82-1; 1,3,5-trichlorobenzene, 108-70-3; 1,2,3,4-tetrachlorobenzene, 634-66-2; 1,2,3,5-tetrachlorobenzene, 634-90-2; 1,2,4,5-tetrachlorobenzene, 95-94-3; pentachlorobenzene, 608-93-5; hexachlorobenzene, 118-74-1; 2-chlorobiphenyl, 2051-60-7; 2,5-dichlorobiphenyl, 34883-39-1; 2,6-dichlorobiphenyl, 33146-45-1; 2,4,5-trichlorobiphenyl, 15862-07-4; 2,4,6-trichlorobiphenyl, 35693-92-6; 2,3,4,5-tetrachlorobiphenyl, 33284-53-6; 2,2′,4′,5-tetrachlorobiphenyl, 41464-40-8; 2,3,4,5,6-pentachlorobiphenyl, 18259-05-7; 2,2′,4,5,5′-pentachlorobiphenyl, 37680-73-2; 2,2′,3,3′,6,6′-hexachlorobiphenyl, 38411-22-2; 2,2′,3,3′,4,4′-hexachlorobiphenyl, 38380-07-3; 2,2′,4,4′,6,6′-hexachlorobiphenyl, 33979-03-2; 2,2′,3,3′,4,4′,6-heptachlorobiphenyl, 52663-71-5; 2,2′,3,3′,5,5′,6,6′-octachlorobiphenyl, 2136-99-4; 2,2′,3,3′,4,5,5′,6,6′-nonachlorobiphenyl, 52663-77-1; 2,2′,3,3′,4,4′,5,5′,6,6′-decachlorobiphenyl, 2051-24-3.

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Solubility of Uranium Hexafluoride in Some Low-Temperature Solvents

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The solubility of UF_6 has been measured in several liquefied gases. Phosgene, nitrous oxide, and sulfur dioxide are found to be good solvents for UF_6 at temperatures below 0 °C. The results are compared with ideal and regular solution theory. Regular solution theory provides a reasonably good description of the $UF_6/COCl_2$ system but is less good for the UF_6/SO_2 system.

Introduction

The solubility of uranium hexafluoride (UF_6) in liquids has been studied many times, particularly in halocarbons (*1–3*) and fluorinated halogens (*4–8*). A few of these studies have included temperatures below 0 °C (*2*, *5*, *7*) but there has been more interest in the solubilities at temperatures above 0 °C.

It is the purpose of this paper to report the solubilities of UF_6 in liquefied gases of small, inorganic molecules. Solubilities have been measured for temperatures between about 160 and 270 K, and the experimental observations are compared with regular solution theory.

Experimental Methods

The apparatus is a calibrated, 80-mL, graduated, cylindrical, borosilicate glass flask measuring about 4 cm in diameter by 8 cm high and attached to a vacuum system through a glass tube. The flask is surrounded by a clear glass Dewar, and the top of the Dewar is closed with a Styrofoam plug that fits around the vacuum line. A glass-encapsulated magnetic stirring rod is inside the flask. An iron–constantan thermocouple is inserted into a thin-walled glass well that protrudes into the interior of the flask; this thermocouple well is fused to the top of the flask and reaches nearly to the bottom.

The experimental procedure is as follows. The flask is evacuated to about 10^{-4} torr and is heated to drive off any residual water. The bottom of the flask is then cooled with dry ice or liquid nitrogen, and the solute, UF_6, is condensed out from its gas phase. The Dewar is raised into position and closed with the Styrofoam plug. Further cooling of the flask is accomplished by blowing cold N_2 gas into the Dewar through a hole in the Styrofoam plug. The solvent is condensed into the flask, and the mixture of UF_6 and solvent is stirred vigorously. The temperature of the liquid is held constant, and solvent is *slowly* condensed into the flask in small increments. After each addition of solvent, the mixture is stirred for a period that depends on the solvent, the temperature, and the amount of undissolved UF_6. After most of the UF_6 has been dissolved, solvent is added in very small increments and the stirring times between additions are lengthened, to ensure that the solution is saturated with UF_6. Some of the data points reported here have been obtained with a small fraction (<2%) of the total UF_6 still undissolved. When the UF_6 crystals just disappear under bright illumination, the stirring motion is stopped, and the level of the

