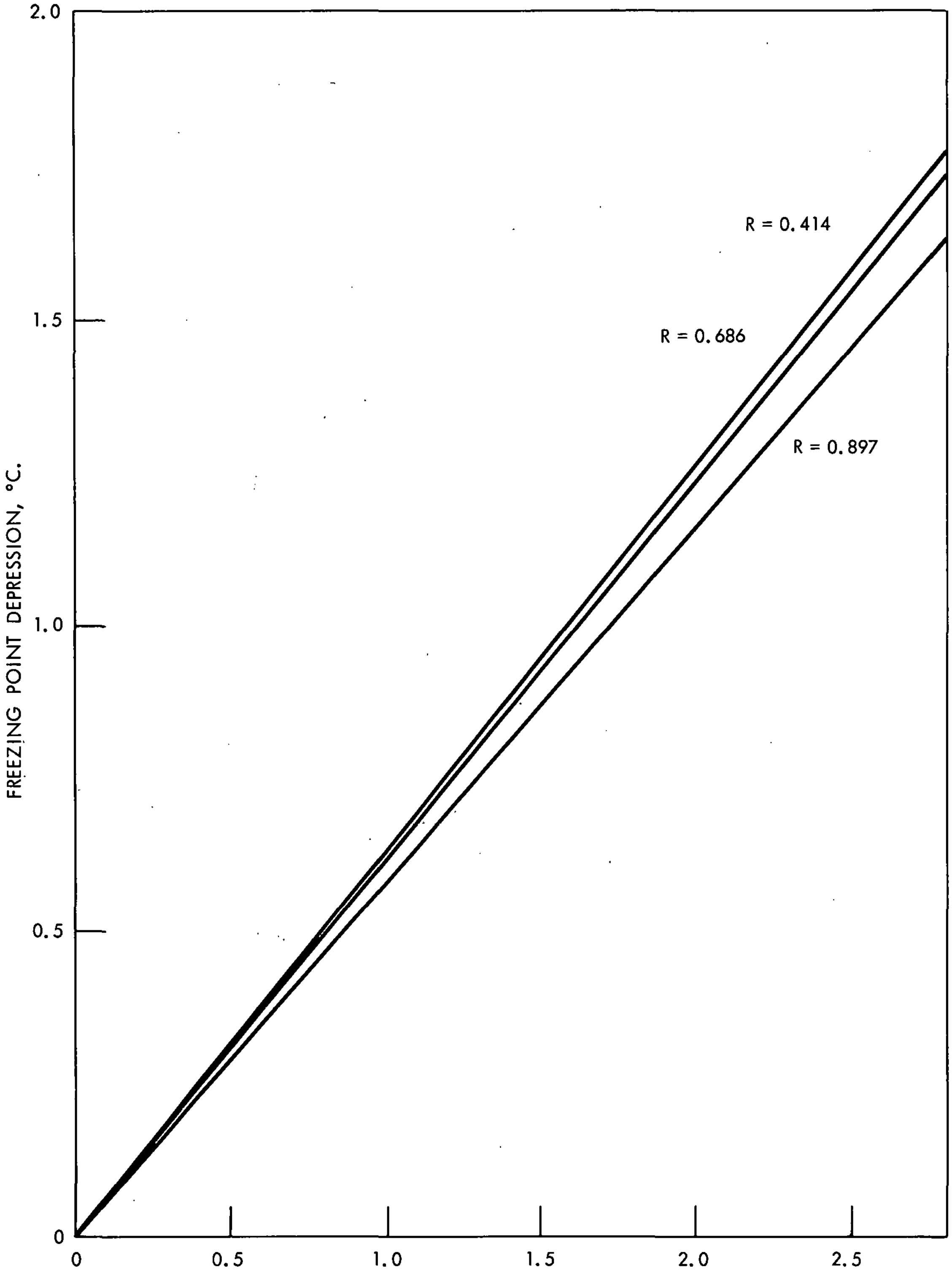

Figure 5

FREEZING POINT CURVES OF THE UF_6-$C_2Cl_2F_4$ IN FREEZING POINT CELLS WITH DIFFERENT LIQUID VOLUME TO TOTAL VOLUME RATIOS

