Total Pressure Measurements for Copper(II) Chloride + Methanol + Ethanol at 303.15 K

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Total pressure is reported as a function of liquid-phase composition for copper(II) chloride + methanol + ethanol at 303.15 K. Pressure was measured as a function of salt concentration along various lines of essentially constant methanol mole fraction (on a salt-free basis). The data were reduced by Barker's method using an empirical expression for the excess Gibbs free energy of the liquid phase. Calculated vapor-phase compositions for the two alcohols indicate that the vapor-phase mole fraction of ethanol is increased (ethanol is salted out) by addition of copper(II) chloride to liquid mixtures of methanol and ethanol.

Introduction

Thermodynamic data for systems comprised of a salt and two or more solvents are of interest because the relative volatilities of the solvent components are changed upon addition of the salt. Hence, salts may be used as separating agents in extractive or azeotropic distillation (Furter, 1976).

The correlation and prediction of vapor–liquid behavior for systems of salts in mixed solvents have been examined by a number of investigators (for example, Hala, 1983; Bekerman and Tassios, 1976; Engles, 1987; Sander et al., 1986). However, the base of experimental data for developing and testing the correlations is small. While many data exist for systems of salt and water, far fewer are available for salts in solvents other than water. Data for salts in mixed solvents are even more scarce.

Furthermore, most of the data are reported for isobaric, rather than isothermal, conditions. For salts in single solvents, this is probably of little consequence since the boiling temperature does not change drastically upon addition of salt. However, for systems of salts and two solvents, the temperatures for isobaric data may differ widely and data reduction is complicated unless it is assumed that the activity coefficients of the species in solution are independent of temperature or unless an explicit temperature dependence is incorporated in the solution model used for data reduction. No such difficulty exists for isothermal data, for which it is (at low to moderate pressure) an excellent assumption to neglect the pressure dependence of the activity coefficients.

In order to add to the data base for systems comprised of a salt in two or more solvents, and also to that for systems of a salt in a single nonaqueous solvent, results are reported here, in the form of total pressure data, for the copper(II) chloride + methanol + ethanol system at 303.15 K.

Experimental Section

Apparatus. The apparatus is a modified version of that used earlier (Bhethanabotla and Campbell, 1991; Thomas et al., 1991) to make total pressure measurements for nonelectrolyte systems. A schematic diagram is shown in Figure 1. The apparatus consists of two glass cells—an equilibrium cell (B) and a reference cell (C) which are connected to either side of a differential pressure transducer (E). The transducer (Omega high linearity wet/wet, PX 820 series) has a range of 0 to 34 kPa in differential pressure.

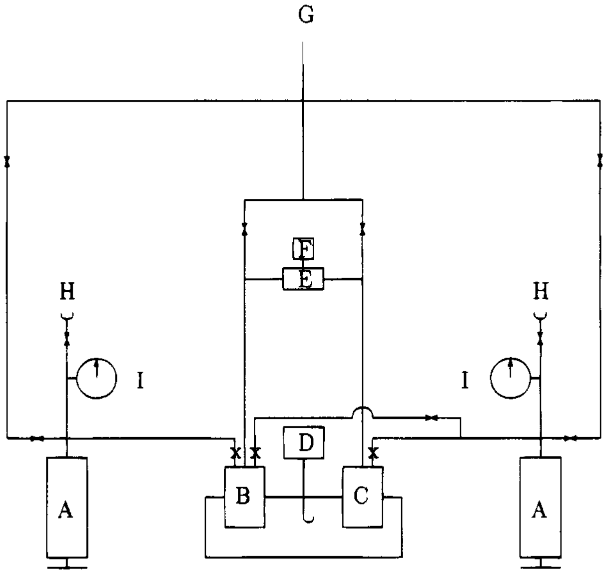

Figure 1. Schematic of apparatus: (A) piston injector, (B) equilibrium cell, (C) reference cell, (D) heater circulator, (E) differential pressure transducer, (F) digital strain indicator, (G) vacuum pump, (H) fill tube, (I) Bourdon gauge.

