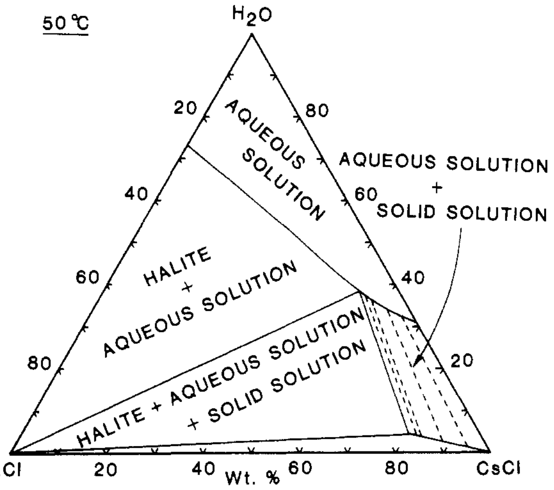

Figure 3. Solubility relations in the system NaCl-CsCl-H_2O at 50 °C. The dashed lines are experimentally determined tie lines (see Figure 1).

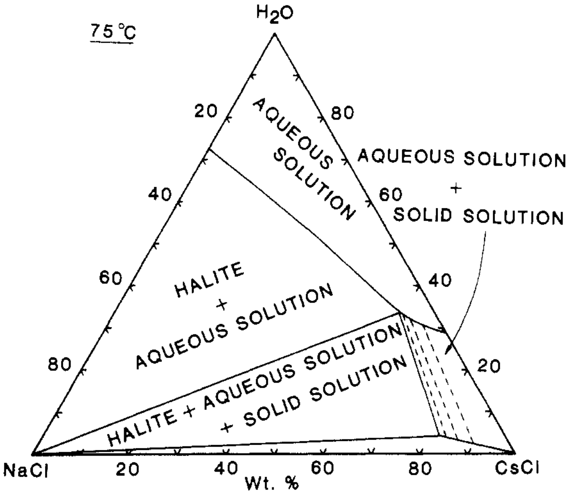

Figure 4. Solubility relations in the system NaCl-CsCl-H_2O at 75 °C. The dashed lines are experimentally determined tie lines (see Figure 2).

gravimetrically (*2*). The reported solution compositions (Table I) are accurate to ±0.3 wt %.

Results are summarized in Figures 1 and 2 (solid symbols and solid lines) for 50 and 75 °C experiments, respectively. The solubilities of CsCl in H_2O are from Clynne and Potter (*4*). Also shown in these figures (open symbols and dashed lines) are the solubility relations given by Plyushchev et al. (*3*). The compositions of their solids were not determined. However, the existence of solid solutions between NaCl and CsCl was proposed. Their solubility data agree with our values to within ±0.5 wt % at 50 °C and within ±0.8 wt % at 75 °C. The orientations of their solid-solution tie lines (dashed lines in Figures 1 and 2) agree very well with ours (solid lines) at 75 °C. However, the agreement at 50 °C is limited to the most CsCl-rich region only. The composition of their eutonic solution (the solution that coexists with two solids) agrees with that of ours to ±0.37 wt % at 50 °C and to ±0.73 wt % at 75 °C.

By combining our results here with the halite solubility data presented earlier (*1*), we were able to determine the solubility relations of the entire system at 50 and 75 °C. These relations are shown in Figures 3 and 4. At these temperatures, the new compound, $CsCl{\cdot}2NaCl{\cdot}2H_2O$, found at 25 °C (*2*) is not stable. However, the solid solutions, $[Cs_{1-x}(Na{\cdot}H_2O)_x]Cl$, remain stable at these temperatures. The maximum values for x are 0.352 at 50 °C and 0.325 at 75 °C, compared with a value of 0.427 at 25 °C (*2*).

Conclusion

The solubility relations in the system NaCl-CsCl-H_2O are determined at 50 and 75 °C. Our solubility data agree very well with the literature values (*3*). However, the relations between the solutions and the coexisting solids reported by Plyushchev et al. (*3*) are modified considerably in this study.

Registry No. Cesium chloride, 7647-17-8; sodium chloride, 7647-14-5.