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Table I. Solubilities and Some Remarks

solvent	temp, K	concn,[a] M	soln color	comments[b]	temp range,[c] K
$COCl_2$	270.8	2.12	pale yellow[d]	slight residue	158.7-270.8
SO_2	264.7	1.43	yellow[d]	slow reaction slight residue	203.9-264.7
N_2O	193.7	0.185	colorless	no reaction	182.4-193.7
O_2	77	<0.003[e]	light blue[f]	residue	
H_2S	223	*e*		reaction residue	
COS	205	<0.072[g]	red-brown	reaction residue	

[a] This concentration is appropriate for the highest temperature at which the solubility was measured for a given solvent. [b] Residues sometimes remained after the solvent and all remaining UF_6 were pumped away. Reactions between UF_6 and the solvent (or some undetected impurity in the solvent) were often observed. [c] Temperature ranges for which solubilities were measured. [d] The color depends on the concentration. [e] Insoluble, within the limitations of this experiment. [f] Pure O_2 is light blue. [g] Detailed measurements were not made on this system, but UF_6 did dissolve.

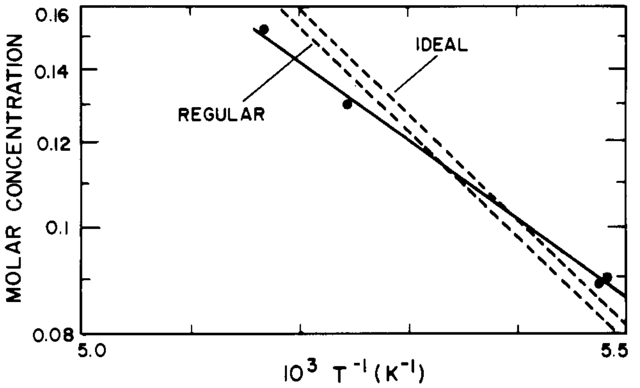

Figure 1. Molar concentration (moles of solute per liter of solution) of UF_6 in liquid N_2O at saturation vs. $10^3/T$. Data are shown as solid circles (●). The solid line is judged to be an adequate representation of the data. The dashed lines are theoretical predictions of the solubility of UF_6 in liquid N_2O.

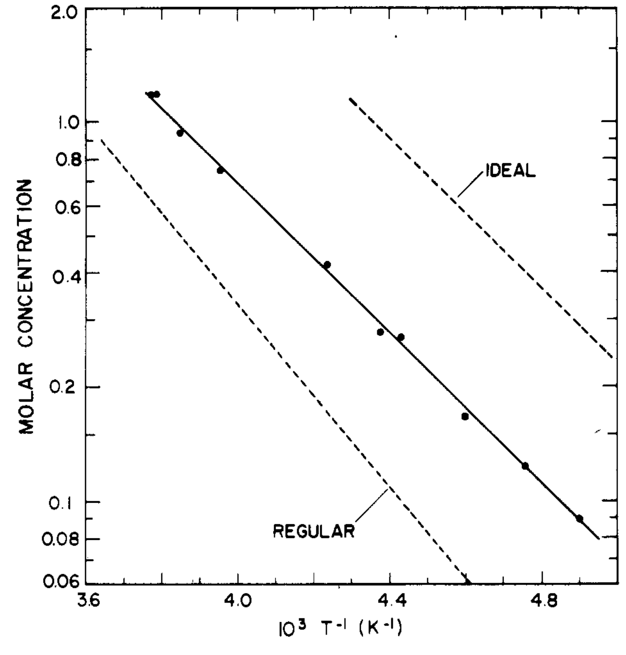

Figure 2. Molar concentration of UF_6 in liquid SO_2 at saturation vs. $10^3/T$. The solid line is judged to be an adequate representation of the data (●). The dashed lines are theoretical predictions of the solubility of UF_6 in SO_2.

solution within the flask is read.

The gases used in this experiment were the best, commercially available grade. Mass-spectral analyses were made of all gases. Before the $COCl_2$ was used, it was passed through a trap filled with MgO to remove a small HCl impurity. The UF_6 used was obtained from the Oak Ridge Gaseous Diffusion Plant and was better than 99.9% pure; our infrared analyses of the UF_6 revealed no impurities.

