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Vapor–Liquid Equilibrium Data for the Systems 2-Methoxyethanol–Ethylbenzene, 2-Methoxyethanol–*p*-Xylene, and 2-Ethoxyethanol–*p*-Xylene

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Experimental vapor–liquid equilibrium data were obtained for the binary systems 2-methoxyethanol–ethylbenzene, 2-methoxyethanol–*p*-xylene, and 2-ethoxyethanol–*p*-xylene at 760 mmHg. The activity coefficients were calculated taking into account the nonideality of the vapor phase. The data satisfied the thermodynamic consistency tests of Herington, Norrish-Twigg, and Black. The correlations of van Laar, Redlich-Kister, Wilson, NRTL, and Wiehe-Bagley, with the constants evaluated by the method of nonlinear least squares, represented the data well.

Isobaric vapor–liquid equilibrium data for the binary systems 2-methoxyethanol–ethylbenzene, 2-methoxyethanol–*p*-xylene, and 2-ethoxyethanol–*p*-xylene were obtained at 760 ± 2 mmHg pressure using a vapor recirculating type of equilibrium still. All these systems formed minimum boiling azeotropes. The azeotropic conditions for 2-methoxyethanol–ethylbenzene and 2-ethoxyethanol–*p*-xylene systems are 118.8 °C, 61.5 mole % 2-methoxyethanol, and 128.5 °C, 51.5 mole % 2-ethoxyethanol, respectively, whereas the corresponding literature values (*11*) are 117 °C, 59.3 mole % 2-methoxyethanol, and 128.6 °C, 54.1 mole % 2-ethoxyethanol, respectively. The system 2-methoxyethanol–*p*-xylene has an azeotropic temperature of 119.3 °C at 64.0 mole % 2-methoxyethanol. Garber and Bovkun (*7*) determined the azeotropic temperature for this system as 119.5 °C at 63 mole % 2-methoxyethanol at 750 mmHg, using a distillation column. Vapor–liquid equilibrium data for the system 2-methoxyethanol–ethylbenzene at 62 mmHg were reported by Jakubiček et al. (*12*). Garber and Bovkun (*8*) have reported vapor–liquid equilibrium data for the systems 2-methoxyethanol–*p*-xylene and 2-ethoxyethanol–*p*-xylene at 60 mmHg pressure.

Experimental Section

Materials. Ethylbenzene obtained from British Drug Houses and *p*-xylene from Riedel were dried by contacting them overnight with pure anhydrous calcium chloride whereas 2-methoxyethanol and 2-ethoxyethanol obtained from Riedel were dried by refluxing for about 4 h with anhydrous potassium carbonate. These were further purified by distillation in a well-insulated 30-mm glass column packed to a height of 1 m with 4 mm helices made from nichrome wire. The heart cuts were collected by maintaining an approximate reflux ratio of 9:1 using a reflux head described by Miller (*16*).

Apparatus. The vapor recirculating type equilibrium still used, incorporates the salient features of the stills reported by Jones (*13*) and Raju et al. (*24*). Figure 1 gives details of the apparatus which essentially consists of the boiler (B), condensate receiver (C), and vaporizer (V). The vapors from the boiler pass through the entrainment trap (E) and then bubble through the condensate in (C) providing thorough mixing of the condensate before they are totally condensed by the condenser (W). The condensate overflows through the capillary tubing (T) into the total vaporizer (V) from where the vapors bubble through the boiling liquid in (B) providing thorough mixing of the liquid in boiler. Double-walled thermometer pockets (P_1 and P_2) were introduced to avoid surface heating of mercury from the neighboring heater elements. The heaters (H_1, H_2, and H_3) having wattages of 125, 125, and 250, respectively, are controlled separately by using a combination of "Sunvic" energy regulators and autotransformers. The still has a capacity of about 100 mL of liquid. The system pressure was kept constant at 760 ± 2 mmHg with the help of an air blower. The fluctuations in the manometer readings were minimized by using two empty vessels with a total capacity of 40 L in the circuit between the blower and the manometer. The air used for maintaining the pressure was dried by passing first through sulfuric acid bubblers and then through U-tubes containing anhydrous calcium chloride. Temperatures were measured with the help of short range "Anschutz" thermometers having an accuracy of 0.1 °C. Stem corrections were applied to the observed thermometer readings. Care was taken to avoid any refluxing in the vapor space above the boiler (B) by keeping the temperature of the vapors slightly higher than the boiler temperatures by means of the heater (H_2). Also superheating of vapors in the vaporizer (V) was minimized by observing, at frequent intervals, wetting of the vaporizer outlet tube with the help of a magnifying lens. The circulation was continued for 3 h so as to ensure complete equilibrium between the vapor and liquid phases. About 7 mL each of the samples of the liquid in the boiler and the condensate receiver was collected through stopcocks K_1 and K_2, respectively, after flushing the stagnant liquid collected just above the stopcocks. Narrow-necked chilled sampling bottles, provided with standard ground joint stoppers,

