The fact that all points in Figure 1 fall on the single line confirms the absence of complexes higher than HF2and of polynuclear species.

The stability constants were evaluated using a least squares technique based on the following mass-balance relationships:

$$C_{\rm H} = [{\rm H}^+] \{ 1 + \beta_1 [{\rm F}^-] + \beta_2 [{\rm F}^-]^2 \}$$
(7)

$$C_{\rm F} = [{\rm F}^-] + [{\rm H}^+] \{\beta_1 [{\rm F}^-] + 2\beta_2 [{\rm F}^-]^2\}$$
(8)

The equilibrium constant  $K_2$  is obtained from

$$\kappa_2 = \beta_2 / \beta_1 \tag{9}$$

For each experimental point, [F-] is obtained from the observed E;  $\beta_1$  and  $\beta_2$  are chosen intuitively; and  $[H^+]$ values are calculated from Equation 7. A new set of  $\beta$ 's is obtained from Equation 8 by minimizing the function

$$\sum_{1}^{k} \{1 - FX_{k}/X_{k}\}^{2}$$
(10)

where

and

$$FX_{k} = \beta_{1}[F^{-}]_{k} + 2\beta_{2}[F^{-}]_{k}^{2}$$
(11)

$$X_{k} = \{(C_{F})_{k} - [F^{-}]_{k}\}/[H^{+}]_{k}$$

The process is repeated using the new  $\beta$ 's to generate new [H<sup>+</sup>] values, and the  $\beta$ 's are further refined until convergence is achieved. The resulting  $\beta$ 's along with their standard deviations are given in Table III. The standard deviations of log  $\beta_1$  and log  $\beta_2$  were obtained by the pit-mapping method of Sillén (14). The variance in log  $K_2$  is taken as the sum of the variances of log  $\beta_1$  and  $\log \beta_2$ .

Figure 2 is a plot of the deviation of FX/X from the theoretical value. Although there is some scatter at low  $\bar{n}$ (high  $C_H/C_F$ ) values, there is no systematic deviation that would indicate the presence of species other than HF and  $HF_2^-$ .

The enthalpies and entropies for Equations 1 and 3 were obtained by conventional methods, and the results are given in Table IV.

## Conclusions

The above results once again confirm the existence of HF and HF2<sup>-</sup> and the absence of other complexes. The temperature coefficients of the equilibrium constants appear to be more accurate than previous estimates (6). The entropy change for Equation 1 of 23.8 equiv. units (molal units) is in good agreement with the value of 22.0 equiv. units (molar units) quoted in ref. 7 for 0.5M NaClO<sub>4</sub> solutions. The major disagreement between all the works listed in Table I appears to be associated with  $\beta_2$  and hence  $K_2$ .

## Literature Cited

- (1) Ostwal, W. J., Prakt. Chem., 32, 300 (1885).
- Ostwal, W. J., Prakt. Chem., 32, 300 (1865).
   Pick, H., Nernst-Festschr., 1912, p 300.
   Davies, C. W., Hudleston, L. J., J. Chem. Soc., 125, 260 (1924).
   Roth, W. A., Lieb. Ann. Chem., 542, 35 (1939).
   Brosset, C., Svk. Kem. Tidskr., 54, 155 (1942).

- (6) Brosset, C., Svk. *Netr. Huski*, 34, 155 (1942).
  (6) Broene, H. H., DeVries, T., *J. Amer. Chem. Soc.*, 69, 1644 (1947).
  (7) Connick, R. E., Hepler, L. G., Hugus, Z. Z., Kury, J. W., Latimer, W. M., and Tsao, M.-S., *ibid.*, 78, 1827 (1956).
  (8) Mesmer, R. E., *Anal. Chem.*, 40, 443 (1968).

- (9) Neumann, G., Sillén, L. G., quoted by Butler, ref. 11.
  (10) Srinivasan, K., Rechnitz, G. A., Anal. Chem., 40, 509 (1968).
  (11) Butler, J. N., "Ion Selective Electrodes," R. A. Durst, Ed., Chap. 5,
- NBS Spec. Pub. 314, 1969. International Critical Tables, Vol 111, McGraw-Hill, New York, N.Y., (12) 1928.
- (13) Bjerrum, J., "Metal Ammine Formation in Aqueous Solution," P. Haase and Son, Copenhagen, 1957
- (14) Sillén, L. G., Acta Chem. Scand., 16, 159 (1962).

Received for review March 5, 1973. Accepted August 13, 1973.

# Isothermal Vapor-Liquid Equilibrium Data of DMSO Solutions by Total Pressure Method. DMSO-Acetone, DMSO-Tetrahydrofuran, and DMSO-Ethyl Acetate Systems

Yoshimasa Sassa, Ryoichi Konishi,<sup>1</sup> and Takashi Katayama<sup>2</sup> Department of Chemical Engineering, Faculty of Engineering Science, Osaka University, Toyonaka, Osaka, Japan

(12)

Isothermal vapor-liquid equilibrium data were obtained by the total pressure method for the binary mixtures of dimethyl sulfoxide (DMSO) with acetone, tetrahydrofuran, or ethyl acetate at 25°, 35°, and 45°C. These data were reduced to obtain excess Gibbs free energies.

The extensive applications of dimethyl sulfoxide (DMSO) as solvent and reaction medium have been markedly increased in recent years (8). In spite of the increased use of DMSO, few phase equilibrium data for the systems containing DMSO have been studied.

<sup>1</sup>Present address, Mitui Senboku Petroleum Chemical Co., Osaka, Japan. <sup>2</sup>To whom correspondence should be addressed.

44 Journal of Chemical and Engineering Data, Vol. 19, No. 1, 1974

As DMSO has a strong polarity, it is highly self-associated and has the ability to make molecular complexes with other compounds. For the fundamental understanding of the effects of the strong molecular interactions on the thermodynamic excess properties, systematic studies of vapor-liquid equilibria for DMSO mixtures have been projected. This paper reports vapor-liquid equilibrium data for the DMSO-acetone, DMSO-tetrahydrofuran, and DMSO-ethyl acetate systems at 25°, 35°, and 45°C.

### Experimental

Total vapor pressures for the three systems studied were measured as a function of composition at constant temperature. A schematic diagram of the apparatus described here (Figure 1) was taken from the studies of Quitzsch and coworkers (12) and Arm and coworkers (1). The apparatus consists of two cells, A and A', approximately 60 ml each for the liquid container; a small null manometer, F; a mercury reservoir, E; a magnetic stirrer, C; and the manometer, D, which is a mercury manometer and can be exchangeable to a dibutyl phthalate manometer for the measurements of low vapor pressures.

After mercury was placed in the reservoir (E) by a hypodermic syringe from the stopcock, dissolved gas in the mercury was removed by evacuating and warming carefully. The liquid sample of known composition was placed in the cell (A) and frozen with liquid nitrogen. A vacuum of about  $10^{-3}$  mm Hg was effected rather quickly to avoid vaporization, the stopcock was closed, and the liquid was melted to remove dissolved gas. These procedures were repeated several times. Finally, the sample was distilled between the two cells for perfect degassing.

After the sample had been degassed, the mercury in the mercury reservoir was introduced to the small null manometer by rotating the equilibrium cell at the taper joint (G). The cell was gradually warmed and thermostated. The liquid in the equilibrium cell was magnetically stirred to assure perfect mixing. Equilibrium was attained in about 30 min. Arms of the small null manometer were adjusted to about the same levels by bleeding dry nitrogen gas into the reference side of the manometer through a capillary. The difference between the mercury (or dibutyl phthalate) level in both arms of the manometer tube (D, 12-mm diameter) was read by a PCTM-1000 cathetometer from Nakamura Optical Co. The small null manometer was read by using a traveling microscope from Shimazu Seisakasho Co., and the difference was added to the pressure difference in the manometer (D). All manometer readings were corrected for gravity and temperature. The liquid composition was corrected by considering the vapor volume.

A calibrated glass thermometer was used for temperature measurements which were accurate within  $0.03^{\circ}$ C. Temperature deviation of  $0.03^{\circ}$ C would cause a maximum pressure deviation of 0.6 mm Hg for the DMSOacetone system at 45°C. The temperature of the bath was thermostated to  $\pm 0.01^{\circ}$ C. This uncertainty in temperature causes a maximum error of  $\pm 0.10\%$  in the vapor pressure. Vapor pressures were measured to within  $\pm 0.10$  mm Hg.

Pure vapor pressures of DMSO were measured by introducing degassed DMSO into the small null manometer and by using the dibutyl phthalate manometer as the manometer (D). Densities of dibutyl phthalate were preliminarily measured as a function of temperature. Vapor pressures of pure DMSO were measured to within  $\pm 0.01$  mm Hg.

Spectrograde DMSO, acetone, tetrahydrofuran, and ethyl acetate from Merck Co. were used. DMSO and acetone were dried over molecular sieves prior to the experiments. In the titration with Carl Fisher reagent, water was present in DMSO to about 0.07 mol %. Physical properties of these materials in this work are summarized in Table I.

## **Results and Data Reduction**

The experimentally determined vapor pressures of DMSO-acetone, DMSO-tetrahydrofuran, and DMSOethyl acetate systems are listed in column 2 of Table II. Two separate runs of *P*-x data were taken for the DMSOacetone system at  $35^{\circ}$ C to ascertain the reproducibility of the data. Average deviation between the two runs was about 0.3% in pressure as shown in Table II. These vapor-pressure data over the whole range of composition were reduced to calculate the excess Gibbs free energy by Barker's method (2) of successive approximation. That is, the total vapor pressures in a binary mixture are calculated from the following equation (11).

$$P_{\text{calcd}} = \frac{x_1 \gamma_1 f_1^{\circ}}{\phi_1} + \frac{x_2 \gamma_2 f_2^{\circ}}{\phi_2}$$
(1),

Based on Equation 1, the coefficients of the three-parameter Redlich-Kister equation (13), the Renon NRTL equation (14), and the Wilson equation (18) were calculated so that the criteria  $\Sigma[(P_{exptl} - P_{calcd})/P_{exptl}]^2 =$  minimum were satisfied. Fugacity coefficients ( $\phi_i$ ) were calculated by using second-virial coefficients as given elsewhere (11). The pure-component second-virial coefficients and the cross-virial coefficients were estimated by the method of Prausnitz et al. (11). The critical properties



Figure 1. Schematic diagram of apparatus

Table I. Physical Constants of Materials Used

	Refract	ive index	Vapor press, mm Hg						
Component	Mea- sured	Lit	Mea- sured	Lit					
		255	25°C						
DMSO	1.4772	1.4773 (10)	0.59	0.582 (9)					
Acetone	1.3564	1.35662 (17)	231.2	229.4 (17)					
Tetrahydrofuran	1.4049	1.4048 (15)	161.7	162.02 (15)					
Ethyl acetate	1.3701	1.37012ª (17)	95.4	94.5 (6)					
<sup>a</sup> At 25.2°C.									

