

Table II. Viscosities (mPa·s) of Aqueous Solutions of Maleic Acid

concn, wt %	temp, °C				
	25	40	60	80	90
0.00	(0.890)	(0.653)	(0.467)	(0.355)	(0.315) ^a
10.08	1.092	0.792	0.595	0.450	0.408
19.61	1.349	0.968	0.696	0.528	0.477
29.98	1.775	1.255	0.861	0.649	0.584
40.15	2.459	1.702	1.115	0.832	0.741
52.22		2.540	1.571	1.144	1.009
61.00			2.245	1.557	1.357
72.07					2.080

^a From ref 4.

The coefficient of the multiple correlation is 0.99995, and the standard deviation of the experimental points from this equation is 0.00087 g/cm³.

Viscosity. Results and Correlation

The viscosities of the aqueous maleic acid solutions for experimental conditions similar to those in the determination of densities were measured. The experimental results are tabulated in Table II.

Several empirical equations have been tested to correlate viscosity in function with concentration and with temperature. The best obtained equation which relates the viscosity μ to the absolute temperature T and the mole fraction of maleic acid x_A is the following

$$\log \mu = -1.0375 + 2.810 \times 10^7 T^{-3} - 5.284 \times 10^{13} T^{-6} + (3.740 - 1.974 \times 10^7 T^{-3} + 1.534 \times 10^{15} T^{-6}) x_A + (-3.151 \times 10^8 T^{-3} + 4.966 \times 10^{15} T^{-6}) x_A^2 \quad (3)$$

The multiple correlation coefficient is 0.9992, and the standard deviation of the experimental points from this equation is 0.0227 mPa·s.

This equation has been selected because we had observed that the relationship between $\log \mu$ and the composition and the temperature was almost in accordance to the equations proposed by Litovitz (7) and Suryanarayana and Venkatesan (8).

Glossary

t	temperature, °C
T	temperature, K
w_A	mass fraction of maleic acid
x_A	mole fraction of maleic acid

Greek Letters

ρ	density, g/cm ³
μ	absolute viscosity, mPa·s

Registry No. Maleic acid, 110-16-7.

Literature Cited

- (1) *International Critical Tables*; McGraw-Hill: New York, 1928; Vol. 3, pp 111-114.
- (2) Weiss, J. M.; Downs, C. R. *J. Am. Chem. Soc.* **1923**, *45*, 1003.
- (3) Delmas, G.; Saint-Romain, P.; Purves, P. *J. Chem. Soc., Faraday Trans. 1* **1975**, *71*, 1181-1187.
- (4) *Handbook of Chemistry and Physics*; CRC Press: Boca Raton, FL, 1981-82.
- (5) Soler, A.; Bódalo, A.; Gómez, D. *Invest. Téc. Pap.* **1979**, *16*, 61, 596-621.
- (6) Ruiz, J.; Gómez, D.; Miñana, A.; Soler, A. *An. Quím.* **1984**, *80*, 573.
- (7) Litovitz, T. A. *J. Chem. Phys.* **1952**, *20*, 1088.
- (8) Suryanarayana, C. V.; Venkatesan, V. K. *Monatsh. Chem.* **1958**, *89*, 824.

Received for review January 27, 1984. Revised manuscript received January 21, 1986. Accepted April 7, 1986.

Determination of Activity Coefficients of Oxygenated Hydrocarbons in Squalane by Gas-Liquid Chromatography

Layla A. Qaddora and George M. Janini*

Department of Chemistry, Kuwait University, Kuwait

A study of the thermodynamics of oxygenated hydrocarbons and aromatic solutes in squalane was conducted using gas-liquid chromatography. Solute infinite-dilution activity coefficients were determined at four temperatures in the range of 30-45 °C. The corresponding excess thermodynamic properties were calculated and the results were examined and discussed according to the regular solution theory and the perturbation theory of solutions.

