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Organic Salt Effect of Tetramethylammonium Bicarbonate on the Vapor−Liquid Equilibrium of the Methanol−Water System

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ABSTRACT: The effect of organic salt tetramethylammonium bicarbonate (TMAC) at salt mole fractions from 0 to 0.159 on the vapor−liquid equilibrium (VLE) of the binary methanol−water system has been experimentally investigated at 101.32 kPa using a modified Rose equilibrium still. The experimental data were correlated by Ohe's preferential solvation model and the Tan−Wilson model, and the average deviations of the vapor-phase mole fraction of this salt-containing methanol−water system are 0.011 and 0.017, respectively. The average deviations of temperature are 1.24 K for Tan's model.

INTRODUCTION

Tetramethylammonium hydroxide (TMAH) is an organic base as strong as NaOH or KOH. It is widely used in developing and washing processes of semiconductor production. Recently, semiconductors have been integrated more densely, and this progress requires a higher purity of TMAH. A more purified TMAH solution was obtained, through electrolysis of the tetramethylammonium bicarbonate (TMAC) in an electrolytic cell with a cation exchange membrane.[1] TMAC is an organic salt, which molecular formula is $(CH_3)_4NHCO_3$.

In the process of synthesizing TMAC, we need methanol as a solvent and at the same time methanol as one byproduct of the synthesis reaction. However, methanol is an impurity to be removed from TMAH through distillation to produce a pure salt−water solution. The correct design of distillation columns requires accurate vapor−liquid equilibrium (VLE) data. A survey of the literature shows that very few papers on the organic salt effect in VLE are available. Burns and Furter[2] reported the organic salt of the tetraethylammonium bromide effect on VLE. Slusher et al.[3,4] reported the organic salt effect of tetrabutylammonium bromide in the system of water + 2-propanol and the organic salt of tetraalkylammonium bromide in the system of water + propanol. Lee et al.[5] studied the vapor pressure and vapor−liquid of the organic salt of benzyltributylammonium chloride in the system of ethanol + water. Kurzin et al.[6−9] successfully correlated the VLE data of the organic salt effect in the solvent mixture system with the electrolyte nonrandom two-liquid (NRTL) model. The methanol−water system containing salt has been studied by Natarajan and Srinivasan,[10] Kurzin et al.,[11] and Yang and Lee.[12] However, there are no studies about the VLE data of the methanol−water system containing TMAC. In this paper, we measured the vapor−liquid phase equilibrium data of the methanol−water containing TMAC system and correlated the equilibrium data with Ohe's preferential solvation model[13] and the Tan−Wilson model.[14]

EXPERIMENTAL SECTION

Materials. Methanol (≥ 0.999 GC purity, Merck), deionized water, and TMAC (made in our own laboratory, mass fraction greater than 0.980; the mass fractions of impurity tetramethylammonium carbonate and water were less 0.005 and 0.015, respectively) were used. Back titration was used for analysis of bicarbonate/carbonate salt mixtures.[15] The salt was desiccated in an oven held at 120 °C at least for 24 h.

Apparatus and Procedure. The equilibrium apparatus was a modified Rose equilibrium still. Mixtures consisting of methanol, water, and TMAC were prepared gravimetrically with an analytical balance with an accuracy of ± 0.1 mg. After putting these mixtures in the equilibrium still, each experiment was kept at the boiling point for 30 min or more to ensure a stationary state.

Composition Analysis of the Samples. Compositions of the condensed vapor and salt-free liquid phase (water + methanol) were analyzed using a BFRL SP-2100A GC with a thermal conductivity detector. The resulting data were processed using a N2000 chromatography station. The chromatographic column (2 m × 3 mm) was packed with Porapak QS (80-100). The carrier gas was hydrogen, flowing at 40 $mL \cdot min^{-1}$, and the column temperature was 80 °C. The injector and detector temperatures were (120 and 150) °C, respectively. An injection volume of 0.4 μL was used. Vapor-phase compositions were measured by chromatography, and the compositions of the solvent and salt in the liquid phase were measured by the method of combining titration and gas chromatographic analyses.[16] A calibration curve was obtained from a set of gravimetrically prepared standard solutions which was used to calculate the mole fraction of methanol in the unknown samples; each sample was analyzed at least thrice to ensure the accuracy, and the uncertainty of the mole fractions were ± 0.001 in the methanol + water system without salt. However, in the methanol + water + TMAC ternary system, the liquid composition deviation with the method of combining titration and gas chromatographic analyses was below 0.005. The expanded uncertainty of the methanol mole fractions in the vapor phase was ± 0.01, and that of the temperatures was ± 0.1 K.

