

Total Pressure Measurements for Chloroform + Acetone + Toluene at 303.15 K

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Total pressure measurements are reported for chloroform + acetone + toluene and its constituent binary systems at 303.15 K. The results were obtained using a Van Ness apparatus and were fitted to a flexible correlating expression for the excess Gibbs free energy of the liquid phase using Barker's method. This expression represents the measured pressures for the ternary system with an average deviation of 0.06 kPa. The applicability of the NRTL equation to this system is examined, and it is found to predict the ternary results with an average deviation of 0.26 kPa.

Introduction

To represent properties of liquid mixtures in which hydrogen bonding occurs, it has become common to use solution models which contain contributions both from van der Waals forces and from "chemical" effects. The chemical effects are treated by assuming that new species are formed by hydrogen bonding in amounts dictated by chemical equilibria. An assumption about the stoichiometry of any associated complex is required, and models have been published for which the number of species forming a complex range from two to infinity.

For many systems, including that of the present investigation, it is sufficient to assume that the only species present are monomers and dimers. The dimers may be assumed to be formed by self-association, cross-association, or both, depending on the species present. Examples of monomer-dimer models are the combined physical and chemical model proposed by Harris and Prausnitz (1), the equation of state with dimerization proposed by Gmehling et al. (2), and the monomer-dimer form of the APACT equation of state of Ikonomou and Donohue (3).

The majority of association models proposed in the literature have been applied only to pure substances and binary mixtures. To extend these models to higher order systems, experimental data for ternary and higher systems must be available for testing. As part of a continuing study of ternary systems in which one or more complexes are present in the liquid phase, total pressure measurements are reported here for the system chloroform + acetone + toluene and its three constituent binary systems at 303.15 K. This system was chosen because of the presence of the well-known chloroform-acetone complex and the weak chloroform-toluene complex (4).

Many sets of isothermal data have been reported for acetone + chloroform. These are summarized by Apelblat et al. (5) and by Goral et al. (6). Isothermal data for chloroform + toluene have been reported at 318.15 K by Ohta et al. (7) and for acetone + toluene at 318.15 K by Orye and Prausnitz (8), at 313.15 K by Kolasinska et al. (9), and at 308.15, 318.15, and 328.15 K by Kraus and Linek (10). No isothermal data have been located for the system chloroform + acetone + toluene. However, isobaric data at 1.01 bar were reported by Satapathy et al. (11).

Experimental Section

Apparatus and Procedure. The apparatus is identical to that described in detail earlier by Bhethanabotla and Campbell (12) except for the replacement of the pressure gauge with one of 0.001-kPa resolution as described by Pradhan et al. (13). It is of the Van Ness type (14) in which total pressure is measured as a function of overall composition in the equilibrium cell. For binary system measurements, the overall composition in the cell was changed by charging metered amounts of the pure components from their respective piston injectors. Volumes displaced from the piston injectors were converted to moles using saturated liquid volumes of the pure species at room temperature which were obtained from ref 15.

Ternary system measurements were made following the same procedure except that one piston injector contained pure acetone while the other contained a mixture of chloroform and toluene. Hence, a ternary run proceeded along a line of constant mole ratio of chloroform to toluene and is characterized by the parameter C' , defined as

$$C' = \frac{z_1}{z_1 + z_3} \quad (1)$$

where z_1 and z_3 are the overall mole fractions of chloroform and toluene, respectively, in the equilibrium cell. Three such runs were made corresponding to C' values of 0.2642, 0.4883, and 0.7499.

For the ternary runs, it was necessary to know the exact density of the chloroform + toluene mixture in the piston injector at room temperature. This was found using the saturated liquid volumes obtained above in conjunction with excess volume data reported by Rastogi et al. (16).

The experiment consists of the measurement of the pressure and the overall composition in the equilibrium cell. Corrections to obtain the liquid-phase composition are made as part of the data reduction procedure as described earlier (12).

Experimental uncertainties are $\pm 0.1\%$ in pressure, ± 0.02 K in temperature, and between ± 0.0005 and ± 0.001 in mole fraction, the smaller value applying at the extremes in composition.