Journal of Chemical and Engineering Data, Vol. 19, No. 1, 1974 45

## Table II. Isothermal Vapor-Liquid Equilibria

Gr. Namelic         Particle mm Hg         Particle in yn         In yn <t< th=""><th></th><th></th><th></th><th></th><th></th><th></th><th></th><th></th><th></th><th></th><th></th><th></th><th></th><th></th><th></th></t<>																		
x,         mm Hg         mn Hg         y,         In         n </td <td></td> <td>P<sub>exptl</sub>,</td> <td>P<sub>calcd</sub>,</td> <td></td> <td></td> <td></td> <td>G<sup>E</sup>, cal/</td> <td></td> <td></td> <td>P<sub>exptl</sub>,</td> <td>Pcalcd,</td> <td></td> <td></td> <td></td> <td>G<sup>E</sup>, cal/</td>		P <sub>exptl</sub> ,	P <sub>calcd</sub> ,				G <sup>E</sup> , cal/			P <sub>exptl</sub> ,	Pcalcd,				G <sup>E</sup> , cal/			
DMSO(T)-exectore(2) system st 25°C         JEC           0.0000         212.1         2.0.4         0.0000         0.795         0.0000         5.97         0.0000         1.597         0.0001         1.597         0.0001         1.597         0.0001         1.597         0.0001         1.597         0.0001         0.516         0.0211         1.183         0.0211         1.18         0.0211         1.18         0.0211         1.18         0.0211         1.18         0.0211         1.18         0.0211         0.010         0.0107         1.201         0.000         0.3017         1.201         0.0388         1.78         0.016         0.5421         1.60         0.5421         1.60         0.5421         1.60         0.5491         1.05         0.017         0.911         0.946         0.5471         1.20         0.9921         1.02         0.138         0.992         0.12         0.138         0.992         0.12         0.138         0.9921         1.011         0.138         0.9921         0.12         0.138         0.992         0.12         0.138         0.992         0.12         0.12         0.992         0.12         0.12         0.12         0.992         0.12         0.12         0.992         0.12         <	×ı	mm Hg	mm Hg	Уı	ln γı	$\ln \gamma_2$	mol	ر	1	mm Hg	mm Hg	<b>y</b> 1	ln γı	In y2	mol			
0.0000         231.2          0.0000         0.7544         0.0005         0.1754         0.0000         1.133         0.0001         1.133         0.0001         1.133         0.0001         1.133         0.0011         1.1133         0.0011         1.1133         0.0011         1.1133         0.0011         1.1133         0.0011         1.1133         0.0011         1.1133         0.0011         1.1133         0.0011         1.1133         0.0011         1.1133         0.0011         0.1131         0.0021         0.317         200.6         20.6         0.0021         0.317         200.6         20.6         0.0021         0.317         200.6         20.6         0.0021         0.311         0.021         0.321         1.33.5         1.33.2         0.0011         0.1110         0.021         0.311         0.011         0.321         1.33.5         1.33.2         0.0011         0.1110         0.021         0.0111         0.021         0.011         0.0111         0.0111         0.021         0.011         0.021         0.011         0.0111         0.021         0.011         0.0111         0.021         0.011         0.0111         0.0111         0.0111         0.0111         0.0111         0.0111         0.0111         <	DMSO(1)-acetone(2) system at 25°C											35°C						
0.0689         210.6         210.4         0.0098         10.0080         10.0089         222.2         22.5         9         0.0217         11.33         0.0214         11.33         0.0214         11.33         0.0214         11.33         0.0214         11.33         0.0214         11.33         0.0214         11.33         0.0214         11.33         0.0214         11.33         0.0224         11.34         0.0026         0.1234         0.0026         0.0124         11.33         0.0244         0.014         0.0244         0.014         0.0244         0.014         0.0244         0.0244         0.0244         0.0241         0.124         0.0026         0.1234         0.0244	0.0000	231.2		0.0000	0.7954	0.0000	0	0.0	000	245.9		0.0000	1.5977	0.0000	0			
$ \begin{array}{c} 1.977 \\ 0.298 $	0 0989	210.6	210.4	0.0005	0.6018	0 0098	41	0 0	1999	227 2	225 9	0 0017	1 1122	0.0241	81			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0 1977	193 5	192 1	0.0010	0 4500	0.0360	70	0.0	2006	213 7	212 5	0.001/	0 7824	0.0241	136			
$ \begin{array}{c} 0.000 \\ 0.000 $	0.2008	174 2	174 4	0.0015	0.3265	0.0767	90	0.2	1017	200 6	200 6	0.0020	0.7024	0.001/	170			
$ \begin{array}{c} 0.000 \\ 0.001 \\ 0.001 \\ 0.001 \\ 0.002 \\ 0.000 $	0.2000	156 6	167 1	0.0010	0.0200	0.0707	100	0.0	0017	107 0	100 5	0.0032	0.0400	0.1034	105			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.4000	137.0	130 7	0.0020	0.2311	0.1200	100	0.3	000	172 0	170.0	0.0036	0.364/	0.24/3	185			
$ \begin{array}{c} 0.0007 & 118.2 & 118.9 & 0.0034 & 0.197 & 0.1807 & 0.182 & 84 & 0.7001 & 135.5 & 133.5 & 0.0071 & 0.0161 & 0.4967 & 114 \\ 0.7937 & 71.5 & 71.5 & 0.0009 & 0.0250 & 0.4231 & 63 & 0.7982 & 104.2 & 103.8 & 0.098 & 0.0201 & 0.7461 & 112 \\ 0.906 & 33.1 & 39.2 & 0.0013 & 0.0000 & 0.556 & 35 & 0.3992 & 22.2 & 62.4 & 0.1176 & 0.0120 & 0.7461 & 112 \\ 0.000 & 159 & \dots & 1.0000 & 0.0000 & 0.6518 & 0 & 1.0000 & 1.119 & \dots & 1.0000 & 0.0000 & 0.6518 & 0 & 1.000 & 1.119 & 0.0001 & 0.520 & 63 \\ 0.0090 & 340.3 & \dots & 0.0000 & 0.7158 & 0.0001 & 0 & 0.0000 & 564.8 & \dots & 0.0000 & 1.0217 & 0.0227 & 61 \\ 0.0090 & 340.3 & \dots & 0.0000 & 0.1158 & 0.0001 & 0 & 0.0000 & 564.8 & \dots & 0.0000 & 1.0217 & 0.0227 & 115 \\ 0.0000 & 140.3 & 0.0000 & 0.4265 & 0.0399 & 67 & 0.2018 & 315.6 & 313.9 & 0.037 & 0.0578 & 0.0759 & 0.0775 & 155 \\ 0.0001 & 234.8 & 234.8 & 0.0007 & 0.5784 & 0.0681 & 38 & 0.3991 & 276.4 & 276.9 & 0.0086 & 0.3270 & 0.3271 & 184 \\ 0.0001 & 234.8 & 234.8 & 0.0007 & 0.0224 & 0.1125 & 38 & 0.3991 & 276.4 & 276.9 & 0.0086 & 0.4320 & 1.027 & 81 \\ 0.0001 & 234.8 & 206.7 & 0.0095 & 0.5111 & 0.1565 & 100 & 0.5032 & 257.0 & 25.1 & 0.001 & 0.1569 & 0.4828 & 13 \\ 0.0001 & 1.73 & 10.768 & 0.0097 & 0.5127 & 54 & 0.5991 & 196.0 & 0.0008 & 0.4828 & 13 \\ 0.0001 & 1.73 & 10.768 & 0.0007 & 0.5127 & 54 & 0.5993 & 89.5 & 69.7 & 0.0226 & 0.0124 & 0.4890 & 0.889 & 10 \\ 0.0000 & 1.78 & 0.0000 & 0.140 & 0.4097 & 0.5217 & 54 & 0.0993 & 89.5 & 69.7 & 0.0226 & 0.0014 & 0.4898 & 10 \\ 0.0000 & 1.19 & \dots & 1.0000 & 0.0000 & 0.6216 & 0 & 0.0000 & 1.623 & 0.0000 & 1.623 & 0.0000 & 1.623 & 0.0000 & 1.624 & 0.0890 & 0.0266 & 17 \\ 0.0000 & 1.19 & \dots & 1.0000 & 0.0000 & 0.6217 & 0 & 0.0000 & 1.624 & 0.0000 & 0.064 & 0.5926 & 17 \\ 0.0001 & 1.19 & \dots & 1.0000 & 0.0000 & 0.6217 & 0 & 0.0000 & 1.623 & 0.0116 & 0.727 & 0.032 & 1.000 & 0.0000 & 1.623 & 0.0116 & 7.7 & 0.000 & 1.623 & 0.0116 & 7.7 & 0.000 & 1.626 & 0.789 & 10.000 & 0.0000 & 0.644 & 0.0993 & 0.426 & 0.0000 & 0.0000 & 0.000 & 0.000 & 0.0000 & 0.0000 & 0.0000 & 0.0000 & 0.0000 & 0.0000 & 0.0000 & 0.0000 & 0$	0.5019	137.9	138.7	0.0020	0.10074	0.1903	102	0.5	029	1/3.0	1/3.4	0.0046	0.2540	0.3542	180			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.601/	119.2	118.9	0.0034	0.0974	0.2011	90	0.6	800	155.5	155.9	0.0056	0.161/	0.46/6	1/4			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.69/4	97.8	97.5	0.0046	0.055/	0.3382	84	0.7	011	133.5	133.2	0.00/1	0.0911	0.5991	149			
0.9000 39.1 39.2 0.0139 0.0061 0.5386 35 0.8992 62.2 62.4 0.0176 0.0110 0.9220 63 38°C, Run 1 570 0.0000 39.3 0.0000 0.75578 0.0000 0.6518 0.109 0.355, 9 334.7 0.0020 1.5249 0.0000 0.000 0.0000 317.1 318.1 0.0007 0.5578 0.0061 38 0.0000 335, 9 334.7 0.0022 0.7569 0.0775 135 0.1979 290.6 282.2 0.0013 0.4265 0.309 67 0.3019 295.5 295.4 0.0040 0.5321 0.1524 169 0.4001 234.8 234.4 0.0027 0.2246 0.1162 98 0.3991 276.4 275.9 0.0040 0.332 0.758 1.5 0.4001 234.8 234.8 0.002 0.3246 0.162 98 0.3991 276.4 275.9 0.0040 0.3370 0.2378 184 0.6019 177.3 176.8 0.0047 0.033 0.1216 10 0.5022 251.0 2521 0.1524 0.4004 0.4526 173 0.603 0.173 10.758 0.0040 0.2239 0.403 63 0.7994 150.8 105.8 105.9 0.0044 0.2239 0.403 63 0.7994 150.8 105.8 105.9 0.0044 0.4239 0.4238 14 0.7975 105.8 105.9 0.0049 0.0239 0.403 63 0.7994 150.8 105.8 0.0022 0.0404 0.8266 134 0.7975 105.8 155.8 0.0047 0.2349 0.403 63 0.7994 150.8 105.8 0.002 0.1748 0.428 15 0.0000 1.19 1.0000 0.0000 0.6215 0 1.0000 2.20 1.0000 0.1005 0.1228 120 0.4010 0.2228 0.403 0.402 0.0000 0.6215 0 1.0000 2.20 1.0000 0.1000 1.683 0 0.0000 0 349.3 0.0000 0.7180 0.0000 0 0 0.419 0.0320 0.1342 0.644 66 0.392 0.4051 0.1000 2.20 1.0000 0.108 7.2 20 0.200 349.3 0.0000 0.6215 0 0.0000 95.4 0.0000 1.0528 0.406 1 0.203 0.442 223.6 22.4 0.003 0.124 0.4986 1 0.023 0.4000 0 0.442 223.6 2.00.003 0.1342 0.644 66 0.2913 77.1 77.1 0.001 0.5678 0.1046 1 0.294 150 0.442 223.6 2.00.03 0.1242 0.644 66 0.2913 77.1 77.1 0.001 0.5678 0.1046 1 0.294 16.42 17.7 1 0.000 0.0000 0.6217 0 0.403 0.528 8 0.099 0.44 17.4 0.001 0.5876 0.1295 16 0.442 223.6 0.0030 0.125 0 0.104 0 15.5 0.003 0.125 0 0.004 0.237 9 0.443 0.423 0.003 0.140 0.1469 0.023 0 0.000 0 0.423 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0.99 0 0.44 0 0 0.99 0 0.44 0 0.99 0 0.44 0 0 0.99 0 0.44 0 0 0.99 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	0.7973	71.5	71.5	0.0069	0.0250	0.4291	63	0.7	992	104.2	103.8	0.0098	0.0420	0.7467	112			