Table I. Refractive Index-Composition Data at 30 °C

2-Methoxyethanol–ethylbenzene		2-Methoxyethanol–*p*-xylene		2-Ethoxyethanol–*p*-xylene	
Mole fraction, 2-methoxyethanol	Refractive index	Mole fraction, 2-methoxyethanol	Refractive index	Mole fraction, 2-ethoxyethanol	Refractive index
0.0000	1.4908	0.0000	1.4908	0.0000	1.4908
0.0709	1.4859	0.0808	1.4854	0.0658	1.4856
0.1473	1.4808	0.1467	1.4807	0.1192	1.4815
0.2183	1.4754	0.2157	1.4758	0.1795	1.4769
0.2802	1.4708	0.2889	1.4706	0.2447	1.4720
0.4035	1.4613	0.3454	1.4663	0.2949	1.4682
0.4168	1.4602	0.4052	1.4616	0.3536	1.4636
0.4531	1.4571	0.4606	1.4571	0.4033	1.4596
0.5082	1.4524	0.5053	1.4533	0.4639	1.4545
0.5674	1.4471	0.5613	1.4483	0.5125	1.4506
0.6088	1.4431	0.6073	1.4438	0.5537	1.4470
0.6565	1.4385	0.6551	1.4392	0.6085	1.4422
0.6996	1.4342	0.7003	1.4348	0.6549	1.4381
0.7449	1.4293	0.7488	1.4296	0.7098	1.4328
0.7838	1.4251	0.7838	1.4256	0.7474	1.4295
0.8243	1.4206	0.8235	1.4212	0.7952	1.4251
0.8594	1.4165	0.8655	1.4160	0.8368	1.4209
0.9004	1.4114	0.9009	1.4121	0.8763	1.4169
0.9308	1.4076	0.9343	1.4076	0.9233	1.4121
0.9689	1.4026	0.9678	1.4031	0.9597	1.4084
1.0000	1.3985	1.0000	1.3985	1.0000	1.4042

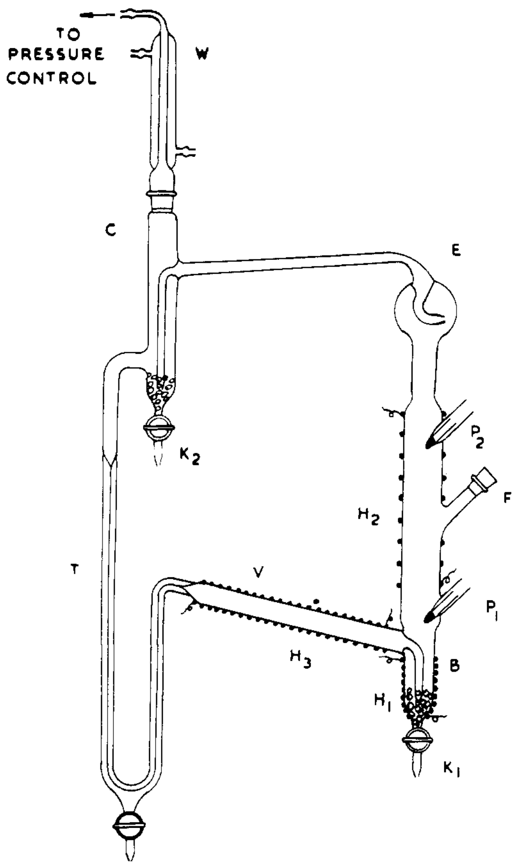

Figure 1. Equilibrium still: B, boiler; C, condensate receiver; E, entrainment trap; F, feed inlet; H_1, H_2, H_3, heating cords; K_1, K_2, sampling cocks; P_1, P_2, thermometer pockets; T, capillary tube; V, vaporizer; W, condenser.