Materials. Chloroform, acetone, and toluene were all obtained from Aldrich Chemical (HPLC grade) and had purities of 99.9% or better. The acetone and toluene were used without purification except for degassing as described by Bhethanabotla and Campbell (12). The chloroform, which contained a small amount of amylene stabilizer, was purified

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Table 1. Vapor Pressures P_i^{sat} , Saturated Liquid Volumes V_i^{L} , and Second Virial Coefficients for Single Components B_{ii} and Mixtures B_{ij} Used for Chloroform + Acetone + Toluene at 303.15 K

	chloroform	acetone	toluene
$P_i^{\text{sat}}/\text{kPa}$	32.421	37.946	5.033
$V_i^{\text{L}}/(\text{cm}^3 \text{mol}^{-1})$	81.2	74.5	107.4
$B_{ii}/(\text{cm}^3 \text{mol}^{-1})$	-1168	-1835	-2567
Second Virial Cross Coefficients, $B_{ij}/(\text{cm}^3 \text{mol}^{-1})$			
chloroform + acetone		-793	
chloroform + toluene		-1718	
acetone + toluene		-1136	

Table 2. Values of Parameters Appearing in Eqs 2 and 4 and Resulting Average Deviations ΔP_{av} and Maximum Deviations ΔP_{max} for Chloroform + Acetone + Toluene and Its Constituent Binary Systems at 303.15 K

	acetone (i) + toluene (j)	chloroform (i) + toluene (j)	chloroform (i) + acetone (j)
A_{ij}	0.7507	-0.3598	-0.6832
A_{ji}	0.6752	-0.2646	-0.7218
α_{ij}	0.2780	0.1425	0.5027
α_{ji}	0.2938	22.2653	3.0636
$\Delta P_{\text{av}}/\text{kPa}$	0.027	0.021	0.016
$\Delta P_{\text{max}}/\text{kPa}$	0.084	0.084	0.032
Chloroform (1) + Acetone (2) + Toluene (3)			
C_{11}		0.0104	
C_{22}		-0.2341	
C_{33}		-0.0622	
$\Delta P_{\text{av}}/\text{kPa}$		0.055	
$\Delta P_{\text{max}}/\text{kPa}$		0.175	

by contacting with type 13X molecular sieves followed by filtering and degassing. The averages of the pure component vapor pressures measured in this study at 303.15 K are reported in Table 1. The values for chloroform and acetone are 0.112 kPa (0.3%) higher and 0.065 kPa (0.2%) lower than those reported in ref 15. The value for toluene is 0.138 kPa (2.8%) higher than that reported in ref 15 and is in agreement with that reported by Mahl et al. (17).

Data Reduction. Data were reduced using Barker's method (18) and a flexible correlating equation for the excess Gibbs free energy. Unweighted regressions were performed in which the sum of the squares of the differences between calculated and experimental pressures was minimized. The binary systems were represented by the four-parameter modified Margules equation proposed by Abbott and Van Ness (19):

$$\frac{G^E}{RT} = x_i x_j \left[A_{ji} x_i + A_{ij} x_j + \frac{\alpha_{ij} \alpha_{ji} x_i x_j}{\alpha_{ij} x_i + \alpha_{ji} x_j} \right] \quad (2)$$

The ternary system was represented by the form advocated by Abbott et al. (20):

$$\frac{G^E}{RT} = \left(\frac{G^E}{RT} \right)_{1,2} + \left(\frac{G^E}{RT} \right)_{1,3} + \left(\frac{G^E}{RT} \right)_{2,3} + F(x_1, x_2, x_3) \quad (3)$$

where the expressions used for $(G^E/RT)_{ij}$ are those obtained from the fits of the modified Margules equation to the three constituent binary systems. The function F was taken to be

$$F = x_1 x_2 x_3 (C_0 - C_{11} x_1 - C_{22} x_2 - C_{33} x_3) \quad (4)$$

where

$$C_0 = (1/2) \sum_i \sum_j A_{ij} \quad i \neq j \quad (5)$$

A detailed description of the fitting procedure is given by Bhethanabotla and Campbell (12). Second virial coefficients

