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Solid−Liquid Equilibria in Three Binary Mixtures Containing Diphenyl Carbonate

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ABSTRACT: This study aims to measure the solid−liquid equilibria (SLE) of systems containing diphenyl carbonate (DPC) for production of high-purity DPC as required for a nonphosgene-based polycarbonate production process. SLE data for three binary systems: methanol + DPC, dimethyl carbonate (DMC) + DPC, and phenol + DPC, were determined at atmospheric pressure by visually observing the melting points of a wide range of compositions. An experimental apparatus to allow both accurate control of temperature and visual observation of melting was designed for this study, and the experimental apparatus and procedure were verified by measuring the SLE of the system 2-methyl-1*H*-imidazole + 1-dodecanol. The binary mixtures DMC + DPC were simple eutectic systems, and the eutectic point was determined. From the experimental SLE for the two binary mixtures methanol + DPC and phenol + DPC, a solid addition compound containing a 1:1 ratio of methanol or phenol and DPC can be expected. The experimental SLE data of the three binary mixtures were represented by the semi-empirical equation of Ott and Goates. A nonrandom two-liquid (NRTL) model was also used for the data reduction of the systems DMC + DPC.

INTRODUCTION

Polycarbonate (PC) is a widely used engineering plastic which has excellent features, such as transparency, impact resistance, thermal stability, dimensional stability, and flame resistance. Therefore, PC has been widely applied in various applications including CDs and DVDs, electric and electronic appliances, automobiles, and mobile phones.[1−4] Most PC production has been via the phosgene process, which uses carbon monoxide and chloride as raw materials. However, this phosgene process has a number of environmental drawbacks.[1−5] In particular, highly toxic phosgene is used as a monomer, and a large quantity of the low-boiling-point solvent methylene chloride is required for polymerization. In addition, a very large quantity of water must be used to remove phosgene and chlorine (from methylene chloride) from the product PC. To overcome these environmental problems, a new process for PC production starting from carbon dioxide, ethylene oxide, and bisphenol A has been developed and has attracted much attention as it offers much greener and more sustainable chemistry.[1] Diphenyl carbonate (DPC) ($C_{13}H_{10}O_3$, CAS Registry No. 102-09-0) is an important intermediate material in this new process. The purity of the DPC directly influences the downstream PC quality, with high-purity DPC being critical for PC optical media applications. One of the options for purification of DPC is a crystallization process.[6−8] In the design and development of this crystallization process, knowledge of solid−liquid equilibrium (SLE) data is important.

This study aims to measure the SLE data for binary systems containing DPC. In this study, we have focused on the transesterification reaction for production of DPC. This reaction is based on methanol, dimethyl carbonate (DMC), phenol, and DPC. Thus, we have measured the SLE for three binary mixtures: methanol + DPC; DMC + DPC; and phenol + DPC, at atmospheric pressure by visually observing the melting points of a wide range of compositions. Before the measurements of these three binary mixtures, the experimental apparatus and procedure were checked by measuring the SLE for the system 2-methyl-1*H*-imidazole + 1-dodecanol, which covers a similar temperature range to that expected for the three binary mixtures. The experimental SLE data of the three binary mixtures were represented by a polynomial equation of the composition suggested by Ott and Goates.[9] The NRTL model[10] was also used to correlate the experimental data of those systems which showed simple eutectic behavior.

EXPERIMENTAL SECTION

Materials. The chemicals used in this study were: DPC (99 % purity), 2-methyl-1*H*-imidazole (99 % purity), and 1-dodecanol (98 % purity) from Sigma-Aldrich Co., Milwaukee, WI, United States; special grade methanol, first grade DMC, and special grade phenol (99 % purity) from Wako Pure Chemical Industry Ltd., Japan. Methanol and DMC were dried with molecular sieves: 3A for methanol and 13X for DMC and 1-dodecanol. Other substances were used without further purification. DPC and 2-methyl-1*H*-imidazole were dried at 323 K for 24 h under vacuum before use. The purities of methanol, DMC, and 1-dodecanol were checked by gas chromatography, and were found to be better than 99.9 % for methanol and DMC and better than 99 % for 1-dodecanol. The melting points and heats of fusion[7,8,11,12] of the compounds studied are summarized in Table 1, which also includes literature values of melting points[7,8,12−19] for comparison. As shown in Table 1, the experimental melting point temperatures for 2-methyl-1*H*-imidazole, DMC, and phenol are close to the literature values. In the case of DPC, differences among the literature values[7,8,18,19] are