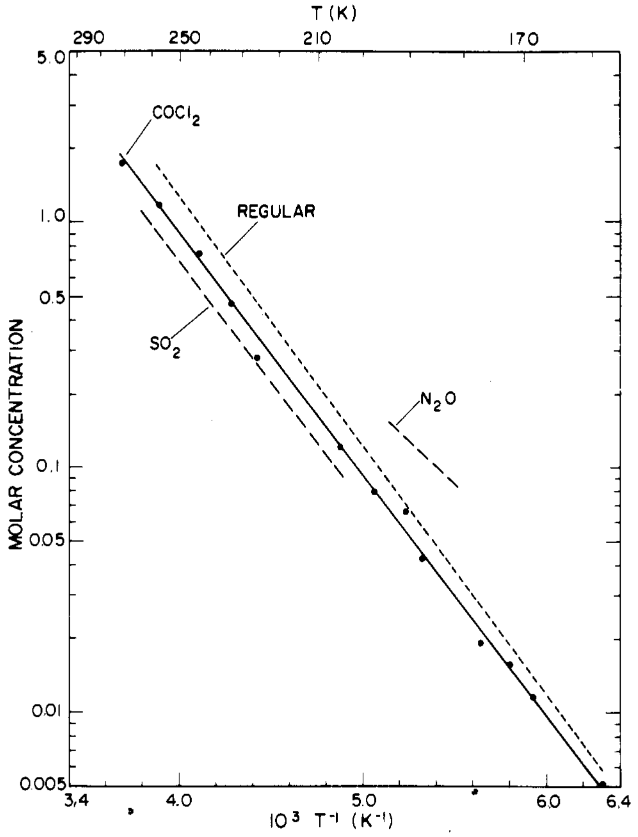

Figure 3. Molar concentration of UF_6 in liquid $COCl_2$ at saturation vs. $10^3/T$. The solid line is judged to be an adequate representation of the data (●). The long-dashed lines are experimental solubilities reproduced from Figures 1 and 2 for comparison. The short-dashed line is the solubility predicted by regular solution theory for $UF_6/COCl_2$.

Temperatures quoted herein should be accurate to ±0.5 °C. The total uncertainty in the volume measurements is ±3%. The amount of UF_6 condensed into the flask is known within ±3%, and at least 98% of the UF_6 has gone into solution for any data point. The maximum possible errors in the concentrations reported here should thus be ±8%, and the reproducibility of the data should be about ±5%.

Results and Discussion

The liquefied gases used in this investigation are given in Table I, together with some experimental observations. UF_6 has been dissolved in N_2O, $COCl_2$, SO_2, and COS. The COS solution was red-brown, and the UF_6 *may* have been dissolved

Table II. Solubility of UF_6

T, K	concn, mol/L	T, K	concn, mol/L
	$COCl_2$ as Solvent		
158.7	0.00502	197.7	0.0802
158.7	0.00508	205.0	0.122
168.7	0.0116	226.0	0.282
169.5	0.0152	233.2	0.467
169.5	0.0156	243.4	0.737
177.2	0.0193	256.8	1.17
187.7	0.0428	270.8	1.73
191.2	0.0670		
	SO_2 as Solvent		
203.9	0.0894	235.8	0.415
210.2	0.123	252.2	0.755
217.2	0.166	259.4	0.950
225.6	0.267	263.7	1.18
228.2	0.278	264.7	1.18
	N_2O as Solvent		
182.4	0.0896	190.7	0.130
182.7	0.0886	193.7	0.152

as U(V), which has a characteristic reddish color.

The solubilities of UF_6 in liquid N_2O, SO_2, and $COCl_2$ have been measured over as wide a temperature range as possible with our apparatus. The data are presented in Figures 1–3 and in Table II. There are too few data points for N_2O to be sure that the appropriate curve is indeed straight, but the solubilities of UF_6 in $COCl_2$ and SO_2, when plotted against $1/T$, lie along straight lines over a fairly wide range of temperatures.

The other lines shown in Figures 1–3 have been computed from regular or ideal solution theory for comparison with the data (*9*, *10*). The equations used for regular solution theory are

$$RT \ln (a_2/x_2) = V_2\phi^2(\delta_1 - \delta_2)^2$$

$$\phi = x_1V_1/(x_1V_1 + x_2V_2)$$

$$\delta_k = [(\Delta H_k{}^{\mathrm{v}} - RT)/V_k]^{1/2}$$

$$\ln a_2 = (\Delta H_0{}^{\mathrm{f}}/R)(1/T_0 - 1/T) \qquad (1)$$

For ideal solutions, $\delta_1 - \delta_2 = 0$. The numerals 1 and 2 label quantities appropriate for solvent and solute (UF_6), respectively. $T \equiv$ temperature in Kelvin. $x_k \equiv$ mole fraction of component k in solution. $R \equiv 1.988$ cal/(cm mol K). $V_1 \equiv$ molar volume of the solvent. $V_2 \equiv$ molar volume of the (hypothetical) liquid solute. $\Delta H_k{}^{\mathrm{v}} \equiv$ heat of vaporization of component k. T_0 and $\Delta H_0{}^{\mathrm{f}}$ are respectively the melting point of the solute and the heat of fusion of the solute at T_0. Table III lists values used for the parameters appearing in these equations. Note that solutions of UF_6 in $COCl_2$ and in N_2O are expected to be nearly ideal.