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Table 1. Vapor−Liquid Equilibrium Data for Methanol (1) + Water (2) at 101.32 kPa

no.	T/°C	x_1	y_1	no.	T/°C	x_1	y_1
1	64.69	1.000	1.000	9	76.97	0.339	0.687
2	65.30	0.969	0.987	10	83.42	0.152	0.526
3	66.14	0.894	0.958	11	87.00	0.104	0.439
4	67.16	0.833	0.936	12	94.70	0.038	0.183
5	68.57	0.732	0.898	13	97.03	0.023	0.113
6	71.43	0.591	0.836	14	98.87	0.005	0.024
7	73.45	0.483	0.776	15	100.00	0.000	0.000
8	75.60	0.392	0.721				

Table 2. Bubble-Point Data for Methanol (1) + TMAC (3) and Water (2) + TMAC (3) at 101.32 kPa

	methanol (1) + TMAC (3)		water (2) + TMAC (3)	
no.	T/K	x_3	T/K	x_3
1	353.60	0.175	395.65	0.203
2	351.88	0.159	393.00	0.185
3	349.45	0.136	388.90	0.159
4	347.10	0.113	382.90	0.114
5	345.02	0.091	380.35	0.093
6	343.55	0.076	378.10	0.076

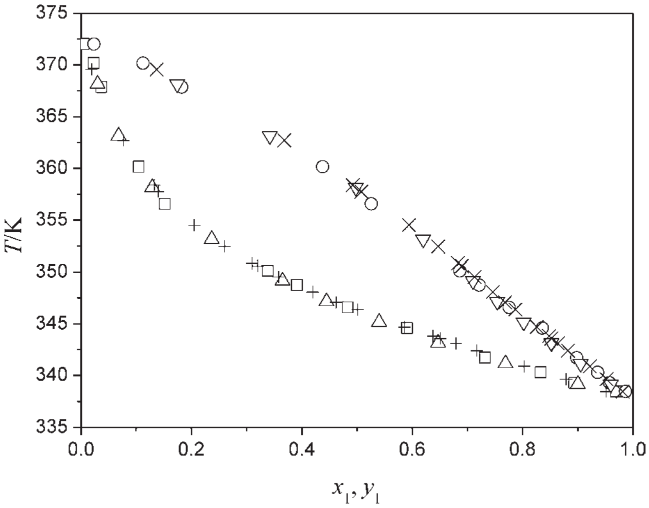

Figure 1. Experimental and literature temperature−composition diagram for methanol (1) + water (2) at 101.32 kPa: ○, $T-y_1$; □, $T-x_1$ from this experiment; ▽, $T-y_1$; △, $T-x_1$ from Kato et al.;[17] ×, $T-y_1$; +, $T-x_1$ from Kurihara et al.[18]

■ RESULTS AND DISCUSSION

Results of Measurements. VLE data for the binary systems methanol + water, water + TMAC, and methanol + TMAC as well as the VLE for the water + methanol + TMAC ternary system were measured at 101.32 kPa, and the results are listed in Tables 1 and 2. In this two tables, x_1 represents the methanol mole fraction, x_2 represents the water mole fraction, and x_3 represents the salt mole fraction in the liquid phase, respectively; x_1' is the mole fraction of methanol, x_2' is the mole fraction of water on a salt-free basis liquid phase, y_1 is the mole fraction of methanol in the vapor phase, and T is the equilibrium temperature.