were used for collecting the samples. The equilibrated samples were analyzed by the refractive index method using a Bausch and Lomb Abbe-3L refractometer having an accuracy of ±0.0002 in refractive index readings. The analyses were carried out at 30 ± 0.1 °C maintaining the prism temperatures using a "Precision Scientific" water circulating bath. Table I gives the refractive index-composition data for the three systems.

Results and Discussion

The activity coefficients were calculated using eq 1 and 2.

$$\gamma_1 = \frac{\pi y_1}{P_1{}^0 x_1} \exp\left[\frac{(\pi - P_1{}^0)(B_{11} - V_1)}{RT} + \frac{\pi \delta_{12} y_2{}^2}{RT}\right] \qquad (1)$$

$$\gamma_2 = \frac{\pi y_2}{P_2{}^0 x_2} \exp\left[\frac{(\pi - P_2{}^0)(B_{22} - V_2)}{RT} + \frac{\pi \delta_{12} y_1{}^2}{RT}\right] \qquad (2)$$

where

$$\delta_{12} = 2B_{12} - B_{11} - B_{22} \qquad (3)$$

Table II gives the vapor–liquid equilibrium data for the three systems 2-methoxyethanol-ethylbenzene, 2-methoxyethanol–*p*-xylene, and 2-ethoxyethanol–*p*-xylene. The pure component properties were estimated by carefully selected correlations from literature (*2, 17*). The vapor pressures and second virial coefficients for ethylbenzene and *p*-xylene were estimated by the correlations of RPME (*14, 15*) and Pitzer-Curl (*23*), respectively. The vapor pressures of 2-methoxyethanol and 2-ethoxyethanol were estimated by Antoine equation (*1*) with the constants taken from Gallaugher and Herbert (*6*) and Pick et al. (*22*), respectively, whereas for second virial coefficients the method of O'Connell and Prausnitz (*20, 21*) was used. In the absence of experimental data on virial coefficients for 2-methoxyethanol and 2-ethoxyethanol, the values for the association constant, η, in the O'Connell–Prausnitz correlation were assumed to be 0.55 and 0.50, respectively. The values of homomorph acentric factors, ω_H, were taken as 0.252 and 0.290, respectively. The cross virial coefficients, B_{12}, for the binary mixtures were evaluated by employing the mixing rules suggested by O'Connell and Prausnitz (*20*). The liquid molar volumes for all the components were estimated by the surface tension correlation (*9*). Thermodynamic consistency tests of Herington (*10*), Black (*3, 4*) and Norrish–Twigg (*19*) were well satisfied by the data for all three systems. No attempt was made to estimate the heat of mixing term in the Gibbs–Duhem equation. Watson's correlation (*28*) was used for estimating the heats of vaporization at system temperatures required in the Norrish–Twigg test. The activity coefficient–composition data above 0.1 mole fraction 2-methoxy- and 2-ethoxyethanol are well represented by the correlations of van Laar (*5*), Redlich–Kister (*25, 26*), Wilson (*30*), NRTL (*27*), and Wiehe–Bagley (*29*). The data in the dilute region

