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Measurement and Correlation of Solubilities of Potassium Chloride and Potassium Sulfate in Aqueous Glycerol Solutions

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ABSTRACT: The solubilties of potassium chloride and potassium sulfate in aqueous glycerol solutions have been measured at temperatures ranging from (289 to 349) K using a dynamic method. The solute-free mass fractions of glycerol range from 0.0000 to 0.8001. The solubilities of KCl and K_2SO_4 in aqueous glycerol solutions increased with temperature and decreased with addition of glycerol. The experimental data were correlated with the electrolyte nonrandom two liquid (E-NRTL) model. The binary interaction parameters in the E-NRTL model were obtained simultaneously using the solubility data of the ternary systems. The calculated solid−liquid equilibrium (SLE) temperatures with the E-NRTL model were in good agreement with the experimental results. The root-mean-square deviations of solubility temperature varied from (0.36 to 1.05) K.

INTRODUCTION

Because of the depletion of the world's petroleum reserves and the increasing environmental concerns, there is a great demand for alternative sources of petroleum-based fuel. Biodiesel seems to be an ideal diesel fuel substitution because it is biodegradable, nontoxic, and burns with low sulfur, carbon monoxide, unburned hydrocarbons, particulate matter, and aromatic-free emission profile.[1−3]

The traditional commercial production process for biodiesel is known as alkali-catalyzed transesterification. A vegetable oil or animal fat reacts with an alcohol (methanol or ethanol) in presence of a catalyst to produce biodiesel and glycerol (a byproduct).[4] The most common catalysts used are strong mineral bases, such as KOH. The obtained biodiesel requires further purification to remove impurities to provide qualified fuels. Wet washing,[5,6] which involves one or more steps of water-biodiesel contact, is widely used, leading to generating large amounts of highly polluted wastewater. The wastewater is at high pH because of significant levels of residual KOH and a high chemical oxygen demand-(COD) value because of high contents of oil and grease (O&G), methanol, and glycerol.[7] Acid such as concentrated HCl or H_2SO_4 can be introduced before wet washing to neutralize residual KOH.[3] As a result, KOH is transformed to KCl or K_2SO_4 and is removed in the wet washing step.

Under the conventional process, about 20 L of raw biodiesel wastewater is discharged per 100 L biodiesel produced.[8] Currently, several strategies are being pursued to treat biodiesel wastewater. Suehara[8] utilized microorganism to treat the wastewater based on the biological method. Siles[9] attempted the anaerobic codigestion of glycerol and wastewater to produce methane at laboratory scale. Ito[10] utilized wastewater as a source of carbon for oil degrading yeast to produce hydrogen and ethanol. Electrochemical methods, such as electrocoagulation process[7] or combined processes of chemical recovery and electrochemical treatment[1], were also proposed and the effects of these methods were evaluated. However, none of these methods is widely used in industrial level processes. Thus, other effective methods suitable to industrial application need to be studied. As mentioned above, high contents of O&G, methanol, glycerol, and salt (KCl or K_2SO_4) are present in wastewater. O&G can be effectively removed by an electrocoagulation process,[7] and methanol can be easily recovered by a vacuum flash process. Then, glycerol and salt are the main substances remaining in the wastewater. Solubility data of KCl and K_2SO_4 in aqueous glycerol mixed solvents are essential for further biodiesel wastewater treatment. Unfortunately, the data are very scarce up to now. Kraus[11] have reported the solubility data of KCl in aqueous glycerol solutions, but the data were restricted to 25 °C.

In this work, the solubilities of KCl and K_2SO_4 in aqueous glycerol solutions have been investigated by a dynamic method at temperatures ranging from (289 to 349) K. The E-NRTL model,[12] which was proposed by Chen and has been successfully used to describe vapor−liquid equilibrium,[13−15] solid−liquid equilibrium,[16,17] and liquid−liquid equilibrium,[18] was employed to correlate the experimental data. The binary interaction parameters of the E-NRTL model were obtained.

EXPERIMENTAL SECTION

Materials. KCl and K_2SO_4 were purchased from Guangfu Chemical Reagents Co., Tianjin, China, and had a purity of 99.5 % and 99.0 % mass fraction, respectively. Prior to the measurements, the salts were dried in an oven at 433 K for 2 days. Glycerol with a purity greater than 99.0 % mass fraction was obtained from Guangfu Chemical Reagents Co., Tianjin, China. Deionized water was supplied by Nankai Chemical Reagents Co., Tianjin, China. Glycerol and deionized water were used without further purification.