TABLE IV

FREEZING POINT EQUATIONS FOR OBSERVED SOLUTES AT MEASURED LIQUID VOLUME TO TOTAL VOLUME RATIOS

Solute	Liquid to Total Volume	Equation
Trichloroheptafluorobutane	0.862	$\Delta T = 59.37\ M_f$
	0.707	$\Delta T = 63.94\ M_f$
	0.396	$\Delta T = 73.36\ M_f$
Perfluorodimethylcyclohexane	0.889	$\Delta T = 56.97\ M_f$
	0.708	$\Delta T = 62.33\ M_f$
	0.416	$\Delta T = 79.42\ M_f$
Dichlorotetrafluoroethane	0.897	$\Delta T = 58.31\ M_f$
	0.686	$\Delta T = 62.08\ M_f$
	0.414	$\Delta T = 62.96\ M_f$
Hydrogen Fluoride (3)	0.926	$\Delta T = 51.4\ M_f$
	0.715	$\Delta T = 45.3\ M_f$
	0.415	$\Delta T = 36.9\ M_f$

Freezing Point Curve Equations

If the ΔT-mole fraction constants of the equations in table IV are plotted against their corresponding volume ratios for each binary system, smooth curves may be fitted through the three observed points to a volume ratio of one, representing a cylinder containing liquid only. Figure 6 is a plot of the curves obtained in this manner. The extrapolations of the curves to a ratio of one yield the equation constants in table V.

TABLE V

FREEZING POINT EQUATIONS FOR THE VOLUME RATIO OF ONE

Solute	Equation*
Trichloroheptafluorobutane	$\Delta T = 55.2\ M_f$
Perfluorodimethylcyclohexane	$\Delta T = 55.6\ M_f$
Dichlorotetrafluoroethane	$\Delta T = 54.6\ M_f$
Hydrogen Fluoride	$\Delta T = 53.3\ M_f$

*ΔT = 64.052°C minus the freezing point of the solution.
M_f = mole fraction of solute in the solution

The arithmetic average of the equation constants in table V is 54.7 with a 95 percent confidence interval of ± 1.9. Assuming an infinitely dilute solution, this average corresponds to the molal freezing point constant: 19.2 ± 0.7 degrees centigrade per mole of solute per 1000 grams of uranium hexafluoride. This empirical constant compares to a theoretical constant of 17.4 derived from the heat of fusion of uranium hexafluoride (1); and to an ideal constant of 16.6 based on Raoult's law of partial pressures.

APPLICATION TO METHODS OF ANALYSIS

Determining Molecular Weights of Solutes

All of the binary solutions observed in this study were nonideal. With each, the vapor above the liquid contained a proportionately greater amount of solvent (uranium hexafluoride) than did the liquid. The hydrogen fluoride system studied by Wertz and Hedge (3) was reversed from these, with a proportionately greater amount of solute (hydrogen fluoride) in the vapor than in the liquid. Corrections for liquid-vapor composition differences for the observed binary systems are obtained by applying the freezing point depression constants corresponding to known liquid volume to total volume ratios as found in figure 6.