Both cells are suspended in a constant-temperature bath which is maintained at 303.15 K by a temperature controller (D) (Fisher Model 7300). Solvents are charged to the equilibrium and reference cells from a piston injector (A), which was manufactured by Ruska Instrument Corp. (Model 2200). Only the piston injector on the right side of Figure 1 was used in the present study. The piston injector has a fill tube (H) through which degassed solvents are charged to the apparatus and a Bourdon pressure gauge (I), which is used to maintain a consistent pressure in the piston injector. The entire apparatus is evacuated through a vacuum port (G). A close-up view of the reference cell and equilibrium cell is shown in Figure 2, where it may be seen that the contents of each cell are agitated by a submersible magnetic stirrer (Troemner Inc., Model 700 HV Series 1).

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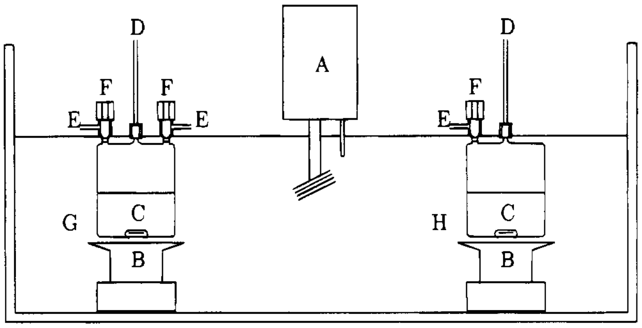

Figure 2. Equilibrium cell and reference cell.(A) heater circulator, (B) submersible stirrer, (C) stirringbar, (D) line to pressure transducer, (E) line to piston-injector, (F) Teflon valves, (G) equilibrium cell, (H) reference cell.

The pressure transducer and all lines between it and the two glass cells are wrapped with heating tape and maintained at 333.15 K by an Omega CN 91210 temperature controller. The heating tape prevents condensation of the solvent vapors in the lines and also provides a stable temperature environment for the pressure transducer.

Materials. Methanol (HPLC grade) and copper(II) chloride were obtained from Aldrich Chemical Co. and had purities of 99.9+ and 99.999% by mass, respectively. Absolute ethanol was obtained from Aaper Alcohol and had a purity of 99.9+% by mass. Potassium chloride, lithium chloride, and water were used in testing the apparatus. The potassium chloride and lithium chloride had purities of 99.98 and 99.99%, respectively, by mass and were obtained from Aldrich. The water was distilled and deionized. All solvents were used as is except that they were degassed as described by Bhethanabotla and Campbell (1991).

Procedure for Binary Salt−Solvent Measurements. For mixtures where the concentration of salt is low, the pressure above a salt + solvent mixture is not much lower than the vapor pressure of the pure solvent. To minimize any effects due to drifting bath temperature, the pressure above the salt + solvent mixture (in the equilibrium cell) is not measured directly but, instead, is measured relative to that above pure solvent in the reference cell. Initially, a known mass of copper(II) chloride is placed in the equilibrium cell and connected to the apparatus. The entire apparatus is then evacuated for approximately 20 h to remove any moisture from the salt. Degassed solvent is charged to the piston injector, and a portion of this is charged to the reference cell. The transducer measures directly the vapor pressure of the solvent in the reference cell since the equilibrium cell at this point contains only the nonvolatile salt. At this point, a measured volume of the solvent in the piston injector is charged to the equilibrium cell, where it is mixed with the salt. An amount of solvent (estimated from solubility data in Janz and Tomkins, 1972) is added so that the mixture is just within the solubility limit of the salt. After equilibrium is reached (15−30 min after solvent addition as evidenced by constancy in pressure), the pressure difference between the reference and equilibrium cells is recorded, and the absolute pressure in the equilibrium cell is calculated from the previously measured solvent vapor pressure. The procedure is repeated by adding additional solvent to the equilibrium cell so as to obtain increments in concentration of approximately 0.5 molal.

Procedure for Ternary Measurements. Ternary measurements are taken along lines of constant salt-free solvent composition. A degassed binary mixture of methanol + ethanol for which the composition is accurately known is prepared using the method described by Bhethanabotla and Campbell (1991). It is charged to the piston injector, and a portion of this is charged to the reference cell, where the total pressure above the methanol + ethanol mixture is measured. The rest of the run proceeds in a manner identical to that described above for the binary measurements. A number of different runs with different solvent compositions are made until the entire range of salt-free methanol + ethanol compositions is covered.