Knowledge of the thermodynamics of nonelectrolytic solutions is of great theoretical and practical importance in physical chemistry and chemical engineering. Static methods are accurate, but in most cases, time consuming. In contrast GLC has been shown to be rapid, efficient, and capable of 1-2% accuracy in the determination of infinite-dilution activity coefficients and related solution thermodynamic properties (1). The accuracy of activity coefficients as measured by GLC depends on the type of systems studied, careful control of the experimental parameters, and the availability of accurate physical data. For systems where the only retention mechanism operative is solute absorption in the bulk of the stationary phase,

the accuracy can be as good as 1% or better.

In this study we sought to apply the GLC method with squalane as stationary phase for the determination of the solution properties of oxygenated aliphatic and aromatic hydrocarbons. The solutes were selected to represent different functional groups. Squalane was chosen because it has been extensively studied by GLC and by static techniques and considerable quantities of thermodynamic properties of a wide range of solutes have been accumulated (2-8). Although most of the studies in the cited literature were conducted at temperatures exceeding those studied here, an attempt was made to compare our results with independent measurements.

Solute activity coefficients were determined at four temperatures. The corresponding thermodynamic properties were calculated and the results were examined and discussed according to the regular solution theory and the perturbation theory of solutions.

Theory

Solute activity coefficients at infinite dilution in the stationary phase are related to GLC measured specific retention volume by the expression (9)

$$\gamma_p^\infty = \frac{1.704 \times 10^7}{MP_2^0 V_g^0} \quad (1)$$

where M is the molar mass of the stationary phase in g mol^{-1} ; P_2^0 is the saturated vapor pressure of the pure solute in Torr, and V_g^0 is the specific retention volume in $\text{cm}^3 \text{g}^{-1}$.

The fugacity-corrected activity coefficient γ_f^∞ can be calculated with (9)

$$\ln \gamma_f^\infty = \ln \gamma_p^\infty - \frac{P_2^0}{RT} (B_{22} - V_2^0) + \frac{\bar{P}}{RT} (B_{12} - V_2^0) \quad (2)$$

where T is the column temperature in kelvin; R is the gas constant; \bar{P} is the average column pressure; V_2^0 is the molar volume of pure solute; B_{22} is the second virial coefficient for pure solute; and B_{12} is the mixed solute-carrier gas second virial coefficient. At moderate pressure, using helium as carrier gas, B_{12} can be safely neglected. V_2^0 is also negligibly small compared to B_{22} . Thus eq 2 can be approximate by

$$\ln \gamma_f^\infty = \ln \gamma_p^\infty - \frac{B_{22} P_2^0}{RT} \quad (3)$$

There are certain assumptions and experimental conditions accompanying the derivation of eq 1 that must be met before it can be used for the derivation of thermodynamic quantities (9).

The infinite-dilution activity coefficient is related to the solute molar excess Gibbs energy by

$$G_2^\circ = RT \ln \gamma_2^\infty = H_2^\circ - TS_2^\circ \quad (4)$$

where H_2° and S_2° are the infinite-dilution excess molar enthalpy and entropy of solution, respectively. By assuming S_2° and H_2° constant within a narrow range of temperature and differentiating $\ln \gamma_2^\infty$ with respect to temperature, both H_2° and S_2° may be determined. Plots of $\ln \gamma_2^\infty$ vs. reciprocal temperature exhibit 1–5% experimental scatter in excess of that of $\ln V_g^0$ vs. temperature because the vapor pressure, fugacity, and the critical data are included in the calculation of the activity coefficients.

Experimental Section

The gas chromatographic experiments were conducted with a set of solutes selected to provide a wide range of chemical type. Solute vapor pressure data were calculated by using the Antoine equation. For most of the solutes, the constants needed for this equation were available in literature (10–15). Solute second virial coefficients were calculated by using the equation of corresponding states by McGlashan and Potter (16).

