ABSTRACT

In this work, solubilities of propane in CS_2, CCl_4, C_6H_6, C_6H_{12}, and C_6F_{14} were measured along with the solubilities of propane in 5 isomers of n-hexane over a temperature range of about 60°C. The experimental solubilities were compared with correlations. It was found that solubility in isomers of hexane is the same for all isomers. It was also found that perfluorohexane does not behave according to regular solution theory.

Diffusivities of propane in cyclohexane, benzene and perfluorohexane were measured at 3 different temperatures and were compared to predictions of 4 correlations. It was found that the diffusivity - viscosity correlation developed by Hayduk and Cheng [30] represented the data best.

ACKNOWLEDGEMENTS

The author expressed his appreciation and gratitude to Dr. W. Hayduk for his valuable assistance and guidance in this work. My sincerest gratitude is extended to Mme France Perron for her fine typing of this thesis and also to Mr. G. Gasparetti for his technical assistance in constructing and maintaining the equipment.

No word of thanks would be complete without mention of my wife Nicole, for the countless number of ways in which she has helped in the evolution of this thesis.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

NOMENCLATURE

A, B	constants in the diffusivity viscosity correlation
C	molar concentration
D	diffusion coefficient (m^2/sec)
D_{21}	diffusion coefficient (cm^2/sec)
$E_{D_{21}}$	activation energy for diffusivity
E_{μ_1}	activation energy for viscosity
F	volumetric flow rate of solvent from infusion pump (cc/sec)
J	molar flux relative to average velocity
K	ratio of molar concentrations $\frac{C_{2_E}}{C_1}$
L	Ostwald coefficient
M	molecular weight
N	molar flux relative to fixed position
N_A	Avogadro's number
P	pressure (mm mercury)
R	gas constant
T	temperature °K
V	volume; V_2' is vapor-free gas volume in Equation 9
$\overline{V}$	partial molar volume (cc/g mole)
X_1	association number of solvent
f	fugacity
h	rate of descent of the bead
k	ratio of mass concentrations $\frac{\rho_{2_E}}{\rho_L}$
n	mass flux relative to fixed position
w	mass fraction
x	mole fraction
z	distance (length) (cm)

ΔE	internal energy change of vaporization
ΔH	enthalpy change of vaporization
ΔS	entropy change
α	Bunsen coefficient
ρ	density
μ	viscosity (cps ; NS/m^2 in Equation 22
ϵ_2	geometric parameter
δ	solubility parameter

Superscript

*	partial (pressure)
o	vapor (pressure)
i	ideal (solubility)

Subscript

O	reference (solubility)
O	at position O (calculation of diffusivity)
1	solvent
2	gas solute
t	total (atmospheric pressure)
r	room temperature
s	measurement temperature
a, b	arbitrary temperature
E	effective (density of solute gas in liquid phase)
L	liquid at position L

INTRODUCTION

The measurement of both solubility and diffusivity are becoming of increasing importance. These measurements are needed for a better understanding of gas-liquid equilibrium and solution theory and also in practical considerations such as the design of gas-liquid mass transfer equipment. In this work, solubilities and diffusivities of propane in different isomers of hexane and in other non-polar solvents were measured.

Because of the dual nature of this work, the presentation is divided into two parts, the first dealing with gas-liquid solubilities and the second dealing with gas liquid diffusivities. The " conclusions" for both sections are combined.

SOLUBILITY OF GASES IN LIQUIDS

Solutions can be characterized mostly by entropy and enthalpy. These two properties define whether or not a solution is ideal. The concept of ideal solution is a theoretical one but it describes the behavior of actual solutions as a first approximation. Deviation from ideality can arise from chemical effects, association, dissociation, solvation and Van der Waals' effects. There are many liquid pairs whose molecular species are so nearly alike in their attractive forces that they mix with negligeable heat effects. Such solutions are called athermal [16]. An athermal solution can be ideal if the entropy of solution is ideal. In the ideal solution an individual molecule of either species requires the same kinetic energy to escape from the solution as it would from its pure liquid. The partial molar entropy of an ideal solution is expressed as:

$$\Delta\bar{S}_1 = - R \ln x_1 \qquad (1)$$

and the fugacity is expressed as:

$$f_1 = f^\circ_1 x_1 \qquad (2)$$

If vapor pressures are substituted for fugacities, an expression of Raoult's law is obtained:

$$p_1 = p^\circ_1 x_1 \qquad (3)$$

The ideal solubility of a gas can be calculated from its saturation pressure by the aid of Raoult's law [16]. As the solubility of a gas in an ideal solvent is equivalent to the concentration to which the liquefied gas must be diluted to in order to obtain a partial pressure of the gas equal to one atmosphere, the ideal solubility is then the ratio of atmospheric pressure to the vapor pressure of the gas. For gases above their critical temperature,

the ideal solubility can be calculated using a fictitious value of vapor pressure extrapolated from measured vapor pressures to the given temperature [16].

Although they have positive heats of mixing, many non-ideal solutions have nearly ideal entropies of mixing. These solutions are called regular solutions.

Hildebrand and Scott [16] developed equations to represent regular solution of gases in liquids:

$$\ln x_2 = \ln x_2^i - \frac{\bar{V}_2}{RT} (\delta_1 - \delta_2)^2 \qquad (4)$$

When the molecules of solvents and solute differ substantially in size, correction terms were added:

$$\ln x_2 = \ln x_2^i - \ln \frac{\bar{V}_2}{V_1} - (1 - \frac{\bar{V}_2}{V_1}) - \frac{\bar{V}_2}{RT} (\delta_1 - \delta_2)^2 \ldots .$$

$$\ldots\ldots\ldots\ldots\ldots \qquad (5)$$

where

$$\delta_1 = \left(\frac{\Delta E_1}{\bar{V}_1}\right)^{\frac{1}{2}} \approx \left(\frac{\Delta H_{vap} - RT}{\bar{V}_1}\right)^{\frac{1}{2}} \qquad (6)$$

Despite the many assumptions made in their derivation, equations 4 and 5 have proven to be at least partially successful for solutions where both solute and solvent were non-polar, or only slightly polar.

Temperature Variation of Solubility

Generally the solubilities of gases decrease with increasing temperature for gases having solubilities greater than $x_2 = 1 \times 10^{-3}$ and solubilities increase with increasing temperature for gases having solubilities less than $x_2 = 1 \times 10^{-3}$.

It was also found that a log-log plot of mole fraction solubility versus temperature gave a straight line, the slope of which were related to the entropy of solution [17]. Furthermore when the solubilities for many gases in the same solvent were plotted against temperature on the same graph paper, it was found that the lines intersected at the solvent critical temperature. The ordinate was coined the reference solubility (x_o) and a correlation was developed relating this reference solubility to the solvent solubility parameter δ_1 [12].

$$\log (10^4 x_o) = 2.265 - 0.134 \delta_1 \qquad (7)$$

Logic defines another point on the solubility curve at the normal boiling point temperature of the gas. At this temperature, the mole fraction solubility of the gas should be one. Extrapolation of the solubility versus temperature data should then be possible to include the gas normal boiling point.

Methods of Expressing Gas Solubility

Battino and Clever made a complete review of the methods of expressing solubility [1]. Here only a few, more widely used methods are defined.

The Bunsen or absorption coefficient α is defined as the volume of gas at standard conditions which is absorbed by a unit volume of solvent at the temperature of the measurement, under a gas partial pressure of 760 millimeters of mercury. When the partial pressure of the gas is not 760 mm. of mercury, it is corrected using Henry's law.

$$\alpha = \left[\frac{V_2}{V_1} \frac{273.15}{T} \frac{P_2^*}{760} \right] \frac{760}{P_2^*}$$

or

$$\alpha = \frac{V_2}{V_1} \frac{273.15}{T} \qquad (8)$$

The correction to standard temperature and pressure assumes ideal gas behavior. The possible error thus introduced can be minimized by using an equation of state but for most gases and conditions, the error will be less than 1%.

The Ostwald coefficient is the ratio of the concentration of solute in the solution phase to the concentration of solute in the gas phase. It can also be considered as the ratio of the volume of gas absorbed to the volume of absorbing liquid at the temperature and pressure of the measurement.

$$L = \frac{C_1}{C_2} = \left(\frac{V_2}{V_1} \right)_{T,P} = \frac{V_2'}{V_1} \frac{P_t}{P_2^*} \qquad (9)$$

where $$P_2^* = P_t - (1 - x_2) P_1^0 \qquad (10)$$

The Ostwald coefficient is nearly independent of pressure at low pressures. This is explained by the fact increasing the pressure of the gas proportionately increases the quantity of gas dissolved (Henry's law) by the same ratio as it decreases the volume that the gas occupies (gas law).

Mole fraction is the most commonly used method of expressing solubility. In terms of the gas and solvent volumes and fluid properties, the mole fraction is:

$$x_2 = \frac{V_1}{V_2} \frac{760}{P_t} \Big/ \left(\frac{V_1}{V_2} \frac{760}{P_t} + \frac{V_1 \rho_1}{M_1} \right) \qquad (11)$$

or

$$x_2 = 1/\left(1 + \frac{\rho_1}{M_1} \frac{V_2}{L} \frac{P_t}{760}\right) \qquad (12)$$

PROPERTIES OF TEST FLUIDS

Propane was purchased in the liquefied form from Matheson of Canada. It had a specified minimum purity of 99.5%. The pertinent properties of propane are listed in Table 1.

The benzene, 2-methylpentane, 3-methylpentane, 2,2-dimethylbutane, 2,3-dimethylbutane and the normal hexane were purchased from the Phillips Petroleum Company and were certified to be minimum 99 mole % pure. The cyclohexane and carbon disulfide were bought from the Matheson, Bell and Coleman Company and were also certified to be minimum 99 mole % pure. The carbon tetrachloride was purchased from J. T. Baker Chemical Company and the perfluorohexane from P.C.R. Inc. and both were specified to be of spectrophotometric quality.

The viscosities of benzene, cyclohexane and perfluorohexane are listed for the pertinent temperatures in Table 2. The density and vapor pressure data for all solvents used are shown in Table 3.

TABLE 1

Gas phase propane molar volume and effective density of dissolved propane

T (°C)	$V_{2\ gas}$ (cc/mole)	$\rho_{dissolved}$ (gm/cc)
-15	20679.5 (6)	0.621 (20)
-10	21193.5	
0	21982.8	
5	22412.4	0.593
10	22839.8	0.586
25	24105.8	0.565 (0.566) [a]
35	24945.1	
40	25366.1	
45	25786.1	0.537
50	26203.2	0.530
70	27879.8	0.501

a) This value was obtained in this laboratory using a high precision density meter. The density of a propane saturated n-hexane solution was measured. With this, the Ostwald coefficient solubility and the density of pure hexane, the density of dissolved propane could be determined. The density of propane dissolved in perfluorohexane was also measured and was found to be very similar (0.569 gm/cc) to that in hydrocarbons.

TABLE 2

Viscosity of Solvents

Solvent	T (°C)	μ (cp)
benzene (19)	10	0.7490
	25	0.6028
	50	0.4360
cyclohexane (19)	10	1.180
	25	0.898
	50	0.606
perfluorohexane (22)	-10	1.170
	5	0.901
	25	0.661

TABLE 3

Density and Vapor Pressure of Solvents

	T	ρ gm/cc	vap. pres. mm Hg
CS_2 [5]	-15	1.3153	60.97
	0	1.2931	126.49
	25	1.2559	359.25
	35	1.2406	506.19
C_6H_6 [19]	10	0.8894	45.62
	25	0.8734	94.99
	50	0.8465	268.65
	70	0.8247	572.77
CCl_4 [5]	-15	1.6611	13.44
	0	1.6326	31.74
	25	1.5843	109.81
	50	1.5353	313.72
C_6H_{12} [19]	10	0.7881	47.47
	25	0.7743	97.75
	50	0.7504	270.01
	70	0.7307	555.02
n-hexane [19]	-15	0.6894	18.88
	0	0.6726	58.79
	25	0.6550	151.58
	50	0.6318	405.93
2, methyl-pentane [19]	-15	0.6851	29.96
	5	0.6666	85.66
	25	0.6485	207.02
	50	0.6250	571.92
3-methyl-pentane [19]	-15	0.6964	25.71
	5	0.6779	76.98
	25	0.6596	187.92
	50	0.6363	490.73
2,2 dimethyl-butane [19]	-15	0.6813	51.35
	5	0.6626	138.67
	25	0.6441	318.94
	40	0.6309	551.94
2,3-dimethyl-butane [19]	-15	0.6934	34.51
	5	0.6751	98.12
	25	0.6579	234.44
	45	0.6388	495.01
perfluoro-n-hexane [22]	-15	1.7986	26.98
	-10	1.7845	36.14
	5	1.7421	83.71
	25	1.6718	220.77
	45	1.6102	498.01

APPARATUS AND PROCEDURE

The apparatus and method used in this work were the same as those described by Hayduk and Cheng [11]. The solubility apparatus (shown in Figure 1) consisted mainly of a gas burette, a contacting chamber in the form of a glass tube spiral, a manometer and a solution collecting burette, all enclosed in a glass jacket through which water at constant temperature was circulated. The solvent was brought into the contacting chamber by the use of a syringe attached to a syringe pump. The gas was maintained at atmospheric pressure by the use of a mercury column controlled by a variable speed mercury lift. The burette for confining the solute gas in the solubility apparatus used for most determinations had a volume of 100 milliliters and the liquid containing burette had a volume of 5 milliliters. For measuring somewhat lower solubilities at higher temperatures, a second apparatus with a 50 milliliter gas burette and a 10 milliliter liquid burette was used.