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Table 1. Melting Point Temperature T_{fus} and Heat of Fusion $\Delta_{\mathrm{fus}}H$ of the Pure Components Studied

	T_{fus}/K		$\Delta_{\mathrm{fus}}H$
component	experimental	literature	kJ·mol^{-1}
2-methyl-1*H*-imidazole	419.38	419.00^{a}	-
		297.50^{a}	
1-dodecanol	296.51	296.84^{b}	31.38^{j}
		296.6^{c}	
methanol	-	175.47^{d}	3.175^{j}
DMC	278.16	278.20^{e}	11.58^{e}
phenol	313.99	314.05^{f}	11.28^{j}
		355.95^{g}	
DPC	353.96	353^{h}	24.30^{g}
		352.07^{i}	

[a] Ref 15. [b] Ref 16. [c] Ref 17. [d] Ref 13. [e] Ref 12. [f] Ref 14. [g] Refs 7 and 8. [h] Ref 18. [i] Ref 19. [j] Ref 11.

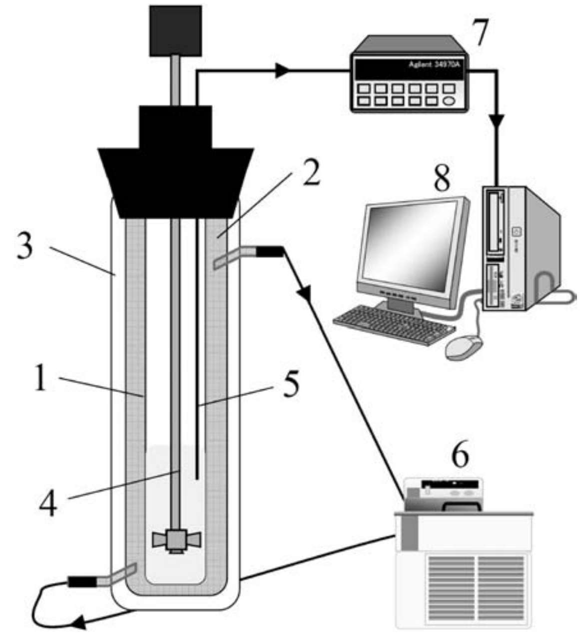

Figure 1. Schematic of the SLE measurement apparatus: 1, equilibrium cell; 2, heating jacket; 3, vacuum jacket; 4, stirrer; 5, thermometer; 6, thermostat; 7, data logger; 8, computer.

Table 2. Experimental SLE Data for the System 2-Methyl-1*H*-Imidazole (1) + 1-Dodecanol (2)

x_1^{L}	T/K	x_1^{L}	T/K	x_1^{L}	T/K
0.0000	296.51	0.3409	316.50	0.5981	372.22
0.0698	295.43	0.3570	322.17	0.6174	374.89
0.0863	295.01	0.3775	326.28	0.6224	375.29
0.1004	294.61	0.4075	334.48	0.6415	379.32
0.1495	293.64	0.4346	339.69	0.6554	381.93
0.1988	292.38	0.4514	343.69	0.7018	387.68
0.2427	291.28	0.4693	348.67	0.7523	393.38
0.2501^{a}	290.94^{a}	0.4922	351.83	0.7944	398.78
0.2574	292.59	0.5148	356.84	0.8467	404.70
0.2692	297.08	0.5305	359.37	0.8984	410.47
0.2837	301.21	0.5646	365.20	0.9493	414.17
0.2981	306.48	0.5826	369.11	1.0000	419.38
0.3123	310.40				

[a] Eutectic point.