The packing material was prepared by coating squalane on Chromosorb W-HP (100–120 mesh) using the solvent slurry technique. The mass percent of liquid phase on the solid support was determined gravimetrically by ashing duplicate samples.

A Perkin-Elmer Sigma 1B gas chromatograph was employed in this study. Columns were held in a precision thermostat and the system was equipped with precision gas flow control and an ionization flame detector. Retention data was directly recorded by the online data station. An average of three measurements was taken for each point. The retention time of methane was used to correct for column dead volume. Flow rates were measured with a soap bubble flow meter. All solute peaks except for the highly polar acetone appeared to be symmetric. Four columns of different lengths but with the same percent loading (10.10%) were used. High liquid-phase loadings on highly inert Chromosorb W-HP were used in order to minimize any contribution to retention from adsorption of the solute

Table I. Solute Specific Retention Volumes

solute	V_g^0 , mL g ⁻¹			
	30 °C	35 °C	40 °C	45 °C
benzene	475	390	323	269
toluene	1550	1230	984	793
<i>n</i> -heptane	1040	807	634	501
cyclohexane	634	517	424	350
ethylbenzene	4130	3190	2490	1950
<i>o</i> -xylene	6000	4600	3560	2770
<i>m</i> -xylene	4940	3800	2950	2310
<i>p</i> -xylene	4810	3701	2880	2250
mesitylene	15400	11400	8580	6490
<i>n</i> -propylbenzene	10600	7990	6070	4660
isopropylbenzene	7830	5950	4570	3540
chlorobenzene	3100	2430	1920	1520
acetone	70.6	60.5	52.2	45.2
ethyl methyl ketone	193	159	132	110
2-pentanone	455	372	307	254
3-pentanone	508	412	336	276
2-heptanone	3950	3020	2330	1810
ethyl acetate	226	183	149	122
<i>n</i> -propyl acetate	587	474	384	314
isopropyl acetate	344	279	227	187
<i>n</i> -butyl acetate	1790	1380	1080	845
phenol	4980	3780	2890	2230
<i>o</i> -cresol	12600	9430	7080	5370
<i>m</i> -cresol	14300	10700	7980	6030
<i>p</i> -cresol	14600	10800	8010	6020

at the gas-liquid or liquid-solid interfaces.

Results and Discussion

Solute specific retention volume (V_g^0) were calculated for all solutes at four temperatures. The (V_g^0) values reported in Table I represent the experimental average of three measurements. (V_g^0) values were calculated from experimentally measured quantities by using the expression

$$V_g^0 = \frac{t_R'}{W_L} F_{tm} \frac{273.2}{T_{tm}} \left(\frac{P_{tm} - P_w}{P_{tm}} \right)^3 \frac{(P_i/P_o)^2 - 1}{2(P_i/P_o)^3 - 1} \quad (5)$$

where t_R' is the solute-corrected retention volume; F_{tm} is the flow rate; W_L is the mass of liquid phase in the column; T_{tm} is ambient temperature; P_{tm} is the atmospheric pressure; P_w is the saturated vapor pressure of water at T_{tm} ; P_i is the column inlet pressure and P_o is the column outlet pressure. The reproducibility of the V_g^0 values for all aromatic solutes was within $\pm 1\%$. The results for this set of solutes also agree with literature values to within $\pm 1\%$ (17). The oxygenated hydrocarbons did not reproduce as well because of the dependence of retention data on sample size. The accuracy in the V_g^0 values for acetone and methyl ethyl ketone which represent the worse scatter of our results was within $\pm 5\%$.

Values of γ_f^∞ calculated with eq 1 and 3 by using smoothed V_g^0 values are listed in Table II. Solute partial excess molar enthalpies (H_2°) and entropies (S_2°) obtained from the slopes and intercepts of plots of $\ln \gamma_f^\infty$ vs. reciprocal temperature in accordance with eq 4 are listed in Table III.