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Vapor-Liquid Equilibria for the Ternary Systems Acetonitrile-2-Butanone-Benzene and Acetonitrile-Methanol-Benzene at 328.15 K

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Isothermal vapor-liquid equilibrium data at 328.15 K are presented for the ternary systems acetonitrile-2-butanone-benzene and acetonitrile-methanol-benzene, and three binary systems: 2-butanone-acetonitrile, 2-butanone-benzene, and acetonitrile-benzene. Experimental data are correlated by the Wilson and UNIQUAC equations and the UNIFAC method.

Introduction

Vapor-liquid equilibrium data at 328.15 K were obtained for the two ternary systems acetonitrile-2-butanone-benzene and acetonitrile-methanol-benzene, and three binary systems: 2-butanone-acetonitrile, 2-butanone-benzene, and acetonitrile-benzene with a Boublik still (*1*). Isothermal vapor-liquid equilibrium data for the binary systems have been published by several investigators: 2-butanone-acetonitrile (*2*); 2-buta-

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Table I. Physical Constants of Compounds

compd	density at 298.15 K, g/cm^3		refractive index at 298.15 K		vapor press. at 328.15 K, kPa	
	exptl	lit.[a]	exptl	lit.[a]	exptl	lit.[a]
acetonitrile	0.7767	0.776 6	1.3416	1.341 63	40.86	40.849[b]
2-butanone	0.7999	0.799 7	1.3762	1.376 4	43.64	43.640
benzene	0.8735	0.873 70	1.4979	1.497 92	43.57	43.592
methanol	0.7868	0.786 64	1.3265	1.326 52	68.30	68.289

[a] Reference *28*. [b] Reference *29*.

Table II. Experimental Vapor-Liquid Equilibrium Data for Three Binary Systems at 328.15 K

x_1	y_1	P, kPa	γ_1	γ_2	ψ_1	ψ_2
2-Butanone (1)-Acetonitrile (2)						
0.079	0.096	42.12	1.191	1.010	0.988	0.943
0.160	0.184	43.21	1.153	1.024	0.985	0.942
0.239	0.264	43.70	1.117	1.032	0.983	0.942
0.336	0.357	43.98	1.079	1.040	0.981	0.943
0.442	0.455	44.09	1.046	1.053	0.978	0.944
0.547	0.551	44.54	1.032	1.081	0.976	0.945
0.640	0.637	44.48	1.016	1.100	0.975	0.947
0.755	0.748	44.33	1.007	1.122	0.974	0.950
0.852	0.845	44.09	1.002	1.140	0.974	0.953
0.902	0.897	43.86	1.000	1.140	0.973	0.955
2-Butanone (1)-Benzene (2)						
0.060	0.072	44.69	1.234	1.012	0.977	0.981
0.159	0.174	45.65	1.148	1.028	0.976	0.981
0.278	0.293	46.50	1.124	1.044	0.974	0.981
0.400	0.400	46.74	1.071	1.072	0.973	0.981
0.499	0.489	46.70	1.048	1.093	0.973	0.982
0.608	0.591	46.37	1.032	1.111	0.973	0.982
0.708	0.688	46.14	1.026	1.133	0.972	0.983
0.809	0.788	45.64	1.018	1.165	0.972	0.984
0.911	0.901	44.64	1.011	1.144	0.973	0.985
Acetonitrile (1)-Benzene (2)						
0.062	0.127	47.88	2.465	1.022	0.971	0.981
0.074	0.149	48.62	2.455	1.024	0.968	0.981
0.125	0.216	50.58	2.177	1.039	0.962	0.981
0.207	0.296	52.41	1.852	1.069	0.954	0.983
0.290	0.355	53.78	1.618	1.124	0.949	0.984
0.336	0.388	54.28	1.536	1.152	0.947	0.985
0.350	0.398	54.46	1.517	1.162	0.946	0.986
0.449	0.455	54.90	1.357	1.254	0.942	0.988
0.554	0.517	54.46	1.236	1.367	0.939	0.992
0.706	0.610	52.68	1.104	1.629	0.937	0.997
0.819	0.700	50.36	1.044	1.958	0.937	1.004
0.953	0.893	44.10	1.007	2.387	0.941	1.017

none-benzene (*3*, *4*); acetonitrile-benzene (*5-7*). The data at the same temperature have been reported for the two constituent binary systems: methanol-acetonitrile (*8*) and methanol-benzene (*9*). Ternary data reduction was made by the Wilson and UNIQUAC equations (*10*) and the UNIFAC method (*11-13*) with only parameters obtained from binary data.