We have assumed that the difference, ΔC_p, between the heat capacities of the liquid and solid solute are zero; in fact (*8*), for UF_6, $\Delta C_p \approx 2.1$ cal/(mol K) at the melting point of UF_6. Since $\Delta C_p \neq 0$, the plot of $\ln x_2$ against T^{-1} should be curved, but for us to discern this curvature experimentally $\Delta C_p \geq 8$ cal/(mol K) would be required. The curvature arising from finite ΔC_p will not, therefore, be observable in our data. The term involving ΔC_p is small (~20 cal/mol at 200 K) compared with $\Delta H_0{}^{\mathrm{f}}$ (~4500 cal/mol) and has been taken to be zero.

For comparison of the calculated and experimental curves, we have estimated effects caused by variation of solvent density with temperature, and we have found the effect to be negligible. Similarly, we have calculated the number density, n_2, of the dissolved UF_6 from $n_2 = V_1/x_2$, and we find that this assumption should cause the calculated curves for n_2 to be ~3% higher than the observed solubility of UF_6 in $COCl_2$ at 270 K; the effect of this assumption is negligible for all other data points.

The slopes of the lines computed from regular solution theory and plotted in Figures 2 and 3 are very similar to the slopes of the lines representing the observations over the temperature range for which we have data. The absolute solubilities of UF_6 in SO_2 and $COCl_2$ are not within estimated experimental error of the theoretical curves. Regular solution theory provides a fairly good (~30%) quantitative estimate of the solubility of UF_6 in $COCl_2$ between 158 and 275 K but predicts that the concentration of UF_6 in SO_2 will be a factor of 2–3 below observed values.

In Figure 1, the measured and predicted absolute solubilities of UF_6 in N_2O agree fairly well, but the slope of the curve representing the observed solubilities differs significantly from the slope of the theoretical curve. The significance of this difference in slopes is hard to assess from our data, because the temperature range of our observations is small and the difference in slopes is not very far outside of the range of estimated uncertainty. Regular solution theory provides a reasonably good (5–10%) estimate of the solubility in UF_6 in N_2O between 182 and 194 K.

Regular solution theory does not represent any of the present binary systems within the estimated experimental uncertainty, but the comparison of theory and experiment may be invalidated by several things. First, values estimated for the solubility parameters depend in some cases on extrapolations which may be in error. In the case of UF_6/N_2O, uncertainties in our data may contribute significantly to the lack of agreement. Finally, the solubility parameters are estimated at 200 K, and somewhat different theoretical curves will, in practice, result if the solubility parameters are evaluated at some other temperature. We conclude that, at least for UF_6/N_2O and $UF_6/COCl_2$, the differences between the theory and the experiment may not be significant.

Conclusions

Liquid $COCl_2$, SO_2, N_2O, and even COS are all reasonably good solvents for UF_6. Regular solution theory adequately represents our data for the solubility of UF_6 in N_2O and $COCl_2$ but is less good for the solubility of UF_6 in SO_2. Although our temperatures are lower than in some previous investigations, it appears that the liquids studied here are better solvents for

Table III. Heats of Fusion ($\Delta H_0{}^{\mathrm{f}}$), Heats of Vaporization (ΔH^{v}), Molecular Weight (M_{r}), Molar Volumes (V_i), and Solubility Parameters (δ_i) Evaluated at T = 200 K

molecule	$\Delta H_0{}^{\mathrm{f}}$, kcal/mol	ΔH^{v}, kcal/mol	M_{r}	V_i, cm^3	δ, cal$^{1/2}$ cm$^{-3/2}$
UF_6	4.587[a]	7.61[b]	352.03	84.42[c]	9.24
$COCl_2$		6.80[d]	98.91	61.62[e]	10.19
SO_2		6.70[f]	64.06	39.74[e]	12.63
N_2O		3.77[g]	44.01	35.91[e]	9.70

[a] Taken from ref *11*. [b] Obtained by subtracting the heat of fusion, 4.587 kcal, from the heat of sublimation, 12.20 kcal (p 423, ref *8*) at 200 K. [c] Computed from the equation for the density in ref *12*. [d] Obtained from data in ref *13* and from ref *11*. [e] Computed from formulas and densities in ref *14*. [f] Estimated by extrapolating the data in ref *15*. [g] Taken from ref *15*.