Because of the large temperature difference between the measurement and ambient temperatures, such as for the measurements at 70°C, a slight addition was made to the apparatus. The syringe and contents was kept at 70°C by circulating water at constant temperature through a syringe jacket fitted over the barrel of the syringe. In addition the syringe needle was insulated with styrofoam. With this slight modification to the apparatus the solvent was fed to the absorption coil at the same temperature as that in the solubility apparatus. Heat transfer effects that were formerly found to cause erroneously high results (up to several percent) were thus eliminated.

The water circulating in the jacket of the solubility apparatus was kept at constant temperature in a circulating bath purchased from the Fisher Scientific Company. The temperature control was effected by a

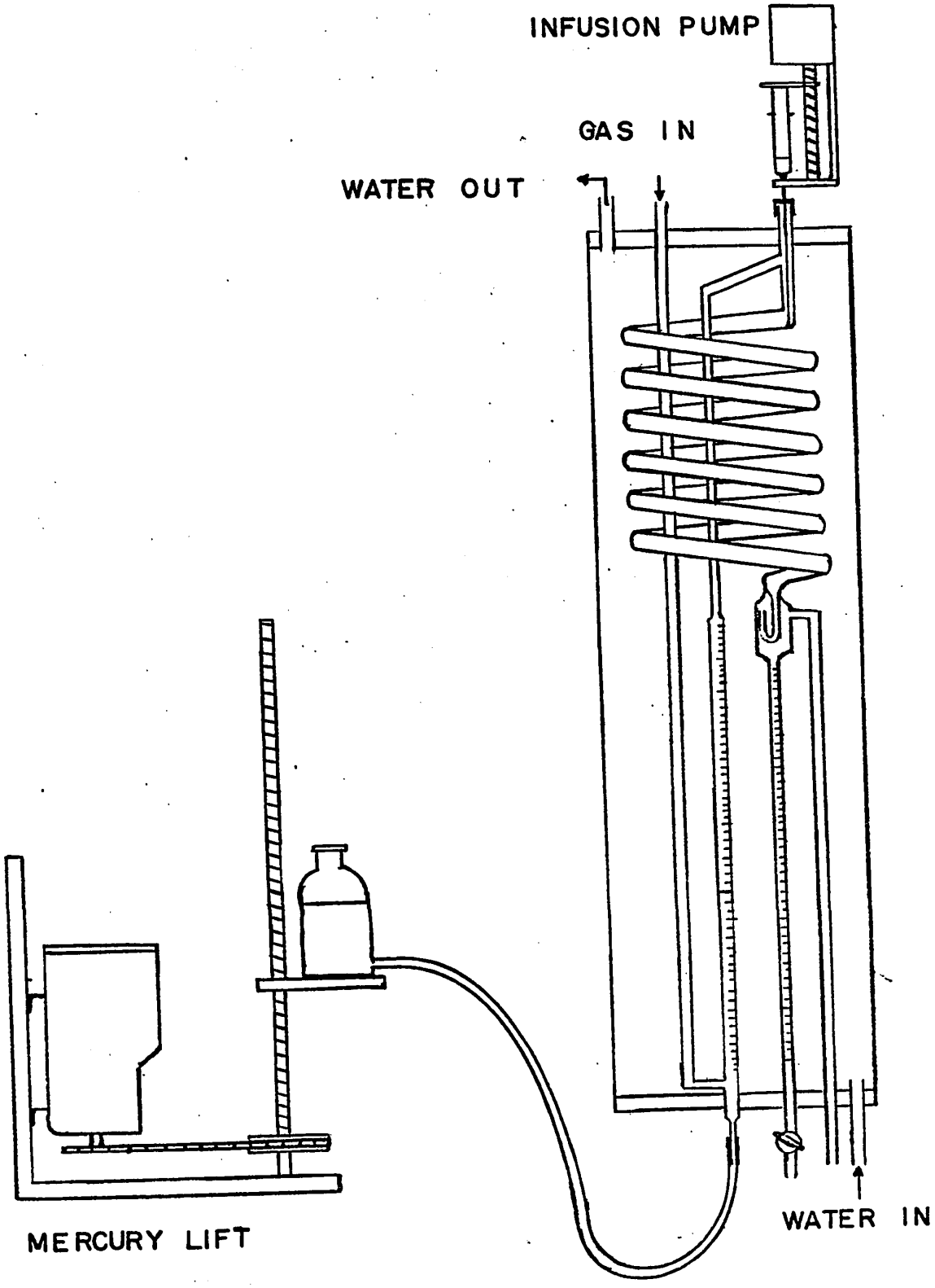

Figure 1 - Hayduk and Cheng [11] - Solubility Apparatus

thermoregulator set at the temperature of the measurement. This thermoregulator controlled a heating element that kept the temperature constant. For temperatures above 40°C, no cooling was necessary since the heat lost by the system was enough to balance the heat input. In the temperature range of 25°C to 40°C, tap water was circulated in a coil inside the bath. To achieve temperatures lower than room temperature a "Bath Cooler" refrigeration unit purchased from Neslab Instruments Inc. was used to cool the solution of water and ethylene glycol that was then circulated in the apparatus. To get to temperatures as low as -15°C, the whole solubility apparatus had to be insulated to minimize the heat intake. For that purpose a glass tube with a diameter about one inch larger than the actual circulation jacket was fitted over the jacket. Also the 1/4" I. D. Tygon tubing connecting the bath and circulation jacket was insulated in the same way using 3/8" I. D. Tygon tubing. With this small addition, the heat intake of the system was minimized and accurate temperature control could be maintained.

The first step in making a solubility determination is the degassing of the solvent. This was done in the so-called degassing apparatus shown in Figure 2. The glass bottle was filled with solvent. A vacuum was then applied to both the solvent bottle and to the long glass column. After about 20% of the liquid had boiled away, the liquid was allowed to flash through the capillary tube into the top of the accumulation column. After enough liquid had been accumulated, the vacuum was released and liquid was withdrawn into a syringe.

While the solvent was being degassed, the solubility apparatus was purged with propane gas. The syringe then containing degassed solvent was then fitted to the syringe pump which previously had been carefully calibrated using distilled water. The needle of the syringe was then inserted through the rubber septum into the contacting chamber of the apparatus.

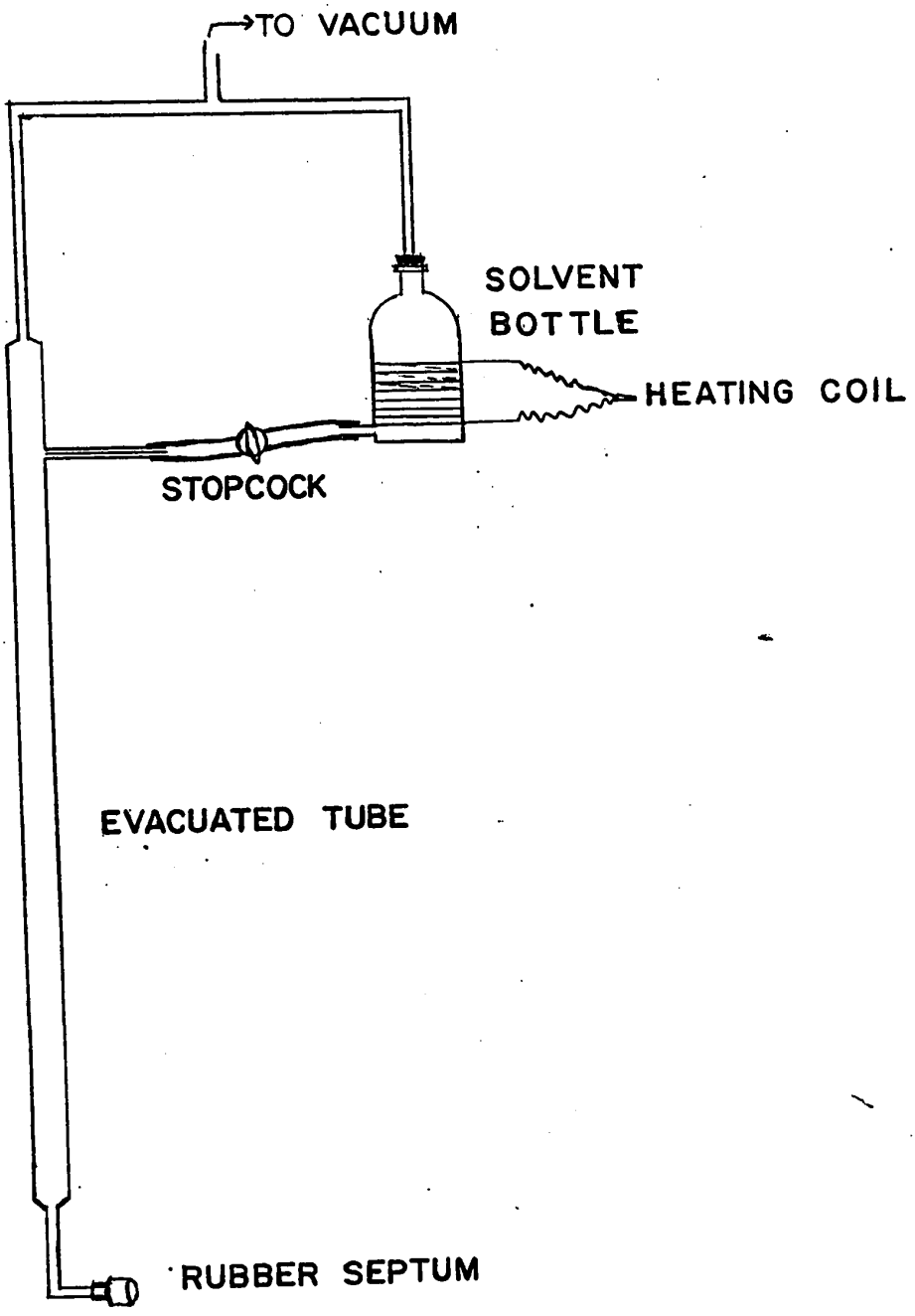

Figure 2 - Degassing Apparatus

The solution burette stopcock was closed and the infusion pump started. Gas was still allowed to flow through the apparatus at a reduced flow rate until steady-state was achieved. Then the mercury lift was started, thus isolating a quantity of propane in the gas burette. The gas feed line was then opened to the atmosphere. The pressure inside the apparatus, monitered on the small manometer, was maintained equal to atmospheric pressure by varying the speed of the mercury lift. Readings of mercury level were recorded at set intervals of time.

RESULTS

From experimental data, a plot of gas volume versus time was drawn. With the occasional points that seriously deviated from a straight line omitted, the best slope was obtained according to a least square fit program. From the slope thus obtained and additional pertinent information, the Ostwald coefficient and mole fraction solubility were calculated using an IBM-360 computer. Figure 3 shows a flow diagram of the computer calculation procedure.

The results thus obtained are listed in Table 4 and Table 5 which show the Ostwald coefficients and mole fraction solubility respectively. The numbers appearing in these tables are computer output and should be treated as such in that the accuracy of the measurements does not go far beyond three significant digits. The solubility measurement results are also represented as the log of mole fraction solubility versus log temperature for each solvent in Figures 4 to 6. These graphs also show the critical temperature of the solvent and normal boiling point temperature of propane. Because of the great similarity in the solubility of the hexane isomers the five sets of data were included on one graph and were drawn as loci of points rather than single points.