Apparatus and Procedure. The solubility was determined using the dynamic method. The laser monitoring observation technique was used to determine the dissolution temperature of a solid−liquid mixture of known composition. The experimental

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Table 1. Experimental Solubility Data for the System of KCl (1) + Glycerol (3) + Water (4)

x_1	T^{exp}	x_1	T^{exp}	x_1	T^{exp}
	K		K		K
$w_3^0 = 0.0000$					
$7.641 \cdot 10^{-2}$	292.96	$8.885 \cdot 10^{-2}$	313.84	$9.944 \cdot 10^{-2}$	333.02
$7.920 \cdot 10^{-2}$	296.73	$9.099 \cdot 10^{-2}$	317.66	$1.017 \cdot 10^{-1}$	336.91
$8.201 \cdot 10^{-2}$	301.50	$9.312 \cdot 10^{-2}$	321.59	$1.041 \cdot 10^{-1}$	340.77
$8.465 \cdot 10^{-2}$	306.72	$9.519 \cdot 10^{-2}$	325.68	$1.065 \cdot 10^{-1}$	345.46
$8.673 \cdot 10^{-2}$	310.30	$9.722 \cdot 10^{-2}$	330.71		
$w_3^0 = 0.1998$					
$7.371 \cdot 10^{-2}$	295.15	$8.319 \cdot 10^{-2}$	310.91	$9.300 \cdot 10^{-2}$	329.88
$7.500 \cdot 10^{-2}$	297.04	$8.414 \cdot 10^{-2}$	312.37	$9.441 \cdot 10^{-2}$	332.24
$7.572 \cdot 10^{-2}$	298.40	$8.564 \cdot 10^{-2}$	315.17	$9.522 \cdot 10^{-2}$	334.27
$7.730 \cdot 10^{-2}$	300.85	$8.669 \cdot 10^{-2}$	317.13	$9.670 \cdot 10^{-2}$	337.08
$7.783 \cdot 10^{-2}$	301.90	$8.774 \cdot 10^{-2}$	319.16	$9.753 \cdot 10^{-2}$	339.36
$7.953 \cdot 10^{-2}$	304.58	$8.895 \cdot 10^{-2}$	321.37	$9.893 \cdot 10^{-2}$	342.42
$8.039 \cdot 10^{-2}$	306.12	$9.081 \cdot 10^{-2}$	325.34	$9.916 \cdot 10^{-2}$	342.66
$8.182 \cdot 10^{-2}$	308.42	$9.199 \cdot 10^{-2}$	327.32	$1.003 \cdot 10^{-1}$	345.90
$w_3^0 = 0.4000$					
$6.678 \cdot 10^{-2}$	293.29	$7.859 \cdot 10^{-2}$	312.04	$8.854 \cdot 10^{-2}$	331.20
$6.985 \cdot 10^{-2}$	297.41	$8.014 \cdot 10^{-2}$	315.03	$9.034 \cdot 10^{-2}$	334.72
$7.102 \cdot 10^{-2}$	299.13	$8.193 \cdot 10^{-2}$	318.16	$9.175 \cdot 10^{-2}$	338.15
$7.290 \cdot 10^{-2}$	302.61	$8.376 \cdot 10^{-2}$	321.28	$9.367 \cdot 10^{-2}$	342.89
$7.534 \cdot 10^{-2}$	306.47	$8.526 \cdot 10^{-2}$	324.42	$9.495 \cdot 10^{-2}$	345.22
$7.677 \cdot 10^{-2}$	309.22	$8.751 \cdot 10^{-2}$	328.83		
$w_3^0 = 0.6001$					
$6.237 \cdot 10^{-2}$	297.11	$7.573 \cdot 10^{-2}$	317.06	$8.482 \cdot 10^{-2}$	335.08
$6.486 \cdot 10^{-2}$	300.70	$7.738 \cdot 10^{-2}$	319.91	$8.669 \cdot 10^{-2}$	339.25
$6.894 \cdot 10^{-2}$	305.40	$7.982 \cdot 10^{-2}$	324.61	$8.766 \cdot 10^{-2}$	342.92
$7.061 \cdot 10^{-2}$	308.99	$8.131 \cdot 10^{-2}$	327.57	$8.986 \cdot 10^{-2}$	346.70
$7.345 \cdot 10^{-2}$	312.98	$8.308 \cdot 10^{-2}$	331.37		
$w_3^0 = 0.8001$					
$5.801 \cdot 10^{-2}$	295.05	$6.910 \cdot 10^{-2}$	314.19	$8.007 \cdot 10^{-2}$	336.11
$5.861 \cdot 10^{-2}$	296.16	$7.119 \cdot 10^{-2}$	317.82	$8.194 \cdot 10^{-2}$	339.53
$6.142 \cdot 10^{-2}$	301.02	$7.298 \cdot 10^{-2}$	321.37	$8.317 \cdot 10^{-2}$	342.86
$6.296 \cdot 10^{-2}$	303.59	$7.430 \cdot 10^{-2}$	323.92	$8.463 \cdot 10^{-2}$	345.19
$6.463 \cdot 10^{-2}$	306.77	$7.670 \cdot 10^{-2}$	328.95	$8.559 \cdot 10^{-2}$	347.30
$6.642 \cdot 10^{-2}$	309.52	$7.840 \cdot 10^{-2}$	332.25		