Table 3. Total Pressure P as a Function of Liquid-Phase Mole Fraction x_i for Chloroform (1) + Acetone (2), Chloroform (1) + Toluene (3), and Acetone (2) + Toluene (3) at 303.15 K

chloroform (1) + acetone (2)		chloroform (1) + toluene (3)		acetone (2) + toluene (3)	
x_1	P/kPa	x_1	P/kPa	x_2	P/kPa
0.0308	37.221	0.0337	5.612	0.0315	7.226
0.0597	36.580	0.0620	6.143	0.0603	8.949
0.1008	35.649	0.1007	6.902	0.0997	11.046
0.1502	34.525	0.1474	7.869	0.1492	13.332
0.1998	33.397	0.2012	9.051	0.1989	15.375
0.2503	32.269	0.2497	10.167	0.2481	17.196
0.3007	31.172	0.3000	11.395	0.2980	18.893
0.3506	30.160	0.3500	12.679	0.3480	20.478
0.4000	29.248	0.3997	14.011	0.3978	21.901
0.4504	28.463	0.4493	15.400	0.4478	23.385
0.5000	27.854	0.4992	16.860	0.4899	24.538
0.5000	27.804	0.5495	18.400	0.5480	26.078
0.5499	27.403	0.5996	19.954	0.5981	27.384
0.5999	27.247	0.5493	18.272	0.5976	27.526
0.6497	27.355	0.5988	19.796	0.6477	28.767
0.6996	27.704	0.6492	21.372	0.6977	30.038
0.7496	28.283	0.6993	22.973	0.7479	31.321
0.7993	29.032	0.7484	24.566	0.7981	32.613
0.8493	29.874	0.7993	26.222	0.8485	33.808
0.8986	30.753	0.8496	27.832	0.8989	35.145
0.9392	31.477	0.8993	29.390	0.9393	36.240
0.9695	31.993	0.9399	30.632	0.9692	37.073
		0.9695	31.529		

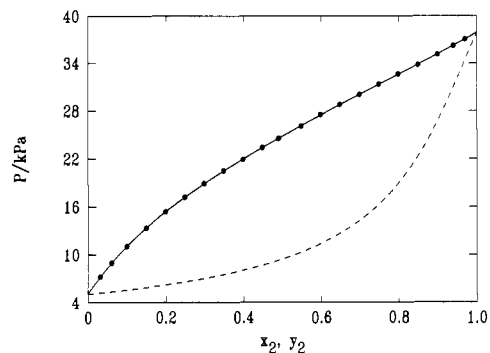


Figure 1. Pressure P vs liquid-phase mole fraction x_2 or vapor-phase mole fraction y_2 for acetone (2) + toluene (3) at 303.15 K. The points are experimental results, the solid curve is the fitted P - x result, and the dashed curve is the predicted P - y result.

and saturated liquid volumes of the pure chemicals are required for data reduction and are included in Table 1. Second virial coefficients were calculated using the correlation of Tsonopoulos (21) with $k_{ij} = 0.13, 0.13,$ and 0.0 for acetone + toluene, acetone + chloroform, and toluene + chloroform, respectively. Saturated liquid volumes at the temperature of the mixing were taken from the same source as the room temperature values used above.

Results

The results of the data reduction procedure are a set of corrected liquid-phase mole fractions for each pressure and values for the parameters appearing in the G^E model. Parameter values and resulting deviations in pressure are given for each system in Table 2. The binary data are all represented by the G^E model to within an average of 0.03 kPa while the ternary data are represented to within an average of 0.06 kPa.

P - x data at 303.15 K for the three constituent binary systems are presented in Table 3 and are shown in Figures 1-3. The only isothermal data at 303.15 K available in the literature are those reported by Rock and Schroder (22) for

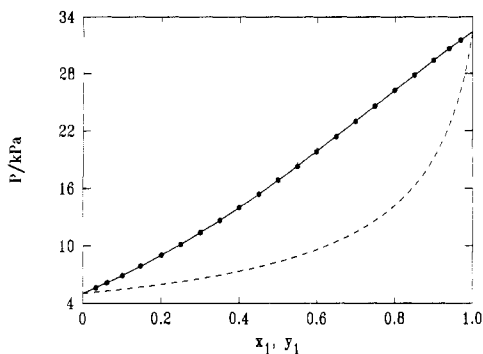


Figure 2. Pressure P vs liquid-phase mole fraction x_1 or vapor-phase mole fraction y_1 for chloroform (1) + toluene (3) at 303.15 K. The points are experimental results, the solid curve is the fitted P - x result, and the dashed curve is the predicted P - y result.