Table II. Vapor-Liquid Equilibrium Data at 760 ± 2 mmHg

t, °C	x_1	y_1	γ_1	γ_2	t, °C	x_1	y_1	γ_1	γ_2
System 2-Methoxyethanol (1)-Ethylbenzene (2)									
124.4	1.000	1.000	—	—	119.1	0.522	0.574	1.291	1.432
123.2	0.966	0.940	1.005	2.567	119.3	0.476	0.556	1.366	1.348
122.7	0.962	0.924	1.008	2.888	119.6	0.441	0.538	1.414	1.303
122.1	0.936	0.890	1.017	2.549	120.1	0.359	0.496	1.573	1.224
120.6	0.862	0.794	1.031	2.303	121.2	0.284	0.460	1.790	1.138
119.7	0.815	0.744	1.052	2.185	122.2	0.228	0.426	1.995	1.092
119.5	0.776	0.714	1.067	2.027	123.5	0.177	0.372	2.157	1.076
119.2	0.739	0.688	1.092	1.916	125.0	0.140	0.314	2.205	1.080
119.1	0.714	0.670	1.102	1.855	126.5	0.110	0.281	2.415	1.048
118.9	0.674	0.648	1.140	1.743	127.5	0.092	0.236	2.330	1.062
118.7	0.676	0.648	1.141	1.764	128.9	0.074	0.190	2.263	1.061
118.9	0.644	0.634	1.166	1.661	130.8	0.049	0.132	2.236	1.054
118.8	0.648	0.640	1.172	1.659	132.9	0.028	0.082	2.284	1.029
119.0	0.634	0.627	1.166	1.642	134.7	0.014	0.046	2.500	1.007
119.0	0.596	0.606	1.200	1.569	136.2	0.000	0.000	—	—
System 2-Methoxyethanol (1)-*p*-Xylene (2)									
124.4	1.000	1.000	—	—	120.6	0.399	0.550	1.552	1.209
122.5	0.942	0.900	1.010	2.633	121.6	0.319	0.514	1.755	1.122
121.0	0.876	0.817	1.032	2.365	122.8	0.249	0.467	1.973	1.077
120.9	0.877	0.814	1.029	2.436	124.4	0.190	0.420	2.211	1.039
120.3	0.834	0.774	1.049	2.236	126.0	0.147	0.347	2.254	1.062
120.2	0.819	0.761	1.055	2.163	126.2	0.146	0.338	2.198	1.068
119.7	0.758	0.710	1.082	1.982	128.2	0.108	0.286	2.364	1.046
119.5	0.696	0.672	/2	1.797	130.4	0.076	0.216	2.375	1.045
119.4	0.664	0.654	1.146	1.721	132.1	0.059	0.172	2.337	1.036
119.3	0.602	0.626	1.214	1.581	134.3	0.038	0.113	2.261	1.024
119.6	0.558	0.608	1.263	1.472	136.4	0.020	0.056	1.916	1.016
119.9	0.490	0.582	1.363	1.350	138.5	0.000	0.000	—	—
System 2-Ethoxyethanol (1)-*p*-Xylene (2)									
135.1	1.000	1.000	—	—	128.6	0.460	0.481	1.268	1.241
134.1	0.968	0.943	0.997	2.002	128.8	0.391	0.440	1.359	1.180
132.8	0.916	0.866	1.007	1.861	129.4	0.306	0.394	1.526	1.104
131.4	0.859	0.788	1.020	1.811	130.4	0.222	0.330	1.717	1.059
130.5	0.812	0.734	1.034	1.738	131.5	0.161	0.273	1.886	1.036
129.9	0.760	0.681	1.046	1.661	132.7	0.120	0.225	2.023	1.021
129.2	0.689	0.628	1.086	1.522	133.7	0.088	0.170	2.022	1.026
128.7	0.625	0.580	1.123	1.446	135.0	0.062	0.126	2.051	1.016
128.6	0.592	0.559	1.146	1.399	136.0	0.044	0.094	2.041	1.008
128.6	0.556	0.538	1.176	1.345	137.2	0.024	0.048	1.935	1.004
128.6	0.526	0.522	1.205	1.309	138.5	0.000	0.000	—	—
128.6	0.505	0.505	1.216	1.287					

Table III. Data Fit in Different Correlations

	2-Methoxyethanol (1)-ethylbenzene (2)		2-Methoxyethanol (1)-*p*-xylene (2)		2-Ethoxyethanol (1)-*p*-xylene (2)	
Correlation	Constants[a]	RMS deviation in y_1	Constants[a]	RMS deviation in y_1	Constants[a]	RMS deviation in y_1
van Laar	A_{12} 1.1962		1.1439		1.1779	
	A_{21} 1.0436	0.013	1.1147	0.013	1.2318	0.009
Redlich-Kister	B 1.1144		1.1169		1.2044	
	C −0.0728		0.0231		0.0262	
	D −0.0016	0.013	−0.1376	0.014	0.0023	0.008
Wilson	A_{12} 0.4075		0.4800		0.4813	
	A_{21} 0.6254	0.013	0.5354	0.014	0.4766	0.009
NRTL	A_{12} 0.4839		0.6248		0.7554	
	A_{21} 0.8575	0.013	0.7157	0.014	0.7059	0.009
Wiehe-Bagley	K 7.9213		7.9739		9.5845	
	p 1.7336	0.014	2.0852	0.015	2.3403	0.008