Experimental Section

Materials. Guaranteed reagent-grade acetonitrile was used directly. C.P. 2-butanone was treated with saturated potassium carbonate solution, then dried over anhydrous sodium sulfate, and fractionally distilled. C.P. benzene was repeatedly recrystallized. C.P. methanol was fractionated in a glass distillation column packed with McMahon packing after storage over anhydrous calcium sulfate. Table I compares values of the densities, refractive indices, and vapor pressures of these materials with literature values.

Apparatus. Isothermal vapor-liquid equilibrium data were obtained with a Boublik still (*1*, *14*). Further details on the still and experimental techniques were described previously (*1*). Vapor and liquid samples were analyzed by a Shimadzu Pulfrich refractometer at 298.15 K for the 2-butanone-benzene and acetonitrile-benzene systems and by a Shimadzu (GC-7A) gas chromatograph connected to a Shimadzu (ITG-2A) digital integrator for the other systems. The errors of observed values are as follows: composition, 0.002 mole fraction; pressure, 13 Pa; temperature, 0.05 K.

Experimental Results and Data Analysis

Table II lists vapor-liquid equilibrium data for the three binary systems. Figures 1-3 compare experimental P-x-y data for the binary systems with those obtained by other authors (*2-7*). The present azeotropic data seem to be compatible with literature values. Tables III and IV include experimental results for the two ternary systems. The liquid-phase activity coefficients were calculated by considering vapor-phase nonideality

$$\gamma_i = \psi_i y_i P/\{\psi_i^s x_i P_i^s \exp[v_i^L(P - P_i^s)/RT]\} \qquad (1)$$

where the vapor pressures of pure components were taken from the experimental values in Table I. The vapor-phase fugacity coefficients were obtained from the virial equation of state truncated after the second term. The generalized method of Hayden and O'Connell (*15*) was used to estimate the second virial coefficients. The liquid molar volumes of each component were calculated by the modified Rackett equation (*16*). Further details on the numerical calculations of these properties are given in the monograph of Prausnitz et al. (*10*).

The activity coefficient for any component i in a multicomponent system is given as follows:

Wilson equation

$$\ln \gamma_i = 1 - \ln \sum_j x_j \Lambda_{ij} - \sum_k \frac{x_k \Lambda_{ki}}{\sum_j x_j \Lambda_{kj}} \qquad (2)$$

where

$$\Lambda_{ij} = (v_j^L/v_i^L) \exp(-\Delta\lambda_{ij}/RT) \equiv (v_j^L/v_i^L) \exp(-a_{ij}/T) \qquad (3)$$

UNIQUAC equation (Anderson and Prausnitz's modification)

$$\ln \gamma_i = \ln (\phi_i/x_i) + 1 - \phi_i/x_i - (Z/2)q_i[\ln (\phi_i/\theta_i) + 1 - \phi_i/\theta_i] - q_i' \ln (\sum_j \theta_j'\tau_{ji}) + q_i' - q_i'\sum_j \frac{\theta_j'\tau_{ij}}{\sum_k \theta_k'\tau_{kj}} \qquad (4)$$

where Z is the coordination number equal to 10 and

$$\tau_{ij} = \exp(-\Delta u_{ij}/RT) \equiv \exp(-a_{ij}/T) \qquad (5)$$

$$\phi_i = x_i r_i / \sum_j x_j r_j \qquad (6)$$

$$\theta_i = x_i q_i / \sum_j x_j q_j \qquad (7)$$

$$\theta_i' = x_i q_i' / \sum_j x_j q_j' \qquad (8)$$

The value of q' for methanol is 0.96 and q' equals q for all other components (*10*, *17*).

Binary Systems. The thermodynamic consistency test proposed by Christiansen and Fredenslund (*18*) was applied to the