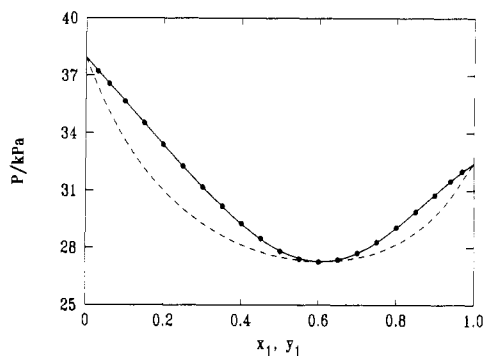


Figure 3. Pressure P vs liquid-phase mole fraction x_1 or vapor-phase mole fraction y_1 for chloroform (1) + acetone (2) at 303.15 K. The points are experimental results, the solid curve is the fitted P - x result, and the dashed curve is the predicted P - y result.

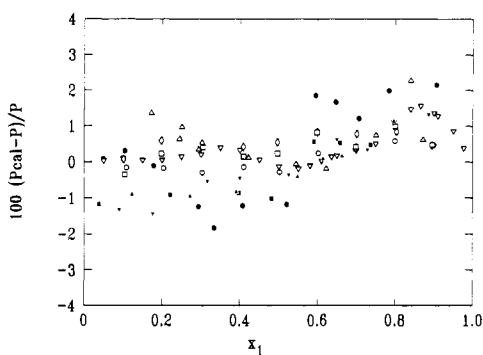


Figure 4. Percent deviations between calculated pressures P_{calcd} and experimental pressures P vs liquid-phase mole fraction x_1 for chloroform (1) + acetone (2): ∇ , Campbell et al. (24), 298.15 K; \blacksquare , \blacktriangle Mueller and Kearns (25), 298.15 and 308.15 K; \circ , Kudryavtseva and Susarev (26), 308.15 K; ∇ , Rabinovich and Nikolaev (27), 298.15 K; \diamond , \square Rock and Schroder (22), 303.15 and 308.15 K; \bullet , \triangle Apelblat et al. (5) 298.15 and 308.32 K.

chloroform + acetone. However, heat of mixing data are available for all three constituent binary systems, and these may be used to compare the total pressure data at 303.15 K reported here to data at other temperatures. For the systems acetone + toluene and chloroform + acetone, the following procedure was used to make these comparisons.

(1) The parameters given in Table 2 were used with eq 2 to yield an expression for G^E at 303.15 K which represents the results reported in this work with the deviations given in Table 2.

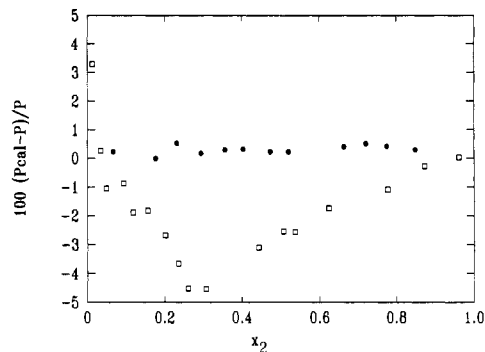


Figure 5. Percent deviations between calculated pressures P_{calcd} and experimental pressures P vs liquid-phase mole fraction x_2 for acetone (2) + toluene (3): \bullet , Kolasinska et al. (9), 313.15 K; \square , Orye and Prausnitz (8), 318.15 K.

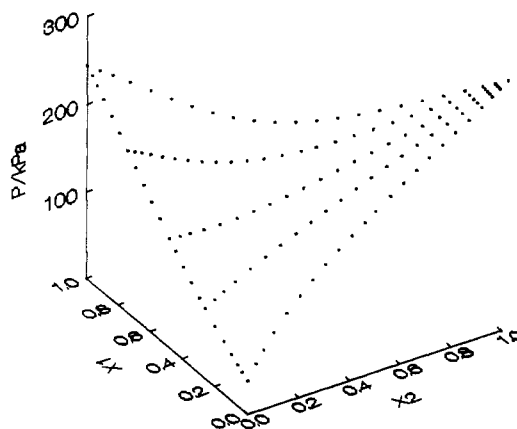


Figure 6. Pressure P vs liquid-phase mole fractions x_1 and x_2 for chloroform (1) + acetone (2) + toluene (3) at 303.15 K.