1.0000         0.99          1.0000         0.0000         0.618         0         1.0000         1.19          1.0000         1.133         0           0.0000         349.3          0.0000         0.7158         0.0000         0         0.0000         336.8          0.0000         1.5249         0.0000         0           0.1077         200.6         289.2         0.0010         0.5778         0.0011         0.4265         0.3019         295.1         295.4         0.0040         0.5321         0.1574         1569         0.0775         135           0.1072         2206.4         226.5         0.0027         0.2264         0.0140         0.5321         251.6         0.0011         0.1560         0.532         255.0         295.9         0.0088         0.2268         0.4204         0.632           0.6019         1.73         175.8         0.0094         0.2239         0.4103         63         0.7949         150.8         0.0226         0.1044         0.8296         63           0.0000         1.19          1.0000         0.0007         0.5421         0.1000         2.20         1.0000         0.0001         80.4	0.9006	39.1	39.2	0.0139	0.0061	0.5356	35	0.8	992	62.2	62.4	0.0176	0.0110	0.9220	63			
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	1.0000	0.59	•••	1.0000	0.0000	0.6518	0	1.0	000	1.19	•••	1.0000	0.0000	1.1313	0			
0.0000 349.3 0.0000 0.7158 0.0000 0 0.0000 35.9 33.4, 7. 0.002 1.7249 0.0000 0 0.0090 37.1 338.1 0.0007 0.5759 0.0081 38 0.1000 33.5 9 33.4, 7 0.0020 1.777 0.0227 81 0.1978 290.6 289.2 0.0013 0.4265 0.0399 67 0.2008 31.5, 6 31.3 9 0.0022 0.7569 0.0775 135 0.2001 234.8 234.8 0.0027 0.2246 0.1152 98 0.3991 27.6 4 27.5 9 0.0040 0.3321 0.1524 169 0.5021 205.8 206.7 0.0035 0.1581 0.1765 10 0.5032 251.0 253.9 0.058 0.2486 0.3420 186 0.6191 177.3 176.8 0.0047 0.0934 0.2458 95 0.0012 227.2 227.6 0.0019 0.0567 0.5893 148 0.7975 105.8 105.9 0.0044 0.0239 0.316 30 0.7994 150.8 150.8 0.0122 0.0126 0.4266 173 0.0000 1.19 1.0000 0.0067 0.5521 0 1.0000 1.200 0.0000 1.0565 0.5803 148 0.7975 105.8 105.9 0.0044 0.0239 0.413 53 0.7994 150.8 150.8 0.0122 0.0104 0.8906 53 0.0000 1.99 0.0061 0.0539 0.413 53 0.7994 150.8 150.8 0.0122 0.0104 0.8906 53 0.0000 1.099 0.0067 0.548 0.0092 40 1.0000 95.4 0.0000 1.2289 112 0.1000 1.19 1.0000 0.0000 0.6215 0 1.0000 95.4 0.0000 1.2839 0 0.2015 257.4 228.3 0.0014 0.4160 0.0323 67 0.0988 80.0 87.8 0.0022 0.9086 0.1288 0.0000 1.0889 0 0.215 227.4 288.3 0.0001 0.0548 0.0922 40 0.0000 95.4 0.0000 1.3239 0.0008 1.2 0.432 223.6 222.4 0.033 0.1329 0.0544 99 0.231 77.1 77.1 0.0041 0.5576 0.1295 156 0.432 223.6 222.4 0.033 0.1328 0.1374 99 0.2913 77.1 77.1 0.0041 0.1576 0.1295 156 0.431 20.8 0.411 0.009 0.0254 0.4127 63 0.9891 47.1 47.1 0.0041 0.0577 0.1296 116 0.599 17.1 177.1 0.0047 0.1056 0.2374 95 0.0386 57.4 55.5 0.0021 0.0444 0.3083 0.1659 10 0.4915 32.4 0.0030 0.1689 0.0324 81 0.638 59.5 55.5 0.0021 0.0444 0.3083 0.1589 10 0.0691 1.19 1.0000 0.0000 0.447 0 0.0990 1.101. 0.554 0.417 63 0.3994 140.4 130.3 0.0044 0.3565 181 0.5991 37.1 177.1 0.0044 0.3056 0.3258 81 0.638 59.5 55.5 0.0021 0.0128 0.9779 166 0.0991 465.7 469.1 0.0090 0.0664 0.521 35 0.8991 25.8 25.8 0.0211 0.0521 0.5790 112 0.0000 1.108 0.1075 0.2246 177 0.0000 1.108 0.0000 0.0000 0.0000 0.0000 0.59 1.10 0.1058 0.1371 0.0000 0.0000 1.238 155 0.0000 1.19 1.0000 0.0000 0.0000 0.0000 0.59 1.0000 0.0000			3	5°C, Run	1							45°C						
0.0990 317.1 318.1 0.0007 0.5578 0.0081 38 0.1000 335.9 334.7 0.0020 1.0717 0.0227 81 0.2999 261.4 261.5 0.0030 0.3144 0.0679 87 0.3019 295.5 295.4 0.0040 0.3321 0.1574 169 0.4001 234.8 234.8 0.0027 0.246 0.1152 98 0.391 276.4 276.9 0.0048 0.2458 0.3420 18 0.5021 205.8 206.7 0.0035 0.1511 0.1765 100 0.5032 253.0 23.9 0.0088 0.2468 0.3420 18 0.5021 205.8 206.7 0.0035 0.1511 0.1765 100 0.5032 253.0 227.6 0.0071 0.1560 0.4760 173 0.6376 144.9 144.7 0.0083 0.0539 0.3216 83 0.7014 194.0 194.0 0.090 0.0875 0.5803 148 0.7975 105.8 105.9 0.0094 0.0239 0.4103 63 0.7944 150.8 150.8 0.0126 0.4011 0.2228 112 0.9000 57.8 57.8 57.8 0.119 0.0057 0.5127 34 0.893 89,5 89.7 0.0226 0.1014 0.19406 68 1.0000 1.19 1.0000 0.0057 0.5127 34 0.893 89,5 89.7 0.0226 0.0101 0.1040 0.9006 1.0893 0 0.0000 349.3 0.0000 0.7180 0.0092 40 0.000 95.4 0.0000 0.0000 1.0893 0 0 0.104 0.4909 0.0236 7 0.1944 315.5 316.7 0.0007 0.3428 0.0092 40 0.0089 88.0 87.8 0.78 0.0000 1.4233 0.0000 0 0.4149 0.325.6 22.4 0.0114 0.1940 0.0236 7 0.1944 315.6 22.4 0.0014 0.4469 0.0232 67 0.0964 88.0 87.8 0.78 0.0000 1.4233 0.0000 1 0.439 0.0365 72 20.125 287.4 288.3 0.0014 0.4469 0.0232 67 0.1944 0.4965 71.4 71.4 0.0051 0.4047 0.2266 177 0.4312 62.22.4 0.002 0.0030 0.1928 0.1379 0.664 181 0.7975 140.1 140.1 0.0066 0.0536 0.3258 81 0.0638 59.5 59.5 0.0073 0.1233 0.4017 171 177.1 0.0079 0.0049 0.0009 0.4561 52.2 0.0000 1.109 0.3044 0.3055 181 0.7085 140.1 140.1 0.0067 0.0356 0.3258 81 0.6038 59.5 59.5 0.0011 0.018 0.7979 16 0.0191 465.7 469.1 0.0007 0.5254 0.4027 47 63 0.663 52.4 52.2 0.0090 0.1017 0.5849 151 0.0000 1.19 1.0000 0.0000 0.599 1.26 0.4039 55. 59.5 0.0211 0.0138 0.7979 16 0.0991 465.7 469.1 0.0007 0.5264 0.4027 47 70 4.830 67.0 67.1 0.0050 0.3744 0.3055 181 0.0000 1.19 1.0000 0.0000 0.669 72.2 0.0000 1.126 0.3370 66 0.0591 0.004 0.4074 0.226 187 0.0000 1.1017 0.5849 1.011 0.0071 0.0050 0.277 45 0.4043 13.3 43.4 0.0025 0.3258 41 0.6038 59.5 59.5 0.0021 0.3030 0.426 0.4094 341.1 34.0 0.0050 0.0364 0.5217 3 0.7699 34.4 4.15 0.0073 0.3026 0.	0.0000	349.3	• • •	0.0000	0.7158	0.0000	0	0.0	000	364.8	•••	0.0000	1.5249	0.0000	0			
0.1978 290.6 299.2 0.0013 0.4265 0.0309 67 0.2008 315.6 313.9 0.002 0.7669 0.0775 135 0.2999 261.4 261.5 0.0020 0.3144 0.0679 87 0.3019 295.5 295.4 0.0040 0.3730 0.2378 184 0.6019 177.3 176.8 0.0027 0.2246 0.1162 98 0.3991 276.4 276.9 0.0088 0.3730 0.2378 184 0.6019 177.3 176.8 0.0047 0.0484 0.2458 95 0.6012 227.2 227.6 0.0071 0.1560 0.4626 173 0.6874 144.9 144.7 0.0653 0.6339 0.4216 83 0.7014 194.0 194.0 0.0090 0.0675 0.5003 148 0.9005 7.8 57.8 0.0094 0.2239 0.4103 63 0.7944 194.0 194.0 0.0090 0.0675 0.5003 148 0.9005 57.8 57.8 0.0190 0.0057 0.5127 34 0.8993 89.5 89.7 0.0226 0.0104 0.9006 63 0.900 349.3 0.0000 0.7160 0.0000 0.625 0 1.0000 2.20 1.0000 1.0001 0.728 112 0.9003 349.3 0.0000 0.7180 0.0000 4 0.0233 67 0.0984 88.0 87.8 0.0000 1.423 0.0000 7 0.5124 0.484 0.424 0.4646 8 0.194 0.9046 48.0 97.4 0.0000 1.423 0.0000 7 0.2015 287.4 288.3 0.0014 0.4169 0.0323 67 0 0.0089 88.0 87.8 0.0000 1.423 0.0006 7 0.2012 287.4 288.3 0.0014 0.4169 0.0323 67 0 0.0984 88.0 87.8 0.0002 0.1047 0.1256 122 0.432 223.6 222.4 0.0030 0.1928 0.1374 99 0.2131 77.1 77.1 0.0041 0.5876 0.1295 156 0.4316 208 0.929 100.4 71.4 71.4 0.0651 0.1040 0.0266 177 0.5379 177.1 177.1 0.0047 0.1056 0.324 9 0.430 67.0 67.1 0.0000 0.1823 0.4171 71 0.5979 177.1 177.1 0.0047 0.1056 0.324 9 0.430 67.0 67.1 0.0059 0.3044 0.3065 131 0.7063 140.1 0.0066 0.5358 81 0.663 52.4 52.2 0.0030 1.0579 0.264 177 0.5979 170.1 177.1 0.0047 0.1056 0.324 95 0.4430 67.0 67.1 0.0000 0.1167 0.5849 151 0.0001 1.38 104.1 0.0097 0.0254 0.324 95 0.430 67.0 67.0 67.1 0.0010 0.0000 1.230 0 0.0000 1.123 0.0157 1.12 0.597 170.1 177.1 0.0047 0.1056 0.324 95 0.430 67.0 67.0 67.1 0.0001 0.3183 0.477 1.579 180.1 13.8 104.1 0.0097 0.0254 0.427 3 0.6991 14.5 0.1021 0.0159 0.3044 0.3055 131 0.000 0.59 1.0000 0.0000 1.230 0 0.000 0.1230 0.000 0.59 0.000 1.139 0.0064 0.2517 37 0.6991 44.4 1.5 0.0211 0.038 0.939 12.2 0.000 0.107 0.0050 0.0660 0.556 0.258 81 0.6063 52.4 52.2 0.0003 0.0000 1.277 0 0.599 14.1 1.21.9 0.0000 0.0000 1.267 0 0.0000 1.279 0.2214 12.7 121.9 0.00	0.0990	317.1	318.1	0.0007	0.5578	0.0081	38	0.1	000	335.9	334.7	0.0020	1.0717	0.0227	81			
0.2999         261.4         261.5         0.0027         0.2244         0.1679         87         0.3019         295.5         295.4         0.0040         0.3320         0.2378         1124           0.5021         205.8         206.7         0.0027         0.2248         0.1162         10         0.5032         253.0         253.9         0.0088         0.2468         0.3400         153           0.6019         17.3         176.8         0.0094         0.0293         0.4103         63         0.714         194.0         194.0         0.0268         0.2468         0.3400         66         13           0.6906         57.8         57.8         0.0190         0.0097         0.512         0         1.0000         1.0126         0.0401         0.2228         112           0.0000         57.8         57.8         0.0100         0.0007         0.5125         0         1.0000         0.0226         0.0140         0.4900         0.0000         0.0000         1.0893         0.000         0.0140         0.4900         1.0893         0.0140         0.4900         0.237         0.0226         0.0020         1.0893         0.000         0.0000         1.0898         0.0140         0.4900	0.1978	290.6	289.2	0.0013	0.4265	0.0309	67	0.2	800	315.6	313.9	0.0032	0.7569	0.0775	135			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.2999	261.4	261.5	0.0020	0.3144	0.0679	87	0.3	019	295.5	295.4	0.0040	0.5321	0.1524	169			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.4001	234.8	234.8	0.0027	0.2246	0.1162	98	0.3	991	276.4	276.9	0.0048	0.3730	0.2378	184			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.5021	205.8	206.7	0.0035	0.1511	0.1765	100	0.5	032	253.0	253.9	0.0058	0.2458	0.3420	186			
