Phase Diagrams for Liquid−Liquid and Liquid−Solid Equilibrium of the Ternary Poly(ethylene glycol) Dimethyl Ether 2000 + Sodium Carbonate + Water System

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The complete phase diagram for the poly(ethylene glycol) dimethyl ether 2000 ($PEGDME_{2000}$) + Na_2CO_3 + H_2O system at 298.15 K was determined. Experimental liquid−liquid equilibrium phase diagrams, tie lines, and plait points were obtained for the ternary system. Compositions of the liquid−liquid and the liquid−liquid−solid equilibria were determined from calibration curves of refractive index of the solutions, and atomic absorption (AA) and X-ray diffraction analyses were made on the solids. Binodal curves were described using the Merchuk equation at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K, and the fitting parameters were obtained for the corresponding temperatures. The effects of temperature on the binodal curve were also studied, and it was observed that the area of the biphasic region increased slightly with increase in temperature. Also, the tie lines were fitted to both the Othmer−Tobias and Bancroft and Setschenow-type equations. Correlation coefficients for all equations are reported.

Introduction

The aqueous two-phase partitioning method of liquid−liquid extraction is useful for separating materials of biological origin.[1] The aqueous two-phase systems (ATPSs) can be formed by combining either two incompatible polymers or a polymer and a salt in water above a certain critical concentration.[1−3] Poly(ethylene glycol) (PEG), which is a hydrophilic polymer, is often used in aqueous two-phase partitioning studies. In laboratory-scale separations, the most commonly used aqueous systems are comprised of the polymers PEG and dextran, while for large-scale enzyme extraction, aqueous PEG + salt systems are used.[4] The later systems are more attractive because of their greater selectivity, lower viscosity, lower cost, rapid phase disengagement, and availability of commercial separators, which allow a faster and continuous protein separation.[5,6] The most common inorganic salts employed by different research groups on the aqueous two-phase extraction technique consist of selective cations (ammonium, potassium, or sodium) and anions (phosphate, sulfate, or carbonate).

Zaslavsky[3] presented a useful summary of experimental liquid−liquid data and equilibrium diagrams for systems formed by PEG, inorganic salts, and water. The principal inorganic salts discussed include ammonium phosphate, ammonium, sodium, magnesium sulfates, potassium and sodium carbonates, and sodium and potassium hydroxides.[3] However, there are few data published on the complete phase diagram consisting of liquid−liquid and liquid−solid equilibrium in the literature for aqueous systems containing a polymer and a salt. There are reports on the complete phase diagrams for a few aqueous PEG + salt systems containing Na_2SO_4[7,8] and Na_2CO_3.[9] Also, we reported experimental data and the complete phase diagram for the poly(ethylene glycol) dimethyl ether 2000 ($PEGDME_{2000}$) + Na_2SO_4 system in our previous work.[10]

This study presents liquid−liquid equilibrium data for the new aqueous poly(ethylene glycol) dimethyl ether 2000 ($PEGDME_{2000}$) (1) + Na_2CO_3 (2) + H_2O (3) system at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K that have not been published previously. The effect of temperature on the binodal curve was studied. In this work, the complete phase diagram of the $PEGDME_{2000}$ (1) + Na_2CO_3 (2) + H_2O (3) system has also been studied at T = 298.15 K. The obtained results are necessary for design and optimization of extraction and crystallization processes. Ethylene glycol, diethylene glycol, and also PEG have been used as antisolvents for crystallization of Na_2CO_3, previously.[11,12] It seems that the information provided in this work may be useful for crystallization of Na_2CO_3 by addition of $PEGDME_{2000}$ as an antisolvent.

Experimental Section

Materials. Poly(ethylene glycol) dimethyl ether 2000 ($PEGDME_{2000}$) and sodium carbonate (Na_2CO_3) with a purity of min. 99 % by mass were supplied from Merck, Germany. The $PEGDME_{2000}$ and Na_2CO_3 were used without further purification, and double distilled deionized water was used. Previously,[10] the number average molar mass and polydispersity of the $PEGDME_{2000}$ were determined to be 2305 $g \cdot mol^{-1}$ and 1.0579, respectively, using gel permission chromatography (GPC, Agilent, USA).