Table III. Experimental Vapor-Liquid Equilibrium Data for the Ternary System Acetonitrile (1)-2-Butanone (2)-Benzene (3) at 328.15 K

x_1	x_2	x_3	y_1	y_2	y_3	P, kPa	γ_1	γ_2	γ_3	ψ_1	ψ_2	ψ_3
0.327	0.347	0.326	0.343	0.295	0.362	49.73	1.280	0.965	1.277	0.948	0.970	0.989
0.243	0.463	0.294	0.262	0.407	0.331	48.74	1.294	0.979	1.267	0.951	0.970	0.987
0.297	0.258	0.445	0.326	0.213	0.461	50.53	1.364	0.952	1.207	0.949	0.969	0.987
0.448	0.305	0.247	0.438	0.261	0.301	49.22	1.177	0.963	1.392	0.944	0.971	0.993
0.105	0.550	0.345	0.125	0.510	0.365	47.09	1.391	0.998	1.147	0.958	0.971	0.984
0.338	0.106	0.556	0.373	0.083	0.544	52.74	1.428	0.941	1.190	0.947	0.968	0.986
0.548	0.358	0.094	0.523	0.339	0.138	46.81	1.091	1.018	1.606	0.943	0.975	1.000
0.152	0.304	0.544	0.194	0.268	0.538	49.02	1.552	0.987	1.115	0.958	0.970	0.983
0.549	0.154	0.297	0.513	0.125	0.362	51.13	1.164	0.948	1.448	0.941	0.971	0.994
0.298	0.559	0.143	0.305	0.518	0.177	46.34	1.166	0.983	1.329	0.949	0.973	0.991
0.104	0.456	0.440	0.130	0.418	0.452	47.60	1.479	0.997	1.124	0.960	0.971	0.984
0.451	0.109	0.440	0.447	0.084	0.469	52.89	1.280	0.929	1.304	0.943	0.968	0.990
0.444	0.455	0.101	0.436	0.427	0.137	46.98	1.128	1.011	1.484	0.944	0.974	0.996
0.055	0.706	0.239	0.065	0.676	0.259	46.01	1.351	1.008	1.148	0.959	0.972	0.985
0.239	0.065	0.696	0.307	0.052	0.641	52.06	1.650	0.949	1.102	0.953	0.968	0.984
0.697	0.250	0.053	0.655	0.252	0.093	45.22	1.037	1.051	1.866	0.942	0.979	1.006
0.053	0.293	0.654	0.080	0.279	0.641	47.92	1.810	1.044	1.077	0.967	0.972	0.981
0.652	0.056	0.292	0.581	0.045	0.374	51.85	1.123	0.952	1.547	0.939	0.971	0.997
0.292	0.647	0.061	0.298	0.623	0.079	44.89	1.126	0.991	1.350	0.949	0.974	0.993
0.110	0.154	0.736	0.168	0.141	0.691	48.97	1.865	1.025	1.055	0.963	0.971	0.982
0.766	0.099	0.135	0.684	0.092	0.224	48.17	1.046	1.029	1.877	0.939	0.977	1.005
0.155	0.740	0.105	0.167	0.709	0.124	45.17	1.202	0.991	1.233	0.954	0.973	0.989
0.056	0.066	0.878	0.105	0.068	0.827	47.42	2.236	1.119	1.024	0.971	0.973	0.981
0.894	0.051	0.055	0.828	0.055	0.117	44.88	1.013	1.120	2.262	0.941	0.983	1.014
0.086	0.860	0.054	0.093	0.843	0.064	44.74	1.197	1.004	1.225	0.956	0.973	0.988

Table IV. Experimental Vapor-Liquid Equilibrium Data for the Ternary System Acetonitrile (1)-Methanol (2)-Benzene (3) at 328.15 K

x_1	x_2	x_3	y_1	y_2	y_3	P, kPa	γ_1	γ_2	γ_3	ψ_1	ψ_2	ψ_3
0.397	0.203	0.400	0.262	0.391	0.347	77.75	1.224	2.204	1.557	0.921	0.970	0.989
0.255	0.330	0.415	0.173	0.475	0.352	83.69	1.354	1.765	1.635	0.921	0.966	0.987
0.513	0.287	0.200	0.320	0.439	0.241	76.99	1.140	1.731	2.159	0.917	0.968	0.996
0.209	0.495	0.296	0.147	0.539	0.314	86.69	1.451	1.378	2.122	0.920	0.962	0.989
0.313	0.166	0.521	0.223	0.380	0.397	78.06	1.331	2.632	1.368	0.925	0.971	0.985
0.078	0.434	0.488	0.060	0.548	0.392	88.97	1.640	1.642	1.640	0.926	0.963	0.983
0.497	0.049	0.454	0.408	0.158	0.434	63.93	1.265	3.073	1.412	0.931	0.982	0.989
0.250	0.654	0.096	0.198	0.634	0.168	80.83	1.524	1.144	3.304	0.920	0.962	1.000
0.118	0.193	0.689	0.095	0.464	0.441	83.90	1.624	2.962	1.230	0.929	0.968	0.981
0.669	0.093	0.238	0.473	0.233	0.294	64.27	1.089	2.395	1.850	0.925	0.980	0.997
0.058	0.644	0.298	0.050	0.602	0.348	90.21	1.861	1.229	2.424	0.925	0.961	0.986
0.671	0.275	0.054	0.449	0.455	0.096	67.30	1.074	1.645	2.819	0.921	0.974	1.009
0.101	0.765	0.134	0.093	0.669	0.238	84.69	1.865	1.079	3.492	0.924	0.960	0.995
0.767	0.140	0.093	0.539	0.307	0.154	61.67	1.038	2.011	2.402	0.925	0.980	1.007
0.893	0.042	0.065	0.739	0.128	0.133	51.01	1.021	2.338	2.469	0.934	0.991	1.012
0.501	0.056	0.443	0.402	0.175	0.423	64.23	1.242	2.991	1.418	0.931	0.982	0.989
0.053	0.899	0.048	0.064	0.811	0.125	78.82	2.294	1.036	4.813	0.931	0.960	1.004