$\begin{array}{c} 0.6376 & 144. 9 & 144. 7 & 0.0663 & 0.639 & 0.3216 & 83 & 0.7014 & 194. 0 & 144. 0 & 0.099 & 0.0375 & 0.893 & 148 \\ 0.7975 & 105. 8 & 105. 9 & 0.094 & 0.0239 & 0.4103 & 63 & 0.7994 & 150. 8 & 150. 8 & 0.0126 & 0.0010 & 0.7228 & 112 \\ 0.9008 & 57. 8 & 57. 8 & 57. 8 & 0.190 & 0.0007 & 0.5127 & 34 & 0.8993 & 89. 5 & 89. 7 & 0.0226 & 0.0104 & 0.8966 & 63 \\ 1.0000 & 1.19 & \dots & 1.0000 & 0.0000 & 0.6215 & 0 & 1.0000 & 2.20 & \dots & 1.0000 & 0.0000 & 1.4893 & 0 \\ \hline 0.0104 & 315.5 & 316.7 & 0.0007 & 0.5458 & 0.0092 & 40 & 0.0988 & 88.0 & 87.8 & 0.0021 & 1.6988 & 0.0186 & 0.0000 & 1.4232 & 0.0000 & 0.4432 & 23.6 & 22.4 & 0.0014 & 0.4169 & 0.0323 & 67 & 0.0988 & 88.0 & 87.8 & 0.0021 & 1.6988 & 0.0186 & 72 & 0.0001 & 0.4432 & 23.6 & 22.2 & 0.0021 & 0.0598 & 0.0186 & 72 & 0.0021 & 0.0092 & 0.0000 & 0.4432 & 23.6 & 22.2 & 0.0021 & 0.0598 & 0.0186 & 72 & 0.0324 & 0.0030 & 0.1428 & 0.1374 & 99 & 0.2131 & 77.1 & 0.0041 & 0.5676 & 0.1295 & 156 & 0.4330 & 67. & 67.1 & 0.0059 & 0.3044 & 0.0645 & 122 & 0.4391 & 77.1 & 0.0041 & 0.0056 & 0.2374 & 95 & 0.6633 & 59.5 & 59.5 & 0.0073 & 0.1823 & 0.4517 & 171 & 0.5797 & 171 & 0.0047 & 0.1056 & 0.2374 & 95 & 0.6633 & 59.5 & 59.5 & 0.0073 & 0.1823 & 0.4517 & 171 & 0.0011 & 140.1 & 0.0066 & 0.6526 & 0.2328 & 81 & 0.6633 & 59.5 & 59.5 & 0.0073 & 0.1823 & 0.4517 & 171 & 0.0011 & 140.1 & 0.0066 & 0.6237 & 35 & 0.6633 & 52.4 & 52.2 & 0.0090 & 0.1107 & 728 & 0.138 & 0.9730 & 166 & 0.0114 & 0.138 & 0.9730 & 166 & 0.0114 & 0.1065 & 0.2374 & 0.0295 & 131. & 0.0000 & 0.5264 & 0.0074 & 37 & 0.0899 & 14.4 & 41.5 & 0.0121 & 0.0521 & 0.7579 & 116 & 0.0091 & 45.7 & 499.1 & 0.0090 & 0.5264 & 0.0074 & 37 & 0.0899 & 14.4 & 41.5 & 0.0121 & 0.0521 & 0.7579 & 116 & 0.0000 & 1.571 & 0.0000 & 0.5967 & 0.0284 & 0.0133 & 1.030 & 0.0630 & 5.204 & 100.00 & 0.131 & 0.0000 & 0.0001 & 1.192 & 0.052 & 0.7633 & 0.0613 & 122 & 0.570 & 0.133 & 0.076 & 0.2384 & 0.572 & 0.013 & 0.0285 & 1312 & 0.0572 & 0.0131 & 0.0461 & 0.1525 & 0.0284 & 0.0176 & 72 & 0.777 & 0.4431 & 10.56 & 0.0075 & 0.2384 & 0.0055 & 0.3284 & 0.0055 & 0$	0.6019	177.3	176.8	0.0047	0.0948	0.2458	95	0.6	012	227.2	227.6	0.0071	0.1560	0.4526	173			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.6976	144.9	144.7	0.0063	0.0539	0.3216	83	0.7	014	194.0	194.0	0.0090	0.0875	0.5803	148			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.7975	105.8	105.9	0.0094	0.0239	0.4103	63	0.7	994	150.8	150.8	0 0126	0 0401	0 7228	112			
0.0000         1.19          1.0000         0.000         0.0215         0.000         1.0000         0.000         1.0000         1.0000         0.0000         1.4323         0.0000         0.0086         0.0021         0.0000         1.0000         0.0000         1.0000         0.0000         1.4323         0.0000         1.0000         0.0001         0.4401         0.0041         0.0051         0.4040         7.1         7.1         7.1         0.0011         0.4047         0.0021         0.4323         0.4517         1.0000         0.0031         0.4401         0.2524         0.1224         0.0011         0.4047         0.224         0.1244         0.0025         0	0 9008	57.8	57.8	0.0190	0 0057	0 5127	34	0.7	003	89.5	89.7	0.0120	0.0104	0 8906	63			
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	1.0000	1.19		1.0000	0.0000	0.6215	0	1.0	000	2.20		1.0000	0.0000	1.0893	0			
0.0000         349.3          0.0000         0.7180         0.0000         0         0.0000         1.4823         0.0000         0           0.1040         315.5         316.7         0.0007         0.5458         0.0092         40         0.0088         88.0         87.8         0.0002         1.4533         0.0000         0           0.2105         287.4         288.3         0.0140         0.1594         81.9         82.0         0.0021         1.6576         0.12951         156           0.4312         223.6         222.4         0.0030         0.1328         0.1633         0.1659         100         0.4445         77.1         77.1         0.0447         0.2265         181           0.7855         140.1         1.40.07         0.0056         0.3278         81         0.6633         52.4         0.0121         0.0521         0.7579         116           0.8014         103.8         104.1         0.0066         0.5254         0.4127         63         0.6693         52.4         0.011         0.0261         0.7579         116           0.8014         13.38         0.0020         0.6536         0.3274         0.9991         45.6         0.0211	Run 2																	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0 0000	349 3		0 0000	0 7180	0 0000	0			DIVISO	(1)-ethyla	acetate(2	) system a	it 25°C				
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.0000	315 5	316.7	0.0000	0.5458	0.0000	40	0.0	000	95.4	•••	0.0000	1.4323	0.0000	0			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.1040	207 /	200.2	0.0007	0.0400	0.0032	67	0.0	988	88.0	87.8	0.0020	1.0598	0.0186	72			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.2010	20/.4	200.3	0.0014	0.7140	0.0323	07	0.1	954	81.9	82.0	0.0032	0.7908	0.0645	122			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.29/2	201.3	262.1	0.0020	0.3142	0.0004	00	0.2	913	77.1	77.1	0.0041	0.5876	0.1295	156			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.4432	223.0	222.4	0.0030	0.1928	0.13/4	99	0.4	045	71.4	71.4	0.0051	0.4047	0.2266	177			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.4916	208.9	208.8	0.0035	0.1603	0.1659	100	0.4	830	67.0	67.1	0.0059	0.3044	0.3065	181			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.5979	177.1	177.1	0.004/	0.1005	0.23/4	95	0.6	038	59.5	59.5	0.0073	0 1823	0 4517	171			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	0.7085	140.1	140.1	0.0066	0.0536	0.3258	81	0.6	963	52.4	52.2	0.0090	0.1107	0 5849	151			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.8014	103.8	104.1	0.0097	0.0254	0.4127	63	0.7	969	41 4	41 5	0 0121	0.0521	0.3043	116			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.9015	58.0	57.9	0.0190	0.0064	0.5217	35	0.7	001	25.8	25.9	0.0121	0.0321	0.7373	66			
45°C         35°C           0.0000         514.0         0.0000         0.6697         0.0000         0         0.0000         151.6          0.0000         1.3710         0.0000         72           0.0991         465.7         469.1         0.0009         0.5264         0.0074         37         0.0989         140.4         139.3         0.0024         1.0210         0.0176         72           0.1980         427.5         425.4         0.0017         0.4055         0.0284         65         0.1955         130.3         130.0         0.0040         0.7653         0.0613         122           0.3001         383.1         383.4         0.0025         0.3010         0.0663         85         0.2914         121.7         121.9         0.0052         0.2956         181           0.5024         300.1         300.9         0.0455         0.1461         0.1662         97         0.4331         105.8         105.6         0.0075         0.2956         0.831         171           0.6021         257.2         256.7         0.0061         0.0920         0.2328         94         0.6039         93.0         93.3         0.0094         0.1765         0.4371	1.0000	1.19		1.0000	0.0000	0.6474	0	1 0	000	0.50	23.0	1 0000	0.0138	1 2200	00			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$				45°C				1.0	000	0.35	•••	35%	0.0000	1.2360	U			
0.0000       914.0       1.1.       0.0000       0.0001       1.91.0       1.1.1       0.0000       1.91.0       0.0000       1.9210       0.0000       0.0000       1.9210       0.0000       1.11       0.0000       0.0000       0.0000       1.11       1.0000       0.0000       0.0000       0.0000       0.0000       0.0000       0.0000       0.0000       0.0000       0.0000       1.11       0.0000       0.0000       1.11       0.0000       0.0000       0.0000       1.11       1.0000       0.0000       0.0000       0.0000       0.0000       0.0000       0.0000       0.0000       0.0000       0.0000       0.000	:0.000	514 0		0,0000	0 6697	0 0000	0	0.0	იიი	151 6		0 0000	1 2710	0 0000	0			
0.1991       403.7       409.1       0.0009       0.3204       0.0074       0.7       0.1980       427.5       425.4       0.0017       0.4055       0.0284       65       0.1955       130.3       130.0       0.0040       0.7653       0.0613       122         0.3001       383.1       383.4       0.0025       0.3010       0.0630       85       0.2914       121.7       121.9       0.0052       0.5700       0.1238       155         0.4004       343.1       343.0       0.0034       0.2161       0.1087       96       0.4047       112.2       112.5       0.0065       0.3927       0.2180       178         0.5024       300.1       300.9       0.0045       0.1461       0.1662       97       0.4831       105.8       105.6       0.0075       0.2954       0.2956       181         0.6021       257.2       256.7       0.0061       0.9220       0.2328       94       0.6039       93.0       93.3       0.0015       0.1067       0.5668       151         0.9099       83.4       83.3       0.0224       0.0233       0.3927       62       0.7970       64.5       64.6       0.0115       0.1067       0.5000       0.7343	0.0000	J14.0	460 1	0,0000	0.0057	0.0000	27	0.0	000	1/0 /	120.2	0.0000	1.3/10	0.0000	70			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.0991	400.7	409.1	0.0009	0.0204	0.0074	57	0.0	989	120.2	139.3	0.0024	1.0210	0.01/6	12			