VLE results for methanol (1) + water (2) at 101.32 kPa are shown in Figure 1 and compared with the results reported by Kato et al.[17] and Kurihara et al.[18] Good agreement between our results and the literature can be seen.

Results of Correlation. The first method for the prediction of salt effects was proposed by Johnson and Furter[19] using a semiempirical equation. It is quite important to explain the mechanism of this effect and to build a method of correlation and prediction. The selective effect of salt can influence the volatilities of the two liquid components, for there is a difference in the interactions of each liquid component with salt ions. The salt may increase or decrease the volatility of a component, namely, the effect of "salting-in" and "salting-out".

The Tan−Wilson equation gives considerably fewer solvent−solute interaction parameters compared with the approach proposed by other workers based on the consideration of ions and disassociation molecules of the dissolved solutes. Besides, Tan's solvent−solute interaction parameters are related to the boiling points of the individual solvent component containing the same molar ratio of solute/total solvent as in the mixture.[14,20] Another model called the solvation model had been proposed by Ohe. This method considered the alteration of vapor-phase composition is thought to be caused by a formation of solvates between solvents and salt.[13] The more polar component molecules are usually preferentially attracted by the electrostatic field of the ions; hence, the vapor composition is enriched by the less polar component. Several correlative and predictive models based on the local composition or group-contribution concept have been proposed to calculate the VLE of systems formed by mixed solvents and electrolytes, such as the electrolyte NRTL model proposed by Mock et al.[21] and the modified UNIFAC model by Kikic et al.[22] In this paper the experimental data were correlated by Ohe's solvation model and the Tan−Wilson model.

Vapor pressures P_i^0 were calculated with the Antoine equation:

$$\ln(P_i^0/\mathrm{kPa}) = A + \frac{B}{(T_{si}/\mathrm{K}) + C} \tag{1}$$

The water and methanol Antoine constants obtained from Albert et al.[23] and Ambrose et al.[24] are summarized in Table 3.

When the vapor phase is assumed to have ideal gas behavior, the VLE relation is

$$y_i P = P_i^0 \gamma_i x_i \tag{2}$$

where P_i^0 is the vapor pressure of solvent i at equilibrium temperature (i = 1, 2), y_i is the vapor-phase mole fraction, and x_i is the liquid-phase mole fraction without salt. γ_i is the activity coefficient of component i, and P is the total pressure in the system. For a binary salt-containing system, the salt was considered one of nonvolatile components, so in the vapor phase $y_{\mathrm{solvent}} = 1$ and $y_{\mathrm{salt}} = 0$.

According to Ohe's solvation model, the vapor pressure of a solvent at a given temperature is depressed by adding nonvolatile substances of salt. As the concentration of solvent is decreased by the number of solvated molecules, the actual solvent composition participating in the VLE is changed. Assuming that a salt

Table 3. Antoine Coefficients *A*, *B*, and *C*

component	temperature range/K	Antoine coefficients *A*	*B*	*C*	ref
water	284 to 441	16.2886	−3816.44	−46.13	23
methanol	257 to 364	16.5725	−3626.55	−34.29	23
	353.4 to 512.63	16.4831	−3614.17	−34.85	24

forms the solvate with the first component, the actual composition x_{1a} is given by

$$x_{1a} = \frac{x_1 - Sx_3}{(x_1 - Sx_3) + x_2} \tag{3}$$

where S is the preferential solvation number, which is the ratio of the number of solvent molecules to salt in the preferential solvate.