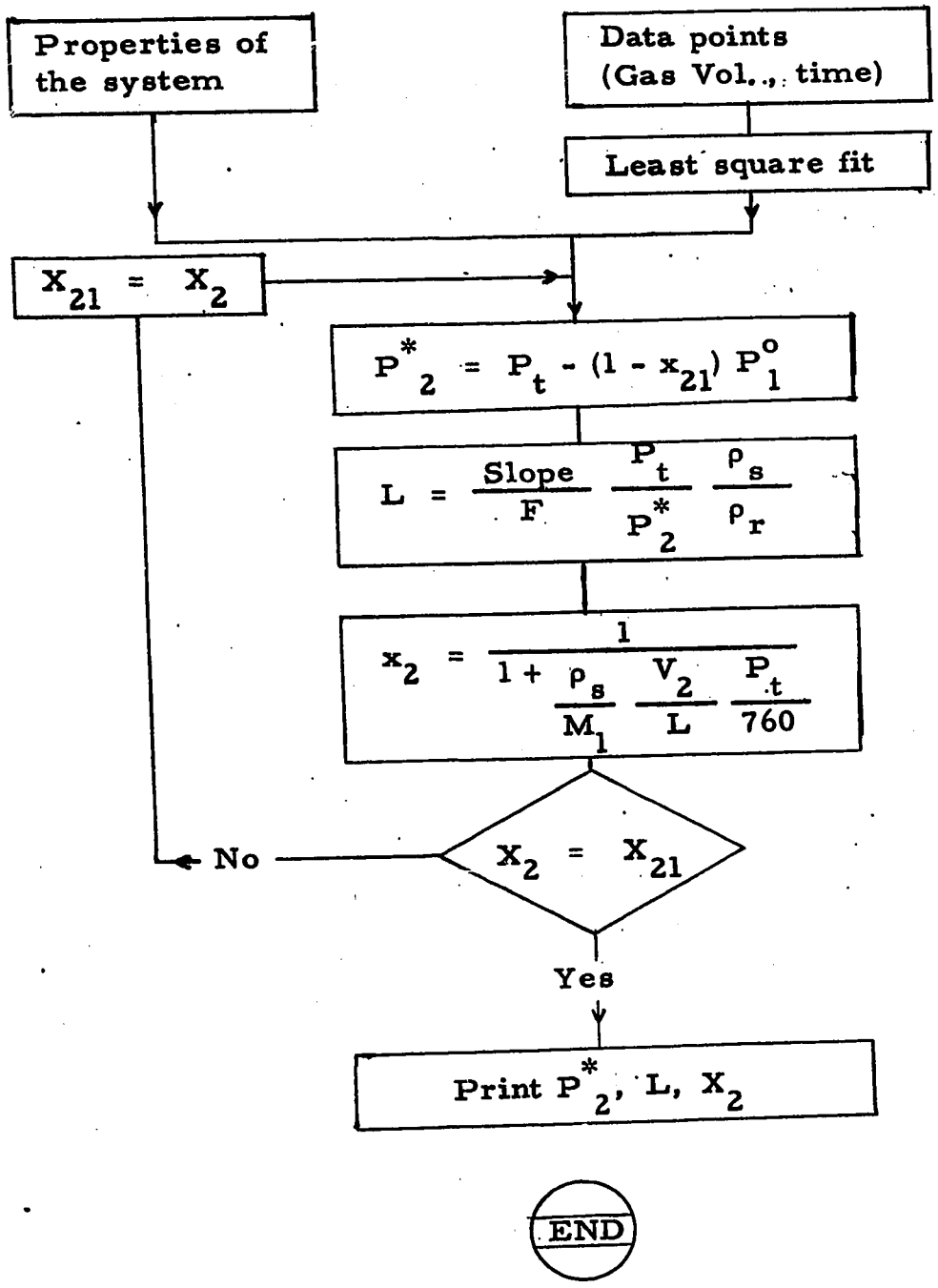

Figure 3 - Flow Diagram of the Computer Calculation Procedure

TABLE 4

SOLUBILITY OF PROPANE ,REPORTED AS OSTWALD COEFFICIENT

TEMP.('C)	L	L	L	AVERAGE
CARBON DISULFIDE				
-15	61.541	61.2075	0	61.3742
0	34.2265	34.3972	0	34.3118
25	18.5528	18.322	0	18.4374
35	14.4533	14.6303	14.5409	14.5415
BENZENE				
10	22.9067	23.0829	0	22.9948
25	15.9828	16.101	0	16.0419
50	10.7599	10.7736	0	10.7667
70	7.5708	7.6402	0	7.6055
CARBON TETRACHLORIDE				
-15	73.5509	77.3017	77.243	76.0318
0	42.5944	42.4717	0	42.533
25	21.7832	22.0938	21.7546	21.8772
50	13.3998	13.6854	13.615	13.5667
CYCLO-HEXANE				
10	33.3403	33.8181	33.3552	33.5045
25	22.621	22.5471	0	22.584
50	13.9727	13.3793	13.5104	13.6208
70	9.33	9.6584	0	9.4942
PERFLUORO-HEXANE				
-15	15.4591	15.502	0	[illegible]
5	9.0175	9.0394	0	9.02845
25	5.9054	5.901	0	5.9032
45	4.0143	3.9673	0	3.9908

Table 4...cont'd...

SOLUBILITY OF PROPANE ,REPORTED AS OSTWALD COEFFICIENT

TEMP.('C)	L	L	L	AVERAGE
N-HEXANE				
-15	83.4555	83.3987	83.3772	83.4105
5	40.4389	40.4154	0	40.4271
25	23.58	23.6458	0	23.6129
50	13.8767	13.84	0	13.8583
2-METHYLPENTANE				
-15	84.6145	84.3891	0	84.5018
5	40.4659	40.368	0	40.4169
25	24.1205	23.9623	0	24.0414
50	14.0436	14.1801	0	14.1118
3-METHYLPENTANE				
-15	83.8702	84.564	0	84.2171
5	43.2818	43.2383	0	43.26
25	23.163	23.3533	23.3923	23.3029
50	13.7674	13.9077	0	13.8375
2,2-DIMETHYLBUTANE				
-15	86.3738	86.9411	0	86.6574
5	41.8976	41.9767	0	41.9371
25	23.6507	23.9666	0	23.8086
40	15.0235	15.1009	0	15.0622
2,3-DIMETHYLBUTANE				
-15	86.8055	85.707	0	86.2562
5	41.9637	41.9533	41.9364	41.9511
25	24.2327	24.1209	0	24.1768
45	14.1945	14.2594	0	14.2269

TABLE 5

MOLE FRACTION SOLUBILITY DATA

TEMP. (°C)	X2	X2	X2	AVERAGE
CARBON DISULFIDE				
-15	.1464	.1457	0	.14605
0	.08397	.08443	0	.0842
25	.04483	.04434	0	.044585
35	.03424	.0347	.03445	3.44633E-2
BENZENE				
10	.08095	.08158	0	.081265
25	.05665	.05623	0	.05644
50	.036951	.036957	0	.036954
70	.02552	.02551	0	.025515
CARBON TETRACHLORIDE				
-15	.2495	.2564	.25625	.25405
0	.1548	.1544	0	.1546
25	.08096	.08206	.08091	.08131
50	.04908	.05016	.04991	4.97167E-2
CYCLO-HEXANE				
10	.1352	.1368	.13525	.13575
25	.09252	.09228	0	.0924
50	.05617	.05483	.05545	5.54833E-2
70	.037543	.03545	.03843	.037141
PERFLUORO-HEXANE				
-15	.1239	.1242	0	.124
5	.07305	.07321	0	.07313
25	.048	.04804	0	.04802
45	.03102	.03067	0	.030845

Table 5...cont'd...

MOLE FRACTION SOLUBILITY DATA

TEMP.('C)	X2	X2	X2	AVERAGE
N-HEXANE				
-15	.33737	.33724	.3348	.33647
5	.18819	.18808	0	.188135
25	.11543	.11571	0	.11557
50	.06747	.06749	0	.06748
2-METHYLPENTANE				
-15	.34247	.3419	0	.342185
5	.18983	.1896	0	.189715
25	.117833	.117879	0	.117856
50	.06911	.06996	0	.069535
3-METHYLPENTANE				
-15	.33724	.33902	0	.33813
5	.1987	.19884	0	.19877
25	.11178	.1128	.11294	.112507
50	.06693	.06769	0	.06731
2,2-DIMETHYLBUTANE				
-15	.34553	.34701	0	.34627
5	.19521	.19547	0	.19534
25	.11553	.11657	0	.11605
40	.075133	.075665	0	.075399
2,3-DIMETHYLBUTANE				
-15	.34253	.33977	0	.34115
5	.19112	.19107	.191	.191063
25	.11618	.11476	0	.11547
45	.070437	.070735	0	.070586

DISCUSSION OF RESULTS

As a check on the accuracy of apparatus and method used, some of the results obtained in this work were compared with available literature values. As can be deduced from Table 6, the results obtained in this project are in close agreement with those formerly obtained in this laboratory and other published values. Also it was found that duplicate or triplicate determinations showed very good reproducibility estimated at less than 0.7%.

In Figure 7, the logarithm of the mole fraction solubility at 25°C was plotted against the solubility parameter of solvents, δ_1. The observed relation for all substances except perfluorohexane could be used to predict solubility of propane in other solvents, provided the solubility parameter of the solvents were known. Here, perfluorohexane exhibits a propane solubility that is not in accordance with the trend shown by other solvents. But looking at a similar diagram for different gases published by Kobatake and Hildebrand [17], it can be noticed that, as the solubility of the gas increases, the line of $\log x_2$, versus δ_1 shows an increasing curvature with a maximum in solubility for $\delta_1 \approx 6.5$. This is consistent with the results obtained here, even though the curvature is not pronounced enough to account for such a low solubility of propane in perfluorohexane.

The results obtained in this work were compared with the mole fraction solubility at 25°C predicted by the regular solution theory (Equations 4 and 5) and also by ideal solution theory (Equation 3) This comparison is shown in Table 7. Equation 5 seems to predict solubilities that agree relatively well with experimentally determined solubilities for the solvents CS_2, C_6H_6, CCl_4 and C_6H_{12}. It seems that the correction for the difference in molecular volumes of solute and solvent is not needed for the system propane/n-hexane since Equation 4 gives the better agreement