Data Reduction. The raw data consist of mass of salt in the equilibrium cell, volumes of solvent (or solvent mixture for the ternary runs) displaced from the piston injector to the equilibrium cell, and pressure difference between the reference and equilibrium cells. Moles of each solvent species are determined from the volumetric data using liquid densities at room temperature taken from Stephan and Hildwein (1987). Since the liquids in the piston injector are only lightly compressed, saturated liquid densities were used. For ternary runs in which the piston injector contained a methanol + ethanol mixture, the effect of the excess volume on liquid density was accounted for using the mixture molar volumes reported by Stephan and Hildwein (1987).

This information is sufficient to determine pressure as a function of overall composition in the equilibrium cell. Since the density of the vapor phase in the equilibrium cell is low, the overall composition and the composition of the liquid phase are essentially equal. However, corrections are made to determine the exact composition of the liquid phase using a procedure that will be described shortly.

The maximum uncertainty in bath temperature (verified with a mercury thermometer with a calibration traceable to the National Institute of Standards and Technology) is ±0.02 K, although the effect of this uncertainty is minimized by using a reference cell. The pressure transducer was calibrated by connecting it in parallel to a mercury manometer which was read with a cathetometer. The absolute uncertainty in pressure is estimated to be ±0.03 kPa. Uncertainties in overall composition within the equilibrium cell arise due to uncertainties in the mass of salt (less than 0.001 g), liquid densities from the literature (estimated to be less than 0.001 g/cm^3), and piston injector reading (0.001 cm^3 per injection). These uncertainties translate to an uncertainty in overall cell composition of 0.0005 or less in mole fraction for the binary salt + solvent mixtures. For the ternary runs, there is an additional uncertainty in the composition of the solvent mixture (estimated to be 0.0002 in mole fraction), so the uncertainty in overall cell composition is slightly higher than 0.0005 mole fraction.

Corrections from overall cell composition to liquid-phase composition were made using a modification of Barker's method (Barker, 1953). In the standard Barker's method, an expression for the excess Gibbs free energy of the liquid phase is assumed and parameter values in the expression are obtained by regressing pressure versus liquid-phase mole fraction such that the best fit to pressure is obtained. In the present study, the data fed to the regression program consisted of measured pressure versus number of moles of each component inside the equilibrium cell. The algorithm used in data reduction is as follows.

Initially, all mass in the equilibrium cell is assumed to be in the liquid phase, and the best set of parameters in the assumed G^E function are found. The vapor-phase compositions are also calculated in this step. From a knowledge of the volume of the equilibrium cell and the volume of liquid in the equilibrium cell, the moles of each component in the vapor phase may be calculated. The

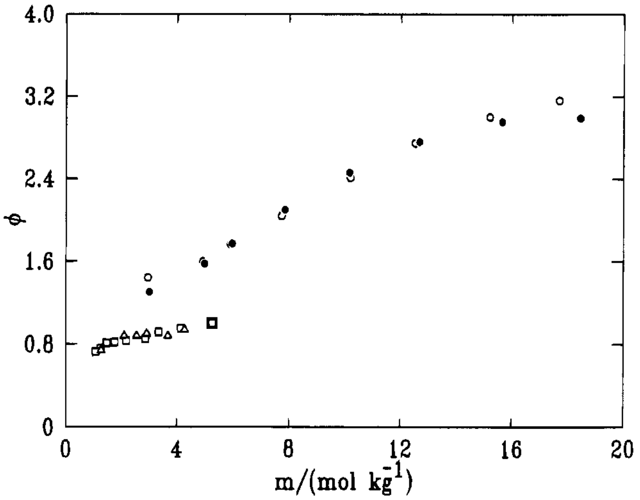

Figure 3. Osmotic coefficient ϕ vs salt molality m for lithium chloride + water at 303.15 K and for potassium chloride + water at 313.15 K: ○, LiCl + H_2O, this study; ●, LiCl + H_2O, Patil et al. (1990); □, KCl + H_2O, this study; △ KCl + H_2O, Patil et al. (1991).

liquid-phase compositions are then corrected and another regression is done. The procedure is repeated until the liquid-phase compositions stabilize. Output from the regression consists of a corrected set of liquid compositions, a calculated vapor composition and pressure for each liquid composition, and a set of parameters in the excess Gibbs free energy model. For the ternary runs, a similar procedure is applied to the reference cell as well since it contains a mixture of methanol and ethanol.