(2) It was assumed that, for a small range in temperature, the excess Gibbs free energy at any temperature T for the two binary systems could be represented by

$$\left(\frac{G^E}{RT}\right)_T = \left(\frac{G^E}{RT}\right)_{303.15} + x_i x_j \left[A \left(\frac{1}{T} - \frac{1}{303.15} \right) x_j + B \left(\frac{1}{T} - \frac{1}{303.15} \right) x_i - D \left(\frac{1}{T} - \frac{1}{303.15} \right) x_i x_j \right] \quad (6)$$

Using the Gibbs-Helmholtz relation, the corresponding expression for the excess enthalpy H^E is given by

$$H^E/R = x_i x_j [A x_j + B x_i - D x_i x_j] \quad (7)$$

Values for A , B , and D in eqs 6 and 7 were obtained for acetone + toluene from Orye and Prausnitz (8) and represent their heat of mixing data at 318.15 K with an average deviation of 6.4 J mol⁻¹. For chloroform + acetone, the parameters were obtained by fitting eq 7 to the heat of mixing results of Kearns (23) at 298.15 K (average deviation of 9.6 J mol⁻¹).

(3) Expressions for activity coefficients of individual species at any temperature were then obtained from eq 6 by

$$\ln \gamma_k = \left(\frac{\partial n G^E / RT}{\partial n_k} \right)_{T, P, n_{i \neq k}} \quad (8)$$

These expressions were then used to calculate pressure P_{calcd} for each liquid composition in a data set from the literature. In order to minimize any effects of pure component impurities, the pure component vapor pressures reported with the data set in question were used in all computations. The pressures calculated in this fashion were compared to the measured pressures from each set.

Table 4. Total Pressure P for Chloroform (1) + Acetone (2) + Toluene (3) at 303.15 K as a Function of Liquid-Phase Mole Fractions x_1 and x_2 for Given Values of the Parameter C' , Eq 1

$C' = 0.2642$			$C' = 0.4883$			$C' = 0.7499$		
x_1	x_2	P/kPa	x_1	x_2	P/kPa	x_1	x_2	P/kPa
0.2552	0.0307	11.623	0.4723	0.0303	16.687	0.7354	0.0179	24.458
0.2474	0.0606	12.654	0.4579	0.0600	17.084	0.7214	0.0367	24.310
0.2372	0.0997	13.920	0.4378	0.1015	17.657	0.7039	0.0601	24.131
0.2240	0.1500	15.451	0.4142	0.1501	18.373	0.6802	0.0918	23.902
0.2109	0.1999	16.921	0.3895	0.2009	19.204	0.6553	0.1251	23.710
0.1979	0.2496	18.353	0.3654	0.2507	20.105	0.6292	0.1600	23.573
0.1847	0.2996	19.757	0.3415	0.2998	21.062	0.6017	0.1969	23.501
0.1716	0.3496	21.133	0.3172	0.3498	22.116	0.5733	0.2348	23.534
0.1584	0.3997	22.501	0.2929	0.3997	23.223	0.5429	0.2755	23.677
0.1452	0.4497	23.837	0.2685	0.4497	24.377	0.5112	0.3179	23.965
0.1320	0.4999	25.166	0.2441	0.4998	25.566	0.4775	0.3628	24.411
0.1188	0.5499	26.466	0.2198	0.5497	26.776	0.4420	0.4103	25.023
0.1188	0.5500	26.512	0.2196	0.5500	26.798	0.4043	0.4606	25.822
0.1055	0.6003	27.799	0.1954	0.5997	28.011	0.3750	0.4998	26.538
0.0924	0.6499	29.059	0.1468	0.6992	30.457	0.3375	0.5498	27.535
0.0792	0.7000	30.323	0.1224	0.7493	31.696	0.3377	0.5496	27.548
0.0661	0.7497	31.583	0.1710	0.6497	29.239	0.3001	0.5997	28.626
0.0527	0.8003	32.872	0.0978	0.7996	32.936	0.2627	0.6496	29.742
0.0396	0.8501	34.119	0.0728	0.8508	34.213	0.2252	0.6996	30.905
0.0264	0.9001	35.417	0.0487	0.9001	35.449	0.1876	0.7498	32.097
0.0158	0.9403	36.454	0.0293	0.9400	36.449	0.1501	0.7997	33.287
0.0077	0.9707	37.264	0.0149	0.9696	37.196	0.1130	0.8492	34.470
						0.0747	0.9004	35.708
						0.0448	0.9402	36.662
						0.0230	0.9693	37.354