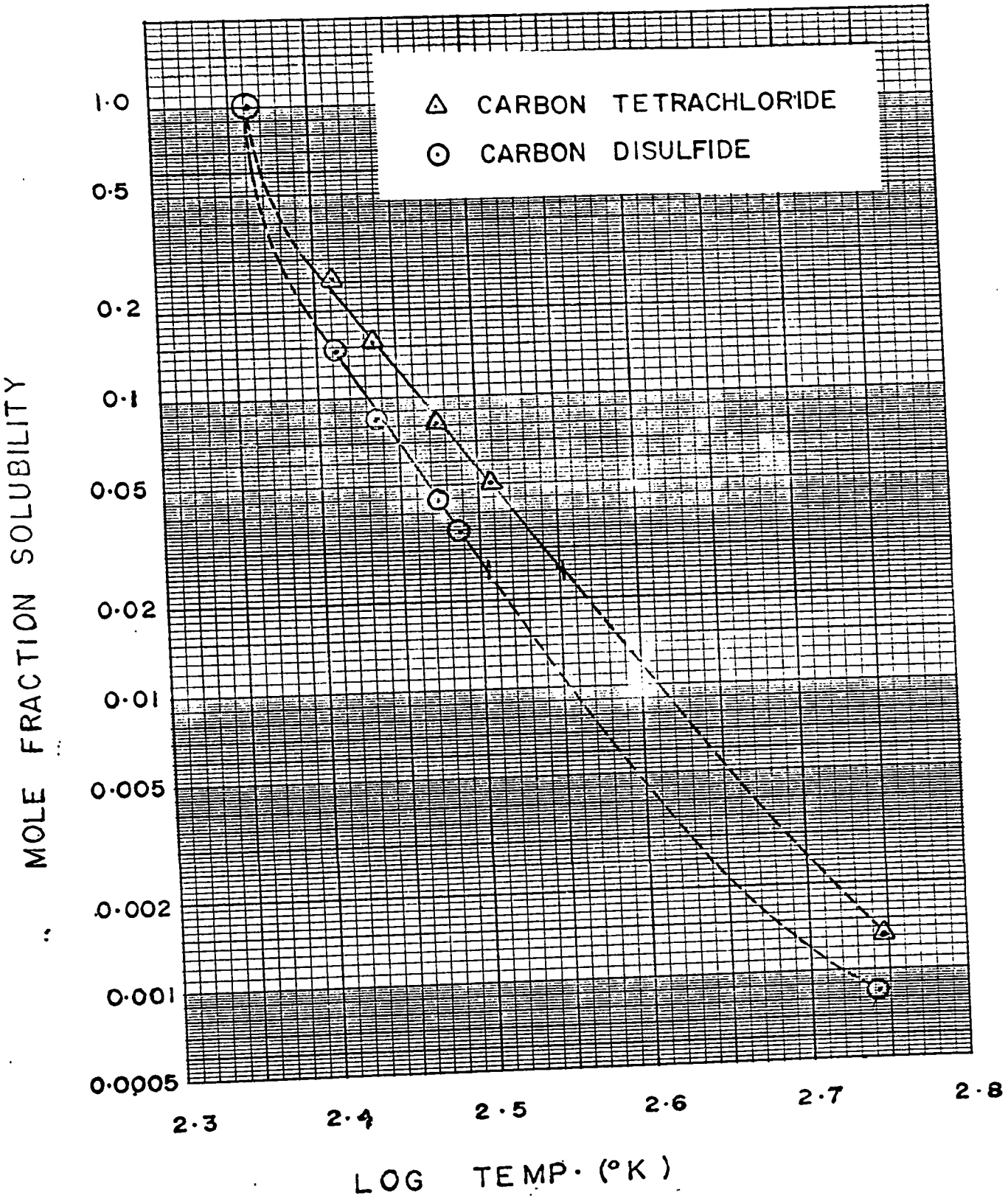

Figure 4 - Log Solubility versus Log Temperature for the solvents carbon tetrachloride and carbon disulfide

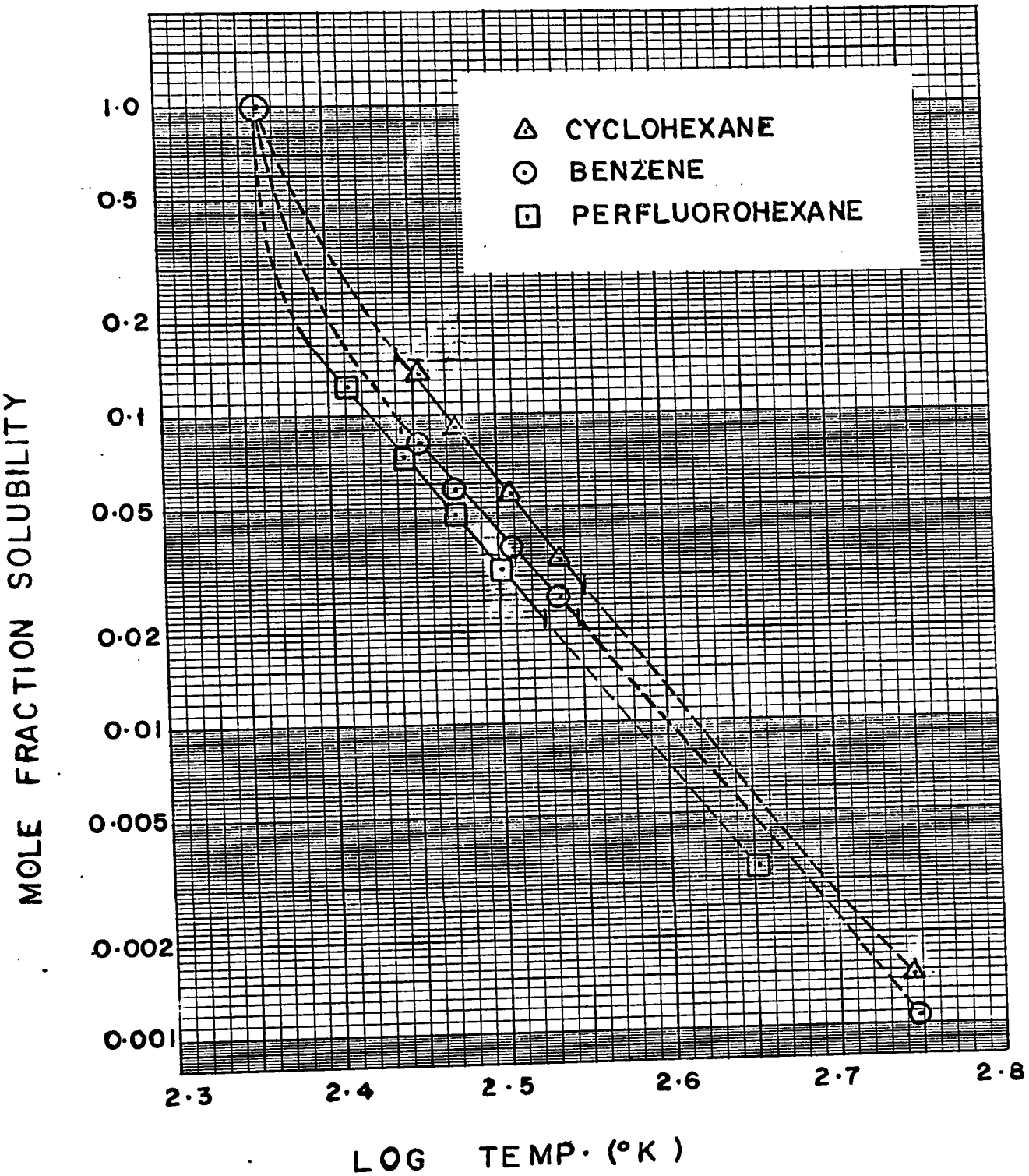

Figure 5 - Log Solubility versus Log Temperature for the solvents cyclohexane, benzene and perfluorohexane.

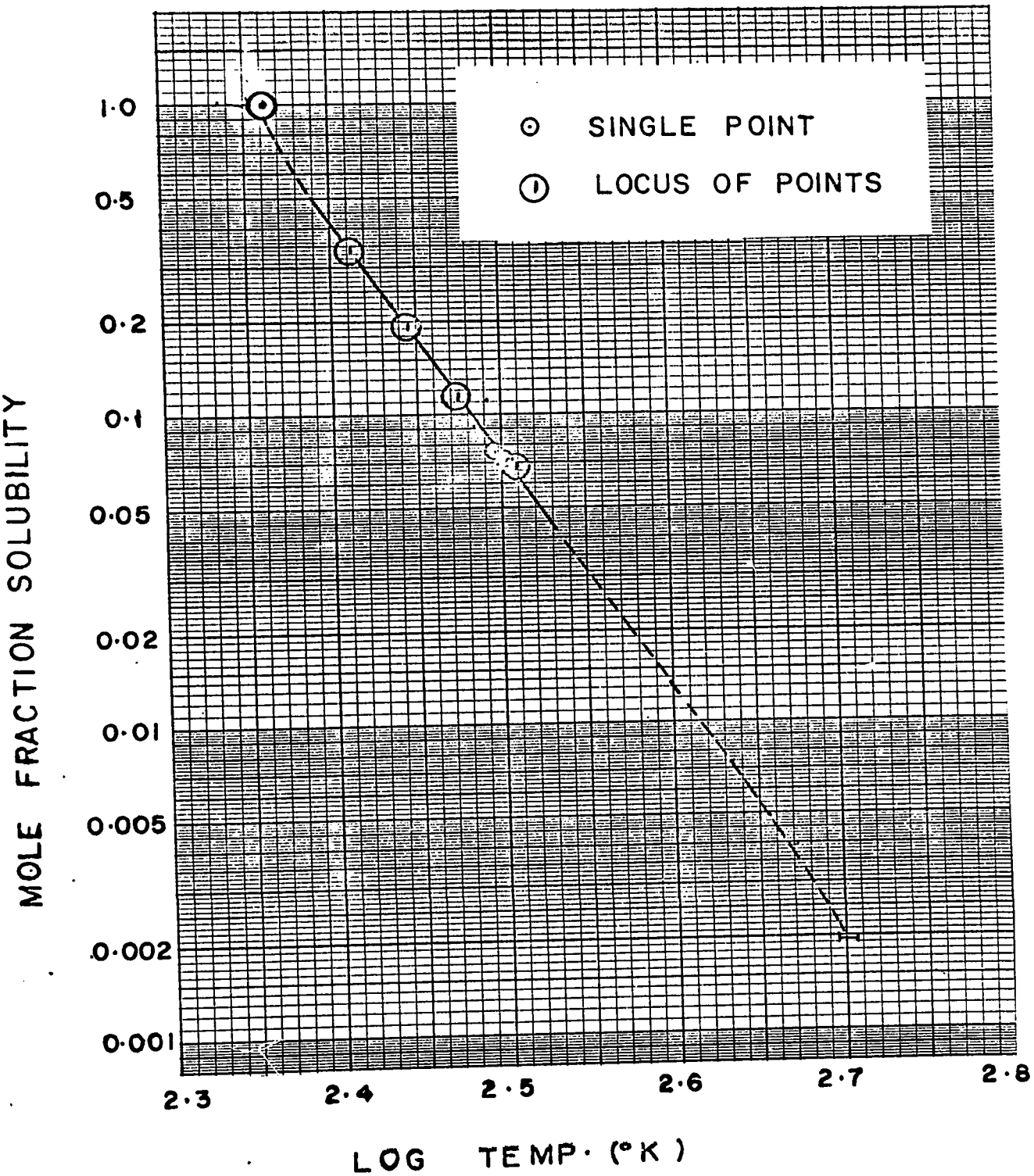

Figure 6 - Log Solubility versus Log Temperature for isomers of hexane

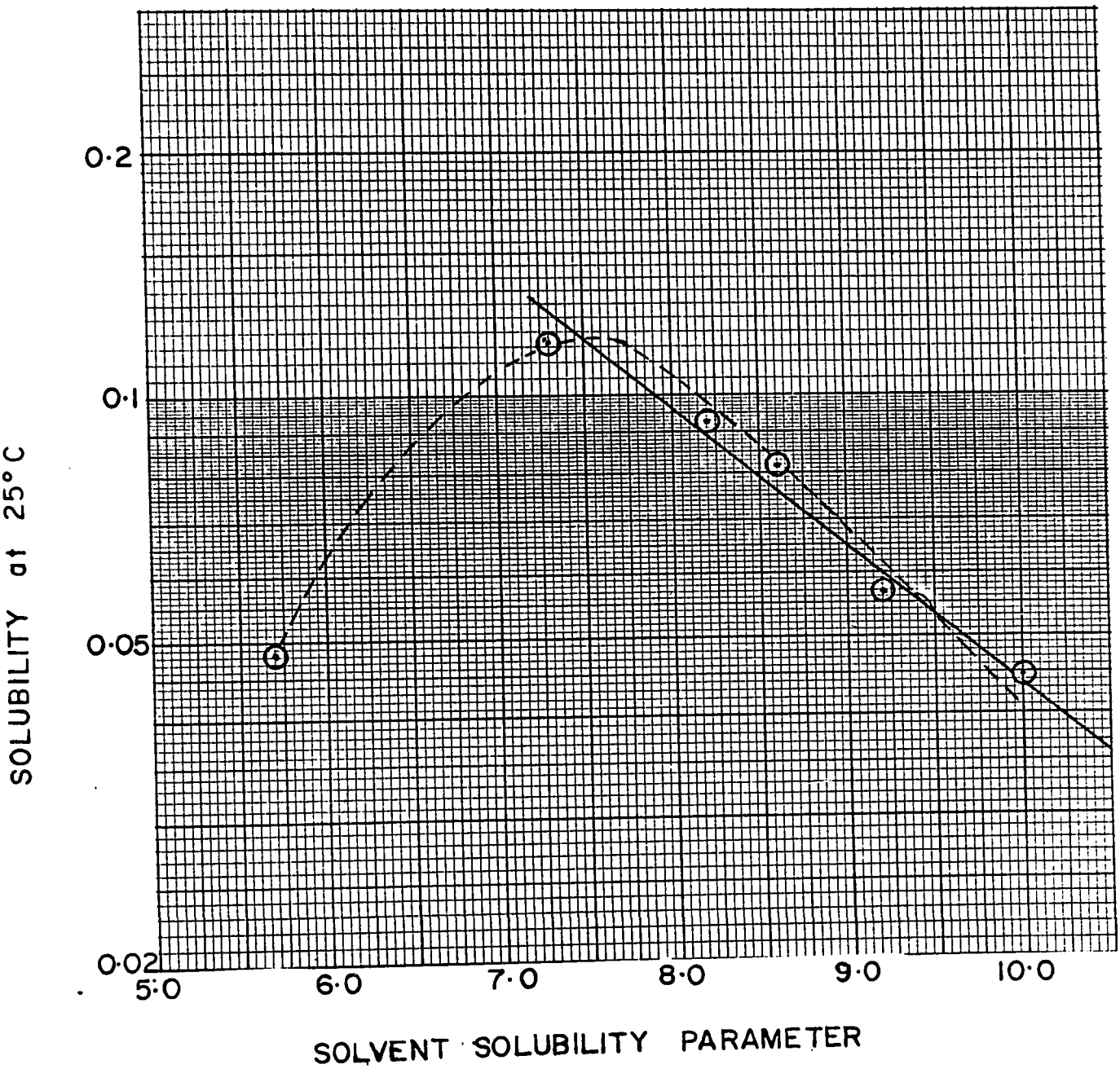

Fig. 7 Solubility at 25°C versus solvent solubility parameter

TABLE 6

Comparison of solubilities obtained in this work with literature values

System	Mole fraction solubility This work	Literature (ref.)
propane/n-hexane at 25°C	.1156	.116 (24) .115 (14) .1166 (15)
propane/benzene at 25°C	.0564	.0568 (24)