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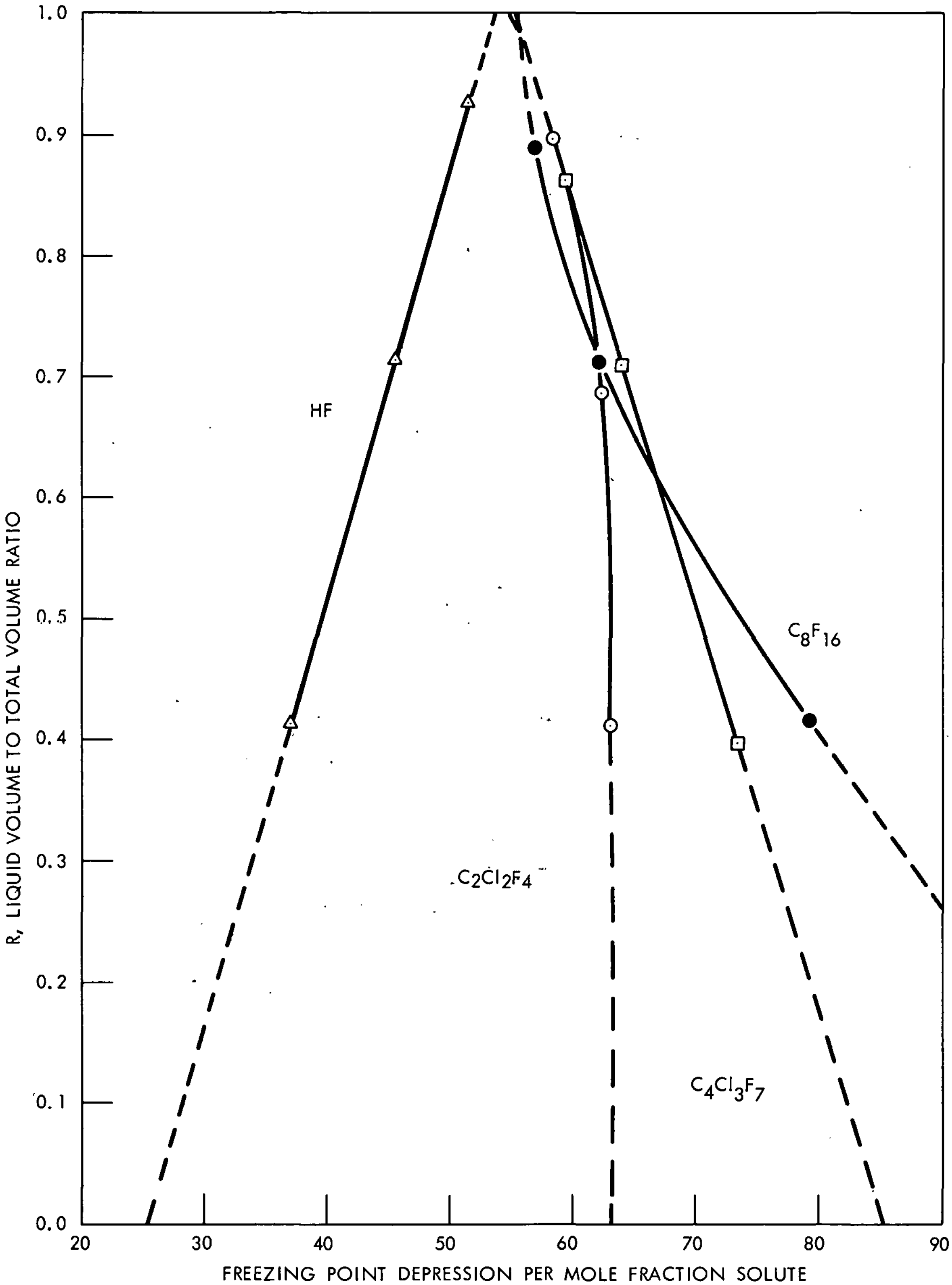