method has been described in detail previously.[19,20] Predetermined amounts of solute and solvents for the solubility measurement were weighed on an analytical balance (Gibertini, Crystal 200, Italy) with an accuracy of ± 0.0001 g and transferred into a vessel. The mixtures were heated slowly with continuously stirring. The heating rate would be less than 0.1 K per 30 min when the system would arrive at a balance. The temperature of the system was measured by a platinum resistance thermometer Pt-100 (calibrated with an accuracy of ± 0.01 K) and was controlled by a refrigerated/heating circulator (Julabo FP45-HE, Germany, temperature stability ± 0.01 K). The temperature at which the last crystal disappeared was taken as the equilibrium temperature. The uncertainty of the measured temperature was ± 0.2 K. The uncertainty of the composition of solubility was ± 0.001 mol fraction.

Table 2. Experimental Solubility Data for the System of K_2SO_4 (2) + Glycerol (3) + Water (4)

x_2	T^{exp}	x_2	T^{exp}	x_2	T^{exp}
	K		K		K
$w_3^0 = 0.0000$					
$1.080 \cdot 10^{-2}$	290.57	$1.471 \cdot 10^{-2}$	310.78	$1.822 \cdot 10^{-2}$	331.85
$1.149 \cdot 10^{-2}$	294.01	$1.526 \cdot 10^{-2}$	313.90	$1.882 \cdot 10^{-2}$	335.89
$1.216 \cdot 10^{-2}$	297.50	$1.588 \cdot 10^{-2}$	317.55	$1.943 \cdot 10^{-2}$	339.85
$1.271 \cdot 10^{-2}$	300.55	$1.648 \cdot 10^{-2}$	321.05	$2.006 \cdot 10^{-2}$	344.15
$1.320 \cdot 10^{-2}$	303.15	$1.707 \cdot 10^{-2}$	324.70	$2.036 \cdot 10^{-2}$	346.35
$1.415 \cdot 10^{-2}$	307.62	$1.767 \cdot 10^{-2}$	328.16		
$w_3^0 = 0.1999$					
$7.639 \cdot 10^{-3}$	290.01	$1.072 \cdot 10^{-2}$	311.95	$1.328 \cdot 10^{-2}$	332.49
$8.257 \cdot 10^{-3}$	294.30	$1.121 \cdot 10^{-2}$	316.00	$1.383 \cdot 10^{-2}$	336.90
$8.765 \cdot 10^{-3}$	297.76	$1.163 \cdot 10^{-2}$	319.28	$1.415 \cdot 10^{-2}$	339.85
$9.189 \cdot 10^{-3}$	301.00	$1.202 \cdot 10^{-2}$	322.47	$1.455 \cdot 10^{-2}$	344.55
$9.747 \cdot 10^{-3}$	304.59	$1.245 \cdot 10^{-2}$	326.05	$1.498 \cdot 10^{-2}$	348.25
$1.011 \cdot 10^{-2}$	307.85	$1.287 \cdot 10^{-2}$	329.35		
$w_3^0 = 0.3999$					
$5.468 \cdot 10^{-3}$	292.35	$7.201 \cdot 10^{-3}$	311.27	$9.069 \cdot 10^{-3}$	332.12
$5.722 \cdot 10^{-3}$	294.95	$7.506 \cdot 10^{-3}$	314.55	$9.395 \cdot 10^{-3}$	336.53
$5.920 \cdot 10^{-3}$	297.47	$7.816 \cdot 10^{-3}$	318.15	$9.658 \cdot 10^{-3}$	339.15
$6.131 \cdot 10^{-3}$	299.56	$8.146 \cdot 10^{-3}$	322.20	$9.937 \cdot 10^{-3}$	343.55
$6.486 \cdot 10^{-3}$	303.40	$8.434 \cdot 10^{-3}$	325.25	$1.013 \cdot 10^{-2}$	346.30
$6.877 \cdot 10^{-3}$	307.35	$8.772 \cdot 10^{-3}$	329.18		
$w_3^0 = 0.5999$					
$3.210 \cdot 10^{-3}$	289.89	$4.508 \cdot 10^{-3}$	311.07	$5.555 \cdot 10^{-3}$	330.35
$3.436 \cdot 10^{-3}$	293.01	$4.697 \cdot 10^{-3}$	314.09	$5.726 \cdot 10^{-3}$	334.13
$3.668 \cdot 10^{-3}$	297.17	$4.841 \cdot 10^{-3}$	317.43	$5.929 \cdot 10^{-3}$	338.18
$3.871 \cdot 10^{-3}$	300.35	$5.037 \cdot 10^{-3}$	321.19	$6.110 \cdot 10^{-3}$	341.73
$4.107 \cdot 10^{-3}$	304.55	$5.200 \cdot 10^{-3}$	323.95	$6.263 \cdot 10^{-3}$	345.45
$4.315 \cdot 10^{-3}$	307.63	$5.368 \cdot 10^{-3}$	327.40		
$w_3^0 = 0.8000$					
$1.754 \cdot 10^{-3}$	294.15	$2.666 \cdot 10^{-3}$	310.02	$3.148 \cdot 10^{-3}$	325.26
$2.060 \cdot 10^{-3}$	298.59	$2.778 \cdot 10^{-3}$	313.89	$3.254 \cdot 10^{-3}$	329.75
$2.161 \cdot 10^{-3}$	300.15	$2.889 \cdot 10^{-3}$	315.95	$3.379 \cdot 10^{-3}$	335.02
$2.329 \cdot 10^{-3}$	303.75	$2.971 \cdot 10^{-3}$	319.41	$3.472 \cdot 10^{-3}$	339.91
$2.485 \cdot 10^{-3}$	307.60	$3.109 \cdot 10^{-3}$	323.82	$3.576 \cdot 10^{-3}$	345.45