TABLE 7

Comparison of experimental solubility with Hildebrand's theoretical equations

Solvent	δ_1	Mole fraction solubitity at 25°C exp't.	eq. 4	eq. 5	ideal
CS_2	10	.0446	.0274	.0302	.120 [c]
C_6H_6	9.2	.0564	.0509	.0509	.120
CCl_4	8.6	.0813	.0727	.0860	.120
C_6H_{12}	8.2	.0924	.0875	.0894	.120
C_6F_{14}	7.3	.1156	.1135	.1230	.120
C_6H_{14}	5.7 [a]	.0480	.1066	.1495	.120
C_3H_8	6.65 [b]				

a) Solubility parameter calculated by Sargent and Seffl (21)
b) Solubility parameter of propane calculated from correlation by Thomsen and Gjaldbaek (24).
c) Ideal solubility of propane calculated from fugacities by Thomsen and Gjaldbaek (24).

TABLE 8

Comparison of solubility in different hexane isomers

Solvent	Mole fraction solubility at -15°C	25°C
n-hexane	.3365	.1156
2-methylpentane	.3422	.1178
3-methylpentane	.3381	.1125
2,2-dimethylbutane	.3463	.1161
2,3-dimethylbutane	.3418	.1155

for this solvent. Also it can be noted that this last system is close to ideal in that the components of the system are close members of an homologous series.

Though the agreement of the regular solution theory (Equation 5) with solubilities in perfluorinated hydrocarbon solvents is good for gases having lower solubilities such as CO_2 [7], H_2, O_2 and CO [10], N_2 [8] and H_2 and O_2 [4], for the highly soluble gas, propane, Equation 5 yields a 208% difference while Equation 4 yields a 122% difference from experimental values. These findings are similar to those reported in the literature by Thomsen ane Gjaldbaek [23] where for ethane /perfluoroheptane the percentage difference between experiment and Equation 5 and 4 was 140% and 42%, respectively. The same trend was noted by these authors for the relation between the solubility parameters and solubility at 25°C. The solubilities of ethane predicted by Equations 4 and 5 had increasing discrepancies compared to experimental values as the solubility parameter difference between solute and solvent increases.

In this work, the solubility of propane was measured in a number of hexane isomers. It was found that the solubility of propane in these isomers at 25°C had a mean of 0.1156 mole fraction with a standard deviation of 0.0017 about the mean. This seems to indicate that the position of the methyl group has little or no influence on the solubility. A list of mole fraction solubilities of propane in the different hexane isomers is given in Table 8. Perhaps the similarity of propane solubilities is better seen in Figure 6 where the points for the different isomers had to be grouped together and only the range could be indicated. Further evidence that this observation can be generalized to other types of solvents was supplied when the solubility of propane was found to be very similar in either ethylbenzene or m-xylene [7]. Solubilities of methane in these two solvents was also found to be very similar. Published data of solubility of inert gases in isomers of octane [2] seem to indicate also that the solubility of

a gas is not dependent on the shape or position of branches in the paraffinic molecules but depends instead mainly on the number of carbons present in the molecule. In these measurements, however, a significant difference in mole fraction solubility was found for all inert gases in 2, 2, 4-trimethylpentane (isooctane) as compared with the other isomers of octane. This difference may be accounted for by the difference in solubility parameters: 6.85 for isooctane and 7.55 for normal octane.

When mole fraction solubility is plotted against temperature on a log-log graph paper, the points form a straight line, as can be seen from Figures 4 to 6. Such a linear behaviour is characteristic of regular solutions. For most solvents, this line passes through the point defined by reference solubility (Equation 7) and critical temperature of solvent. Carbon disulfide however shows a rather large discrepancy in this respect but this solvent shows the same type of behavior for a number of other gases as well [13]. It is suggested that the reference solubility correlation with solubility parameter may have some drawback for carbon disulfide.

Also the log mole fraction solubility - log temperature curve can be extrapolated to the normal boiling point of propane (-42.07°C). At this temperature the mole fraction solubility should be unity. For a solution of propane in n-hexane or in its isomers, this extrapolation can be carried out with confidence to unity at -42.07°C since the extrapolation is almost linear. But the linearity decreases for other solvents. The liquid-liquid solubility of propane in the different solvents is not known but Hildebrand and Scott [16] give evidence to the fact that when two liquids have a difference between solubility parameters of more than about 2.4, a mixture of the two liquids would not be completely miscible at a temperature of about -40°C. For a value of solubility parameter of propane (5.95) the difference in solubility parameters between it and the solvent used in this work would be large enough to suggest that for cyclohexane, carbon tetrachloride, benzene and carbon disulfide

coexisting immiscible liquid phases may occur at -42°C. Further experiments would be needed to find out the exact liquid-liquid solubility of propane in the various solvents and thus determine the point to which the solubility - temperature curve should be extrapolated.

Though perfluorohexane has properties (such as vapor pressure, viscosity, boiling point...) similar to those of normal hexane, the solubility of propane in this solvent is much lower than can be accounted for by the regular solution theory. On the other hand, perfluorohexane behaves like most other solvents in that it has a linear temperature coefficient of solubility and the log x_2 - log T line passes through the reference solubility - critical temperature point (Figure 5).

DIFFUSIVITY OF GASES IN LIQUIDS

Molecular diffusion is concerned with the movement of individual molecules through a substance resulting from a concentration differential. The rate of diffusion is commonly described in terms of a molar flux and a diffusion coefficient as defined by Fick's law [34]:

$$J_2 = - D_{21} \frac{\partial C_2}{\partial Z} \qquad (13)$$

The flux of constituent 2 is measured relative to the average molar velocity of all constituents. By considering the effect of bulk flow the flux can be converted to one relative to a fixed position:

$$N_2 = (N_2 + N_1) \frac{C_2}{C} - D_{21} \frac{\partial C_2}{\partial Z} \qquad (14)$$

The diffusivity is characteristic of the diffusing component and its solvent. The mechanism of diffusion in liquids is still unknown and only semi-empirical correlations, based on known physical properties of the substances are available to predict diffusion coefficients for gas-liquid and liquid-liquid systems.

One of the best known correlations is the Wilke-Chang correlation [35] developed in 1955:

$$D_{21} = \frac{7.4 \times 10^{-8} (X_1 M_1)^{1/2} T}{\mu_1 V_2^{0.6}} \qquad (15)$$

The main drawback of this equation is the use of the association number which has to be determined experimentally for associated solvents. It has a value of 1 for unassociated substances, 2.6 for water, 1.9 for methanol and 1.5 for ethanol [31]. Also, the accuracy of the equation decreases as the solvent viscosity increases. It is noted that the Wilke-Chang equation was derived using predominantly experimental liquid-liquid and solid-liquid diffusion coefficients. Therefore its application to gas-liquid systems is

somewhat of an extrapolation of the data from which it was derived.

Scheibel [33] derived a correlation that did not include the association parameter originally defined by Wilke and Chang but which was very similar to the Wilke-Chang equation:

$$D_{21} = 8.2 \times 10^{-18} T \left[1 + \left(\frac{3 V_2}{V_1} \right)^{2/3} \right] / \mu_1 V_2^{1/3} \quad (16)$$

More recently Akgerman and Gainer developed a new theoretical equation [25, 26]:

$$D_{21} = \frac{k T}{\epsilon_2 \mu_1} \left(\frac{N_A}{V_1} \right)^{1/3} \left(\frac{M_1}{M_2} \right)^{1/2} \exp \left(\frac{E_{\mu_1} - E_{D_{21}}}{RT} \right) \ldots$$

$$\ldots\ldots\ldots\ldots \quad (17)$$

In this equation, the geometrical parameter is calculated from the solute and solvent molar volumes:

$$\epsilon_2 = 6 \left(\frac{V_2}{V_1} \right)^{1/6} \quad (18)$$

The activation energy difference is given by the following equations:

$$E_{\mu_1} - E_{D_{21}} = E_{11} \left[1 - \left(\frac{E_{22}}{E_{11}} \right)^{1/(\epsilon_2 + 1)} \right] \quad (19)$$

$$E_{22} = - \left(R \ln \frac{\mu_{T_a}}{\mu_{T_b}} + \frac{R}{2} \ln \frac{T_a}{T_b} \right) \Big/ \left(\frac{1}{T_b} - \frac{1}{T_a} \right) \ldots$$

$$\ldots\ldots\ldots\ldots \quad (20)$$

$$E_{11} = 5875.3 \, M_1^{-0.186} \quad (21)$$

A simpler correlation for diffusivity of gases in liquid solvents was proposed by Hayduk and Cheng [30]. The proposition was made that there was not enough evidence supporting the inclusion of the absolute temperature or molar volumes in correlations for diffusion coefficients

and that a simple relation for the diffusivity of a substance could be expressed in terms of solvent viscosity by an equation of the form:

$$D = A \mu^{B} \tag{22}$$

Later the values of 0.591×10^{-10} and -0.545 were found for A and B respectively for diffusivities of propane in different solvents [29].

The method used in this work for measuring diffusion coefficients was the steady-state capillary cell method described earlier by Malik and Hayduk [32] and later improved by Hayduk and Cheng [30]. The mathematical analysis for this method has been described earlier [32] and will be presented here with a few additions and refinements.