Since $x_1 = x_1'(1 - x_3)$, $x_2 = x_2'(1 - x_3)$, and $x_1' + x_2' = 1$, eq 3 is rewritten as follows:

$$x'_{1a} = \frac{x'_1(1 - x_3) - Sx_3}{(1 - x_3) - Sx_3} \tag{4}$$

From eq 4, we can obtain

$$S = \frac{1 - x_3}{x_3}\,\frac{x'_1 - x'_{1a}}{1 - x'_{1a}} \tag{5}$$

Therefore, the solvation number can be calculated by determining x_{1a}' from the measured values using the VLE relation obtained without adding a salt. When a salt forms the salvation with the second component, the following three equations can be derived in a similar manner.

$$x_{1a} = \frac{x_1}{x_1 + (x_2 - Sx_3)} \tag{6}$$

$$x'_{1a} = \frac{x'_1(1 - x_3)}{(1 - x_3) - Sx_3} \tag{7}$$

$$S = \frac{1 - x_3}{x_3}\,\frac{x'_{1a} - x'_1}{x'_{1a}} \tag{8}$$

The relation of $\ln(s/x_2')$ and x_3 is as follows:

$$\ln(S/x'_2) = A - B\sqrt{x_3} \tag{9}$$

Based on Figure 2, the parameters A and B can be obtained by linear regression, and the values are presented in Table 4. The preferential solvation number at different salt concentrations can be obtained with eq 9, and then the vapor fraction y_i at different equilibrium conditions can be calculated through the relation of VLE without salt. The deviations between the calculated and the experimental results of the vapor-phase mole fraction Δy_1 are listed in Table 5. The y_1−x_1' diagram for methanol (1) + water (2) at different mole fractions of TMAC is shown in Figure 3. An obvious difference can be observed in this figure. For instance, the addition of TMAC increases the mole fractions of methanol in the vapor phase. Moreover, the different salt concentrations have the same effect in VLE in the whole concentration range. At all salt concentrations, the salting-out effect on methanol rises with increasing salt concentration, as predicted by the relationship of the preferential solvation number as a function of the salt mole fraction and the liquid mole fraction, the relations are linear and are shown in Figure 4.

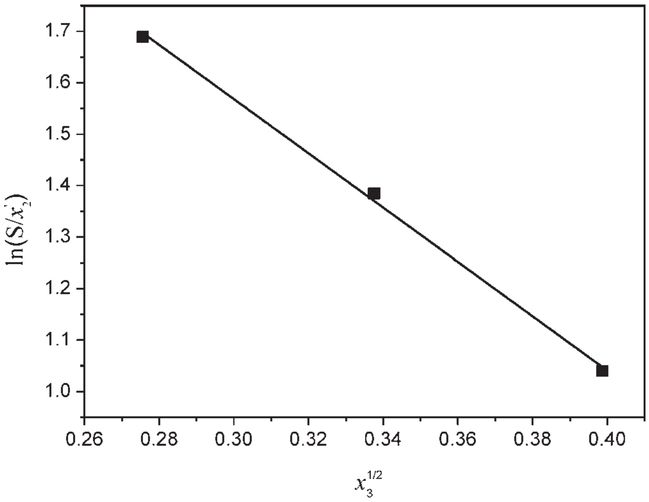

Figure 2. Relationship of solvated number and salt concentrations at 101.32 kPa.

Table 4. Results of the Correlation with Ohe's Preferential Salvation Model and the Tan−Wilson Model

x_3	Ohe's model A	B	Δy_1	Tan−Wilson model A_{s1}	A_{s2}	Δy_1	ΔT/K
0.159	3.151	5.276	0.012	1.256	1.190	0.013	1.64
0.114			0.011	1.435	1.402	0.024	1.34
0.076			0.009	1.708	1.710	0.012	0.75
mean			0.011			0.017	1.24

The Tan−Wilson model gives the activity coefficient of solvent component i in the binary solvent mixtures containing a dissolved salt as

$$\ln \gamma_{is(\text{Tan-Wilson})} = -\ln(A_{si}x'_i + A_{ij}x'_j) + x'_j\phi \tag{10}$$

where γ_{is} is the activity coefficient of solvent component i in the solvent−solute solution, s is the salt; A_{si} is the solute−solvent interaction parameter; A_{ij} is the solvent−solvent interaction parameter as similarly defined by Wilson[25] for the salt-free systems.