■ RESULTS AND DISCUSSION

Solid−liquid equilibrium data for the systems KCl + glycerol + water and K_2SO_4 + glycerol + water were measured at various mixed solvent compositions. The experimental results are given in Tables 1 and 2, in which T^{exp} is the measured absolute temperature, x_1 and x_2 are mole fraction solubility of KCl and K_2SO_4 in aqueous glycerol solutions, respectively, and w_3^0 is mass fraction of glycerol in the solute-free solution.

To verify the reliability of the measurement, the solubilities of KCl and K_2SO_4 in water were measured first and compared with published data.[21−23] The comparison results are shown in Figures 1 and 2, respectively. It is clear from Figures 1 and 2 that the experimental results show good agreement with the literature data.

The solubilities of KCl and K_2SO_4 in aqueous glycerol solvent mixtures are shown in Figures 3 and 4, respectively. It can be

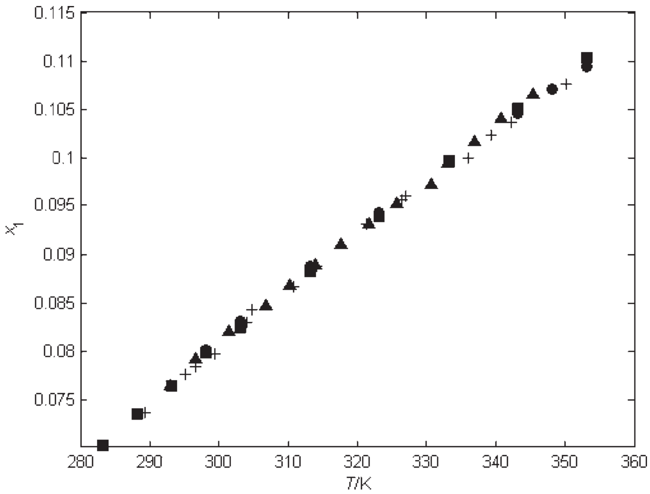

Figure 1. Solubility of KCl in water: ■, literature data;[21] •, literature data;[22] +, literature data;[23]▲, present study.