Examination of the data in Tables II and III reveals the following: For the aromatic hydrocarbons $\gamma_f^\infty < 1$ indicating better solvation for this set of solutes in squalane (i.e., a stronger than ideal bond is formed between solute and solvent). Since the values of H_2° and S_2° are both positive (except for *n*-heptane), the unfavorable positive H_2° is overwhelmed by the favorable positive S_2° . For the oxygenated hydrocarbons the situation is reversed. The favorable entropic factor is overwhelmed by the unfavorable enthalpic factor leading to positive deviation from Raoult's law. For *n*-heptane, which differs in nature from all other solutes since it is a normal-chain hydrocarbon, both S_2° and S_2° are negative. Since $\gamma_f^\infty < 1$, the unfavorable negative S_2° is overwhelmed by the favorable

Table II. Solute Fugacity-Corrected Activity Coefficients at Infinite Dilution

solute	γ_i^{∞}			
	30 °C	35 °C	40 °C	45 °C
benzene	0.717	0.704	0.692	0.682
toluene	0.713	0.704	0.697	0.691
<i>n</i> -heptane	0.672	0.683	0.696	0.709
cyclohexane	0.528	0.525	0.523	0.521
ethylbenzene	0.771	0.763	0.755	0.750
<i>o</i> -xylene	0.752	0.744	0.738	0.733
<i>m</i> -xylene	0.736	0.728	0.722	0.717
<i>p</i> -xylene	0.712	0.711	0.706	0.703
mesitylene	0.768	0.764	0.762	0.761
<i>n</i> -propylbenzene	0.817	0.807	0.801	0.795
isopropylbenzene	0.824	0.812	0.803	0.795
chlorobenzene	0.836	0.815	0.795	0.783
acetone	2.04	1.95	1.86	1.79
ethyl methyl ketone	1.77	1.73	1.70	1.67
2-pentanone	1.80	1.75	1.70	1.65
3-pentanone	1.70	1.65	1.61	1.57
2-heptanone	1.91	1.85	1.80	1.75
ethyl acetate	1.50	1.48	1.46	1.44
<i>n</i> -propyl acetate	1.57	1.52	1.46	1.42
isopropyl acetate	1.53	1.50	1.47	1.44
<i>n</i> -butyl acetate	1.49	1.46	1.43	1.41
phenol	15.1	12.8	10.9	9.37
<i>o</i> -cresol	6.72	5.54	4.59	3.83
<i>m</i> -cresol	13.3	12.0	10.9	9.91
<i>p</i> -cresol	14.4	12.1	10.3	8.72

Table III. Solute Excess Thermodynamic Data

solute	H_2^e , kJ mol ⁻¹	S_2^e , J K ⁻¹ mol ⁻¹
benzene	2.73	11.8
toluene	1.65	8.27
<i>n</i> -heptane	-2.87	-6.15
cyclohexane	0.746	7.77
ethylbenzene	1.50	7.12
<i>o</i> -xylene	1.42	7.07
<i>m</i> -xylene	1.40	7.17
<i>p</i> -xylene	0.760	5.32
mesitylene	0.531	3.95
<i>n</i> -propylbenzene	1.45	6.47
isopropylbenzene	1.88	7.82
chlorobenzene	3.53	13.2
acetone	7.01	17.2
ethyl methyl ketone	3.26	5.99
2-pentanone	4.58	10.2
3-pentanone	4.56	10.6
2-heptanone	4.68	10.1
ethyl acetate	2.00	3.25
<i>n</i> -propyl acetate	5.49	14.3
isopropyl acetate	3.07	6.69
<i>n</i> -butyl acetate	3.03	6.61
phenol	25.7	62.1
<i>o</i> -cresol	30.1	83.5
<i>m</i> -cresol	15.5	29.7
<i>p</i> -cresol	26.8	66.2

negative S_2^e . Cyclohexane and *n*-heptane show the best solvation into squalane out of all other solutes since they are both close to the nature of the solvent. *o*-Cresol shows the best solubility among the dihydroxybenzene isomers because of its weak polarity with respect to *p*- and *m*-cresol.