$$\text{where } \phi = \frac{A_{12}}{A_{s1}x'_1 + A_{12}x'_2} - \frac{A_{21}}{A_{s2}x'_2 + A_{21}x'_1}$$

where x_1' is the mole fraction of methanol in the liquid phase, expressed on a salt-free basis, x_2' is the mole fraction of water, and the solute−solvent interaction parameter, A_{si}, as

$$A_{si} = \frac{\nu_{is}}{\nu_i}\exp\left[-\left(\frac{\lambda_{is} - \lambda_{ss}}{R(T + 273.15)}\right)\right] = P_i^0/\pi \tag{11}$$

where ν_{is} is the molar volume of component i containing salt s; ν_i is the molar volume of component i. λ_{is} and λ_{ss} are related to the potential energy of the i−s and s−s pairs of molecules, respectively. P_i^0 is the saturation vapor pressure of the solvent component i calculated from the eq 1 at the bubble point, T_{si}, of the solvent component i containing the same concentration of solute at the system pressure, π. A_{12} and A_{21} are calculated from

Table 5. Vapor−Liquid Equilibrium Data for the Salt-Containing Systems at 101.32 kPa

				Tan−Wilson		Ohe's model
no.	T/K	x_1'	y_1	Δy_1	ΔT/K	Δy_1
		Methanol (1) + Water (2) + TMAC (3)				
		Mole Fraction of TMAC = 0.159				
1	388.65	0.000	0.000	0.000	0.00	0.000
2	374.65	0.050	0.408	0.014	−7.51	0.022
3	368.05	0.100	0.577	−0.002	−2.95	0.005
4	365.42	0.143	0.655	−0.016	−1.38	−0.015
5	363.85	0.200	0.702	0.036	−1.49	−0.011
6	363.65	0.250	0.753	0.035	−1.36	−0.021
7	362.40	0.333	0.819	0.008	−0.28	−0.023
8	359.95	0.500	0.880	−0.007	0.24	0.006
9	358.24	0.667	0.921	0.014	−0.043	0.011
10	355.43	0.800	0.944	0.001	−0.01	0.015
11	352.60	1.000	1.000	0.026	−0.71	0.000
mean				0.014	1.64	0.012
		Mole Fraction of TMAC = 0.114				
1	382.90	0.000	0.000	0.000	0.00	0.000
2	373.15	0.050	0.390	0.059	−5.03	0.024
3	366.65	0.100	0.559	0.021	−1.73	0.011
4	363.03	0.143	0.619	0.019	−0.90	0.013
5	360.95	0.200	0.675	0.021	−0.89	0.008
6	359.16	0.250	0.740	−0.021	0.84	−0.018
7	357.74	0.333	0.798	−0.029	1.08	−0.013
8	354.45	0.500	0.867	−0.050	1.69	0.012
9	353.35	0.667	0.922	−0.006	0.18	0.007
10	351.80	0.800	0.943	0.037	−1.05	0.014
11	347.11	1.000	1.000	0.000	0.00	0.000
mean				0.024	1.34	0.011
		Mole Fraction of TMAC = 0.076				
1	379.10	0.000	0.000	0.000	0.00	0.000
2	371.45	0.050	0.364	0.024	−2.64	0.025
3	366.45	0.100	0.538	0.008	−1.07	0.012
4	362.06	0.143	0.606	−0.003	0.14	0.010
5	359.15	0.200	0.666	−0.008	0.36	0.003
6	357.86	0.250	0.720	−0.015	0.63	−0.014
7	355.40	0.333	0.776	−0.029	1.09	−0.008
8	352.34	0.500	0.858	−0.029	0.96	0.010
9	350.09	0.667	0.920	−0.007	0.22	0.004
10	346.92	0.800	0.943	−0.013	0.38	0.010
11	343.55	1.000	1.000	0.000	0.01	0.000
mean				0.012	0.75	0.009

salt-free Wilson equation. A_{12} is the methanol−water interaction parameter and A_{21} is the water−methanol interaction parameter. The values of A_{si} are listed in Table 4.

The data were regressed to obtain the two solvent−solvent interaction parameters, $\lambda_{ij}-\lambda_{ii}$, by an minimized examination of the variation of the standard derivations during optimization. λ_{ij} and λ_{ii} are related to the potential energy of the $i-j$ and $i-i$ pairs of molecules, respectively. The regressed parameters of $\lambda_{ij}-\lambda_{ii}$ were listed in Table 6.