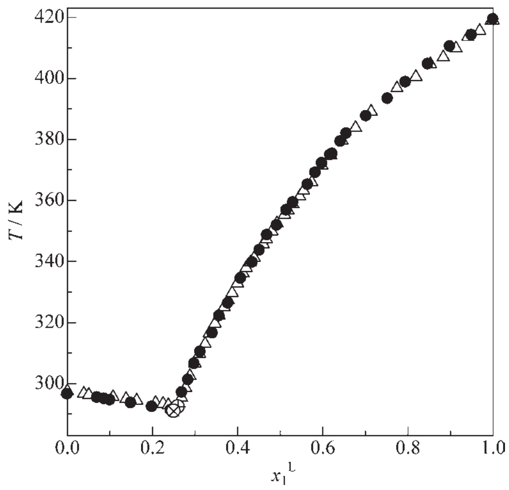

Figure 2. Experimental SLE data for the system 2-methyl-1*H*-imidazole (1) + 1-dodecanol (2). ●, this study; ⊗, eutectic point; △, Domańska and Kozlowska;[15] ⊕, eutectic point from Domańska and Kozlowska.[15]

observed. Our experimental melting point is in reasonable agreement with the data of Erman et al.[18] Our experimental melting point for 1-dodecanol is different to that reported by Domańska and Kozlowska in 2002[15] but is reasonably close to that of Domańska et al. in 2004 and 2011.[16,17]

Apparatus and Procedures. The SLE of the three binary systems were determined using a synthetic, visual technique. The experimental apparatus for measuring SLE was designed for this study and is shown in Figure 1. The apparatus consists of an equilibrium cell (1) with double jackets (heating (2) and vacuum (3)), stirrer (4), thermometer (5), thermostat (6), and data acquisition system (7, 8). Glass apparatus used consists of a double jacketed vessel. The volume of sample is about 30 cm^3. The vacuum cell (3) prevents the condensation of water vapor from air and thus enables visual observation inside the equilibrium cell (1). Temperature control in the equilibrium cell is performed by the heating jacket (2) controlled via the thermostat (5). The temperature is determined with a platinum resistance thermometer. The accuracy is estimated to be ± 0.01 K. The mole fraction is determined gravimetrically (Mettler digital balance model AX504 with a sensitivity of 0.1 mg and a maximum load of 510 g). The uncertainty in the mole fraction was estimated to be ± 0.0001.

For our measurements, a mixture (ca. 30 cm^3) with known composition was charged into the equilibrium cell. Next, the equilibrium cell was cooled by the thermostat, and the mixture was fully crystallized. The mixture was then heated slowly at a defined rate (3 K·h^{-1}). The melting point for a given composition is determined by visual detection of the temperature at which the solid phase just disappears. The uncertainty in melting temperature is estimated to be within ± 0.5 K.

RESULTS AND DISCUSSION

Prior to measurement of SLE for the mixtures containing DPC, the apparatus and procedure for SLE measurement designed in this study were checked by measuring a binary SLE available in the literature. A suitable test system must cover

Table 3. Experimental SLE Data for the System Methanol (1) + DPC (2)

x_1^{L}	T/K	x_1^{L}	T/K	x_1^{L}	T/K
0.0000	353.96	0.5415	327.43	0.8045	323.20
0.1005	346.00	0.5762	326.25	0.8074	322.83
0.2034	339.30	0.6589	325.22	0.8541	320.91
0.3029	335.53	0.6665	325.21	0.8634	319.66
0.3566	332.67	0.7054	324.03	0.8787	316.03
0.4034	329.84	0.7396	323.20	0.9003	313.16
0.4765	328.20	0.7658	323.22	0.9191	308.29
0.4947	328.15				

Table 4. Experimental SLE Data for the System DMC (1) + DPC (2)

x_1^{L}	T/K	x_1^{L}	T/K	x_1^{L}	T/K
0.0000	353.96	0.7059	306.69	0.9390	274.81
0.1148	347.03	0.7974	294.30	0.9447	275.45
0.2080	342.44	0.8999	275.12	0.9507	275.91
0.3037	337.98	0.9021	274.35	0.9602	276.51
0.4103	331.49	0.9050	273.60	0.9695	277.08
0.5112	324.37	0.9073[a]	273.42[a]	0.9901	277.69
0.6028	316.65	0.9202	273.60	1.0000	278.16

[a] Eutectic point.