The excess Gibbs free energy model applied here was similar to that used by Oh and Campbell (1992). For the methanol (2) + ethanol (3) binary, the four-suffix Margules equation is used:

$$G^E/RT = x_2x_3(A_{32}x_2 + A_{23}x_3 - \alpha_{23}x_2x_3) \qquad (1)$$

For the copper chloride (1) + methanol (2) binary, a two-suffix Margules equation is combined with the term proposed by Hala (1983):

$$G^E/RT = E_{12}x_1^{3/2}x_2 + A_{12}x_1x_2 \qquad (2)$$

The expression for the copper chloride (1) + ethanol (3) system is

$$G^E/RT = E_{13}x_1^{3/2}x_3 + A_{13}x_1x_3 \qquad (3)$$

For the ternary system it was assumed that

$$G^E/RT = (G^E/RT)_{12} + (G^E/RT)_{13} + (G^E/RT)_{23} - Cx_1x_2x_3 \qquad (4)$$

where $(G^E/RT)_{ij}$ is the excess Gibbs function for the ij binary system and C is a single adjustable ternary parameter. Expressions for the activity coefficients γ_2 and γ_3 of methanol and ethanol, respectively, are obtained from eq 4 in the usual manner. Pressure and vapor-phase composition are calculated for a given liquid composition through

$$P = \gamma_2x_2P_2^{sat}/\Phi_2 + \gamma_3x_3P_3^{sat}/\Phi_3 \qquad (5)$$

$$y_2 = \gamma_2x_2P_2^{sat}/\Phi_2P \qquad (6)$$

where

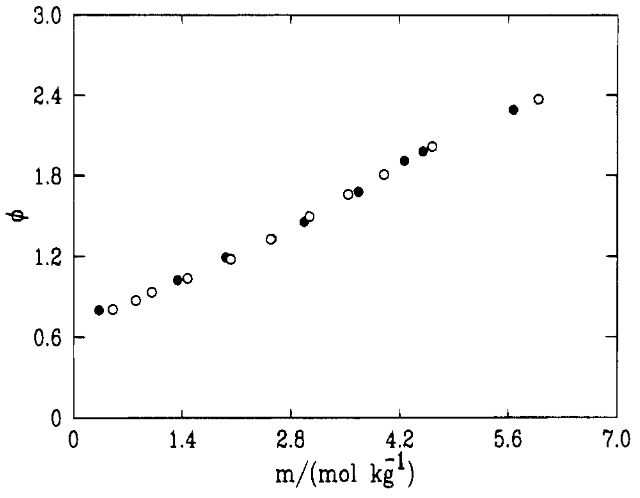

Figure 4. Osmotic coefficient ϕ vs salt molality m for lithium chloride + methanol at 303.15 K: ○, this study; ●, Skabichevskii (1969).

$$\Phi_j = (\phi_j/\phi_j^{sat}) \exp[-V_j^L(P - P_j^{sat})/RT] \qquad (7)$$

In eq 7, ϕ_j is the fugacity coefficient of solvent component j in the vapor phase and ϕ_j^{sat} is the fugacity coefficient of pure solvent component j at the temperature of the mixture and its own vapor pressure. Both are calculated using the two-term virial equation (pressure series). Second virial coefficients were calculated to be −1830, −2658, and −2200 $cm^3 mol^{-1}$ for B_{22}, B_{33}, and B_{23}, respectively, using the method of Tsonopoulos (1974). Saturated liquid volumes V_j^L (40.97 and 58.97 $cm^3 mol^{-1}$ for methanol and ethanol, respectively) for use in the Poytning correction term in eq 7 were obtained from Stephan and Hildwein (1987).

A nonlinear regression program based on Marquardt's method was used for parameter estimation. An extended fit to pressure was used as recommended by Oh and Campbell (1992). In this procedure, binary parameters A_{23}, A_{32}, and α_{23} for the methanol + ethanol system are obtained from an unweighted fit to the methanol + ethanol binary total pressure data. The salt−solvent binary parameters A_{12}, A_{13}, E_{12}, and E_{13}, and the ternary parameter C are then obtained simultaneously by a regression of ternary data and binary data for the two salt + solvent binary systems. Data were weighted by the method of maximum likelihood as described by Oh (1993).