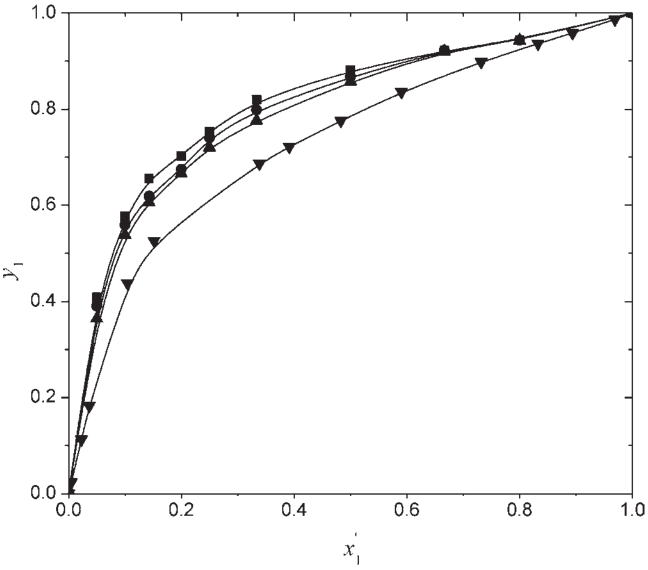

Figure 3. Equilibrium vapor−liquid composition diagram for methanol (1) + water (2) + TMAC at TMAC mole fractions (■, 0.159; ●, 0.114; ▲, 0.076; ▼, 0.000) and 101.32 kPa.

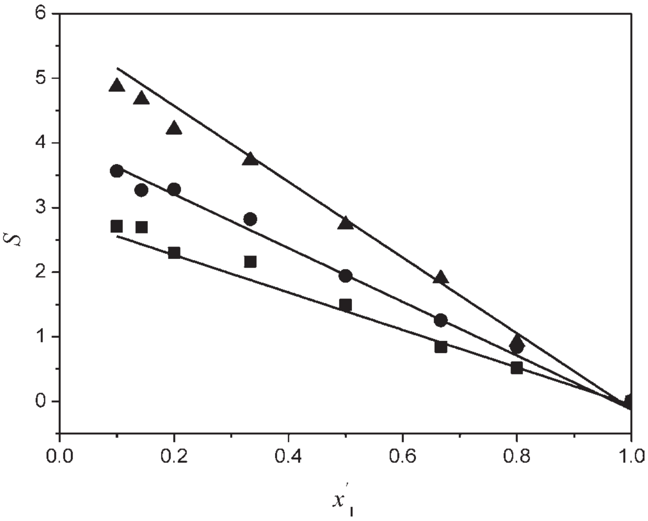

Figure 4. Relationship of the preferential solvation number S and methanol concentration at TMAC mole fractions (▲, 0.159; ●, 0.114; ■, 0.076) and 101.32 kPa. The solid line is the correlated results.

An average deviation was calculated as

$$\Delta y = (1/N)\sum_{i}^{N}|y_{ic} - y_{ie}| \quad (12)$$

where y_c is the equilibrium vapor mole fraction composition calculated from the correlation, y_e is the corresponding experimental equilibrium vapor composition values for the same equilibrium liquid composition, and N is the number of experimental data points. The regressed parameters for the Wilson model are given in Table 6. The deviations between the calculated results using the Tan−Wilson model and the experimental results of vapor-phase mole fraction Δy_1 and temperature change ΔT at different mole fractions of TMAC are listed in Table 5.

Table 6. Results of the Correlation with Wilson Models for Methanol (1) + Water (2)[a]

model type	ΔT/K	Δy	model parameter	units
Wilson	0.23	0.008	$\lambda_{12}-\lambda_{11}$ = 846.087	K
			$\lambda_{21}-\lambda_{22}$ = 2103.559	K

[a] $\Delta T = (1/N)\Sigma_i^N|T_{ci} - T_{ei}|$. $\Delta y = (1/N)\Sigma_i^N|y_{ci} - y_{ei}|$, N = number of data points.