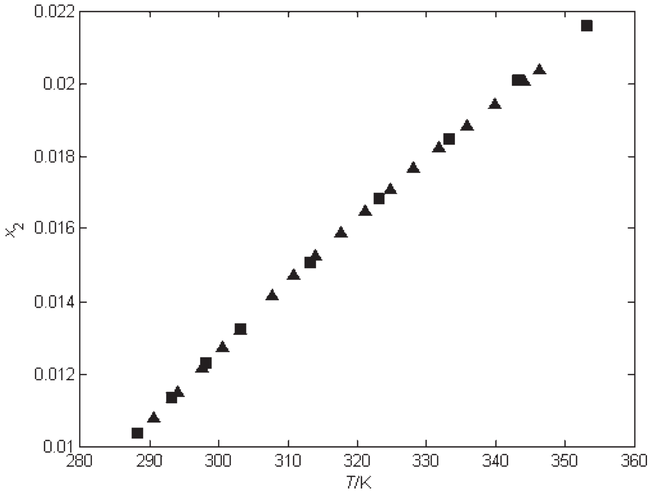

Figure 2. Solubility of K_2SO_4 in water: ■, literature data;[21] ▲, present study.

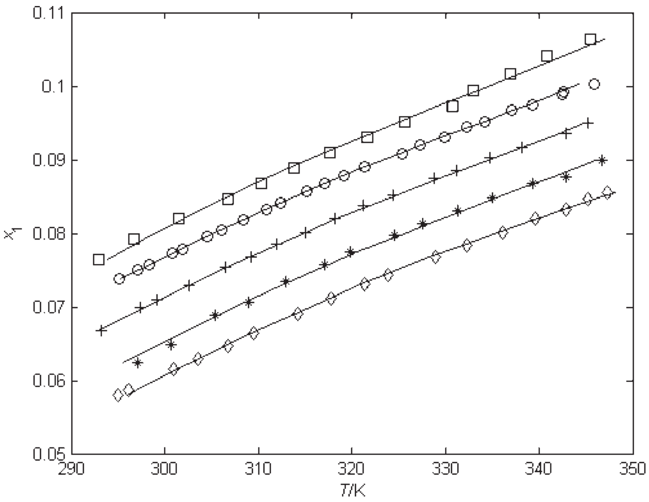

Figure 3. Mole fraction solubilities x_1 of KCl in binary glycerol + water solvent mixtures. Mass fraction of glycerol on the solute-free basis is: □, 0.0000; ○, 0.1998; +, 0.4000; *, 0.6001; ◇, 0.8001; —, solubility curve calculated from the E-NRTL model.

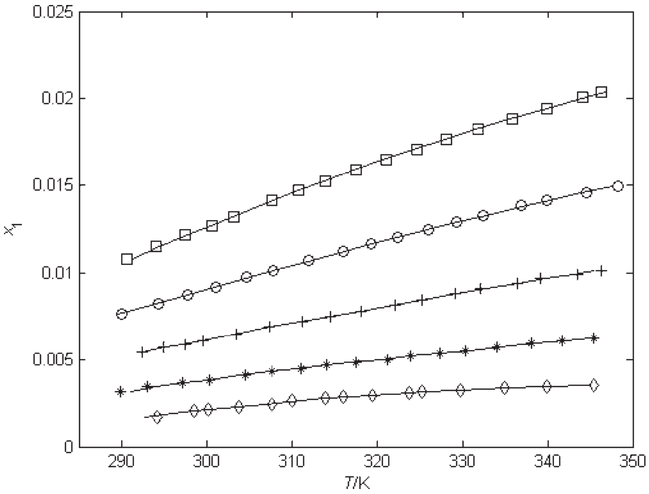

Figure 4. Mole fraction solubilities x_2 of K_2SO_4 in binary glycerol + water solvent mixtures. Mass fraction of glycerol on the solute-free basis is: □, 0.0000; ○, 0.1999; +, 0.3999; *, 0.5999; ◇, 0.8000; —, solubility curve calculated from the E-NRTL model.

observed that the solubilities of KCl and K_2SO_4 both increase with temperature and decrease with addition of glycerol. The highest and the lowest solubility are obtained in pure water and 80 wt % glycerol solution for both systems within the studied solvent compositions. The solubilities of K_2SO_4 in aqueous glycerol solutions are weekly dependent on temperature when w_3^0 is more than 60 wt %.