Apparatus and Procedure. The experimental apparatus employed is essentially similar to the one used previously.[13,14] A glass vessel, volume 50 cm^3, was used to carry out the phase equilibrium determinations. This glass vessel was provided with an external jacket to circulate water using a thermostat. The temperature was controlled within ± 0.05 K. Sodium carbonate (1.5 $mol \cdot kg^{-1}$) stock solutions were prepared by dissolving appropriate amounts of solid in distilled water. Stock solutions were not kept for a long time and used right away as provided. The binodal curves were determined by the cloud point method. A sodium carbonate solution of known concentration was titrated with the $PEGDME_{2000}$ solution or vise versa, until the solution turned turbid. An analytical balance (Shimadzu, 321-34553,

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Table 1. Binodal Data for the $PEGDME_{2000}$ (1) + Na_2CO_3 (2) + H_2O (3) System at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K

T = 288.15 K		T = 293.15 K		T = 298.15 K		T = 308.15 K		T = 318.15 K	
100 w_1	100 w_2	100 w_1	100 w_2	100 w_1	100 w_2	100 w_1	100 w_2	100 w_1	100 w_2
25.78	3.55	25.34	3.55	27.58	3.30	27.88	3.01	22.54	3.70
21.55	4.20	21.26	4.20	22.21	4.01	20.44	4.02	19.30	4.10
16.71	5.10	17.20	4.88	17.17	4.83	14.84	5.03	15.38	4.66
13.19	5.94	13.67	5.72	12.26	5.90	10.13	6.06	11.91	5.23
9.83	6.77	9.74	6.66	8.23	6.84	7.48	6.66	8.82	5.82
6.83	7.58	6.47	7.53	5.59	7.57	5.26	7.25	6.29	6.37
5.57	8.00					3.91	7.70	4.45	6.89
								3.13	7.37
								2.34	7.74

Table 2. Experimental Phase Equilibrium Compositions for the $PEGDME_{2000}$ (1) + Na_2CO_3 (2) + H_2O (3) System at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K

top phase: polymer-rich phase		bottom phase: salt-rich phase	
100 w_1	100 w_2	100 w_1	100 $_2$
	T = 288.15 K		
28.10	3.16	2.74	10.94
31.88	2.64	2.36	12.37
35.15	2.17	1.97	13.83
38.39	1.88	1.50	15.37
41.17	1.60	1.34	17.09
43.29	1.43	1.29	18.65
	T = 293.15 K		
28.47	3.01	3.04	10.71
32.65	2.57	2.58	12.60
35.11	2.31	2.31	13.59
38.79	2.01	2.10	14.77
42.53	1.72	1.72	16.03
44.65	1.61	1.82	17.35
	T = 298.15 K		
30.72	2.74	2.41	10.88
33.66	2.42	1.86	12.50
38.70	1.93	1.50	15.28
41.73	1.75	1.18	16.35
45.09	1.50	1.15	17.66
	T = 308.15 K		
31.48	2.60	1.80	10.12
34.59	2.30	1.23	11.61
38.31	2.10	1.17	13.04
46.00	1.74	1.27	15.02
48.23	1.68	1.18	16.46
	T = 318.15 K		
26.66	3.20	1.33	8.98
32.43	2.53	0.97	10.43
36.21	2.20	0.82	11.48
40.11	1.87	0.80	12.88
43.99	1.65	0.71	14.13