Figure 6

FREEZING POINT DEPRESSION CONSTANTS VERSUS THE LIQUID VOLUME TO TOTAL VOLUME RATIO

TABLE VI

EQUATION CONSTANTS OF OBSERVED SOLUTES
FOR DIFFERENT LIQUID VOLUME TO TOTAL VOLUME RATIOS

Liquid Volume to Total Volume Ratio	$C = \Delta T/M_f$			
	$C_4Cl_3F_7$	C_8F_{16}	$C_2Cl_2F_4$	HF
0.00	85.1	104.8	63.1	25.4
0.36	74.4	81.8	63.0	35.5
0.38	73.8	80.7	63.0	36.1
0.40	73.2	79.4	63.0	36.7
0.42	72.6	73.2	62.9	37.4
0.44	72.0	72.0	62.9	37.9
0.46	71.4	75.6	62.9	38.4
0.48	70.8	74.4	62.9	39.0
0.50	70.2	73.1	62.8	39.5
0.52	69.6	71.9	62.8	40.0
0.54	69.0	70.7	62.8	40.6
0.56	68.4	69.5	62.7	41.2
0.58	67.8	68.4	62.6	41.7
0.60	67.2	67.3	62.5	42.3
0.62	66.6	66.3	62.4	42.8
0.64	66.0	65.3	62.3	43.4
0.66	65.4	64.4	62.2	44.0
0.68	64.8	63.5	62.1	44.5
0.70	64.2	62.6	62.0	45.0
0.72	63.6	61.8	61.8	45.6
0.74	63.0	61.1	61.7	46.2
0.76	62.4	60.4	61.4	46.8
0.78	61.8	59.8	61.2	47.4
0.80	61.2	59.1	60.9	47.9
0.82	60.6	58.6	60.5	48.5
0.84	60.0	58.1	60.1	49.1
0.86	59.4	57.6	59.6	49.7
0.88	58.8	57.2	58.6	50.2
0.90	58.2	56.8	58.3	50.7
1.00	55.2	55.6	54.6	53.3

Table VI was compiled from the curves in figure 6. The constants in table VI correspond to C in the general equation, $\Delta T = CM_f$, where ΔT is the freezing point depression in degrees centigrade and M_f is the mole fraction of solute. These constants are applicable, at their corresponding ratios of liquid volume to total volume, to a minimum of 2.5 mole percent of solute in solution. The constants reflect the gross compositions of the observed binary systems in terms of their freezing point depressions.

Using the molal freezing point depression constant and assuming a solution with only a liquid phase, the molecular weight of a solute can be determined. By substituting 19.2 for K_f (molal freezing point constant), 0.01 for W_2 (weight of solute), and 100 for W_1 (weight of solvent), into the Clausius - Clapeyron equation, $M_2 = 1000\ K_f \frac{W_2}{W_1 \Delta T}$; the equation becomes $M_2 = (1000)(19.2)\ \frac{0.01}{100\ \Delta T}$. This is equivalent to $M_2 = 1.92/\Delta T$. Figure 7 is a logarithmic plot of this equation with molecular weights versus cryoscopic constants as ΔT/0.01 weight percent solute. Superimposed on the plot are the individual solute values with a straight line drawn through them. The two lines are not significantly different. To determine the molecular weight of a solute in solution with uranium hexafluoride, only the freezing point depression and weight percent solute values need be known. Thus, if 0.01 weight percent of solute in uranium hexafluroide produces a freezing point depression of 0.0068°C, the solute has a molecular weight of 287.5. Conversely, if the molecular weight is known to be 287.5, the freezing point depression is 0.0068°C.

The Determination of Gross, Liquid, and Vapor Composition of Binary Systems at Their Freezing Points

The gross, liquid, and vapor compositions of ideal binary solutions are identical; for non-ideal solutions, they are different. The disparity between the phases can be predicted from empirical cryoscopic data and liquid volume to total volume ratios. Figure 8 is a plot of the trichloroheptafluorobutane - uranium hexafluoride system. It is constructed by substituting ΔT into the trichloroheptafluorobutane equations from table IV and solving for M_f. The line labeled R = 0 is obtained from substitutions in a similar equation using C for $C_4Cl_3F_7$ from table VI at a liquid volume to total volume ratio of zero; e.g., for R = 0, $\Delta T = 85.1\ M_f$. The line labeled R = 1 is obtained by using the equation, $\Delta T = 54.7\ M_f$, which contains the molal freezing point constant on a mole fraction basis, and applies to dilute solutions of all solutes in uranium hexafluoride.