Table 5. Experimental SLE Data for the System Phenol (1) + DPC (2)

x_1^{L}	T/K	x_1^{L}	T/K	x_1^{L}	T/K
0.0000	353.96	0.5053	325.70	0.8902	307.23
0.1021	348.53	0.5200	325.10	0.8981	306.76
0.2014	343.03	0.5489	325.09	0.8989[a]	306.63[a]
0.2996	337.52	0.5992	323.79	0.9001	306.85
0.4000	332.13	0.6493	322.48	0.9048	307.44
0.4553	328.29	0.6996	320.69	0.9201	308.63
0.4808	326.46	0.7518	317.99	0.9300	309.29
0.4905	325.88	0.7979	314.52	0.9498	310.43
0.4952[a]	325.61[a]	0.8484	309.98	1.0000	313.99
0.4999[b]	325.72[b]				

[a] Eutectic point. [b] 1:1 (phenol: DPC) congruently melting solid addition compound.

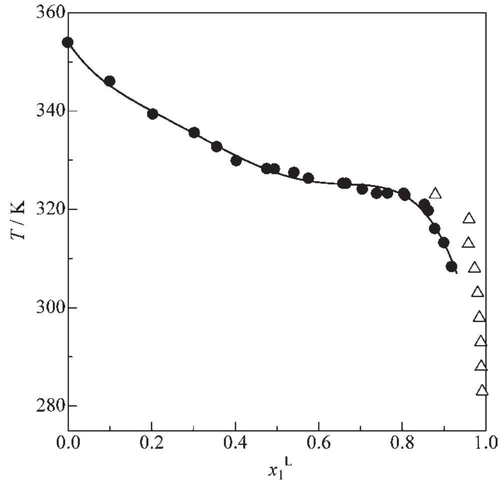

Figure 3. Experimental SLE data for the system methanol (1) + DPC (2). ●, this study; △, Wei and Pei;[7,8] —, eq 1.

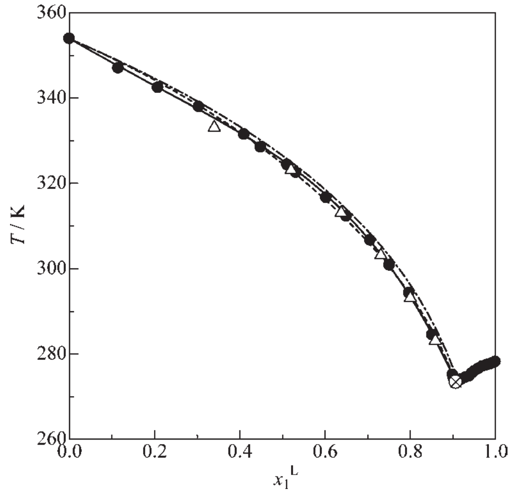

Figure 4. Experimental SLE data for the system DMC (1) + DPC (2). ●, this study; ⊗, eutectic point; △, Li et al.;[21] —, eq 1; - - -, NRTL; —•—, ideal solution.

a wide temperature range, since the temperature range of melting points for the three binary mixtures containing DPC is expected to vary widely, considering the melting points of pure components used. Therefore, in this study, the system 2-methyl-1*H*-imidazole + 1-dodecanol, with a temperature range of SLE from (292 to 419) K, was selected. The SLE data of this mixture have been reported by Domańska and Kozlowska.[15] The experimentally determined melting point temperatures for the system 2-methyl-1*H*-imidazole (1) + 1-dodecanol (2) are listed in Table 2, and a comparison of these data with literature values is presented in Figure 2. Our experimentally determined eutectic point is $x_1^{\mathrm{L}} = 0.2501$ and $T = 290.94$ K, which agrees reasonably well with that found by Domańska and Kozlowska ($x_1^{\mathrm{L}} = (0.26 \pm 0.02)$ and $T = (292.35 \pm 0.1)$ K). In particular, a good agreement between the two data sets was obtained for the composition of the eutectic point. The composition of our experimental eutectic point is within the error of that of the literature value. On the other hand, in the 1-dodecanol rich region, which contains the eutectic point, our experimental melting temperatures were up to 1.4 K lower than the literature values. This discrepancy is consistent with that for the melting point of 1-dodecanol mentioned in the Experimental Section. Therefore, the difference in the melting point of 1-dodecanol seems to be due to this difference between the experimental and literature SLE data. In view of these considerations, the experimental apparatus designed in this study was applied for measurements of SLE in three binary mixtures containing DPC.