J. Chem. Eng. Data 1984, 29, 193-199

UF_6 than many of the halocarbons and fluorinated halogens studied in the past (*1*, *4*). Thus, reasonably good, low-temperature solvents exist for uranium hexafluoride, even among rather simple solvent molecules.

Registry No. UF_6, 7783-81-5; $COCl_2$, 75-44-5; N_2O, 10024-97-2; SO_2, 7446-09-5.

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Heat Capacities of Some Binary and Ternary Aqueous Nonelectrolyte Systems

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Heat capacities of a variety of binary and ternary aqueous nonelectrolyte systems were measured and the data fitted to $C_{p,\phi} = C_{p,\phi}{}^{\circ} + c_2m + c_3m^2$. The systems studied were aqueous solutions of acetamide, acetone, *p*-dioxane, dimethylformamide, ethyl acetate, formamide, hexamethylenetetramine, methyl formate, *s*-trioxane, and combinations of two of the above solutes. Internal consistency of data is very good and c_2 coefficients agree with literature values. However, small discrepancies of $C_{p,\phi}{}^{\circ}$ with literature values are observed, indicating a problem with the calorimeter.

Introduction

Heat capacities of binary and ternary aqueous solutions of myoinositol (IN), D-mannitol (MAN), cyclohexanol (CHEX), formamide (F), *N*,*N*-dimethylformamide (DMF), *s*-trioxane (T), and hexamethylenetetramine (HMT) were measured to complement recent enthalpy studies (*1*-*5*). Heat capacity measurements contribute to an understanding of solutions: details of structural effects have been found (*6*, *7*), solute-solvent and solute-solute interactions have been uncovered (*8*-*12*), evidence of pseudophase transitions in binary aqueous organic systems (*13*) has been cited, and means have been provided for evaluating both the temperature dependence of excess thermodynamic properties (*14*) and isothermal compressibilities from isentropic compressibilities (*15*).

Much of the interest in heat capacity measurements has arisen in the past decade as a result of the widespread use of flow calorimeters, particularly those of the Picker type (*16*). Flow calorimetry offers a number of advantages over earlier techniques: higher sensitivity, elimination of vapor-space corrections, short response times, small sample size, and rapid collection of data. These inherent advantages, together with some elegant design features, make possible the use of "the general principle that it is preferable to measure heat capacities as a function of temperature and integrate to obtain enthalpies at various temperatures than to derive heat capacities from the temperature dependence of enthalpies" (*15*). This type of calorimeter is now being used for measurements at high temperatures (*17*-*23*). With the great importance now attached to heat capacity measurements, it is essential that the accuracy of the data should not be called into question. The results presented here give rise to some concern over this matter.

Experimental Section

Materials. The purification of myoinositol (*1*), D-mannitol (*1*), cyclohexanol (*1*), formamide (*2*), *N*,*N*-dimethylformamide (*2*), *s*-trioxane (*3*), and hexamethylenetetramine (*5*) used by one of us (I.R.T.) has been previously reported. The purification of acetamide, acetone, dioxane, ethyl acetate, formamide, hexamethylenetetramine, D-mannitol, methyl formate, and *s*-trioxane (used by S.K.S.) has also been reported elsewhere (*24*). Details of solution preparation and handling are also available (*1*-*3*, *5*).

Apparatus and Procedure. Measurements were taken with a Sodev Model CP-C Picker-type flow heat capacity microcalorimeter (*16*, *25*, *26*). Essentially, two liquids (1 and 2) with heat capacities per unit volume of σ_1 and σ_2 are maintained at the same temperature and flow rate as they enter twin cells; here they are simultaneously heated in such a manner that their final temperatures are identical. If W_1 and W_2 are the electrical powers supplied to produce the temperature rise, then, under ideal circumstances (i.e., no heat losses), we have

$$\frac{W_2 - W_1}{W_1} = \frac{\sigma_2 - \sigma_1}{\sigma_1} \qquad (1)$$

The instrumentation is arranged so that the difference in applied

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