A solution in a 785 ml freezing point cell containing 0.040 moles of trichloroheptafluorobutane and 7.124 moles of uranium hexafluoride is represented by the dashed lines in figure 8. The liquid volume of the solution above is (7.124)(352)/3.67 = 682 ml, assuming an infinitely dilute solution at 64.052°C (4). The liquid volume to total volume ratio is then 682/785 = 0.87. The gross composition as mole fraction solute

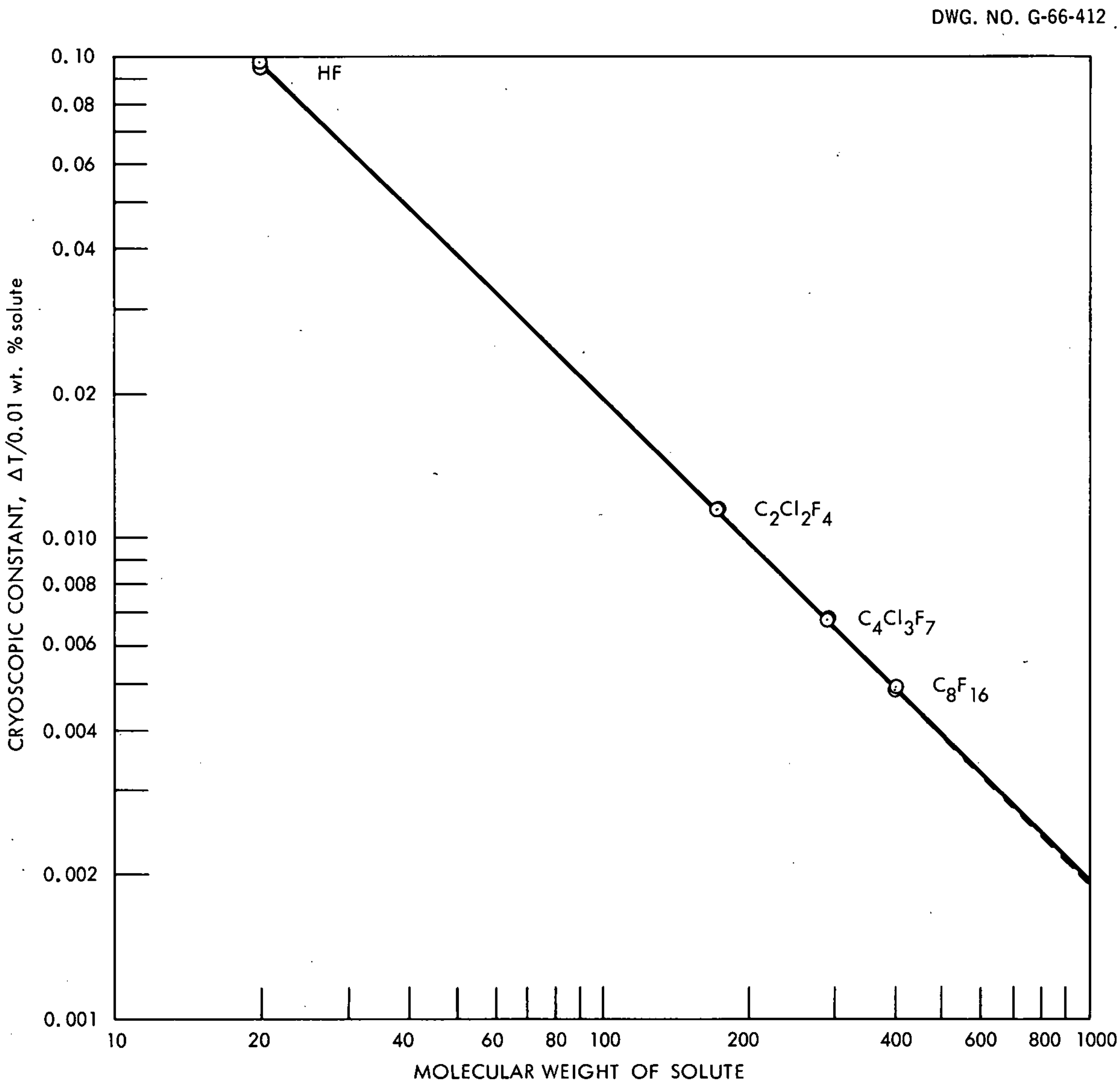

Figure 7

GRAPHICAL RELATIONSHIP OF A SOLUTE'S MOLECULAR WEIGHT AND ITS CRYOSCOPIC CONSTANT

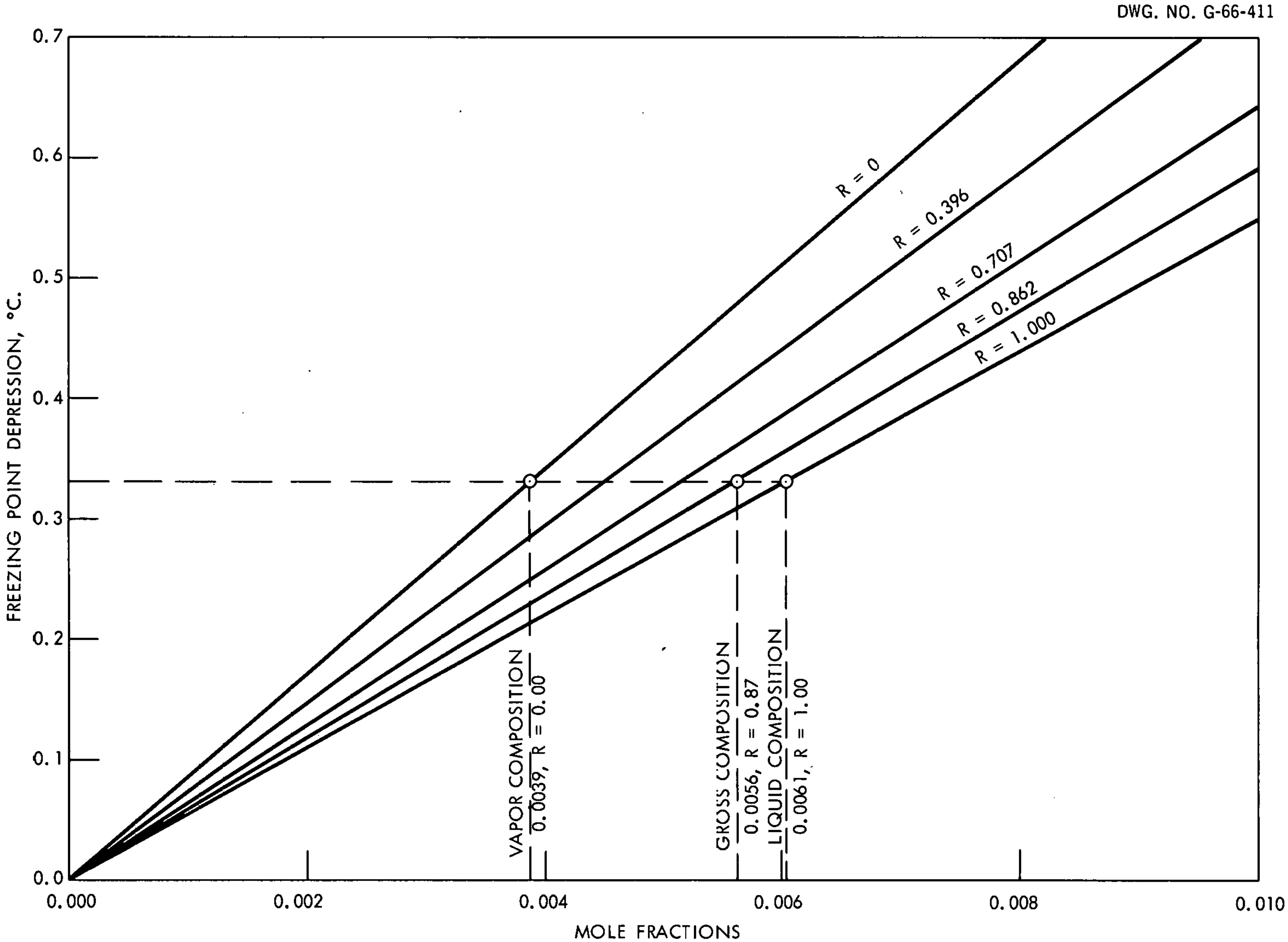

Figure 8

GROSS, VAPOR, AND LIQUID COMPOSITION OF THE SYSTEM UF_6-$C_4Cl_3F_7$

is 0.040/7.124 = 0.0056. The freezing point depression is 0.33°C. The intersection of the horizontal dashed line from the freezing point depression of 0.33°C with the volume ratio line of R = 0 is the mole fraction of solute in the vapor, 0.0039; the intersection with R = 0.87 is the gross composition, 0.0056; and the intersection with R = 1.0 is the liquid composition of 0.0061. If samples are withdrawn from this solution so that the liquid volume to total volume ratio becomes 0.396, the proportion of uranium in the cylinder correspondingly increases and the uranium composition of the liquid and vapor phases readjust. The composition of the remaining solution after withdrawing the samples is determined by the following procedure: Trace the horizontal dashed line in figure 8 until the intersection with the R = 0.396 curve is obtained. From this intersection, proceed downward until the intersection with