The experimental values of SLE in the three binary mixtures: methanol (1) + DPC (2), DMC (1) + DPC (2), and phenol (1) + DPC (2), are listed in Tables 3 to 5, respectively. These experimental SLE data are plotted in Figures 3 to 5 along with the literature values. For the mixture methanol (1) + DPC (2), the SLE data at mole fraction of methanol x_1^{L} over 0.88 (temperature range: (283 to 323) K) have been reported by Wei and Pei.[7,8] However, data for mole fraction of methanol $x_1^{\mathrm{L}} =$ (0 to 0.87) are not available. Therefore, in this study, we have measured the SLE data of this system in the region of $x_1^{\mathrm{L}} =$ (0 to

0.92). According to a report by Hoyle et al.,[20] DPC may aggregate in polar solvents such as methanol. Therefore, this mixture also might form the molecular aggregates. In addition, as well as the binary system phenol + DPC mentioned later, a 1:1 (methanol: DPC) congruently melting solid addition compound might form, because the temperature change of the experimental melting point is slightly different before and after $x_1^L = 0.5$. However, in this study, a characterization of the structure of the solid phase could not be performed. Comparing our experimental values to those of Wei and Pei,[7,8] we have observed a decrease in temperature of the SLE from $x_1^L = 0.81$, while Wei and Pei observed a decrease from $x_1^L = 0.88$. Data by Wei and Pei are measured by a static analytical method, and the compositions of the SLE are determined by a spectrophotometric technique. In the methanol rich region of this mixture, the SLE temperatures decrease steeply with increasing x_1^L. Therefore, it would appear that the choice of experimental method influenced the data obtained.

Next, for the DMC (1) + DPC (2) system, the SLE data over the entire composition range were determined. From the experimental results, this system is identified as a simple eutectic system, as found for the system methanol (1) + DPC (2). The calculated results of the SLE assuming ideal solubility are also shown in Figure 4. The SLE of this system shows nearly ideal behavior. On the basis of the experimental SLE data, the eutectic point ($x_1^L = 0.9073$, $T = 273.42$ K) was determined. Li et al.[21] have reported values in the region $x_1^L = (0.34 \text{ to } 0.86)$. When our experimental SLE data are compared with the data by Li et al., good agreement is obtained.

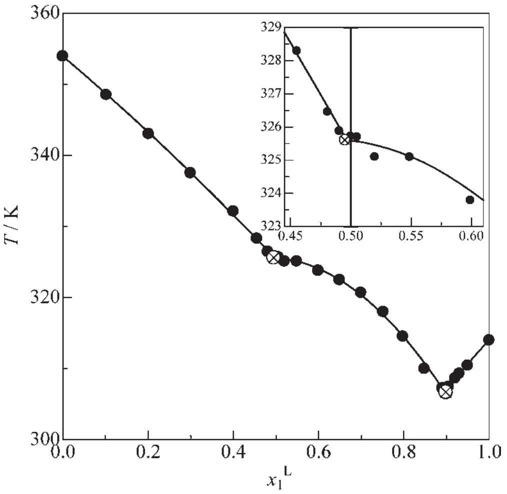

Figure 5. Experimental SLE data for the system phenol (1) + DPC (2). ●, this study; ⊗, eutectic point; —, eq 1.