0.3001 383.1 383.4 0.0023 0.3010 0.0630 89 0.2914 121.7 121.9 0.0052 0.5700 0.1238 155 0.4004 343.1 343.0 0.0034 0.2161 0.1087 96 0.4047 112.2 112.5 0.0065 0.3927 0.2180 178 0.5024 300.1 300.9 0.0045 0.1461 0.1662 97 0.4831 105.8 105.6 0.0075 0.2954 0.2956 181 0.6021 257.2 256.7 0.0060 0.0920 0.2328 94 0.6039 93.0 93.3 0.0094 0.1765 0.4371 171 0.6978 209.8 209.5 0.0081 0.0525 0.3062 82 0.6965 81.8 81.5 0.0115 0.1067 0.5668 151 0.7978 152.5 152.8 0.0122 0.0233 0.3927 62 0.7970 64.5 64.6 0.0157 0.0500 0.7343 116 0.9009 83.4 83.3 0.0244 0.0056 0.4926 34 0.8992 39.8 39.8 0.0275 0.0131 0.9415 65 1.0000 2.20 1.0000 0.0000 0.5991 0 1.0000 1.19 1.0000 0.0000 1.1942 0 DMSO(1)-Tetrahydrofuran(2) system at 25°C 45°C 0.0000 161.7 0.0000 1.6300 0.0000 0 0.0000 233.0 0.0000 1.3404 0.0000 0 0.0998 149.8 149.2 0.0013 1.1393 0.0244 80 0.0990 215.3 214.4 0.0029 0.9955 0.0173 72 0.2005 141.2 140.5 0.0020 0.8027 0.0829 137 0.1956 199.5 199.8 0.0047 0.7451 0.0601 123 0.3015 133.0 132.9 0.0024 0.5644 0.1621 168 0.2916 186.9 187.1 0.0062 0.5546 0.1211 156 0.3986 124.9 125.1 0.0029 0.3966 0.2520 183 0.4049 173.0 172.3 0.0078 0.3823 0.2128 178 0.5026 115.0 115.4 0.0034 0.2626 0.3615 185 0.4834 160.9 161.3 0.0091 0.2877 0.2883 182 0.6006 103.8 104.2 0.0041 0.1675 0.4782 173 0.6042 141.4 141.9 0.0114 0.1722 0.4258 172 0.7009 89.6 89.3 0.0052 0.0946 0.6139 148 0.6967 124.0 123.5 0.0141 0.1044 0.5521 152 0.7989 70.3 70.0 0.0072 0.0438 0.7665 112 0.7972 97.4 97.5 0.0192 0.0491 0.7158 117 0.8990 42.2 42.3 0.0129 0.0115 0.9487 63 0.8993 60.0 60.0 0.0338 0.0129 0.9191 61 0.7989 70.3 70.0 0.0072 0.0438 0.7665 112 0.7972 97.4 97.5 0.0192 0.0491 0.7158 117 0.8990 42.2 42.3 0.0129 0.0115 0.9487 63 0.8993 60.0 60.0 0.0338 0.0129 0.9191 66	0.1980	427.5	425.4	0.0017	0.4000	0.0284	00	0.1	900	130.3	130.0	0.0040	0.7653	0.0613	122			
0.4004 343.1 343.0 0.0034 0.2161 0.1087 96 0.407 112.2 112.5 0.0065 0.3927 0.2180 178 0.5024 300.1 300.9 0.0045 0.1461 0.1662 97 0.4831 105.8 105.6 0.0075 0.2954 0.2956 181 0.6021 257.2 256.7 0.0060 0.0920 0.2328 94 0.6039 93.0 93.3 0.0094 0.1765 0.4371 171 0.6978 209.8 209.5 0.0081 0.0525 0.3062 82 0.6965 81.8 81.5 0.0115 0.1067 0.5668 151 0.7978 152.5 152.8 0.0122 0.0233 0.3927 62 0.7970 64.5 64.6 0.0157 0.0500 0.7343 116 0.9009 83.4 83.3 0.0244 0.0056 0.4926 34 0.8992 39.8 39.8 0.0275 0.0131 0.9415 65 1.0000 2.20 1.0000 0.0000 0.5991 0 1.0000 1.19 1.0000 0.0000 1.1942 0 DMSO(1)-Tetrahydrofuran(2) system at 25°C 45°C 45°C 0.0000 161.7 0.0000 1.6300 0.0000 0 0.0000 233.0 0.0000 1.3404 0.0000 0 0.0998 149.8 149.2 0.0013 1.1393 0.0244 80 0.0990 215.3 214.4 0.0029 0.9955 0.0173 72 0.2005 141.2 140.5 0.0020 0.8027 0.0829 137 0.1956 199.5 199.8 0.0047 0.7451 0.0601 123 0.3015 133.0 132.9 0.0024 0.5644 0.1621 168 0.2916 186.9 187.1 0.0062 0.5546 0.1211 156 0.3986 124.9 125.1 0.0029 0.3966 0.2520 183 0.4049 173.0 172.3 0.0078 0.3823 0.2128 178 0.5026 115.0 115.4 0.0034 0.2626 0.3615 185 0.4834 160.9 161.3 0.0091 0.2877 0.2883 182 0.5026 115.0 115.4 0.0034 0.2626 0.3615 185 0.4834 160.9 161.3 0.0091 0.2877 0.2883 182 0.5026 115.0 115.4 0.0034 0.2626 0.3615 185 0.4834 160.9 161.3 0.0091 0.2877 0.2883 182 0.5026 115.0 115.4 0.0034 0.2626 0.3615 185 0.4834 160.9 161.3 0.0091 0.2877 0.2883 182 0.5026 115.0 115.4 0.0034 0.2626 0.3615 185 0.4834 160.9 161.3 0.0091 0.2877 0.2883 182 0.5026 115.0 115.4 0.0034 0.2626 0.3615 185 0.4834 160.9 161.3 0.0091 0.2877 0.2883 182 0.5006 103.8 104.2 0.0041 0.1675 0.4782 173 0.6042 141.4 141.9 0.0114 0.1722 0.4258 172 0.7009 89.6 89.3 0.0052 0.0946 0.6139 148 0.6967 124.0 123.5 0.0141 0.1044 0.5521 152 0.7989 70.3 70.0 0.0072 0.0438 0.7665 112 0.7972 97.4 97.5 0.0192 0.04491 0.7158 117 0.8990 42.2 42.3 0.0129 0.0115 0.9487 63 0.8993 60.0 60.0 0.0338 0.0129 0.9191 66	0.3001	383.1	383.4	0.0025	0.3010	0.0030	85	0.2	914	121.7	121.9	0.0052	0.5/00	0.1238	155			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.4004	343.1	343.0	0.0034	0.2161	0.108/	96	0.4	04/	112.2	112.5	0.0065	0.3927	0.2180	178			
0.6021       257.2       256.7       0.0060       0.0920       0.2328       94       0.6039       93.0       93.3       0.0094       0.1765       0.4371       171         0.6978       209.8       209.5       0.0081       0.0525       0.3062       82       0.6965       81.8       81.5       0.0115       0.1067       0.5668       151         0.9009       83.4       83.3       0.0244       0.0056       0.4926       34       0.8992       39.8       39.8       0.0275       0.0131       0.9415       65         1.0000       2.20        1.0000       0.5991       0       1.0000       1.19        1.0000       0.0000       1.1942       0         DMSO(1)-Tetrahydrofuran(2) system at 25°C        45°C        45°C        0.0000       1.3404       0.0000       0         0.0005       141.2       140.5       0.0020       0.8027       0.0829       137       0.1956       199.5       199.8       0.0047       0.7451       0.0601       123         0.3015       133.0       132.9       0.0024       0.5644       0.1621       168       0.2916       186.9       187.1       0.0062	0.5024	300.1	300.9	0.0045	0.1461	0.1662	9/	0.4	831	105.8	105.6	0.0075	0.2954	0.2956	181			
0.6978       209.8       209.5       0.0081       0.0525       0.3062       82       0.6965       81.8       81.5       0.0115       0.1067       0.5668       151         0.7978       152.5       152.8       0.0122       0.0233       0.3927       62       0.7970       64.5       64.6       0.0157       0.0500       0.7343       116         0.9009       83.4       83.3       0.0244       0.0056       0.4926       34       0.8992       39.8       39.8       0.0275       0.0131       0.9415       65         1.0000       2.20        1.0000       0.5991       0       1.0000       1.19        1.0000       0.0000       1.1942       0         DMSO(1)-Tetrahydrofuran(2) system at 25°C         0.0000       161.7        0.0000       1.6300       0.0000       0       0.0000       233.0        0.0000       1.3404       0.0000       0         0.2005       141.2       140.5       0.0020       0.8027       0.829       137       0.1956       199.5       199.8       0.0047       0.7451       0.0601       123         0.3015       133.0       132.9       0.0024	0.6021	257.2	256.7	0.0060	0.0920	0.2328	94	0.6	039	93.0	93.3	0.0094	0.1765	0.4371	171			
0.7978       152.5       152.8       0.0122       0.0233       0.3927       62       0.7970       64.5       64.6       0.0157       0.0500       0.7343       116         0.9009       83.4       83.3       0.0244       0.0056       0.4926       34       0.8992       39.8       39.8       0.0275       0.0131       0.9415       65         1.0000       2.20        1.0000       0.0000       0.5991       0       1.0000       1.19        1.0000       0.0000       1.1942       0         DMSO(1)-Tetrahydrofuran(2) system at 25°C         0.0000       161.7        0.0000       1.6300       0.0000       0       0.0090       215.3       214.4       0.0000       1.3404       0.0000       0         0.2005       141.2       140.5       0.0020       0.8027       0.0829       137       0.1956       199.5       199.8       0.0047       0.7451       0.0601       123         0.3015       133.0       132.9       0.0024       0.5644       0.1621       168       0.9       187.1       0.0062       0.5546       0.1211       156         0.3086       124.9       125.1       0.0034 <td>0.6978</td> <td>209.8</td> <td>209.5</td> <td>0.0081</td> <td>0.0525</td> <td>0.3062</td> <td>82</td> <td>0.6</td> <td>965</td> <td>81.8</td> <td>81.5</td> <td>0.0115</td> <td>0.1067</td> <td>0.5668</td> <td>151</td>	0.6978	209.8	209.5	0.0081	0.0525	0.3062	82	0.6	965	81.8	81.5	0.0115	0.1067	0.5668	151			
0.9009       83.4       83.3       0.0244       0.0056       0.4926       34       0.8992       39.8       39.8       0.0275       0.0131       0.9415       65         1.0000       2.20        1.0000       0.0000       0.5991       0       1.0000       1.19        1.0000       0.0000       1.1942       0         DMSO(1)-Tetrahydrofuran(2) system at 25°C       45°C         0.0000       161.7        0.0000       1.6300       0.0000       0       0.0990       215.3       214.4       0.0029       0.9955       0.0173       72         0.2005       141.2       140.5       0.0020       0.8027       0.0829       137       0.1956       199.5       199.8       0.0047       0.7451       0.0601       123         0.3015       133.0       132.9       0.0024       0.5644       0.1621       168       0.2916       186.9       187.1       0.0062       0.5546       0.1211       156         0.3026       115.0       115.4       0.0034       0.2626       0.3615       185       0.4834       160.9       161.3       0.0091       0.2877       0.2883       182         0.6006	0.7978	152.5	152.8	0.0122	0.0233	0.3927	62	0.7	970	64.5	64.6	0.0157	0.0500	0.7343	116			
1.0000       2.20        1.0000       0.0000       0.5991       0       1.0000       1.19        1.0000       0.0000       1.1942       0         DMSO(1)-Tetrahydrofuran(2) system at 25°C       45°C         0.0000       161.7        0.0000       1.6300       0.0000       0       0.0000       233.0        0.0000       1.3404       0.0000       0         0.0998       149.8       149.2       0.0013       1.1393       0.0244       80       0.0990       215.3       214.4       0.0029       0.9955       0.0173       72         0.2005       141.2       140.5       0.0020       0.8027       0.0829       137       0.1956       199.5       199.8       0.0047       0.7451       0.0601       123         0.3015       133.0       132.9       0.0024       0.5644       0.1621       168       0.2916       186.9       187.1       0.0062       0.5546       0.1211       156         0.5026       115.0       115.4       0.0034       0.2626       0.3615       185       0.4834       160.9       161.3       0.0091       0.2877       0.2883       182         0.6006       103.8	0.9009	83.4	83.3	0.0244	0.0056	0.4926	34	0.8	992	39.8	39.8	0.0275	0.0131	0.9415	65			