By comparing the solubility data between the two ternary systems, we find that the solubilities of K_2SO_4 in aqueous glycerol solutions are much lower than that of KCl. In 80 wt % glycerol solution, less than 1.3 g K_2SO_4 is present in per 100 g mixed solvent in the studied temperature range. It means H_2SO_4 is more beneficial than HCl to be used to neutralize wastewater from the point of view of separating the salt with aqueous glycerol solution. K_2SO_4 in biodiesel wastewater can be easily separated from aqueous glycerol solution by evaporating water to concentrate the aqueous glycerol solution.

The phase equilibrium between the solvent phase and the salt's crystal phase can be described by the solubility product. The solubility product defined by activity coefficient and mole fraction of aqueous ions is expressed as follows:

$$K_s = a_+^{\nu+} a_-^{\nu-} = (\gamma^*{}_+ x_+)^{\nu+} (\gamma^*{}_- x_-)^{\nu-} \tag{1}$$

where a, γ, and x denote the activity, the activity coefficient, and the solubility in units of mole fraction, respectively. The superscript ν is the stoichiometric coefficient of the ion. The subscripts + and − refer to cation and anion, respectively. Equation 1 can be used not only for aqueous solutions but also for organic or mixed solvent electrolyte solution.[24] The solubility product is expressed as a function of temperature

$$\ln K_s = Ax_0 + B(1 - x_0) + (Cx_0 + D(1 - x_0))/T \tag{2}$$

where T is absolute temperature in Kelvin; A, B, C, and D are constants obtained by regressing solubility data. x_0 is solute-free mole fraction of glycerol in the aqueous solutions.

Table 3. E-NRTL Model Parameters for the KCl + Glycerol + Water and K_2SO_4 + Glycerol + Water Systems (Defined by eq 4)

i	j	a_{ij}	a_{ij}	b_{ij}	b_{ji}	$\alpha_{ij} = \alpha_{ij}$
				K	K	
KCl	glycerol	40.473	47.955	−8511.8	−20429	0.14878
KCl	water	3418.2	39.293	53.236	−15957	0.2[a]
glycerol	water	59.677	4.0677	−18700	213.44	0.3
K_2SO_4	glycerol	1.7076	−9.7097	210.82	−218.44	0.41345
K_2SO_4	water	−21.629	−9.3118	11386	−841.20	0.2[a]

[a] Obtained from ref 18.

To estimate the SLE temperature of the dissolution of salts in aqueous glycerol solution, one requires the evaluation of the solubility product and a thermodynamic model that allows to estimate the activity coefficients of all species considered in solution. In this work, the E-NRTL model is used to calculate the activity coefficients in eq 1 on the basis of complete dissociation assumption.

The activity coefficient of ion i is represented by the combination of the Pitzer−Debye−Hückel equation and the NRTL equation as follows:

$$\ln \gamma_i^* = \ln \gamma_i^{*\mathrm{PDH}} + \ln \gamma_i^{*\mathrm{lc}} \tag{3}$$

The former term on the right side of the above equation is the activity coefficients accounting for the electrostatic interactions of ions and the later represents the local composition NRTL contribution. The expressions for $\ln \gamma_i^{*\mathrm{PDH}}$ and $\ln \gamma_i^{*\mathrm{lc}}$ have been described by Chen.[12] In the original model, the NRTL binary interaction parameters τ_{ij} are taken as model parameters to be regressed from experimental data, while in this work, τ_{ij} are expressed as a function of temperature as follows:

$$\tau_{ij} = a_{ij} + b_{ij}/T \tag{4}$$

where a_{ij} and b_{ij} are constants describing the temperature dependence of τ_{ij}.

Besides, though a value of 0.2 is satisfactory for solvent-salt nonrandomness factor in regressing data of single-solvent electrolyte system, it is critical in representing the phase equilibrium of mixed-solvent electrolyte systems as suggested by Chen.[18] Therefore, the value of the glycerol-salt nonrandomness factor is also regressed in this work and the values of glycerol−water and water-salt nonrandomness factor are fixed to 0.3 and 0.2, respectively.

The optimum algorithm applied in the model parameters estimation is the Nelder−Mead Simplex Method. The root-mean-square deviation σ between the experimental equilibrium temperature and calculated equilibrium temperature is taken as the object function, which is defined by

$$\sigma = \left[\sum_{i=1}^{N} (T^{\exp} - T)^2/(N-1)\right]^{0.5} \tag{5}$$

where N is the number of experimental data points; $T^{\exp}$ is the experimental equilibrium temperature; and T is the calculated equilibrium temperature calculated from eqs 1 to 4.