Finally, for the system phenol (1) + DPC (2), melting point temperatures were also obtained over entire composition range (Figure 5). In Figure 5, an expansion of the region around $x_1^L = 0.5$ is also shown. As expected from Figure 5, this mixture has two eutectic points, and a maximum temperature value is obtained at $x_1^L = 0.4999$. These results suggest that a 1:1 (phenol/DPC) congruently melting solid addition compound forms. This behavior is consistent with previous literature by Shafer et al.[22]

DATA REDUCTION

Polynomial Equation. The experimental SLE data for the three binary mixtures were correlated using the semi-empirical equation of Ott and Goates:[9]

$$T = T^* \left[1 + \sum_{j=1}^{\mathrm{NP}} a_j (x_1 - x_1^*)^j\right] \tag{1}$$

where x_1 is the mole fraction of the first component, T is the SLE temperature of the binary mixture, T^* is the melting temperature of the pure substance or eutectic point, x_1^* is the value of mole fraction at which $T = T^*$, a_j are the coefficients to be determined by regression of the experimental SLE data, and NP is the number of a_j. The coefficients, a_j, of eq 1 were estimated so that the following objective function (F_{obj}) was minimized for each system by means of the Marquardt algorithm:[23]

$$F_{\mathrm{obj}} = \sum_{k=1}^{\mathrm{NDP}} (T_{\mathrm{exptl}} - T_{\mathrm{calcd}})_k^2 \tag{2}$$

where T_{exptl} and T_{calcd} are the experimental and calculated melting point temperature, respectively, and NDP is the number of data points per system. The determined parameters and absolute deviations of the liquid mole fraction obtained with eq 1 are summarized in Table 6.

Table 6. Determined Parameters and Deviations between Experimental and Calculated SLE of the Three Binary Systems Using eq 1

$x^L_{1,\mathrm{min}}$	$x^L_{1,\mathrm{max}}$	x_1^*	T^*/K	a_1	a_2	a_3	a_4	a_5	$\lvert\Delta x_1^L\rvert_{\mathrm{av}}$[a]
				Methanol (1) + DPC (2) System					
0.0000	0.9191	0.0000	353.96	−0.3541	1.4288	−4.2848	5.9602	−2.9480	0.0183
								overall	0.0183
				DMC (1) + DPC (2) System					
0.0000	0.9073	0.0000	353.96	−0.1597	0.0135	0.0812	−0.2281		0.0034
0.9073	1.0000	1.0000	278.16	0.0120	−4.8625	−31.9557			0.0026
								overall	0.0031
				Phenol (1) + DPC (2) System					
0.0000	0.4952	0.0000	353.96	−0.1436	−0.0356				0.0025
0.4952	0.8989	0.4952	325.61	−0.0102	−0.3357	−0.0007			0.0073
0.8989	1.0000	1.0000	313.99	0.1984	−0.2744				0.0016
								overall	0.0045

[a] $\lvert\Delta x_1^L\rvert_{\mathrm{av.}} = \sum_{k=1}^{\mathrm{NDP}} \lvert x^L_{1,\mathrm{exptl}} - x^L_{1,\mathrm{calcd}}\rvert_k/\mathrm{NDP}$, where NDP is the number of data points.

NRTL. In the binary system with simple eutectic mixtures, DMC + DPC, the NRTL model was applied for correlation of the experimental SLE data. In this study, we adopted the approximated expression of the condition of the SLE of eutectic systems,[24] which does not consider the difference in the molar heat capacities of the pure components $\Delta C_{P,i}$:

$$\ln x_i^{\mathrm{L}} = -\frac{\Delta_{\mathrm{fus}} H_i}{RT}\left(1 - \frac{T}{T_{\mathrm{fus},\,i}}\right) - \ln \gamma_i^{\mathrm{L}} \qquad (3)$$

where x_i^{L} and γ_i^{L} are the mole fraction and activity coefficient, respectively, of component i in the liquid phase, $\Delta_{\mathrm{fus}}H_i$ and $T_{\mathrm{fus},i}$ are the molar enthalpy of fusion and melting temperature of component i, T is the temperature, and R is the gas constant. In this study, the experimental results of $T_{\mathrm{fus},i}$ and the literature values of $\Delta_{\mathrm{fus}}H_i$ were used. Those values are listed in Table 1. The activity coefficient in the liquid phase, γ_i^{L}, was expressed by the NRTL model. A constant value of 0.2 was adopted for α_{12}, the nonrandomness parameter of the NRTL model. Binary interaction parameters $g_{ij} - g_{jj}$ for the NRTL model were determined by using the following objective function:

$$F_{\mathrm{obj}} = \sum_{k=1}^{\mathrm{NDP}} \left(\frac{\gamma_{i,\,\mathrm{exptl}}^{\mathrm{L}} - \gamma_{i,\,\mathrm{calcd}}^{\mathrm{L}}}{\gamma_{i,\,\mathrm{exptl}}^{\mathrm{L}}}\right)_k^2 \qquad (4)$$

where $\gamma_{\mathrm{exptl},i}^{\mathrm{L}}$ and $\gamma_{\mathrm{calcd},i}^{\mathrm{L}}$ are the experimental and calculated γ_i^{L}, respectively. The estimated parameters are $g_{12} - g_{22} = 7.29384 \cdot 10^3$ $\mathrm{J \cdot mol^{-1}}$ and $g_{21} - g_{11} = -4.64671 \cdot 10^3$ $\mathrm{J \cdot mol^{-1}}$, and the absolute deviations of the liquid mole fraction $|\Delta x_1^{\mathrm{L}}|_{\mathrm{av}}$ is 0.0070. The correlated results of the NRTL model are shown in Figures 2 and 3. The deviations $|\Delta x_1^{\mathrm{L}}|_{\mathrm{av}}$ indicate that the NRTL model gives reasonable correlations with the experimental results.

CONCLUSIONS

The SLE data for three binary mixtures containing DPC: methanol + DPC, DMC + DPC, and phenol + DPC, were determined by visually observing the melting points of a wide range of compositions. In the mixtures DMC + DPC, a simple eutectic system was formed, and the eutectic point was determined from the experimental SLE data. Formation of a molecular aggregate or a solid addition compound with a 1:1 ratio of methanol or phenol and DPC was expected from the experimental SLE data for two systems methanol + DPC and phenol + DPC. Further investigation for a confirmation of the structure of the solid phase of methanol + DPC is required in the future. The experimental SLE data for the three binary mixtures were correlated by the semi empirical equation of Ott and Goates. For the system DMC + DPC, the NRTL model was also used for data reduction. Reasonable correlation accuracy was obtained with both models.

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REFERENCES

(1) Fukuoka, S.; Kawamura, M.; Komiya, K.; Tojo, M.; Hachiya, H.; Hasegawa, K.; Aminaka, M.; Okamoto, H.; Fukawa, I.; Konno, S. A Novel Non-Phosgene Polycarbonate Production Process Using By-product CO_2 as Starting Material. *Green Chem.* **2003**, *5*, 497–507.

(2) Haubrock, J.; Raspe, M.; Versteeg, G. F.; Kooijman, H. A.; Taylor, R.; Hogendoorn, J. A. The Reaction from Dimethyl Carbonate to Diphenyl Carbonate. 1. Experimental Determination of the Chemical Equilibria. *Ind. Eng. Chem. Res.* **2008**, *47*, 9854–9861.

(3) Haubrock, J.; Wermink, W.; Versteeg, G. F.; Kooijman, H. A.; Taylor, R.; van Sint Annaland, M.; Hogendoorn, J. A. Reaction from Dimethyl Carbonate (DMC) to Diphenyl Carbonate (DPC). 2. Kinetics of the Reactions from DMC via MPC to DPC. *Ind. Eng. Chem. Res.* **2008**, *47*, 9862–9870.

(4) Gong, J.; Ma, X.; Wang, S. Phosgene-Free Approaches to Catalytic Synthesis of Diphenyl Carbonate and its Intermediates. *Appl. Catal., A* **2007**, *316*, 1–21.

(5) Ono, Y. Catalysis in the Production and Reactions of Dimethyl Carbonate, an Environmentally Benign Building Block. *Appl. Catal., A* **1997**, *155*, 133–166.

(6) Kuze, S.; Okumura, R.; Suwabe, Y. Preparation of Polycarbonate by Melt Method without Discoloration. Eur. Patent EP0561363, 1993.