DMSO(1)-Tetrahydrofuran(2) system at 25°C         45°C           0.0000         161.7          0.0000         1.6300         0.0000         0         0.0000         233.0          0.0000         1.3404         0.0000         0           0.0998         149.8         149.2         0.0013         1.1393         0.0244         80         0.0990         215.3         214.4         0.0029         0.9955         0.0173         72           0.2005         141.2         140.5         0.0020         0.8027         0.0829         137         0.1956         199.5         199.8         0.0047         0.7451         0.0601         123           0.3015         133.0         132.9         0.0024         0.5644         0.1621         168         0.2916         186.9         187.1         0.0062         0.5546         0.1211         156           0.3986         124.9         125.1         0.0029         0.3966         0.2520         183         0.4049         173.0         172.3         0.0078         0.3823         0.2128         178           0.5026         115.0         115.4         0.0034         0.2626         0.3615         185         0.4834         <	1.0000	2.20	•••	1.0000	0.0000	0.5991	0	1.0	000	1.19	•••	1.0000	0.0000	1.1942	0			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		DMSO(1)	)–Tetrahy	drofuran(	2) system	at 25°C						45°C						
0.0998       149.8       149.2       0.0013       1.1393       0.0244       80       0.0990       215.3       214.4       0.0029       0.9955       0.0173       72         0.2005       141.2       140.5       0.0020       0.8027       0.0829       137       0.1956       199.5       199.8       0.0047       0.7451       0.0601       123         0.3015       133.0       132.9       0.0024       0.5644       0.1621       168       0.2916       186.9       187.1       0.0062       0.5546       0.1211       156         0.3986       124.9       125.1       0.0029       0.3966       0.2520       183       0.4049       173.0       172.3       0.0078       0.3823       0.2128       178         0.5026       115.0       115.4       0.0034       0.2626       0.3615       185       0.4834       160.9       161.3       0.0091       0.2877       0.2883       182         0.6006       103.8       104.2       0.0041       0.1675       0.4782       173       0.6042       141.4       141.9       0.0114       0.1722       0.4258       172         0.7009       89.6       89.3       0.0052       0.0946       0.6139 <td>0,0000</td> <td>161.7</td> <td></td> <td>0.0000</td> <td>1.6300</td> <td>0.0000</td> <td>0</td> <td>0.0</td> <td>000</td> <td>233.0</td> <td>•••</td> <td>0.0000</td> <td>1.3404</td> <td>0.0000</td> <td>0</td>	0,0000	161.7		0.0000	1.6300	0.0000	0	0.0	000	233.0	•••	0.0000	1.3404	0.0000	0			
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.0998	149.8	149.2	0.0013	1.1393	0.0244	80	0.0	990	215.3	214.4	0.0029	0.9955	0.0173	72			
0.3015       133.0       132.9       0.0024       0.5644       0.1621       168       0.2916       186.9       187.1       0.0062       0.5546       0.1211       156         0.3986       124.9       125.1       0.0029       0.3966       0.2520       183       0.4049       173.0       172.3       0.0078       0.3823       0.2128       178         0.5026       115.0       115.4       0.0034       0.2626       0.3615       185       0.4834       160.9       161.3       0.0091       0.2877       0.2883       182         0.6006       103.8       104.2       0.0041       0.1675       0.4782       173       0.6042       141.4       141.9       0.0114       0.1722       0.4258       172         0.7009       89.6       89.3       0.0052       0.0946       0.6139       148       0.6967       124.0       123.5       0.0141       0.1044       0.5521       152         0.7989       70.3       70.0       0.0072       0.0438       0.7665       112       0.7972       97.4       97.5       0.0192       0.0491       0.7158       117         0.8990       42.2       42.3       0.0129       0.0115       0.9487	0.2005	141.2	140.5	0.0020	0.8027	0.0829	137	0.1	956	199.5	199.8	0.0047	0.7451	0.0601	123			
0.3986       124.9       125.1       0.0029       0.3966       0.2520       183       0.4049       173.0       172.3       0.0078       0.3823       0.2128       178         0.5026       115.0       115.4       0.0034       0.2626       0.3615       185       0.4834       160.9       161.3       0.0091       0.2877       0.2883       182         0.6006       103.8       104.2       0.0041       0.1675       0.4782       173       0.6042       141.4       141.9       0.0114       0.1722       0.4258       172         0.7009       89.6       89.3       0.0052       0.0946       0.6139       148       0.6967       124.0       123.5       0.0141       0.1044       0.5521       152         0.7989       70.3       70.0       0.0072       0.0438       0.7665       112       0.7972       97.4       97.5       0.0192       0.0491       0.7158       117         0.8990       42.2       42.3       0.0129       0.0115       0.9487       63       0.8993       60.0       60.0       0.0338       0.0129       0.9191       66         1.0000       0.59        1.0000       1.0600       1.0000 <t< td=""><td>0.3015</td><td>133.0</td><td>132.9</td><td>0.0024</td><td>0.5644</td><td>0.1621</td><td>168</td><td>0.2</td><td>916</td><td>186.9</td><td>187.1</td><td>0.0062</td><td>0.5546</td><td>0.1211</td><td>156</td></t<>	0.3015	133.0	132.9	0.0024	0.5644	0.1621	168	0.2	916	186.9	187.1	0.0062	0.5546	0.1211	156			
0.5026       115.0       115.4       0.0034       0.2626       0.3615       185       0.4834       160.9       161.3       0.0091       0.2877       0.2883       182         0.6006       103.8       104.2       0.0041       0.1675       0.4782       173       0.6042       141.4       141.9       0.0114       0.1722       0.4258       172         0.7009       89.6       89.3       0.0052       0.0946       0.6139       148       0.6967       124.0       123.5       0.0141       0.1044       0.5521       152         0.7989       70.3       70.0       0.0072       0.0438       0.7665       112       0.7972       97.4       97.5       0.0192       0.0491       0.7158       117         0.8990       42.2       42.3       0.0129       0.0115       0.9487       63       0.8993       60.0       60.0       0.0338       0.0129       0.9191       66         1.0000       0.59        1.0000       1.1674       0       1.0000       2.20        1.0000       0.0000       1.1686       0	0.3986	124.9	125.1	0.0029	0.3966	0.2520	183	0.4	049	173.0	172.3	0.0078	0.3823	0.2128	178			
0.6006         103.8         104.2         0.0041         0.1675         0.4782         173         0.6042         141.4         141.9         0.0114         0.1722         0.4258         172           0.7009         89.6         89.3         0.0052         0.0946         0.6139         148         0.6967         124.0         123.5         0.0114         0.1722         0.4258         172           0.7989         70.3         70.0         0.0072         0.0438         0.7665         112         0.7972         97.4         97.5         0.0192         0.0491         0.7158         117           0.8990         42.2         42.3         0.0129         0.0115         0.9487         63         0.8993         60.0         60.0         0.0338         0.0129         0.9191         66           1.0000         0.59          1.0000         1.1674         0         1.0000         2.20          1.0000         0.0000         1.1686         0	0.5026	115.0	115.4	0.0034	0.2626	0.3615	185	0.4	834	160.9	161.3	0.0091	0.2877	0.2883	182			
0.7009         89.6         89.3         0.0052         0.0946         0.6139         148         0.6967         124.0         123.5         0.0141         0.1044         0.5521         152           0.7989         70.3         70.0         0.0072         0.0438         0.7665         112         0.7972         97.4         97.5         0.0192         0.0491         0.7158         117           0.8990         42.2         42.3         0.0129         0.0115         0.9487         63         0.8993         60.0         60.0         0.0338         0.0129         0.9191         66           1.0000         0.59          1.0000         1.1674         0         1.0000         2.20          1.0000         0.0000         1.1686         0	0.6006	103.8	104.2	0.0041	0.1675	0.4782	173	0.6	042	141.4	141.9	0.0114	0.1722	0,4258	172			
0.7989 70.3 70.0 0.0072 0.0438 0.7665 112 0.7972 97.4 97.5 0.0192 0.0491 0.7158 117 0.8990 42.2 42.3 0.0129 0.0115 0.9487 63 0.8993 60.0 60.0 0.0338 0.0129 0.9191 66 1.0000 0.59 1.0000 0.0000 1.1674 0 1.0000 2.20 1.0000 0.0000 1.1686 0	0.7009	89.6	89.3	0,0052	0,0946	0.6139	148	0.6	967	124.0	123.5	0.0141	0.1044	0.5521	152			
0.8990 42.2 42.3 0.0129 0.0115 0.9487 63 0.8993 60.0 60.0 0.0338 0.0129 0.9191 66 1.0000 0.59 1.0000 0.0000 1.1674 0 1.0000 2.20 1.0000 0.0000 1.1686 0	0.7989	70.3	70.0	0.0072	0.0438	0.7665	112	0.7	972	97.4	97.5	0.0192	0.0491	0.7158	117			
1.0000 0.59 1.0000 0.0000 1.1674 0 1.0000 2.20 1.0000 0.0000 1.1686 0	0.8990	42.2	42.3	0.0129	0.0115	0.9487	63	0.8	993	60.0	60.0	0.0338	0.0129	0.9191	66			
	1,0000	0.59		1.0000	0.0000	1.1674	0	1.0	000	2.20		1.0000	0.0000	1.1686	0			