One can arrive at a number of different equations relating diffusivity and the absorption rate of the gas, depending on the initial assumptions. When the mass density of solute in the liquid phase is considered approximately equal to that of the solvent, Fick's first law of diffusion in the following form can be applied:

$$n_2 = -\rho D_{21} \frac{d \bar{w}_2}{d \bar{z}} + \bar{w}_2 (n_2 + n_1) \tag{23}$$

The displacement of the column in the capillary can be expressed as:

$$\frac{n_2}{\rho_{2_E}} = - \frac{(n_2 + n_1)}{\rho_L} \tag{24}$$

Replacing $(n_2 + n_1)$ into Equation 23, the result is:

$$n_2 = - \frac{\rho_L}{\rho_{2_E}} n_2 \bar{w}_2 - \rho D_{21} \frac{d \bar{w}_2}{d \bar{z}} \tag{25}$$

If $k = \frac{\rho_L}{\rho_{2_E}}$ then the following differential equation results:

$$n_2 (1 + k w_2) = - \rho D_{21} \frac{d w_2}{d z} \tag{26}$$

Integration of Equation 26 yields an expression for the diffusivity as follows:

$$D_{21} = \frac{n_2 L}{\rho_{2_E} \ln \left[\frac{1 + k w_{2_O}}{1 + k w_{2_L}} \right]} \tag{27}$$

Equation 27 is further simplified for $k \approx 1$:

$$D_{21} = \frac{n_2 L}{\rho_L \ln \left[\frac{1 + w_{2_O}}{1 + w_{2_L}} \right]} \tag{28}$$

For a constant molar concentration along the diffusion path in the capillary, equations similar to Equations 27 and 28 can be derived through a similar procedure. These equations are:

$$D_{21} = \frac{N_2}{C_{2_E}} \frac{L}{\ln \left[\frac{1 + K x_{2_O}}{1 + K x_{2_L}} \right]} \tag{29}$$

and

$$D_{21} = \frac{N_2}{C} \frac{L}{\ln \left[\frac{1 + x_{2_O}}{1 + x_{2_L}} \right]} \tag{30}$$

If neither assumption is at least approximately valid, as found to be the case for diffusivities of propane in perfluorohexane, the assumption of a linearly varying concentration profile in the capillary can be made. Then the diffusion coefficient can be expressed as:

$$D_{21} = \frac{n_2 \; L}{\rho_{2_E} \ln \left[\frac{1 + \rho_{2_O} / \rho_{2_E}}{1 + \rho_{2_L} / \rho_{2_E}} \right]} \tag{31}$$

Similarly the diffusion coefficient can be expressed in terms of molar concentrations:

$$D_{21} = \frac{N_2 \; L}{C_{2_E} \ln \left[\frac{1 + C_{2_O} / C_{2_E}}{1 + C_{2_L} / C_{2_E}} \right]} \tag{32}$$

The mass flux and molar flux can be expressed in terms of the experimentally determined rate of bead descent:

$$n_2 = h \times \frac{\rho_2}{760} \times \frac{M_2}{V_2 \text{ (gas)}} \tag{33}$$

$$N_2 = \frac{h}{V_2 \text{ (gas)}} \times \frac{\rho_2}{760} \tag{34}$$

In order to decide which assumption was more appropriate for a particular dissolved gas-solvent system, the percent difference between effective molar volumes of solute and solvent was calculated and compared to the percent difference between effective mass densities of solute and solvent. Table 9 shows the results of these calculations. The mass or molar concentration that yielded the smaller percentage difference was subsequently used as a basis for calculation of diffusivity. For benzene, at all temperatures of determination, and for cyclohexane at 50°C, the molar concentrations yielded the smaller percent difference so the assumption of constant molar concentration was made whereas, for cyclohexane at 10°C and 25°C, the mass concentration percentage difference

TABLE 9

Comparison of % Difference in Concentrations [a]

TEMP.('C)	PROPANE	BENZENE	% DIFF.
10	.586	.8894	51.7747
	75.2389	87.8345	16.7408
25	.565	.8734	54.5841
	78.0354	89.4435	14.6192
50	.53	.8465	59.717
	83.1887	92.2859	10.9356

TEMP.('C)	PROPANE	CYCLOHEXANE	% DIFF.
10	.586	.7881	34.488
	75.2389	106.788	41.9325
25	.565	.7743	37.0443
	78.0354	108.692	39.2851
50	.53	.7504	41.5849
	83.1887	112.154	34.8182

TEMP.('C)	PROPANE	PERFLUOROHEXANE	% DIFF.
-10	.586	1.7845	204.522
	75.2389	189.442	151.788
5	.565	1.7421	208.336
	78.0354	194.053	148.673
25	.53	1.6718	215.434
	83.1887	202.213	143.078

---

a for each system the first line is the calculation for mass density and the second line, the calculation for molar concentrations.

was smaller and the constant mass concentration assumption was made. For systems with perfluorohexane, both percentage differences were very large, therefore the assumption of a linearly varying concentration was used. When this assumption is made, either the mass or molar concentration can be used for calculations.

APPARATUS AND PROCEDURE

The method used in this research project to measure propane diffusivities was the steady-state capillary cell method described first by Malik and Hayduk [32] and later refined by Hayduk and Cheng [30]. This cell, shown in Figure 8, consisted of an upper, small diameter (1 mm.), thick wall capillary tubing sealed into a small, tubular glass reservoir. Stopcocks located on either side of this reservoir permitted filling of the cell. Used in conjunction with this cell was a constant temperature, full-visibility bath (Neslab Instruments Inc. Model TEV-40) into which the cell was immersed, and a cathetometer (Precision Tool and Instruments Company) which was used to measure the rate of descent of the saturated bead of liquid. An electric timer was used in conjunction with the cathetometer to measure the rate.

Diffusivity determinations were started by degassing the solvent, the procedure for which was explained in the Solubility section of this thesis. The only difference in the degassing apparatus was that the rubber septum at the bottom of the accumulation tube was replaced by a piece of Tygon tubing closed by a pinchcock. The cells were cleaned and the stopcocks carefully greased using water resistant grease (Vaseline) on the extremities and hydrocarbon resistant grease (Nonaq Stopcock Grease, Fisher Scientific Co.) in the center. The cells were then filled with degassed solvent by fitting the Tygon tubing of the accumulation column to the cell and opening the stopcocks. When enough solvent had been purged through to insure that there was no dissolved air contamination, the stopcocks on either side of the cell were closed and the cell fitted into the constant temperature bath. The capillary tubing of the diffusion cell was then connected to a glass tee arrangement through which propane gas was permitted to flow. The level of liquid was then adjusted to obtain a diffusion path of about 3 centimeters in length. After the cell had been immersed in the bath for a period of

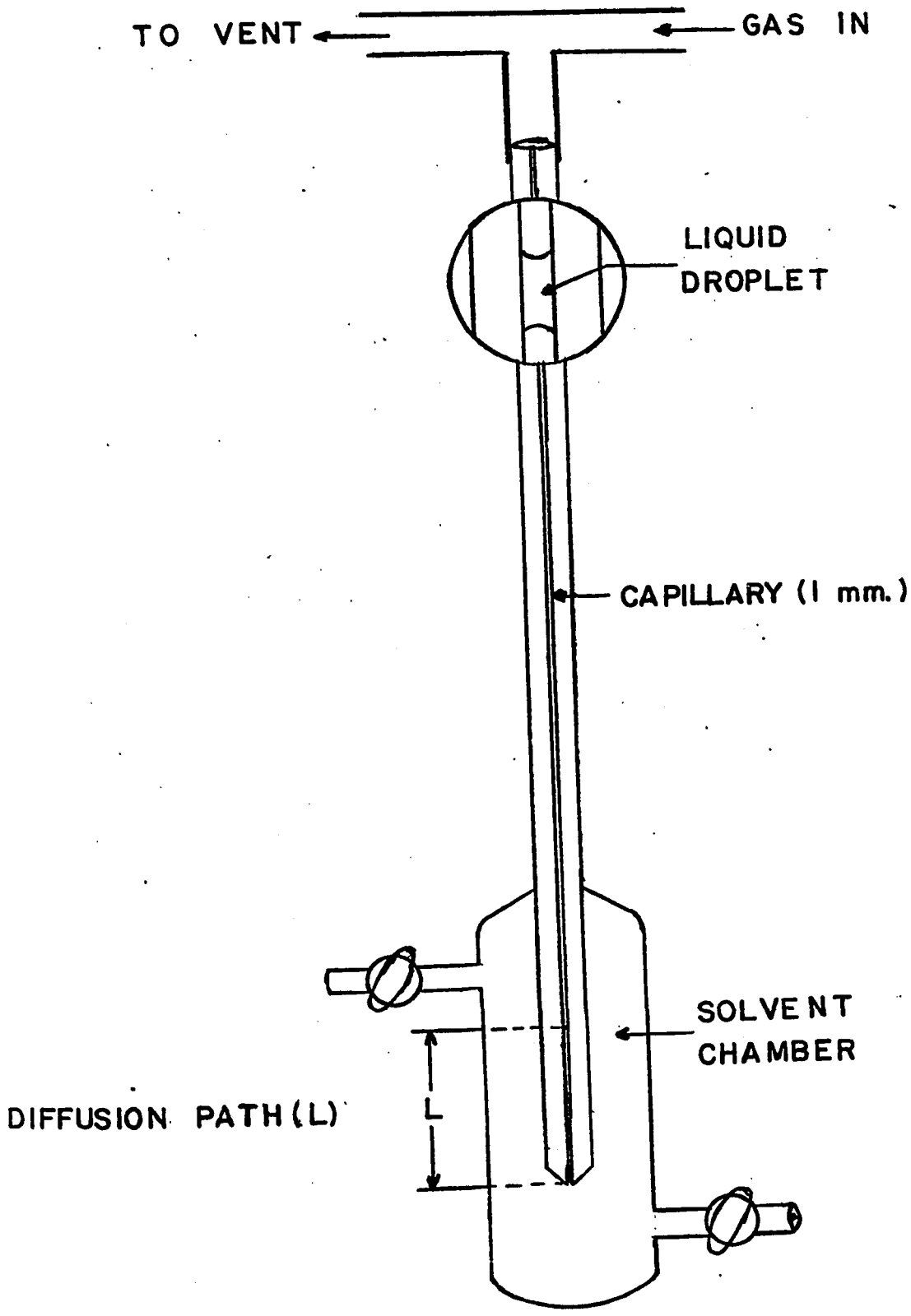

Figure 8 - Diffusivity Cell

about 36 hours or until steady-state was reached, a droplet of propane-saturated solvent was injected into the top of the capillary by means of a syringe. The rate of descent of the bead was measured at 4-10 minutes intervals during a period of about 2 hours.

RESULTS AND DISCUSSION

The results of the diffusivity determinations are reported in Table 10, in which are listed the measured diffusivities of propane in benzene and cyclohexane at 10, 25 and 50°C and in perfluorohexane at -10, 5 and 25°C. Also included in Table 10 is the diffusion coefficient of propane in normal hexane at 25°C, which was measured as a check on the experimental procedure. The raw experimental data for these measurements are given in the appendix. The diffusivity value for propane in hexane at 25°C measured in this work was 4.68×10^{-9} m^2/sec. compared to $4.87 \pm 0.20 \times 10^{-9}$ m^2/sec reported by Hayduk et al [29]. Bromfield [27] reported a value of 4.68×10^{-9} m^2/sec. and Castaneda [28] reported one of 4.48×10^{-9} m^2/sec. The value obtained in this work is favorably compared with the values obtained earlier in this laboratory.

The experimental diffusion coefficients were compared to ones predicted by the Wilke-Chang correlation (Equation 15), the Scheibel correlation (Equation 16) the Akgerman-Gainer equation (Equation 17) and the diffusivity - viscosity correlation (Equation 22). This comparison is presented in the form of a table (Table 11). The diffusivity plotted against solvent viscosity on log-log graph paper are shown in Figure 9. As can be seen from this graph and from Table 11, the correlation developed by Hayduk and Cheng [6] represents most of the data accurately, especially for normal hexane, cyclohexane and benzene. All the other correlations tested here show a fit that is, in every case, further away from experimental measurements. Even if it has better agreement, not even the viscosity - diffusivity correlation represents the diffusivities of propane in perfluorohexane well. This may be due to the fact that perfluorohexane molecules are very large and that the association between them is almost non-existent. It would be difficult to predict any of the properties of perfluorohexane