In a previous work (Oh and Campbell, 1992), liquid-phase mole fractions used in the above equations were calculated on the assumption that the salt was completely dissociated. Since then, we have also tested procedures in which analytical mole fractions (mole fractions based on assumption of no dissociation) are used. Tests (Oh, 1993) on literature data do not show a clear advantage of one approach over the other, and both methods yield similar results. For the system presented here the use of analytical mole fractions (no dissociation) yielded slightly lower deviations in pressure, and it is these results that are reported here. Hence, all mole fractions reported in the tables are calculated on this basis. It should be noted that when dissociation is not considered, the first term in eqs 2 and 3, which Hala designed to model the Debye−Hückel equation, has no theoretical significance and is simply an empirical term.

Results

As a test of the apparatus and procedure, three binary salt + solvent systems (potassium chloride + water at

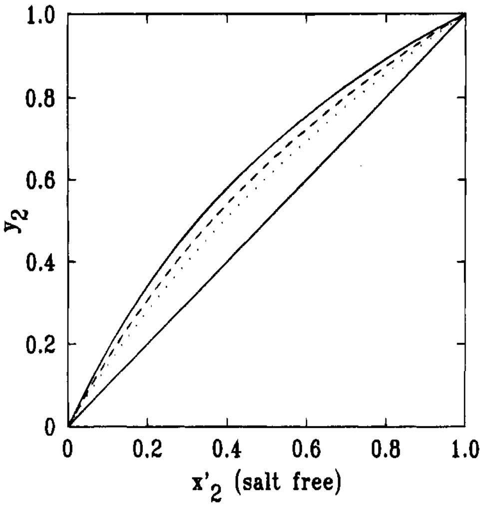

Figure 5. Calculated vapor-phase mole fraction y_2 vs liquid-phase mole fraction (on salt-free basis) x_2' for copper(II) chloride (1) + methanol (2) + ethanol (3) at 303.15 K. The solid line is for x_1 = 0, the dashed line is for x_1 = 0.05, and the dotted line is for x_1 = 0.10.

Table 1. Total Pressure P as a Function of Liquid-Phase Mole Fraction x_i for Copper(II) Chloride (1) + Methanol (2) and Copper(II) Chloride (1) + Ethanol (3) at 303.15 K

copper(II) chloride (1) + methanol (2)		copper(II) chloride (1) + ethanol (3)	
x_2	P/kPa	x_3	P/kPa
0.8745	16.988	0.8304	8.016
0.8828	17.488	0.8412	8.254
0.8966	18.240	0.8580	8.579
0.9103	18.922	0.8767	8.909
0.9242	19.563	0.8954	9.222
0.9388	20.161	0.9220	9.592
0.9536	20.695	0.9348	9.778
0.9687	21.167	0.9557	10.008
0.9842	21.625	0.9774	10.240
1.0000	21.978	1.0000	10.480

Table 2. Total Pressure P as a Function of Liquid-Phase Mole Fractions x_2 and x_3 for Copper(II) Chloride (1) + Methanol (2) + Ethanol (3) at 303.15 K

x_2	x_3	P/kPa	x_2	x_3	P/kPa
0.0803	0.7547	8.708	0.3501	0.4990	10.966
0.0813	0.7641	8.968	0.3540	0.5048	11.293
0.0828	0.7793	9.320	0.3602	0.5139	11.796
0.0845	0.7953	9.693	0.3669	0.5236	12.313
0.0863	0.8121	10.030	0.3738	0.5334	12.813
0.0881	0.8289	10.355	0.3811	0.5435	13.316
0.0900	0.8465	10.672	0.3886	0.5538	13.770
0.0921	0.8649	10.974	0.3966	0.5645	14.238
0.0943	0.8838	11.277	0.4050	0.5753	14.703
0.0965	0.9035	11.580[a]	0.4133	0.5867	15.189[a]
0.1731	0.6676	9.422	0.4720	0.4067	12.963
0.1750	0.6750	9.661	0.4801	0.4138	13.483
0.1783	0.6883	10.106	0.4890	0.4215	14.040
0.1817	0.7018	10.517	0.4981	0.4291	14.560
0.1854	0.7158	10.921	0.5078	0.4370	15.074
0.1892	0.7303	11.308	0.5178	0.4448	15.566
0.1932	0.7454	11.682	0.5283	0.4527	16.060
0.1975	0.7608	12.055	0.5389	0.4611	16.560[a]
0.2020	0.7768	12.417			
0.2066	0.7934	12.801[a]	0.5226	0.3470	13.092
			0.5294	0.3515	13.424
0.2218	0.6202	9.730	0.5391	0.3581	14.042
0.2249	0.6276	10.004	0.5483	0.3642	14.606
0.2302	0.6447	10.657	0.5583	0.3706	15.161
0.2328	0.6519	10.911	0.5687	0.3772	15.703
0.2376	0.6655	11.383	0.5796	0.3839	16.231
0.2423	0.6784	11.784	0.5909	0.3906	16.731
0.2474	0.6922	12.185	0.6023	0.3977	17.253[a]
0.2530	0.7066	12.581			
0.2585	0.7207	12.988	0.6510	0.2352	14.928
0.2640	0.7360	13.417[a]	0.6618	0.2392	15.578
			0.6734	0.2433	16.188
0.2758	0.5701	10.309	0.6849	0.2473	16.813
0.2788	0.5766	10.619	0.6971	0.2515	17.370
0.2838	0.5872	11.077	0.7096	0.2556	17.891
0.2891	0.5984	11.533	0.7227	0.2597	18.386
0.2948	0.6102	11.971	0.7359	0.2641	18.855[a]
0.3007	0.6221	12.427			
0.3067	0.6342	12.857	0.7703	0.1206	16.451
0.3132	0.6467	13.286	0.7839	0.1227	17.174
0.3201	0.6596	13.768	0.7959	0.1246	17.774
0.3270	0.6730	14.142[a]	0.8092	0.1265	18.385
			0.8227	0.1285	18.941
			0.8367	0.1304	19.477
			0.8510	0.1323	19.949
			0.8656	0.1344	20.363[a]