Table 5. Parameters for the NRTL Equation and Resulting Average Deviations ΔP_{av} and Maximum Deviations ΔP_{max} for Chloroform + Acetone + Toluene and Its Constituent Binary Systems at 303.15 K

	acetone (i) + toluene (j)	chloroform (i) + toluene (j)	chloroform (i) + acetone (j)
τ_{ij}	0.1946	1.6416	2.5730
τ_{ji}	0.4879	-1.3554	-1.9615
α_{ij}	0.30	0.30	0.30
$\Delta P_{av}/\text{kPa}$	0.107	0.025	0.142
$\Delta P_{max}/\text{kPa}$	0.199	0.085	0.241
Chloroform (1) + Acetone (2) + Toluene (3)			
$\Delta P_{av}/\text{kPa}$		0.256	
$\Delta P_{max}/\text{kPa}$		0.680	

These comparisons are shown for chloroform + acetone in Figure 4. Since many literature data are available for this system, it was felt necessary only to compare the present results to those obtained in the temperature range of 298.15–308.32 K. The data considered are those of Rock and Schroder (22) at 303.15 and 308.15 K, Campbell et al. (24) at 298.15 K, Mueller and Kearns (25) at 298.15 and 308.15 K, Kudryavtseva and Susarev (26) at 308.15 K, Rabinovich and Nikolaev (27) at 298.15 K, and Apelblat et al. (5) at 298.15 and 308.32 K. Figure 4 indicates that the pressures predicted using the data of the present study fall within the range of literature values for liquids rich in acetone but are higher than the average of those reported in other studies by a maximum of approximately 1.0% for liquids rich in chloroform.

The comparisons for acetone + toluene are shown in Figure 5. Kraus and Linek (10) reported vapor–liquid equilibrium measurements for this system at 308.15, 318.15, and 328.15 K but did not include toluene vapor pressures within this temperature range. Hence, a comparison to their results was not made. Instead, comparisons were made to the data of Orye and Prausnitz (8) at 318.15 K and those of Kolasinska et al. (9) at 313.15 K. Figure 5 indicates good agreement between the present study and that of Kolasinska et al. However, the present study predicts pressures that are as much as 5% lower than those reported by Orye and Prausnitz.

Isothermal data for chloroform + toluene at 318.15 K have been reported by Ohta et al. (7). Since they did not measure

pure component vapor pressures, the analysis used for the other two constituent binary systems could not be performed. However, since they reported numerical values for the liquid-phase activity coefficients, a comparison of the present results to theirs can be made. The following procedure was used. First, values of parameters A , B , and D in eqs 6 and 7 were obtained from Rastogi et al. (16), who found that eq 7 was able to represent their heat of mixing data at 308.15 K with a standard deviation of 6.4 J mol⁻¹. Next, the activity coefficients reported by Ohta et al. were used to calculate values of G^E/RT as a function of liquid-phase composition at 318.15 K. Equation 2 was then fitted to these results to determine values of the parameters A_{ij} , A_{ji} , α_{ij} , and α_{ji} at 318.15 K. All of the parameters were then used in eq 6 with a new reference temperature of 318.15 K and a temperature of 303.15 K to calculate pressure for each liquid composition of the present study (using the pure component vapor pressures of the present study). The analysis results in calculated pressures that are systematically higher by an average of 0.48 kPa (3.3%) than the measured pressures reported here.

P - x data at 303.15 K for the chloroform + acetone + toluene system are given in Table 4. The P - x surface described by the binary measurements and the three ternary runs is shown in Figure 6. A ternary azeotrope does not exist for this system at 303.15 K.

Ternary vapor–liquid equilibrium data are useful for testing the predictive capability of various solution models. Local composition equations are particularly useful here because all parameters may be determined from fitting the vapor–liquid equilibrium data for the constituent binary systems. As an example, the NRTL equation (28) is considered:

$$G^E/RT = \sum_i x_i \left[\sum_j \tau_{ji} G_{ji} x_j / \sum_k G_{ki} x_k \right] \quad (9)$$

where

$$G_{ij} = \exp(-\alpha_{ij} \tau_{ij}) \quad (10)$$

and where $\alpha_{ij} = \alpha_{ji}$. For a binary system, eq 9 contains three parameters, α_{12} , τ_{12} , and τ_{21} . The parameters α_{ij} were fixed

at 0.30 for all three constituent binary systems. Barker's method was used to determine the parameter values τ_{ij} and τ_{ji} from the binary data of this study with the results shown in Table 5. These parameters were also used to predict pressures for the ternary liquid compositions and were compared to the experimental results as shown in Table 5. The predictions are fair and correspond to an average deviation in pressure of 1.1%. This is consistent with results for other systems examined by Renon and Prausnitz (28).

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