It can be seen that the both models correlate phase equilibrium data well for the salt-containing system. The average error for the mole fraction of the vapor phase at 33 data points by Ohe's model was Δy = 0.011, and that for the Tan−Wilson model was Δy = 0.017; ΔT = 1.24 K. In the correlation of the vapor composition in the salt-containing system, Ohe's model was relatively better than Tan's model. This may be due to the fact that Ohe's model is the half-empirical equation, the data correlation based on the measured salt-free data of VLE.

■ CONCLUSION

New VLE data for the salt-containing systems of water + TMAC and methanol + TMAC as well as the VLE for the water + methanol + TMAC ternary system have been measured at 101.32 kPa. The experimental data for studied systems have been correlated by using Ohe's model and the Tan−Wilson model. The average deviations of the vapor-phase mole fraction of the ternary system are 0.011 and 0.017, respectively. The average deviations of temperature are 1.24 K for the Tan−Wilson model. Experimental studies showed that dissolved TMAC preferentially salted-out methanol from the liquid phase more than water in a ternary solvent mixture.

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■ REFERENCES

(1) Yagi, O.; Shimizu, S. Synthesis of Tetramethylammonium Hydroxide solution Free from Chloride Ion by Electrolysis of Its Formate. *Denki Kagaku* **1993**, *22*, 443–445.

(2) Burns, J. A.; Furter, W. F. Effects of Salts Having Large Organic Ions on Vapor−liquid Equilibrium. *Adv. Chem. Ser.* **1976**, *155*, 99–127.

(3) Slusher, J. T.; Decker, K. J.; Liu, H.; Vega, C. A.; Cummings, P. T.; O'Connell, J. P. Vapor−Liquid Equilibria and Salt Apparent Molar Volumes of the Water + 2-Propanol + Tetrabutylammonium Bromide System. *J. Chem. Eng. Data* **1994**, *39*, 506–509.

(4) Slusher, J. T.; Cummings, P. T. Vapor−Liquid Equilibrium and Density Measurements of Tetraalkylammonium Bromide + Propanol + Water Systems. *J. Chem. Eng. Data* **1995**, *40*, 792–798.

(5) Lee, L. S.; Huang, M. Y.; Hsu, H. L. Vapor Pressure of Ethanol + Benzyltributylammonium Chloride Solution and Vapor−Liquid Equilibrium of Ethanol + Water + Benzyltributylammonium Chloride Mixture at Atmospheric Pressure. *J. Chem. Eng. Data* **1999**, *44*, 528–531.

(6) Kurzin, A. V.; Evdokimov, A. N.; Poltoratskiy, G. M.; Platonov, A. Y.; Gusev, V. E.; Golubeva, Y. M. Isothermal Vapor−liquid Equilibrium Data for the Systems 1,4-Dioxane + Water + Tetrabutylammonium Nitrate and Acetonitride + Water + Tetrabutylammonium Bromide. *J. Chem. Eng. Data* **2004**, *49*, 208–211.

(7) Kurzin, A. V.; Evdokimov, A. N.; Antipina, V. B.; Pavlova, O. S. Measurement and Correlation of Isothermal Vapor−Liquid Equilibrium Data for the System Acetonitrile + Water + Tetrapropylammonium Bromide. *J. Chem. Eng. Data* **2006**, *51*, 1361–1363.

(8) Kurzin, A. V.; Evdokimov, A. N.; Antipina, V. B.; Pavlova, O. S.; Gusev, V. E. Isothermal Vapor−Liquid Equilibrium Data for the System Methanol + Toluene + Triphenylbenzylphosphonium Chloride. *J. Chem. Eng. Data* **2007**, *52*, 2174–2176.

(9) Kurzin, A. V.; Evdokimov, A. N.; Antipina, V. B.; Pavlova, O. S. Vapor−Liquid Equilibrium Data for the System Methanol + Toluene + Tetrabutylammonium Tetraphenylborate. *J. Chem. Eng. Data* **2008**, *53*, 1411–1413.