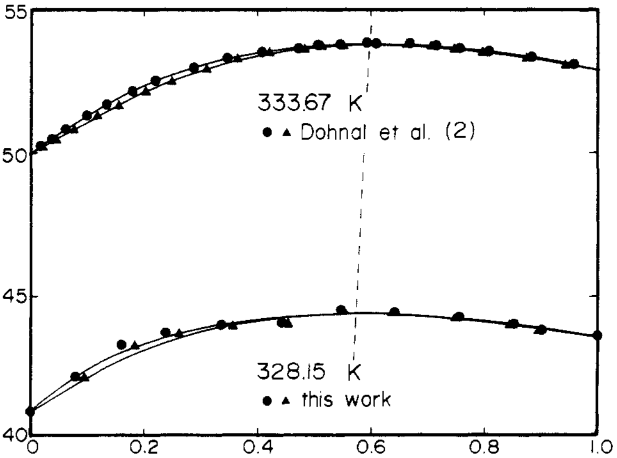

Figure 1. Comparison with published data for 2-butanone (1)-acetonitrile (2) system: (●, ▲) Experimental data. (—) Calculated by the Wilson equation; 333.67 K (ref *2*); 328.15 K (this work). (- - -) Locus of azeotropes.

Table V. Azeotropic Data for Binary Systems at 328.15 K

system	x_1	P, kPa
2-butanone (1)-acetonitrile (2)	0.574	44.43
2-butanone (1)-benzene (2)	0.435	46.70
acetonitrile (1)-benzene (2)	0.456	54.65

present experimental data. The absolute arithmetic mean deviations between the experimental and calculated vapor compositions for the three binary systems at 328.15 K are as follows: 2-butanone-acetonitrile, 0.003; 2-butanone-benzene, 0.006; acetonitrile-benzene, 0.008. These results suggest that the experimental data are thermodynamically consistent.

Azeotropic compositions and pressures obtained by interpolation of experimental values are listed in Table V.

A computer program used for binary parameter estimation is similar to that in detail described by Prausnitz et al. (*10*), which is based on the maximum likelihood principle. Table VI lists binary parameters together with the estimated variance of the fit. The binary parameters, a_{12} and a_{21}, obtained from the Wilson and UNIQUAC equations differ, not only in magnitudes but sometimes even in signs. These do not render contradictory results. There are many similar cases described by Gmehling et al. (*19-27*).

***Ternary Systems*.** Table VII presents the predicted results for the two ternary systems. Calculated values by the Wilson

Table VI. Binary Parameters of Wilson and UNIQUAC (Anderson and Prausnitz's Modification) Equations and Variance of Fit at 328.15 K

system	no. of data points	eq	a_{12}, K	a_{21}, K	σ_F^2	data source
2-butanone (1)-acetonitrile (2)	10	Wilson	39.84	99.54	2.07	this work
		UNIQUAC	−0.05	56.53	2.10	
2-butanone (1)-benzene (2)	9	Wilson	−91.52	226.55	6.68	this work
		UNIQUAC	−151.28	215.57	6.06	
acetonitrile (1)-benzene (2)	12	Wilson	−22.66	426.75	10.65	this work
		UNIQUAC	−65.18	279.09	11.87	
methanol (1)-acetonitrile (2)	12	Wilson	114.47	245.86	4.93	*8*
		UNIQUAC	−19.32	341.04	5.19	
methanol (1)-benzene (2)	9	Wilson	88.42	924.69	4.82	*9*
		UNIQUAC	−62.35	980.24	5.20	