(7) Wei, D. W.; Pei, Y. H. Measurement and Correlation of Solubility of Diphenyl Carbonate in Alkanols. *Ind. Eng. Chem. Res.* **2008**, *47*, 8953–8956.

(8) Wei, D. W.; Pei, Y. H. Solubility of Diphenyl Carbonate in Pure Alcohols from (283 to 333) K. *J. Chem. Eng. Data* **2008**, *53*, 2710–2711.

(9) Ott, J. B.; Goates, J. R. Solid + Liquid) Phase Equilibria in Binary Mixtures Containing Benzene, a Cycloalkane, an *n*-Alkane, or Tetrachloromethane an Equation for Representing (Solid + Liquid) Phase Equilibria. *J. Chem. Thermodyn.* **1983**, *15*, 267–278.

(10) Renon, H.; Prausnitz, J. M. Local Compositions in Thermodynamic Excess Functions for Liquid Mixtures. *AIChE J.* **1968**, *14*, 135–144.

(11) *The Dortmund Data Bank (DDB)*, DDBST Software and Separation Technology; GmbH: Oldenburg, Germany, 2010.

(12) Ding, M. S. Liquid-Solid Phase Equilibria and Thermodynamic Modeling for Binary Organic Carbonates. *J. Chem. Eng. Data* **2004**, *49*, 276–282.

(13) Riddick, J. A.; Bunger, W.; Sakano, T. K. *Organic Solvents Physical Properties and Methods of Purification*, 4th ed.; John Wiley & Sons: New York, 1986.

(14) Andon, R. J. L.; Counsell, J. F.; Herington, E. F. G.; Martin, J. F. Thermodynamic Properties of Organic Oxygen Compounds. Part 7. — Calorimetric Study of Phenol from 12 to 330 K. *Trans. Faraday Soc.* **1963**, *59*, 830–835.

(15) Domańska, U.; Kozlowska, M. Solubility of Imidazoles in Alcohols. *J. Chem. Eng. Data* **2002**, *47*, 8–16.

(16) Domańska, U.; Bogel-Łukasik, E. Solid-Liquid Equilibria for Systems Containing 1-Butyl-3-Methylimidazolium Chloride. *Fluid Phase Equilib.* **2004**, *218*, 123–129.

(17) Domańska, U.; Paduszyński, K.; Dąbska, J. Measurements, Correlations, and Mod. UNIFAC (Do) Prediction of (Solid-Liquid) Phase Equilibria Diagrams in Binary Systems (Aliphatic Ketone + an Alcohol). *J. Chem. Eng. Data*, in press.

(18) Erman, B.; Marvin, D. C.; Irvine, P. A.; Flory, P. J. Optical Anisotropies of Model Analogues of Polycarbonates. *Macromolecules* **1982**, *15*, 664–669.

(19) Sinke, G. C.; Hildenbr, D. L.; McDonald, R. A.; Kramer, W. R.; Stull, D. R. The Heat, Entropy and Free Energy of Formation of Diphenyl Carbonate. *J. Phys. Chem.* **1958**, *62*, 1461–1462.

(20) Hoyle, C. E.; Rufus, I. B.; Shah, H. Solvent effect on the photophysics of bisphenol-A-based polycarbonate and diphenylcarbonate. *Can. J. Chem.* **1995**, *73*, 2062–2068.

(21) Li, W.; Xuan, A.; Wu, Y.; Ai, J.; Liang, L. Measurement and Correlation of Solid-Liquid Equilibrium of Diphenyl Carbonate in Ethanol-Diethyl Carbonate. *Huagong Xuebao* **2009**, *60*, 1357–1361.

(22) Shafer, S. J.; Park, C.; Pressman, E. J. Method of Purifying Diphenyl Carbonate-Phenol Adduct. U.S. Patent 5,756,801, 1998.

(23) Marquardt, D. W. An Algorithm for Least-Squares Estimation of Nonlinear Parameters. *Soc. Ind. Appl. Math.* **1963**, *11*, 431–441.

(24) Gmehling, J.; Kolbe, B. *Thermodynamik*; VCH: Weinheim, Germany, 1992.