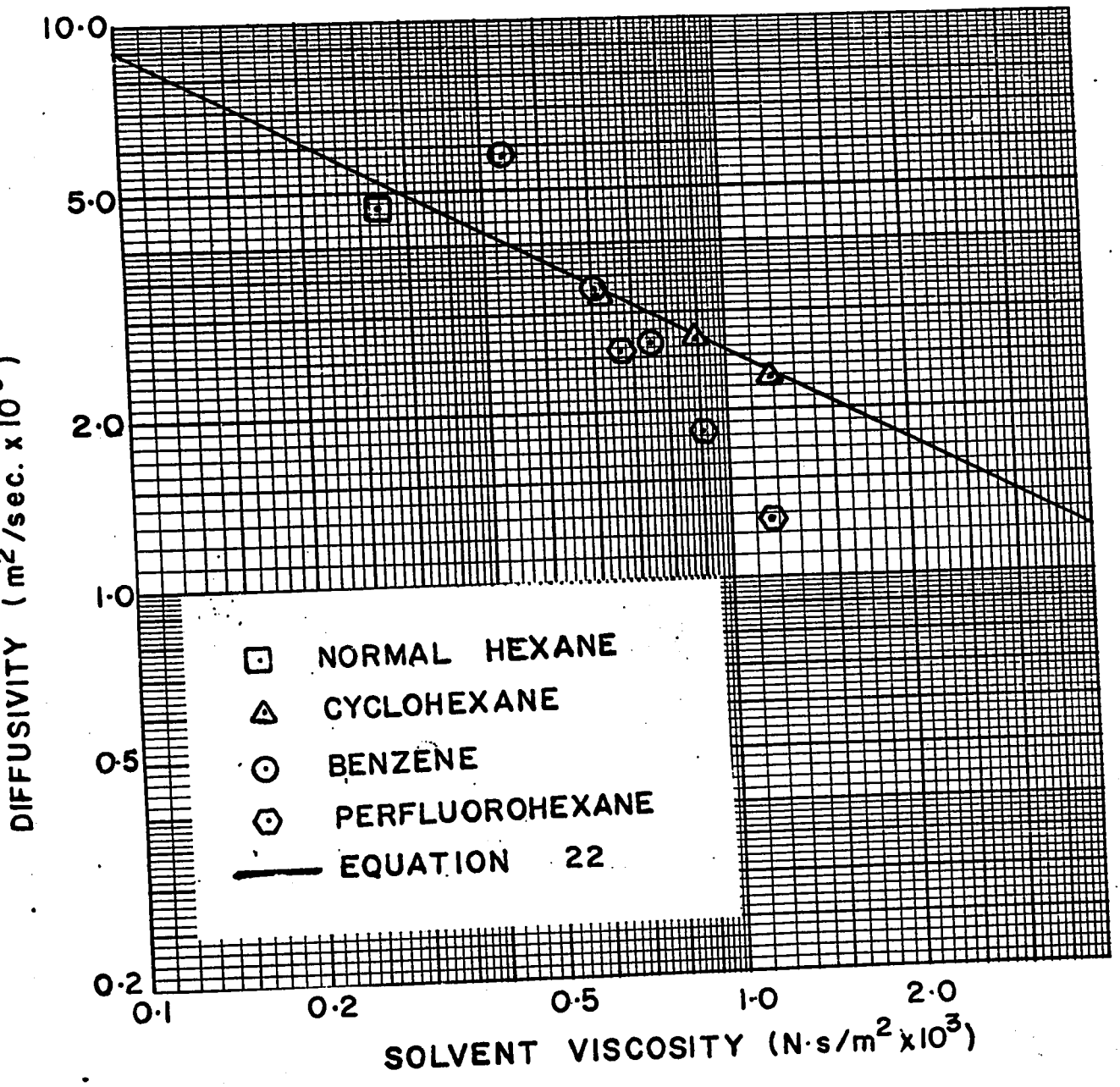

Figure 9 - Diffusivity versus Viscosity

TABLE 10

Results of Diffusivity Determinations (D in $m^2/sec \times 10^{-9}$)

TEMP. (°C)	D	D	D	AVERAGE
NORMAL HEXANE				
25	4.686	4.678	0	4.682
BENZENE				
10	2.645	2.651	2.631	2.64233
25	3.315	3.337	0	3.326
50	5.718	5.784	5.845	5.78233
CYCLO-HEXANE				
10	2.249	2.262	2.327	2.27933
25	2.685	2.681	0	2.683
50	3.278	3.233	3.386	3.299
PERFLUORO-HEXANE				
-10	1.312	1.249	1.264	1.275
5	1.781	1.838	1.857	1.82533
25	2.449	2.662	0	2.5555

TABLE 11

Comparison of propane experimental diffusivities with correlations

Temp. (°C)	Diffusivities ($m^2/sec \times 10^9$) experimental	Wilke-Chang	Scheibel	Akgerman-Gainer	Hayduk et al.
n-hexane					
25	4.682	5.048	2.020	1.925	5.210
benzene					
10	2.642	1.849	1.961	1.900	2.985
25	3.326	2.367	2.565	1.968	3.360
50	5.782	3.414	4.093	2.078	4.009
cyclohexane					
10	2.279	1.2185	1.233	1.615	2.330
25	2.683	1.649	1.699	1.688	2.704
50	3.299	2.549	2.708	1.805	3.351
perfluorohexane					
-10	1.275	2.370	0.927	2.658	2.341
5	1.825	3.164	1.046	2.787	2.699
25	2.556	4.491	1.541	2.956	3.196

unless similar properties had been experimentally measured for homologous compounds. A correlation based on other types of compounds probably would not apply to this solvent.

Experiments were also started for benzene and cyclohexane at 70°C and for perfluorohexane at 45°C, but it was impossible to measure the rate of descent of the bead since, most of the time after initial injection into the capillary, the bead rose back up and out of the capillary. When it did fall, it did so at an accelerated rate for a short period of time and then went back up. Such irregular effects appeared to be associated with the very high vapor pressure of these solvents at those temperatures (572 mm for benzene, 555 mm for cyclohexane and 498 mm for perfluorohexane).

CONCLUSIONS

In this work it was found that, with the exception of perfluorohexane, the solubility of propane decreased with increasing solubility parameter of the solvent. When the results of solubility measurements were compared with those calculated from the regular solution theory, again with the exception of perfluorohexane, there existed a fair agreement. It was also found that the solubility of propane in normal hexane and its isomers was essentially the same and very close to the ideal solubility.

It was found that the position of a methyl group on the main carbon chain of the solvent molecules did not appear to affect the solubility of a gas in that solvent. Even if a solvent with two methyl groups were replaced by a solvent with one ethyl group, the solubility of a gas in the solvent would be expected to remain essentially unchanged.

This work essentially confirms the work done by previous workers in this laboratory with regard to the temperature coefficient of solubility. As predicted, the log solubility - log temperature line was found to pass through the reference solubility of the solvent at its critical temperature.

Finally it was found that the solubility of propane in perfluorohexane appeared to be an exception to all the existing correlations involving regular solution theory.

The diffusion coefficients measured in this work for propane in hydrocarbons could have been predicted by the diffusivity - viscosity correlation to an accuracy of greater than 31% with an average difference of about 9%. The other correlations tested here give estimated values of diffusivity much lower than the experimental values. The diffusivity of propane in perfluorohexane is not represented well by any of the correlations investigated.

The apparatus and method used to measure diffusion coefficients in this work are believed to be accurate and reliable for any gas-liquid system provided the vapor pressure of the solvent is less than about 300 mm. of mercury.

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APPENDIX

Raw experimental data for diffusivity measurements.

R1	=	ρ_1,	solvent density (gm/cc)
R2	=	ρ_{2_E},	density of dissolved propane (gm/cc)
P	=	P_t,	atmospheric pressure (mm Hg)
PO	=	P_1^o,	vapor pressure of solvent (mm Hg)
X2	=	x_2,	mole fraction solubility
M2	=	M_2,	molecular weight of propane (gm/mole)
M1	=	M_1,	molecular weight of solvent (gm/mole)
H	=	h,	rate of descent of bead (cm/sec)
V2	=	V_2,	molar volume of gas at experimental conditions (cc/mole)
L	=	L,	diffusion path length (cm)
D1, K	=		diffusivity calculated using K value (cm^2/sec)
D2	=		diffusivity calculated if K = 1 (cm^2/sec)
D, VAR. CONC.	=		diffusivity calculated using linearly variable concentration.

DIFFUSIVITY, CONSTANT MASS CONCENTRATION
PROPANE-HEXANE @ 25°C MARCH 7 # 1

TIME(SEC)	LENGTH(CM)
0	0
500	.212
1000	.443
1500	.664
2000	.878
2500	1.123
3000	1.374
3500	1.612
4000	1.813

Y= 4.59105E-4 *X + -1.60999E-2

R1= .65501 R2= .565 P= 763.7 P0= 151.58 X2= .1155
M2= 44.09 M1= 86.18 H= 4.59105E-4 V2= 24105.8 L= 2.201

D1,K= 4.68632E-5 D2= 4.68707E-5 D,VAR.CONC.= 4.72356E-5

DIFFUSIVITY, CONSTANT MASS CONCENTRATION
PROPANE -HEXANE @ 25°C MARCH 7 #2

TIME(SEC)	LENGTH(CM)
0	0
500	.21
1000	.424
1500	.647
2000	.86
2500	1.11
3000	1.356
3500	1.594
4000	1.802

Y= 4.56238E-4 *X + -2.32548E-2

R1= .65501 R2= .565 P= 763.7 P0= 151.58 X2= .1155
M2= 44.09 M1= 86.18 H= 4.56238E-4 V2= 24105.8 L= 2.211

D1,K= 4.67822E-5 D2= 4.67897E-5 D,VAR.CONC.= 4.71539E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-BENZENE @ 10°C MAY 5 #1

TIME(SEC)	LENGTH(CM)
1000	.155
1250	.194
1500	.238
1750	.278
2000	.317
2250	.357
2500	.405
2750	.44
3000	.476
3270	.522
3500	.571
3750	.606
4000	.644
4250	.684
4500	.734
4750	.772

Y= 1.64381E-4 *X + -.010739
R1= .889408 R2= .586 P= 753.3 P0= 45.6158 X2= .08128
M2= 44.09 M1= 78.12 H= 1.64381E-4 V2= 22839.8 L= 3.294

D1,K= 2.64458E-5 D2= 2.64362E-5 D,VAR.CONC.= 2.6166E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-BENZENE @ 10°C MAY 4 #2

TIME(SEC)	LENGTH(CM)
0	0
250	.039
500	.083
750	.125
1000	.159
1250	.206
1500	.241
1750	.291
2000	.343

Y= 1.68334E-4 *X + -3.11208E-3
R1= .889408 R2= .586 P= 750.8 P0= 45.6158 X2= .08128
M2= 44.09 M1= 78.12 H= 1.68334E-4 V2= 22839.8 L= 3.225

D1,K= 2.65114E-5 D2= 2.65022E-5 D,VAR.CONC.= 2.62319E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-BENZENE @ 10° C MARCH 21 #2

TIME(SEC)	LENGTH(CM)
0	0
500	.135
1000	.27
1500	.418
2000	.554
2500	.699
3000	.839
3500	.981
4000	1.115
4500	1.255
5000	1.397
5500	1.534
6000	1.668
6500	1.804
7000	1.937

Y= 2.78176E-4 *X + 1.15585E-4

R1= .889408	R2= .586	P= 762.6	P0= 45.6158	X2= .08128
M2= 44.09	M1= 78.12	H= 2.78176E-4	V2= 22839.8	L= 1.936
D1,K= 2.63135E-5		D2= 2.6304E-5	D, VAR.CONC.= 2.60318E-5	

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-BENZENE @ 25°C FEB 26 #1

TIME(SEC)	LENGTH(CM)
0	0
600	.171
1300	.371
2050	.588
2600	.753
3900	1.126
4600	1.319

Y= 2.87912E-4 *X + -7.25337E-4

R1= .8534	R2= .565	P= 772.9	P0= 94.99	X2= .05736
M2= 44.09	M1= 78.12	H= 2.87912E-4	V2= 24105.8	L= 1.702
D1,K= 3.31509E-5		D2= 3.31448E-5	D, VAR.CONC.= 3.29036E-5	

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-BENZENE @ 25°C MARCH 9 #2

TIME(SEC)	LENGTH(CM)
0	0
650	.209
1000	.326
1500	.51
2000	.678
2500	.849
3000	1.016
3550	1.206
4000	1.361
4500	1.538
5000	1.708
5500	1.884
6000	2.051

Y= 3.43467E-4 *X + -9.84074E-3

R1= .8734	R2= .565	P= 772.2	P0= 94.99	X2= .05736
M2= 44.09	M1= 78.12	H= 3.43467E-4	V2= 24105.8	L= 1.469

D1,K= 3.33673E-5 D2= 3.33628E-5 D,VAR.CONC.= 3.3153E-5

DIFFUSIVITY, CONSTANT MASS CONCENTRATION
PROPANE-BENZENE @ 50°C MAY 26 #1

TIME(SEC)	LENGTH(CM)
0	0
250	.051
500	.106
750	.158
1000	.213
1250	.264
1500	.317
1750	.372
2000	.425

Y= 2.12735E-4 *X + -9.56747E-4

R1= .846499	R2= .53	P= 757.1	P0= 268.653	X2= .036936
M2= 44.09	M1= 78.12	H= 2.12735E-4	V2= 26203.2	L= 2.818

D1,K= 5.71832E-5 D2= 5.7184E-5 D,VAR.CONC.= 5.76518E-5

DIFFUSIVITY, CONSTANT MASS CONCENTRATION
PROPANE-BENZENE @ 50°C MAY 20 #1

TIME(SEC)	LENGTH(CM)
500	.055
750	.107
1000	.155
1250	.208
1500	.258
1750	.309
2000	.36
2250	.405
2500	.454
2750	.503

Y= 1.9922E-4 *X + -4.23323E-2
R1= .846499 R2= .53 P= 753.5 P0= 268.653 X2= .03694
M2= 44.09 M1= 78.12 H= 1.9922E-4 V2= 26203.2 L= 3.044