Theoretical Interpretation of Activity Coefficients. We now proceed to analysis of the GLC results according to two solution theories, namely, the regular solution theory and the perturbation theory (18). Common to these theories is the assumption that the deviations from ideality can be considered to be due to two effects: (1) A configurational (conf) effect caused by the difference in size between the solute and solvent molecules, and (2) a thermal (th) effect resulting from differences in the intermolecular interactions of the components of the solution; thus

$$\ln \gamma_2 = \ln \gamma_{2(\text{conf})} + \ln \gamma_{2(\text{th})} \quad (6)$$

Table IV. Configurational Contribution to Nonideality, Interaction, and Solubility Parameters at 30 °C

solute	$-\ln \gamma_2$ (conf)	χ	δ_{2s} (J cm ⁻³) ^{1/2}	δ_1 (J cm ⁻³) ^{1/2}
benzene	0.939	0.607	18.6	22.7
toluene	0.795	0.456	18.1	21.4
<i>n</i> -heptane	0.549	0.152	15.1	16.7
cyclohexane	0.780	0.142	16.7	18.5
ethylbenzene	0.684	0.425	17.9	20.8
<i>o</i> -xylene	0.696	0.412	18.3	21.2
<i>m</i> -xylene	0.682	0.375	17.9	20.7
<i>p</i> -xylene	0.679	0.340	17.8	20.5
mesitylene	0.590	0.327	17.9	20.3
<i>n</i> -propylbenzene	0.587	0.385	17.5	20.1
isopropylbenzene	0.587	0.393	17.3	20.0
chlorobenzene	0.830	0.651	19.3	23.3
acetone	1.10	1.81	19.5	27.4
ethyl methyl ketone	0.932	1.50	18.1	24.6
3-pentanone	0.797	1.33		
ethyl acetate	0.859	1.26	18.2	18.5
<i>n</i> -propyl acetate	0.731	1.18	17.8	22.9
isopropyl acetate	0.717	1.14	17.2	22.1
<i>n</i> -butyl acetate	0.628	1.03	17.6	22.0
phenol	1.05	3.76	24.6	35.9
<i>o</i> -cresol	0.818	2.72	22.6	30.8
<i>m</i> -cresol	0.810	3.40	23.2	32.2
<i>p</i> -cresol	0.810	3.48	23.2	32.3

$\ln \gamma_{2(\text{th})}$ is referred to as the interaction parameter (χ). The configurational contribution may be evaluated according to (19)

$$\ln \gamma_{2(\text{conf})} = \ln (1/r) + (1 + 1/r) \quad (7)$$

where r is the ratio of the molar volume of the solute to that of the solvent, (V_2/V_1). Utilizing eq 7 and using available density values (10, 11), we calculated $\ln \gamma_{2(\text{conf})}$ at 30 °C and the results are listed in Table IV. The interaction parameter (χ) was calculated by using data from Table II and the results are also listed in Table IV. Solutes for which density data and/or activity coefficients were not available are not included.

According to the regular solution theory (20, 21) the interaction parameter (χ) is given by

$$\ln \gamma_{2(\text{th})} = \chi = (V_2/RT)(\delta_1 - \delta_2)^2 \quad (8)$$

where V_2 is the solute molar volume and δ_1 is the solubility parameter of component i . The solute solubility parameters (δ_2) are calculated by using the relationship (20, 21)

$$\delta = \left(\frac{\Delta E_2^v}{V_2} \right)^{1/2} = \left(\frac{\Delta H_2^v - RT}{V_2} \right)^{1/2} \quad (9)$$