[a] Methanol + ethanol binary measurement.

313.15 K, lithium chloride + water at 303.15 K, and lithium chloride + methanol at 303.15 K) for which literature results are available were examined. The results from the present work are compared to those of Patil et al. (1990) in Figure 3 and to those of Skabichevskii (1969) in Figure 4 in the form of osmotic coefficient versus salt molality. The osmotic coefficient ϕ is defined by

$$\phi = -\ln(a)/M_W \nu m \qquad (8)$$

where a is the activity of the solvent, M_W is the molecular weight of solvent (kg mol^{-1}), ν is the number of ions comprising the salt, and m is the salt molality (mol kg^{-1}). The solvent activity is calculated from

$$\ln(a) = \ln(P/P^{sat}) + (P - P^{sat})(B - V^L)/RT \qquad (9)$$

where P^{sat}, B, and V^L are the vapor pressure, second virial coefficient, and saturated liquid volume, respectively, of the pure solvent. The osmotic coefficient is used here as a means of comparison because it greatly magnifies differences between data sets. The comparisons shown in Figures 3 and 4 indicate good agreement between results of the present study and those reported in the literature.

Another check of the apparatus is obtained by examining the results for the methanol + ethanol system. The results of the present study compare to those of Reimers et al. (1992) with an average deviation in pressure of 0.03 kPa, which is within the experimental error of the current investigation.

Finally, the vapor pressures of methanol and ethanol at 303.15 K were found to be 21.978 and 10.480 kPa, respectively, which are in good agreement with the values of 21.861 and 10.462 kPa reported by Ambrose and Sprake (1970).

Total pressure data at 303.15 K for the copper(II) chloride (1) + methanol (2) and copper(II) chloride (1) + ethanol (3) binary systems are given in Table 1. Results at the same temperature for the copper(II) chloride (1) + methanol (2) + ethanol (3) ternary system, which also include the methanol + ethanol results, are given in Table 2. The results of the application of eqs 1–4 to the data are given in Table 3. The resulting average deviation between experimental and calculated pressures is 0.031 kPa, which is within experimental error.

As mentioned earlier, vapor-phase mole fractions are calculated as part of the data reduction procedure. These are plotted in Figure 5 as a function of salt-free liquid-phase composition $x_2' = x_2/(x_2 + x_3)$ for several different

Table 3. Values of Parameters Appearing in Eqs 1–4 for Copper(II) Chloride (1) + Methanol (2) + Ethanol (3) at 303.15 K

parameter	value	parameter	value
A_{23}	−0.0036	E_{12}	−1.3052
A_{32}	−0.0413	A_{13}	−4.6250
α_{23}	−0.0222	E_{13}	−3.2205
A_{12}	−8.3856	C	12.6608

salt mole fractions x_1. This figure indicates that the vapor-phase mole fraction of methanol is decreased upon addition of salt. Hence, methanol is salted in and ethanol is salted out for this system.

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