Table III. Critical Properties and Parameters Characterizing Vapor-Phase Nonideality

	Te, K	P <sub>c</sub> , atm	v <sub>c</sub> , cc/mol	ω	ωΗ	μ, Debye	η
DMSO	707 (9)	57.7 <b>(9)</b>	276.0 (9)	0.425ª	0.187%	4.30 (9)	0.0
Acetone	508.7 (11)	46.6(11)	213.5 (11)	0.309(11)	0.187 (11)	2.88 (11)	0.0(11)
Tetrahydrofuran	538.7°	52.2°	222.5°	0.255ª	0.252%	1.70 (16)	0.28%
Ethyl acetate	523.3(11)	37.8 (11)	286.0(11)	0.373(11)	0.278(11)	1.78 (11)	0.50(11)

• Calculated by the equation of Edmister (4). • Determined from similar substance or homomorph. • Calculated by Fishtine's method (5).



**Figure 2.** Excess thermodynamic functions for DMSO-acetone system at 25°C. Data of  $H^E$ ; Clever and Pigott (3). Brokencurve;  $G^E$  determined from Rayleigh light scattering (3, 7)

and other parameters for estimating the second-virial coefficient are shown in Table III. The pure component fugacity  $(f_i^{\circ})$  was calculated as suggested by Prausnitz et al. (11).