(10) Natarajan, T. S.; Srinivasan, D. Effect of Sodium Nitrate on the Vapor−Liquid Equilibria of the Methanol−Water System. *J. Chem. Eng. Data* **1980**, *25*, 218–221.

(11) Kurzin, A. V.; Evdokimov, A. N.; Antipina, V. B.; Pavlova, O. S. Measurement and Correlation of Isothermal Vapor−Liquid Equilibrium Data for the System Methanol + Water + Ammonium Bromide. *J. Chem. Eng. Data* **2005**, *50*, 2097–2100.

(12) Yang, S. O.; Lee, C. S. Vapor−Liquid Equilibria of Water + Methanol in the Presence of Mixed Salts. *J. Chem. Eng. Data* **1998**, *43*, 558–561.

(13) Ohe, S. Prediction of Salt Effect in Vapor-Liquid Equilibrium: A Method Based on Solvation. *Adv. Chem. Ser.* **1976**, *155*, 53–74.

(14) Tan, T. C. Model for predicting the effect of dissolved salt on the vapour liquid equilibrium of solvent mixtures. *Chem. Eng. Res. Des.* **1987**, *65*, 355–366.

(15) Levitin, G.; Myneni, S.; Hess, D. W. Reactions Between CO_2 and Tetramethylammonium Hydroxide in Cleaning Solutions. *Electrochem. Solid-State Lett.* **2003**, *6*, 101–104.

(16) Fu, J. Q. Isobaric Vapor Liquid Equilibrium for the Methanol + Ethanol + Water + Ammonium Bromide System. *J. Chem. Eng. Data* **1998**, *43*, 403–408.

(17) Kato, M.; Konishi, H.; Hipata, M. Apparatus for Measurement of Isobaric Dew and Bubble Points and Vapor−Liquid Equilibria. *J. Chem. Eng. Data* **1970**, *15*, 501–505.

(18) Kurihara, K.; Nakamichi, M.; Kojima, K. Isobaric Vapor−liquid Equilibria for Methanol + Ethanol +Water and the Three Constituent Binary Systems. *J. Chem. Eng. Data* **1993**, *38*, 446–449.

(19) Johnson, A. I.; Furter, W. F. Salt effect in vapor−liquid equilibrium, Part II. *Can. J. Chem. Eng.* **1960**, *38*, 78–87.

(20) Tan, T. C. A modified NRTL model for predicting the effect of dissolved solute on the vapour−liquid equilibrium of solvent mixtures. *Trans. IChemE* **1990**, *68*, 93–103.

(21) Mock, B.; Evans, L. B.; Chen, C. C. Thermodynamic Representation of Phase Equilibria of Mixed-Solvent Electrolyte Systems. *AIChE J.* **1986**, *32*, 1655–1664.

(22) Kikic, I.; Fermeglia, M.; Rasmussen, P. UNIFAC Prediction of Vapour−Liquid Equilibria in Mixed Solvent-Salt Systems. *Chem. Eng. Sci.* **1991**, *46*, 2775–2780.

(23) Albert, M.; Hahnenstein, I.; Hasse, H.; Maurer, G. Vapor−liquid and Liquid−Liquid Equilibria in Binary and Ternary Mixtures of Water, Methanol, and Methylal. *J. Chem. Eng. Data* **2001**, *46*, 897–903.

(24) Ambrose, D.; Sprake, C. H. S.; Townsend, R. Thermodynamic Properties of Organic Oxygen Compounds. XXXVII. Vapour Pressures of Methanol, Ethanol, Pentan-1-ol, and Octan-1-ol from the Normal Boiling Temperature to the Critical Temperature. *J. Chem. Thermodyn.* **1975**, *7*, 185–190.

(25) Wilson, G. M. Vapour−Liquid equilibrium. XI. A new expression for the excess free energy of mixing. *J. Am. Chem. Soc.* **1964**, *86*, 127–130.