Table VII. Comparison of Prediction of Ternary Vapor-Liquid Equilibria for Two Systems at 328.15 K with Wilson and UNIQUAC (Anderson and Prausnitz's Modification) Equations and UNIFAC Group-Contribution Method

system	no. of data points	root-mean-square deviation in vapor mole fraction			root-mean-square deviation in press., kPa		
		Wilson	UNIQUAC	UNIFAC	Wilson	UNIQUAC	UNIFAC
acetonitrile (1)-2-butanone	25	0.005	0.004	0.008	0.34	0.29	0.59
(2)-benzene (3)		0.003	0.003	0.005			
		0.006	0.005	0.011			
acetonitrile (1)-methanol	17	0.006	0.005	0.004	0.46	0.56	1.58
(2)-benzene (3)		0.007	0.006	0.010			
		0.005	0.005	0.010			

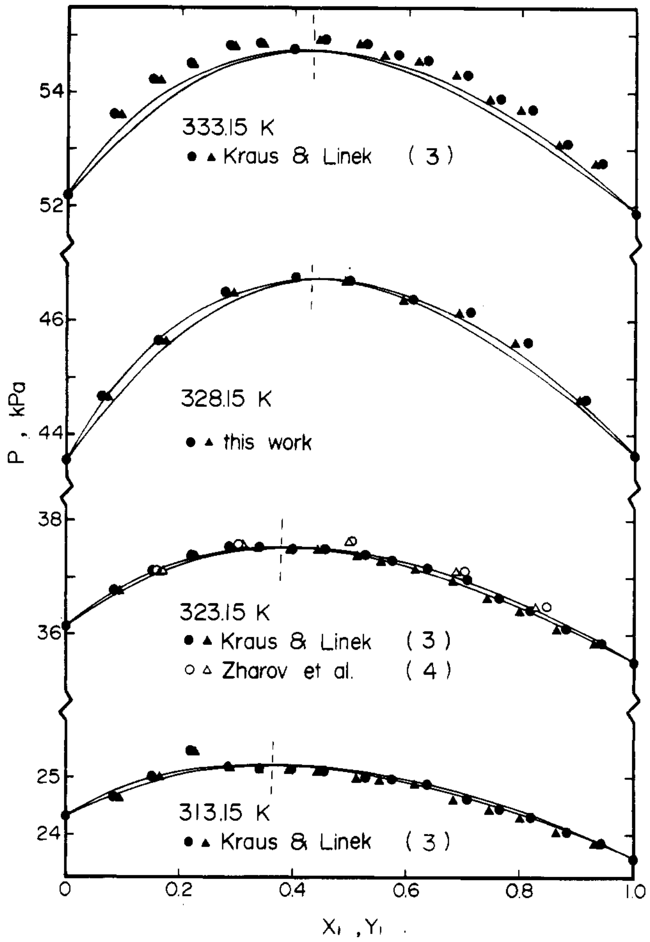

Figure 2. Comparison with published data for 2-butanone (1)-benzene (2) system: (●, ▲, ○, △) Experimental data. (—) Calculated by the Wilson equation; 333.15, 323.15, and 313.15 K (ref *22*); 328.15 K (this work). (- - -) Locus of azeotropes.