where ΔE_2^v and ΔH_2^v are respectively the pure solute molar energy and enthalpy of vaporization, δ_2 values are also listed in Table IV. Unfortunately, δ_1 values for squalane could not be similarly calculated due to lack of enthalpy of vaporization data. Testing of the theory is still possible, however, by determining the δ_1 value using experimental χ values according to eq 8. A single value for δ_1 would indicate the applicability of the regular solution theory. Instead several values of δ_1 were obtained indicating that the theory is not capable of encompassing several classes of solutes. It is to be noted that other workers have also shown it to be only of limited utility (22). However, careful examination of the δ_1 column of Table IV reveals single δ_1 values would be obtained if families of solutes are grouped together. For example, δ_1 from data for the aromatic solutes is 20.85 ± 0.84 and that obtained from the cresols is 31.78 ± 0.88 .

The other solution theory to be considered is the perturbation theory due to Longuet-Higgins (19). According to this theory

$$\chi = \alpha + \beta(T_2^c/(V_2^*)^{1/2}) \quad (10)$$

where α and β are constants for a given solvent and T_2^c/V_2^*

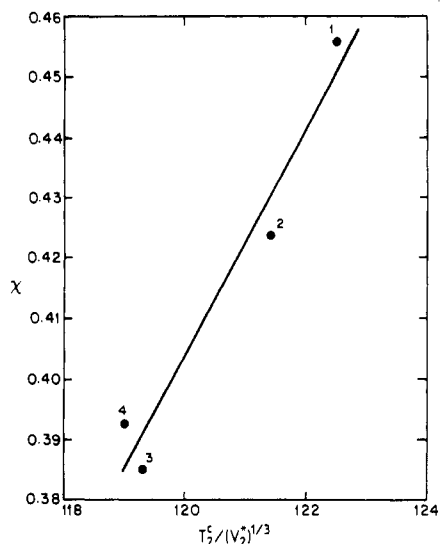


Figure 1. Plot of χ vs. $T_2^c/(V_2^*)^{1/3}$ for monosubstituted benzenes. Solutes: (1) = toluene; (2) = ethylbenzene; (3) = *n*-propylbenzene; and (4) = isopropylbenzene.

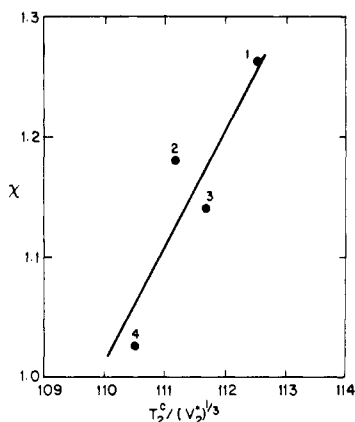


Figure 2. Plot of χ vs. $T_2^c/(V_2^*)^{1/3}$ for alkyl acetates. Solutes: (1) = ethyl acetate; (2) = isopropyl acetate; (3) = *n*-propyl acetate; and (4) = *n*-butyl acetate.

$(V_2^*)^{1/3}$ is a measure of solute potential well depth as introduced by Kreglewski (23, 24). V_2^* is solute molar volume at $0.6T_2^c$. The linear plot of χ vs. $T_2^c/(V_2^*)^{1/3}$ with correlation coefficient close to unity would confirm the universality and applicability of this theory to different classes of solutes. Again our results indicate that this is not so; instead each group of structurally similar solutes will fall on a different straight line as

illustrated in Figures 1 and 2. To construct these figures χ values were obtained from Table IV and V_2^* was calculated from available density and critical temperature data (10, 11). Our results concerning the inadequacies of the perturbation theory are in qualitative agreement with those of Tewari et al. (22) who analyzed the data for several classes of solutes in three different stationary phases.