From Figures 3 and 4 it is possible to observe the good agreement between the experimental equilibrium temperature obtained in this work and the equilibrium temperature calculated by the E-NRTL model for the mixed solvent electrolyte systems.

Table 4. Parameters of K_S for KCl and K_2SO_4 in Aqueous Glycerol Solutions (Defined by eq 2)

system	A	B	C	D
			K	K
KCl + glycerol + water	−14.168	−14.061	−8198.1	−8508.8
K_2SO_4 + glycerol + water	94.457	81.128	−40988	−34143

Table 5. Root-Mean-Square Deviations σ from the Description by the E-NRTL Model (Defined by eq 5)

solute KCl		solute K_2SO_4	
solvents	σ	solvents	σ
	K		K
pure water	1.05	pure water	0.36
water +20 wt % glycerol	0.41	water +20 wt % glycerol	0.37
water +40 wt % glycerol	0.41	water +40 wt % glycerol	0.37
water +60 wt % glycerol	0.84	water +60 wt % glycerol	0.53
water +80 wt % glycerol	0.54	water +80 wt % glycerol	0.58

The values of the model parameters a_{ij}, b_{ij}, and α calculated from the experimental data are listed in Table 3. Parameters A, B, C, and D of eq 2 are listed in Table 4. The root-mean-square deviations for each solvent composition are listed in Table 5, which range from (0.36 to 1.05) K. From Table 5 it can be seen that the E-NRTL model provides a superior results for the K_2SO_4 + glycerol + water system. The results for the KCl + glycerol + water system are also satisfactory with a root-mean-square deviation less than 1.05 K in terms of equilibrium temperature. The results show that the E-NRTL model can be used to describe the solubilities of KCl and K_2SO_4 in aqueous glycerol solutions satisfactorily.

■ CONCLUSIONS

The solubilities of KCl and K_2SO_4 in aqueous glycerol solutions at a temperature range from (289 to 349) K have been investigated by the dynamic method. The salts solubilities of both systems increase with temperature and decrease with addition of glycerol over the range of solvent compositions studied. The highest and the lowest solubility of both systems are obtained in pure water and 80 wt % aqueous glycerol solution. For the K_2SO_4 + glycerol + water system, the temperature dependence is not so pronounced when w_3^0 is more than 60 wt %. The solubilities of KCl in aqueous glycerol solutions are much higher than that of K_2SO_4 at a fixed solvent composition.

The solubility data in aqueous glycerol solutions have been described with the E-NRTL model. Using binary interaction parameters, the E-NRTL model can well describe the two ternary systems over the present studied range of temperature and concentrations. The root-mean-square deviations for each system range from (0.36 to 1.05) K.

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■ REFERENCES

(1) Jaruwat, P.; Kongjao, S.; Hunsom, M. Management of bio diesel wastewater by the combined processes of chemical recovery and electrochemical treatment. *Energy Convers. Manage.* **2010**, *51*, 531–537.

(2) Leung, D. Y. C.; Wu, X.; Leung, M. K. H. A review on biodiesel production using catalyzed transesterification. *Appl. Energy* **2010**, *87*, 1083–1095.

(3) Gerpen, J. V. Biodiesel processing and production. *Fuel Process. Technol.* **2005**, *86*, 1097–1107.

(4) Marchetti, J. M.; Miguel, V. U.; Errazu, A. F. Possible methods for biodiesel production. *Renewable Sustainable Energy Rev.* **2007**, *11*, 1300–1311.

(5) Berrios, M.; Skelton, R. L. Comparison of purification methods for biodiesel. *Chem. Eng. J.* **2008**, *144*, 459–465.

(6) Gonzalo, A.; García, M.; Luis Sánchez, J.; Arauzo, J.; Peña, J. A. Water cleaning of biodiesel. Effect of catalyst concentration, water amount, and washing temperature on biodiesel obtained from rapeseed oil and used oil. *Ind. Eng. Chem. Res.* **2010**, *49*, 4436–4443.

(7) Chavalparit, O.; Ongwandee, M. Optimizing electrocoagulation process for the treatment of biodiesel wastewater using response surface methodology. *J. Environ. Sci.* **2009**, *21*, 1491–1496.