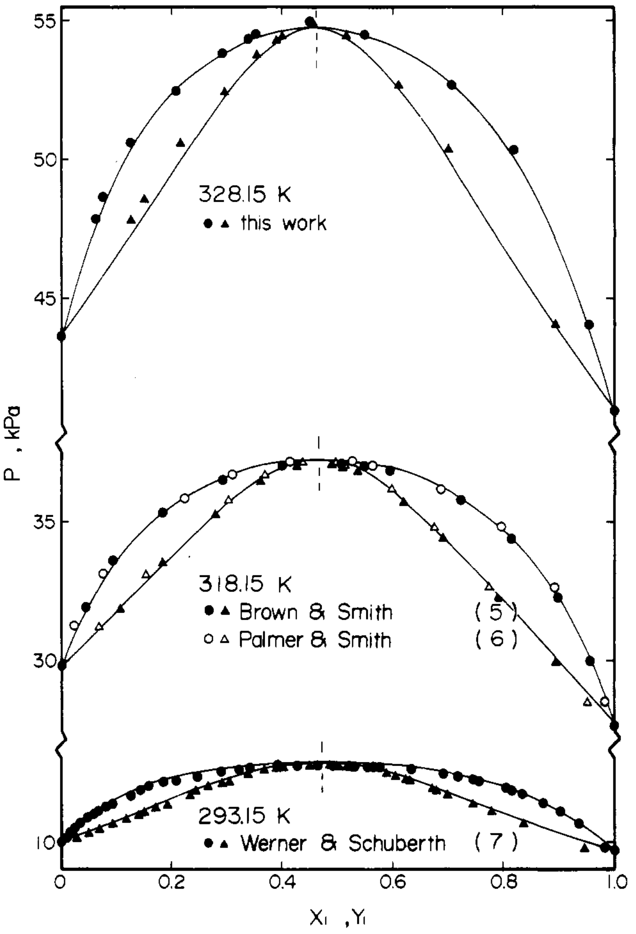

Figure 3. Comparison with published data for acetonitrile (1)-benzene (2) system: (●, ▲, ○, △) Experimental data. (—) Calculated by the Wilson equation; 318.15 and 293.15 K (ref *25*); 328.15 K (this work). (- - -) Locus of azeotropes.

and UNIQUAC equations are close to the experimental data. The table also shows that the UNIFAC method whose parameters are obtained from Skjold-Jørgensen et al. (*13*) works with good accuracy for the systems.

Acknowledgment

We are grateful to T. Murakami and H. Maki for their help in the experimental work and to the Data Processing Center, Kanazawa University, for the use of its facilities.

Glossary

a_{ij}	binary interaction parameter, K
P	total pressure, kPa
P_i^s	vapor pressure of pure component i, kPa
q_i	molecular-geometric area parameter for component i
q_i'	molecular-interaction area parameter for component i
r_i	molecular volume parameter for pure component i
R	gas constant, 8.314 J/(mol K)
T	absolute temperature, K
v_i^L	liquid molar volume of pure component i, cm^3/mol
x_i	liquid-phase mole fraction of component i
y_i	vapor-phase mole fraction of component i
Z	lattice coordination number, a constant set equal to 10

Greek Letters

γ_i	activity coefficient of component i
θ_i	area fraction defined by eq 7
θ_i'	area fraction defined by eq 8
Λ_{ij}	Wilson binary parameter
$\Lambda\lambda_{ij}$	Wilson binary interaction parameter, J/mol
Δu_{ij}	UNIQUAC binary interaction parameter, J/mol
σ_F^2	variance of fit
τ_{ij}	UNIQUAC binary parameter
ϕ_i	volume fraction defined by eq 6
ψ_i	fugacity coefficient of component i
ψ_i^s	fugacity coefficient of pure component i at system temperature and P_i^s

Subscript

i	component

Superscript

s	saturation

Registry No. Acetonitrile, 75-05-8; 2-butanone, 78-93-3; benzene, 71-43-2; methanol, 67-56-1.

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Ultrasonic Velocity in Binary Mixtures of Alkylbenzenes at 25 °C

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Density and ultrasonic velocity values in the frequency range 2.5-15 MHz at 25 °C were measured for binary mixtures of ethylbenzene, *o*-xylene, *m*-xylene, and *p*-xylene. Our data were discussed and compared, where possible, with those found in the literature.

Introduction

Ultrasonic velocity data are of increasing interest in physical chemistry because, as described in the literature (*1-7*), they are the basis of structural studies of liquids. In fact, quantities such as internal pressure and adiabatic compressibility depend on the liquid structure, the extent of molecular interaction (*8*), and the intermolecular free path (*5*) and they may be calculated by using ultrasonic velocity data (*9*). Compressibility is one of the fundamental measurements for calculating reduction parameters in the state equation according to Flory statistics (*1*, *7*, *10*, *11*). In the course of physicochemical investigations of organic systems being carried out in this department (*1*, *12*, *13*), we measured the ultrasonic velocities of mixtures of alkylbenzenes, for use in successive physicochemical studies.

Experimental Section

The interferometric method employed for the measurement of ultrasonic velocity is described elsewhere (*3*, *4*). The

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