[a] Constants obtained by minimizing ln (γ_1/ν_2) function.

were not included in the evaluation of the parameters in view of the likely inconsistencies in composition measurements. The value of constant, α, in the NRTL correlation was taken to be 0.47. The nonlinear least-squares method, as illustrated by Nagahama et al. (*18*), was used by minimizing ln (γ_1/γ_2) function to evaluate the constants in these correlations. The root mean square deviations in vapor composition along with the constants in the correlating equations are given in Table III.

Glossary

B	gas phase second virial coefficient, cm^3 mol^{-1}
P^0	pure component vapor pressure, mmHg
R	gas constant
T	absolute temperature, K
t	system temperature, °C
V	pure component liquid molar volume, cm^3 mol^{-1}
x	mole fraction in liquid phase
y	mole fraction in gas phase
γ	activity coefficient
π	system pressure, mmHg

Subscripts

1	more volatile component
2	less volatile component

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Excess Enthalpy, Volume, and Gibbs Free Energy and Viscosity of Ethyl Acetate–Methyl Cellosolve Mixtures

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Molar excess enthalpies of mixing at 48.2 °C are measured for the ethyl acetate–methyl cellosolve system using an isothermal phase change calorimeter. The molar excess volume is computed from density measurements at 40 °C. The molar excess Gibbs free energies are calculated from isothermal vapor–liquid equilibrium data obtained in a circulation still at 70, 80, and 90 °C. Complete isobaric vapor–liquid equilibrium data at 1 atm pressure are also reported. The activity coefficient data are corrected for vapor phase nonideality, tested for thermodynamic consistency, and correlated by the Wilson equation. The kinematic viscosity of binary mixtures is measured at 40, 50, 60, and 70 °C using an Ostwald viscometer. The viscosity data are correlated by McAllister and excess function models.

The present investigation forms part of a continuing study of excess thermodynamic properties of binary liquid mixtures consisting of methyl cellosolve as one of the components.

Experimental Procedures

Materials. Analytical grade ethyl acetate and methyl cellosolve from British Drug House Co of India were dried over silica and further purified by distillation in a 35-mm glass column packed with 3-mm ceramic insulation beads to a height of 1 m. The column was run at total reflux for 30 min, and the low boiling impurities were drawn off at a very slow rate of rejects. The overhead products were analyzed at various intervals by density and refractive index measurements and the "heart cut" of desired purity was collected for further investigations. The pure component boiling points reported here were obtained in the equilibrium still used in this study. Table I lists the physical properties of the chemicals used.

Apparatus. Calorimetric Measurements. An isothermal phase change calorimeter similar to one described by Dainton (*4*) and further modified by Lakhanpal (*13*) and calibrated in our laboratories by Kalasy (*10*) was used for excess enthalpy measurements at 48.2 °C. The change in the volume of a dilatometric fluid (cetyl alcohol) on phase transformation is a measure of heat change in the mixing process. The conventional bulb breaking method was adopted for mixing the components. The calorimeter was thermostated at 48.2 ± 0.002 °C and the accuracy of any individual measurement of heat change was better than ±0.02 cal mol^{-1}.

Vapor-Liquid Equilibria. The Othmer type of recirculation still (*25*) modified by Mainkar and Mene (*16*) was used to measure the equilibrium data. The still has three separate heating units for the equilibrium chamber to allow for a close approach to adiabatic conditions. For isobaric data the pressure in the still was controlled to 760 ± 1 mmHg using dried air. Two surge tanks between the still and the manostat served to damp out the pressure fluctuations. The data were measured with air present in the condensate returning to the equilibrium still. For isothermal VLE data the still was filled with 230 mL of mixture and heated to boiling. The pressure in the manostat was adjusted for the desired isothermal conditions. After the recirculation had stabilized the still was operated for 2–3 h. The pressure was controlled by a needle valve and indicated by a mercury manometer