Columns 3–7 of Table II show the calculated vapor-liquid equilibria of DMSO-acetone, DMSO-tetrahydrofuran, and DMSO-ethyl acetate systems by the Renon NRTL equation. Table IV shows the coefficients of the Redlich-Kister equation, the NRTL equation, and the Wilson equation and the comparisons of the results by these equations. Although all three equations correlate the data satisfactorily, the NRTL equation gives slightly better fitting in average. The excess Gibbs free energies calculated by these equations give almost the same values.

## Discussion

Columns 5 and 6 of Table II show the activity coefficients of DMSO-acetone, DMSO-tetrahydrofuran, and DMSO-ethyl acetate systems, respectively. These systems are moderately nonideal. The DMSO-acetone system is more ideal than the DMSO-tetrahydrofuran system. The smaller nonideality of the DMSO-acetone system is considered to result from the stronger dipole-dipole interaction between DMSO and acetone (8). The temperature dependency of the activity coefficient is considerable for the DMSO-acetone system but negligibly small for the DMSO-tetrahydrofuran and DMSO-ethyl acetate systems.

The excess enthalpy of the DMSO-acetone system at 25°C has been reported by Clever and Pigott (3). Values of excess entropy have been calculated by combining the excess enthalpy with the excess Gibbs free energy obtained in this work. Figure 2 shows the values of  $G^E$ ,  $H^E$ , and  $TS^E$  of the DMSO-acetone system at 25°C and shows a comparison between the excess Gibbs free energy obtained in this work and those determined from the Rayleigh light scattering (3, 7). As shown in Figure 2, the excess Gibbs free energy obtained in the value determined from the Rayleigh light scattering at equimolar mixture.

#### Nomenclature

 $f_i =$  liquid-phase fugacity of component *i*, mm Hg

 $G^E$  = excess Gibbs free energy, cal/mol

 $H^E$  = excess enthalpy, cal/mol

- P = vapor pressure, mm Hg
- R = gas constant, cal/mol K
- $S^E$  = excess entropy, cal/mol K
- T = temperature, K
- v = liquid molar volume, cc/mol
- $x_i$  = mole fraction of component *i* in the liquid phase
- $y_i =$  mole fraction of component *i* in the vapor phase

#### Greek Letters

- $\gamma_i$  = liquid-phase activity coefficient of component *i*
- $\eta =$  vapor-phase association constant
- $\mu$  = dipole moment, Debye
- $\phi_i$  = vapor-phase fugacity coefficient of component *i*
- $\omega$  = acentric factor
- $\omega_H$  = acentric factor of homomorph of polar component

## Subscripts

- 1 = DMSO component
- 2 = second component, acetone, tetrahydrofuran, or ethyl acetate
- c = critical property

#### Superscript

° = pure component

Table IV. Parameters of Various Equations for Excess Gibbs Free Energy

		Redlich-Kíster, 3-parameterª				Wilson	, 2-param	neter⁵	NRTL, 3-parameter <sup>c</sup>			
System	Temp, °C	В	с	D	$\sigma^d$	<b>A</b> <sub>12</sub>	<b>A</b> <sub>21</sub>	$\sigma^{d}$	$ au_{12}$	$ au_{21}$	α	$\sigma^d$
DMSO(1)-acetone(2)	25	0.6910	0.0651	0.0233	0.37	0.5498	0.8271	0.37	0.3153	0.5585	0.9068	0.36
	35 (Run 1)	0.6553	-0.0455	0.0125	0.24	0.5948	0.8078	0.24	0.2689	0.4916	0.6763	0.26
	(Run 2)	0.6524	-0.0333	0.0303	0.27	0.6321	0.7684	0.45	0.3556	0.4702	1.0147	0.29
	45	0.6247	-0.0345	0.0095	0.33	0.6273	0.7987	0.33	0.2641	0.4470	0.6456	0.33
DMSO(1)-tetrahydro-												
furan(2)	25	1.2506	-0.1919	0.1228	0.56	0.2941	0.6543	0.61	0.6240	1.2174	0.6627	0.35
	35	1.2188	-0.1934	0.1198	0.58	0.2981	0.6756	0.63	0.5991	1.1984	0.6772	0.38
	45	1.1779	-0.1836	0.1077	0.51	0.3128	0.6906	0.53	0.5656	1.1405	0.6825	0.28
DMSO(1)-ethyl												
acetate(2)	25	1.2212	-0.0831	0.1031	0.27	0.4008	0.5499	0.62	0.7352	0.9875	0.6836	0.27
	35	1.1813	-0.0763	0.0917	0.40	0.4177	0.5642	0.62	0.7029	0.9381	0.6894	0.36
	45	1.1514	-0.0741	0.0944	0.32	0.4300	0.5725	0.64	0.6932	0.9188	0.7174	0.29
<sup>a</sup> Redlich-Kister equatio	on (13): G <sup>E</sup>	= x1x9[B +	$C(x_1 - x_2)$	$+ D(x_1 - x_1)$	κ₀)²]							

RT

<sup>b</sup> Wilson equation (18):  $\frac{G^E}{RT} = x_1 \ln (x_1 + A_{12}x_2) + x_2 \ln (x_2 + A_{21}x_1)$ 

• NRTL equation (14):  $\frac{G^{E}}{RT} = x_{1}x_{2} \left[ \frac{\tau_{21} \exp(-\alpha \tau_{21})}{x_{1} + x_{2} \exp(-\alpha \tau_{21})} + \frac{\tau_{12} \exp(-\alpha \tau_{12})}{x_{2} + x_{1} \exp(-\alpha \tau_{12})} \right]$ 

<sup>d</sup> Standard deviation in relative pressure, %; N, number of experimental points:  $100 \times \sqrt{\Sigma \left(\frac{P_{exptl} - P_{calcd}}{P_{exptl}}\right)^2 / N}$ 

## Literature Cited

- (1) Arm, H., Daeniker, H., Schaller, R., Helv. Chim. Acta, 48, 1772 (1965).
- Barker, J. A., Aust. J. Chem., 6, 207 (1953).
   Clever, H. L., Pigott, S. P., J. Chem. Thermodyn., 3, 221 (1971).
   Edmister, W. C., Petrol. Refiner, 37, 173 (1958).
- Fishtine, S. H., Ind. Eng. Chem. Fundam., 2, 149 (1963).
- (6) Geiseler, G., Mehnert, E., Z. Phys. Chem. (Frankfurt), 64, 26 (1969). (7) Haynes, L. L., Schmidt, R. L., Clever, H. L., J. Chem. Eng. Data,
- (7) Hayles, L. L., Schmidt, H. L., Olever, H. L., J. Chem. Eng. Data, 15, 534 (1970).
   (8) Jacob, S. W., Rosenbaum, E. E., Wood, D. C., "Dimethyl Sulfox-ide," Vol I, Marcel Dekker, New York, N.Y., 1971.
   (9) Jose, J., Philippe, R., Clecht, P., Bull. Soc. Chim. Fr., 2860 (1971).
- (10) Lindberg, J. J., Kenttämaa, J., Nissema, A., Suomen Kem., B34, 156 (1961).

- (11) Prausnitz, J. M., Eckert, C. A., Orye, R. V., O'Connell, J. P., "Com-puter Calculations for Multicomponent Vapor-Liquid Equilibria,"
- Prentice-Hall, Englewood Cliffs, N.J., 1967. (12) Quitzsch, K., Hüttig, R., Vogel, H.-G., Geseman, H.-J., Geiseler, G., Z. Phys. Chem. (Leipzig), **223**, 225 (1963).

- (13) Redlich, O., Kister, A. T., *Ind. Eng. Chem.*, **40**, 345 (1948).
  (14) Renon, H., Prausnitz, J. M., *AIChE J.*, **14**, 135 (1968).
  (15) Signer, R., Arm, H., Daeniker, H., *Helv. Chim. Acta*, **52**, 2347 (1969).
- (16) Stecher, P. G., "The Merck Index," 8th ed., Merck & Co., Rahway. N.J., 1968.
- (17) Timmermans, J., "Physico-Chemical Constants of Pure Organic Compounds," Vol.1, Elsevier, New York, N.Y., 1950.
- (18) Wilson, G. M., J. Amer. Chem. Soc., 86, 127 (1964).

Received for review April 2, 1973, Accepted September 19, 1973,

## Permeability of Teflon Polytetrafluoroethylene Resin and Buna-N Butadiene-Nitrile Rubber to Deuterium<sup>1</sup>

## Roy G. Derrick and McIntyre R. Louthan, Jr.

Savannah River Laboratory, E. I. du Pont de Nemours & Co., Aiken, S.C. 29801

The permeability,  $\phi$ , of Teflon and buna-N to deuterium at 0.16-5 atm between 196 and 441K is given by:  $\phi_{\mathrm{Teflon}} =$  2.8  $\times$  10<sup>-4</sup> exp (-4950/*RT*) and  $\phi_{\mathrm{Buna-}N} =$  $1.02 \times 10^{-2} \exp(-6700/RT) \text{ cm}^3$  (NTP gas) cm<sup>-1</sup> atm-1 sec-1. Temperature dependence of deuterium diffusivity, D, in these materials was calculated from measurements of the rate of rise to, and decline from, steady-state permeation. Deuterium solubilities, S, were also calculated.

Teflon polytetrafluoroethylene resin and buna-N, a nitrile rubber, are often used as gasket materials to seal valves, transducers, and/or secondary containers in hydrogen handling systems (1, 3, 5). Systems handling deuterium, an hydrogen isotope, are similarly constructed. Permeation data for Teflon and buna-N are necessary to predict system responses to various deuterium exposures where low-level permeation through the gaskets must be controlled. The permeability and the diffusivity of deuterium in these gasket materials were measured over a range of temperatures.

## Experimental

The permeability was calculated from measurements of the steady-state deuterium flux through membranes exposed to 0.16-5 atm deuterium gas in the test apparatus sketched in Figure 1. Test specimens were  $\sim$  0.050 cm thick by about 4.83 cm in diameter. High-purity

<sup>&</sup>lt;sup>1</sup> Address correspondence to A. E. Symonds, Technical Information Service, Savannah River Laboratory, E. i. du Pont de Nemours & Co., Aiken, S.C. 29801.