Registry No. Squalane, 111-01-3; benzene, 71-43-2; toluene, 108-88-3; *n*-heptane, 142-82-5; cyclohexane, 110-82-7; ethylbenzene, 100-41-4; *o*-xylene, 95-47-6; *m*-xylene, 108-38-3; *p*-xylene, 106-42-3; mesitylene, 108-67-8; *n*-propylbenzene, 103-65-1; isopropylbenzene, 98-82-8; chlorobenzene, 108-90-7; acetone, 67-64-1; ethyl methyl ketone, 78-93-3; 2-pentanone, 107-87-9; 3-pentanone, 96-22-0; 2-heptanone, 110-43-0; ethyl acetate, 141-78-6; *n*-propyl acetate, 109-60-4; isopropyl acetate, 108-21-4; *n*-butyl acetate, 123-86-4; phenol, 108-95-2; *o*-cresol, 95-48-7; *m*-cresol, 108-39-4; *p*-cresol, 106-44-5.

Literature Cited

- (1) Laub, R. J.; Pecsok, R. L. *Physicochemical Application of Gas Chromatography*; Wiley: New York, 1978.
- (2) Desty, D. H.; Swanton, W. T. *J. Phys. Chem.* **1961**, *65*, 766.
- (3) Evered, S.; Pollard, F. H. *J. Chromatogr.* **1960**, *4*, 451.
- (4) Cruickshank, A. J. B.; Everett, D. H.; Westaway, M. T. *Trans. Faraday Soc.* **1965**, *61*, 235.
- (5) Cruickshank, A. J. B.; Windsor, M. L.; Young, C. L. *Proc. R. Soc.* **1967**, *A295*, 271.
- (6) Martire, D. E.; Pollara, L. Z. *J. Chem. Eng. Data* **1965**, *10*, 40.
- (7) Kwantes, A.; Rijnders, W. A. In *Gas Chromatography*; Desty, D. H., Ed.; Butterworths: London, 1958; p 125.
- (8) Martire, D. E.; Pecsok, R. L.; Purnell, J. H. *Trans. Faraday Soc.* **1965**, *61*, 2496.
- (9) Martire, D. E. *Anal. Chem.* **1961**, *33*, 1143.
- (10) Dreisbach, R. R. *Physical Properties of Chemical Compounds*; Adv. Chem. Ser. No. 15; American Chemical Society: Washington DC, 1955.
- (11) Riddick, J. A.; Bunger, W. B. *Organic Solvents-Physical Properties and Methods of Purification*, 3rd ed.; Wiley: New York, 1970; Vol. II.
- (12) Reid, R. C.; Sherwood, T. K. *The Properties of Gases and Liquids - Their Estimation and Correlation*, 2nd ed.; McGraw-Hill: New York, 1966.
- (13) Jordan, T. E. *Vapor Pressure of Organic Compounds*; Interscience: New York, 1954.
- (14) Forman, J. C.; Thodes, G. *AIChE J.* **1958**, *4*, 356.
- (15) Forman, J. C.; Thodes, G. *AIChE J.* **1960**, *6*, 206.
- (16) McGlashan, M. L.; Potter, D. J. B. *Proc. R. Soc.* **1962**, *A267*, 478.
- (17) Janini, G. M.; King, J. W.; Martire, D. E. *J. Am. Chem. Soc.* **1974**, *96*, 5368.
- (18) Ashworth, A. J.; Everett, D. H. *Trans. Faraday Soc.* **1960**, *56*, 1609.
- (19) Longuet-Higgins, H. C. *Discuss. Faraday Soc.* **1953**, *15*, 73.
- (20) Hildebrand, J. H.; Scott, R. K. *Regular Solutions*; Prentice-Hall: Englewood Cliffs, NJ, 1962.
- (21) Hildebrand, J. H.; Scott, R. L. *Solubility of Non-electrolytes*, 3rd ed.; Dover: New York, 1964.
- (22) Tewari, Y. B.; Martire, D. E.; Sheridan, J. P. *J. Phys. Chem.* **1970**, *74*, 2345.
- (23) Kreglewski, A. *J. Phys. Chem.* **1967**, *71*, 2860.
- (24) Kreglewski, A. *J. Phys. Chem.* **1968**, *72*, 1897.

Received for review March 1, 1985. Accepted May 2, 1986.