(8) Suehara, K.; Kawamoto, Y.; Fujii, E.; Kohda, J.; Nakano, Y.; Yano, T. Biological treatment of wastewater discharged from biodiesel fuel production plant with alkali-catalyzed transesterification. *J. Biosci. Bioeng.* **2005**, *100*, 437–442.

(9) Siles, J. A.; Martín, M. A.; Chica, A. F.; Martín, A. Anaerobic co-digestion of glycerol and wastewater derived from biodiesel manufacturing. *Bioresour. Technol.* **2010**, *101*, 6315–6321.

(10) Ito, T.; Nakashimada, Y.; Senba, K.; Matsui, T.; Nishio, N. Hydrogen and ethanol production from glycerol-containing waste discharged after biodiesel manufacturing process. *J. Biosci. Bioeng.* **2005**, *100*, 260–265.

(11) Kraus, K. A.; Raridon, R. J.; Baldwin, W. H. Properties of organic—water mixtures. I. Activity coefficients of sodium chloride, potassium chloride, and barium nitrate in saturated water mixtures of glycol, glycerol, and their acetates. model solutions for hyperfiltration membranes. *J. Am. Chem. Soc.* **1964**, *86*, 2571–2576.

(12) Chen, C. C.; Evans, L. B. A local composition model for the excess Gibbs energy of aqueous electrolyte systems. *AIChE J.* **1986**, *32*, 444–454.

(13) Yan, W.; Rose, C.; Zhu, M.; Gmehling, J. Measurement and correlation of isothermal vapor—liquid equilibrium data for the system acetone + methanol + lithium bromide. *J. Chem. Eng. Data* **1998**, *43*, 585–589.

(14) Kurzin, A. V.; Evdokimov, A. N.; Antipina, V. B.; Gusev, V. E. Isothermal vapor—liquid equilibrium data for the system methanol + benzene + sodium tetraphenylborate. *J. Chem. Eng. Data* **2005**, *50*, 1861–1863.

(15) Vercher, E.; Orchills, A. V.; Vázquez, M. I.; Martínez-Andreu, A. Isobaric vapor-liquid equilibria for 1-propanol + water + lithium chloride at 100 kPa. *Fluid Phase Equilib.* **2004**, *216*, 47–52.

(16) Chen, C. C. Representation of solid-liquid equilibrium of aqueous electrolyte systems with the electrolyte NRTL model. *Fluid Phase Equilib.* **1986**, *27*, 457–474.

(17) Kim, Y.; Haam, S.; Koo, K.; Shul, Y. G.; Son, J.; Jung, J. K. Representation of solid—liquid equilibrium of l-ornithine-l-aspartate + water + methanol system using the Chen model for mixed-solvent electrolyte solution. *J. Chem. Eng. Data* **2001**, *46*, 1387–1391.

(18) Mock, B.; Evans, L. B.; Chen, C. C. Thermodynamic representation of phase equilibria of mixed-solvent electrolyte systems. *AIChE J.* **1986**, *32*, 1655–1664.

(19) Xia, Q.; Zhang, F. B.; Zhang, G. L.; Ma, J. C.; Zhao, L. Solubility of sebacic acid in binary water + ethanol solvent mixtures. *J. Chem. Eng. Data* **2008**, *53*, 838–840.

(20) Chen, S. N.; Xia, Q.; Lu, L. F.; Li, D.; Zhang, F. B.; Zhang, G. L. Solubility of sebacic acid in binary mixtures containing alcohols. *J. Chem. Eng. Data* **2009**, *54*, 776–780.

(21) Stephen, H.; Stephen, T. *Solubilities of Inorganic and Organic Compounds, Vol. 1. Binary Systems. Part 1*; Pergamon Press: NewYork, 1963.

(22) Pinho, S. P.; Macedo, E. A. Solubility of NaCl, NaBr, and KCl in water, methanol, ethanol, and their mixed solvents. *J. Chem. Eng. Data* **2005**, *50*, 29–32.

(23) Sunier, A. A.; Baumbach, J. The solubility of potassium chloride in ordinary and heavy water. *J. Chem. Eng. Data* **1976**, *21* (3), 335–336.

(24) Li, M.; Constantinescu, D.; Wang, L.; Mohs, A.; Gmehling, J. Solubilities of NaCl, KCl, LiCl, and LiBr in methanol, ethanol, acetone, and mixed solvents and correlation using the LIQUAC model. *Ind. Eng. Chem. Res.* **2010**, *49*, 4981–4988.