R = 0.87 is reached. Horizontally extend a line from the latter intersection. The intersection of the extended horizontal line with the R = 0 curve is the vapor composition, 0.0032 mole fractions; the intersection with the R = 0.396 curve is the gross composition, 0.0037 mole fractions; and the intersection with the R = 1 curve is the liquid composition, 0.0047 mole fractions.

The accuracy of estimating the liquid composition by this procedure was validated by successively withdrawing 10-gram liquid samples from a 785 ml cell (figure 1). The cell originally contained 2460 grams of uranium hexafluoride diluted with trichloroheptafluorobutane to a uranium concentration of 67.267 percent. The freezing point depression of this solution was 0.38°C. Sets of five samples were withdrawn as a liquid from the cell and each individual sample of approximately 10 grams was gravimetrically analyzed for uranium. After withdrawing each group of five samples, the cell was weighed to ± 1 gram. Table VII shows the weight of solution remaining in the cylinder after sampling; the liquid volume to total volume ratios of the remaining solution; the calculated gross, vapor, and liquid compositions at the averaged volume ratios obtained by following the procedure described above; and the averaged gravimetric results obtained on each of two sets (10 samples) of liquid withdrawn.

Figure 9 graphically represents the data from table VII. The calculated values are extrapolated to total liquid and to total vapor volume ratios. At a liquid volume to total volume ratio of one, the gross and liquid compositions become identical; and at a volume ratio of zero, the gross and vapor compositions become identical. The dashed line represents the gravimetric uranium results as calculated from a linear least squares fit. The 95% confidence interval about the least squares line does encompass the calculated liquid uranium concentration curve, indicating no significant difference between the calculated and measured values. The confidence interval does not encompass the original uranium concentration line, indicating a difference between the original uranium concentration and the liquid and least squares gravimetric concentrations.

TABLE VII

URANIUM CONCENTRATION DISTRIBUTION COMPARISONS FOR THE SYSTEM CONTAINING 2460 GRAMS OF UF_6 AND 12.8 GRAMS OF C-437

Grams of Solution	Liquid Volume to Total Volume Ratio	Calculated Percent Uranium			Analyzed Percent Uranium in Liquid
		Gross	Vapor	Liquid	
2310	0.800	67.285	67.380	67.248	67.230
2215	0.768	67.295	67.383	67.252	67.269
2095	0.738	67.304	67.386	67.257	67.242
1985	0.689	67.318	67.392	67.267	67.254
1790	0.622	67.336	67.400	67.275	67.348
1640	0.570	67.349	67.405	67.285	67.295
1520	0.528	67.359	67.410	67.291	67.271
1405	0.488	67.367	67.414	67.297	67.335
1315	0.457	67.373	67.416	67.302	67.272
1110	0.385	67.388	67.421	67.312	67.324
990	0.344	67.395	67.425	67.317	67.304
880	0.305	67.402	67.428	67.323	67.296
775	0.269	67.407	67.431	67.327	67.298
680	0.236	67.412	67.433	67.331	67.292
575	0.200	67.417	67.436	67.335	67.311
465	0.161	67.422	67.439	67.339	67.335
385	0.134	67.425	67.440	67.342	67.333
298	0.104	67.430	67.452	67.345	67.338