D1,K= 5.78394E-5 D2= 5.78406E-5 D,VAR.CONC.= 5.83077E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-BENZENE @ 50°C MAY 20 #1

TIME(SEC)	LENGTH(CM)
500	.055
750	.107
1000	.155
1250	.208
1500	.258
1750	.309
2000	.36
2250	.405
2500	.454
2750	.503

Y= 1.9922E-4 *X + -4.23323E-2
R1= .846499 R2= .53 P= 753.5 P0= 268.653 X2= .03694
M2= 44.09 M1= 78.12 H= 1.9922E-4 V2= 26203.2 L= 3.044

D1,K= 5.84452E-5 D2= 5.84452E-5 D,VAR.CONC.= 5.83077E-5

DIFFUSIVITY, CONSTANT MASS CONCENTRATION
PROPANE-CYCLOHEXANE @ 10°C MARCH 15 #3

TIME(SEC)	LENGTH(CM)
2250	1.278
2500	1.388
2750	1.512
3000	1.625
3250	1.742
3500	1.861
3750	1.967
4000	2.058

Y= 4.53301E-4 *X + .262309
R1= .788095 R2= .586 P= 746.8 P0= 47.47 X2= .1352
M2= 44.09 M1= 84.16 H= 4.53301E-4 V2= 22839.8 L= 1.458

D1,K= 2.24858E-5 D2= 2.24984E-5 D,VAR.CONC.= 2.30046E-5

DIFFUSIVITY, CONSTANT MASS CONCENTRATION
PROPANE-CYCLOHEXANE @ 10°C MAY7 #2

TIME(SEC)	LENGTH(CM)
0	0
250	.058
500	.112
750	.173
1000	.223
1250	.271
1500	.326
2000	.426
2250	.477
2500	.531
2750	.58
3000	.634
3250	.682
3500	.73
3750	.78

Y= 2.06826E-4 *X + .010677
R1= .788095 R2= .586 P= 768.4 P0= 47.47 X2= .1352
M2= 44.09 M1= 84.16 H= 2.06826E-4 V2= 22839.8 L= 3.216

D1,K= 2.26181E-5 D2= 2.26319E-5 D,VAR.CONC.= 2.31563E-5

DIFFUSIVITY, CONSTANT MASS CONCENTRATION
PROPANE-CYCLOHEXANE @ 10°C MAY 7 #3

TIME(SEC)	LENGTH(CM)
0	0
250	.051
500	.096
750	.151
1000	.193
1250	.247
1500	.296
1750	.346
2000	.394
2500	.489
2750	.542
3000	.59

Y= 1.96547E-4 *X + 3.80357E-4
R1= .788095 R2= .586 P= 768.4 P0= 47.47 X2= .1352
M2= 44.09 M1= 84.16 H= 1.96547E-4 V2= 22839.8 L= 3.482

D1,K= 2.32718E-5 D2= 2.32859E-5 D,VAR.CONC.= 2.38255E-5

DIFFUSIVITY, CONSTANT MASS CONCENTRATION
PROPANE-CYCLOHEXANE @ 25°C FEB. 27 #2

TIME(SEC)	LENGTH(CM)
1900	.591
2600	.839
3600	1.203
4200	1.418
5600	1.921
6300	2.157
7100	2.454
7800	2.695
8400	2.899
9000	3.109
9700	3.367
10200	3.543
10800	3.762

Y= 3.55275E-4 *X + -7.86153E-2
R1= .77429 R2= .565 P= 774.3 P0= 97.745 X2= .0933
M2= 44.09 M1= 84.16 H= 3.55275E-4 V2= 24105.8 L= 1.58

D1,K= 2.68529E-5 D2= 2.68606E-5 D,VAR.CONC.= 2.72962E-5

DIFFUSIVITY, CONSTANT MASS CONCENTRATION
PROPANE-CYCLOHEXANE @ 25°C MARCH 3 #2

TIME(SEC)	LENGTH(CM)
0	0
500	.138
1000	.262
1500	.407
2000	.539
2550	.687
3000	.821
3500	.963
4000	1.088
4500	1.22
5000	1.357
5700	1.539
6000	1.612

Y= 2.70459E-4 *X + 1.34336E-3
R1= .77429 R2= .565 P= 768.9 P0= 97.745 X2= .0933
M2= 44.09 M1= 84.16 H= 2.70459E-4 V2= 24105.8 L= 2.072

D1,K= 2.68101E-5 D2= 2.68175E-5 D,VAR.CONC.= 2.72492E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-CYCLOHEXANE @ 50°C MAY 14 #4

TIME(SEC)	LENGTH(CM)
0	0
250	.033
500	.064
750	.1
1000	.137
1250	.173
1500	.208
1750	.242
2000	.279
2250	.314
2500	.348
2750	.386
3000	.417
3250	.456
3500	.488
3750	.526
4000	.564
4250	.6

Y= 1.41432E-4 *X + -4.15452E-3
R1= .750439 R2= .53 P= 761 P0= 270.006 X2= .05547
M2= 44.09 M1= 84.16 H= 1.41432E-4 V2= 26203.2 L= 2.963

D1,K= 3.27761E-5 D2= 3.27711E-5 D,VAR.CONC.= 3.24685E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-CYCLOHEXANE @ 50°C MAY 10 #1

TIME(SEC)	LENGTH(CM)
250	.054
500	.096
750	.153
1000	.205
1250	.25
1500	.296
1750	.349
2000	.395
2250	.445
2500	.493
2750	.537
3000	.585
3250	.632
3500	.682
3750	.731
4000	.777
4250	.829
4500	.877

Y= 1.93101E-4 *X + 7.27452E-3
R1= .750439 R2= .53 P= 750.4 P0= 270.006 X2= .05547
M2= 44.09 M1= 84.16 H= 1.93101E-4 V2= 26203.2 L= 2.141

D2= 3.2322E-5 D,VAR.CONC.= 3.20287E-5

D1,K= 3.23265E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-CYCLOHEXANE @ 50°C MAY 11 #1

TIME(SEC)	LENGTH(CM)
2750	.597
3000	.643
3250	.694
3500	.743
3750	.798
4000	.847
4250	.899
4500	.946
4750	1.005
5000	1.055

Y= 2.04466E-4 *X + 3.03953E-2
R1= .750439 R2= .53 P= 745.5 P0= 270.006 X2= .05547
M2= 44.09 M1= 84.16 H= 2.04466E-4 V2= 26203.2 L= 2.118

D2= 3.38519E-5 D,VAR.CONC.= 3.35482E-5

D1,K= 3.3857E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-PERFLUOROHEXANE @ -10°C JUNE 21 #6

TIME(SEC)	LENGTH(CM)
2250	.186
2800	.208
3000	.223
3250	.235
3500	.251
4000	.284
4250	.298
4500	.31
5250	.358

Y= 5.8679E-5 *X + 4.75922E-2

R1= 1.7845 R2= .614 P= 758.6 P0= 36.14 X2= .112
M2= 44.09 M1= 338.06 H= 5.8679E-5 V2= 21125.8 L= 2.929

D1,K= 1.4039E-5 D2= 1.39871E-5 D,VAR.CONC.= 1.31239E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-PERFLUOROHEXANE @ -10°C JUNE 20 #2

TIME(SEC)	LENGTH(CM)
2500	.065
2750	.079
3000	.098
3250	.107
3500	.118
3750	.132
4000	.142
4250	.158
4500	.171
5000	.191
5500	.214
6000	.25
6500	.27

Y= 5.05731E-5 *X + -5.85563E-2

R1= 1.7845 R2= .614 P= 758.8 P0= 36.14 X2= .112
M2= 44.09 M1= 338.06 H= 5.05731E-5 V2= 21125.8 L= 3.234

D1,K= 1.33595E-5 D2= 1.33103E-5 D,VAR.CONC.= 1.24885E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-PERFLUOROHEXANE @ -10°C JUNE 20 #3

TIME(SEC)	LENGTH(CM)
750	.068
1000	.08
1250	.092
1500	.109
1750	.126
2000	.144
2250	.158
2500	.173
2750	.184
3000	.199
3250	.215

Y= 5.98906E-5 *X + 2.09461E-2

R1= 1.7845	R2= .614	P= 757.8	P0= 36.14	X2= .112
M2= 44.09	M1= 338.06	H= 5.98906E-5	V2= 21125.8	L= 2.764

D1,K= 1.35214E-5 D2= 1.34715E-5 D,VAR.CONC.= 1.26409E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-PERFLUOROHEXANE @ 5°C JUNE17

TIME(SEC)	LENGTH(CM)
0	0
250	.009
500	.025
750	.036
1000	.05
1250	.064
1500	.075
1750	.088
2000	.099
2250	.113
2500	.125
2750	.139
3000	.151
3250	.163

Y= 5.06635E-5 *X + -1.11387E-3

R1= 1.7421	R2= .593	P= 757.9	P0= 83.71	X2= .07313
M2= 44.09	M1= 338.06	H= 5.06635E-5	V2= 22412.4	L= 3.055

D1,K= 1.8555E-5 D2= 1.85291E-5 D,VAR.CONC.= 1.78149E-5

DIFFUSIVITY,CONSTANT MOLAR CONCENTRATION
PROPANE-PERFLUOROHEXANE @ 5°C JUNE 16 #3

TIME(SEC)	LENGTH(CM)
3250	.166
3500	.177
3750	.188
4000	.2
4250	.212
4500	.224
5900	.287
6400	.311
6900	.333
7400	.356
7900	.381

Y= 4.60581E-5 *X + .015922
R1= 1.7421 R2= .593 P= 752.6 P0= 83.71 X2= .07313
M2= 44.09 M1= 338.06 H= 4.60581E-5 V2= 22412.4 L= 3.467

D1,K= 1.91411E-5 D2= 1.91151E-5 D,VAR.CONC.= 1.83836E-5

DIFFUSIVITY,CONSTANT MOLAR CONCENTRATION
PROPANE-PERDLUOROHEXANE @ 5°C JUNE 16 #1

TIME(SEC)	LENGTH(CM)
0	0
250	.012
500	.023
750	.034
1000	.045
1250	.056
1500	.07
1750	.081
2000	.094
2250	.104

Y= 4.65211E-5 *X + -4.3627E-4
R1= 1.7421 R2= .593 P= 752.6 P0= 83.71 X2= .07313
M2= 44.09 M1= 338.06 H= 4.65211E-5 V2= 22412.4 L= 3.467

D1,K= 1.93335E-5 D2= 1.93072E-5 D,VAR.CONC.= 1.85684E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-PERFLUOROHEXANE @ 25°C JUNE 11 #3

TIME(SEC)	LENGTH(CM)
0	0
500	.027
1000	.044
1300	.053
1500	.064
2250	.082
2500	.095
2750	.102
6000	.222
6250	.233
6500	.243

Y= 3.63893E-5 *X + 4.84612E-3
R1= 1.6718 R2= .565 P= 752.5 P0= 220.77 X2= .04802
M2= 44.09 M1= 338.06 H= 3.63893E-5 V2= 24105.8 L= 3.908

D1,K= 2.5007E-5 D2= 2.49981E-5 D,VAR.CONC.= 2.44853E-5

DIFFUSIVITY, CONSTANT MOLAR CONCENTRATION
PROPANE-PERFLUOROHEXANE @ 25°C JUNE 11 #1

TIME(SEC)	LENGTH(CM)
0	0
750	.032
1000	.038
1500	.064
2000	.075
2500	.097
3000	.119
3500	.135
4000	.147
4500	.172
5000	.195
5500	.205
6000	.222
6500	.239
7000	.27
7500	.289
8000	.308
8500	.326
8700	.335

Y= 3.79818E-5 *X + 1.18198E-3
R1= 1.6718 R2= .565 P= 754.4 P0= 220.77 X2= .04802
M2= 44.09 M1= 338.06 H= 3.79818E-5 V2= 24105.8 L= 4.072

D1,K= 2.71982E-5 D2= 2.71877E-5 D,VAR.CONC.= 2.66288E-5

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DEGREE M.A.Sc. YEAR GRANTED 1973

(Chemical Engineering)

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