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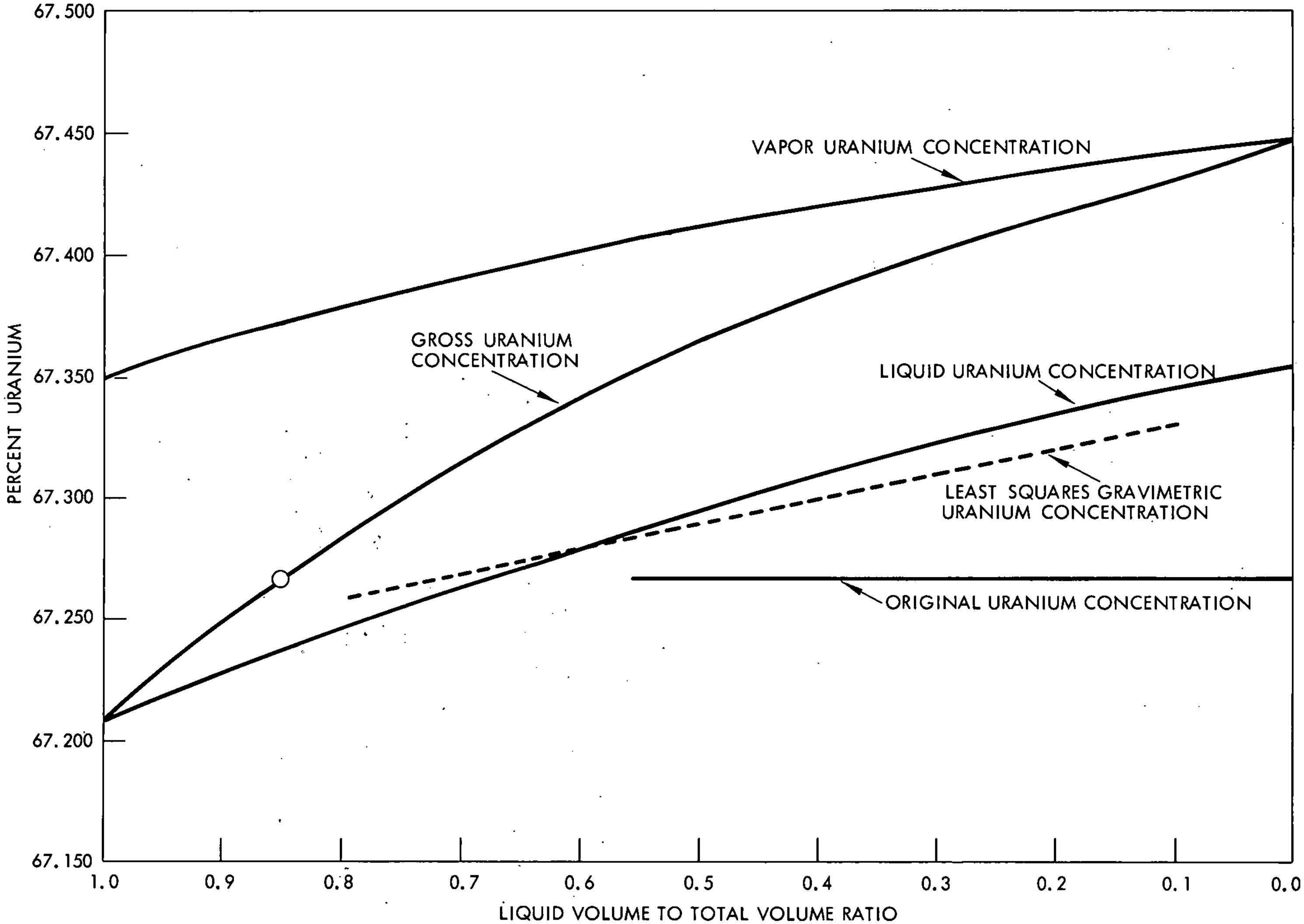

Figure 9

URANIUM CONCENTRATION DISTRIBUTION
FOR THE SYSTEM UF_6(2460 g)-$C_4Cl_3F_7$(12.8 g)

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