Shimadzu Co., Japan) with a precision of ± $1 \cdot 10^{-7}$ kg was used. In accordance with the amount of titrant added until turbidity was observed, the composition of the mixture for each point on the binodal curve was calculated by mass. The maximum uncertainty of mass fraction was found to be 0.002 for both $PEGDME_{2000}$ and Na_2CO_3 by the titration method used. For the determination of the tie-lines, feed samples (about $2 \cdot 10^{-5}$ m^3) were prepared by mixing appropriate amounts of $PEGDME_{2000}$, Na_2CO_3, and H_2O in the vessel. The thermostat was set at a desired temperature, and the samples were stirred for 1 h. Then the samples were placed in thermostat and allowed to settle for at least 48 h to separate into two clear phases. The concentrations of Na_2CO_3 in phases were determined by flame photometry. The concentration of $PEGDME_{2000}$ in both phases was determined by refractive index measurements performed at 298.15 K using a refractometer (ATAGO DR-A1, Japan) with a precision of ± 0.0001. The uncertainty in refractive index measurement method is within ± 0.0002. For dilute aqueous solutions containing a polymer and a salt, the relation between the refractive index, n_D, and the mass fractions of polymer, w_1, and salt, w_2, is given by[15]

$$n_D = n_0 + a_1 w_1 + a_2 w_2 \tag{1}$$

here n_0 is the refractive index of pure water which is set to 1.3325 at 298.15 K. The values of coefficients a_1 and a_2 for the applied system were obtained at 298.15 K as 0.1354 and 0.2136, respectively. However, it should be noted that this equation is only valid for dilute solutions ($w_1 \leq 0.1$ and $w_2 \leq 0.05$) for which linear calibration plots of the refractive index of ternary solutions with a fixed mass fraction of salt or polymer against polymer or salt are obtained. Therefore, before refractive index measurements, it was necessary to dilute the samples to the above mass fraction range. The uncertainty of the mass fraction of $PEGDME_{2000}$ achieved using eq 1 was better than 0.002.

Various series of solutions of the three components were prepared to delimit the three-phase region from the two-phase region. Two liquids and one solid phase coexist in equilibrium in this zone. Once the equilibrium was obtained, the samples of both liquid phases (top and bottom) were analyzed for refractive index and composition. The solid phase was dried at room temperature and analyzed by X-ray diffraction using an automatic, computerized X-ray diffractometer (Siemens Co., model D5000), which showed the solid phase to be monohydrated sodium carbonate.

Results and Discussion

Correlation and Phase Diagrams. For the $PEGDME_{2000}$ (1) + Na_2CO_3 (2) + H_2O (3) system, the binodal data and the tie-line compositions determined experimentally at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K are given in Tables 1 and 2, respectively.

For the studied system, the experimental binodal data were fitted by the least-squares regression method to the following empirical nonlinear expression developed by Merchuk[16]

$$w_1 = a \exp(b w_2^{0.5} - c w_2^3) \tag{2}$$

where a, b, and c are fit parameters. Recently, the above equation has been successfully used for the correlation of binodal data of some aqueous polymer + salt systems.[17,18] Parameters a, b, and c obtained from the correlation of experimental binodal data along with the corresponding standard deviations are given in Table 3. On the basis of obtained standard deviations, we conclude that eq 2 can be satisfactorily used to reproduce the binodal curves of the investigated system. The binodals reproduced from eq 2 at (288.15 and 318.15) K are shown in Figure 1.

The effect of temperature on the phase-forming ability for the studied system is also illustrated in Figure 1. The locus for the experimental binodals shown in Figure 1 demonstrates that by increasing the temperature the two-phase area is expanded which attributed to the decrease in solubility or increase in

Table 3. Values of Parameters of Equation 2 for $PEGDME_{2000}$ (1) + Na_2CO_3 (2) + H_2O (3) at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K

T/K	a	b	10^3c	sd[a]
288.15	1.0288	−6.9376	1.8135	0.16
293.15	1.0785	−7.2567	1.8572	0.19
298.15	1.2767	−8.0566	2.0104	0.19
308.15	1.0101	−7.0136	2.7306	0.21
318.15	1.2116	−7.7352	3.8729	0.05

[a] $\mathrm{sd} = (\sum_{i=1}^{N}[(w_{1,i}^{\mathrm{cal}} - w_{1,i}^{\mathrm{exp}})])/N)^{0.5}$, where N is the number of binodal data. The superscripts "cal" and "exp" refer to the calculated and experimental values, respectively.

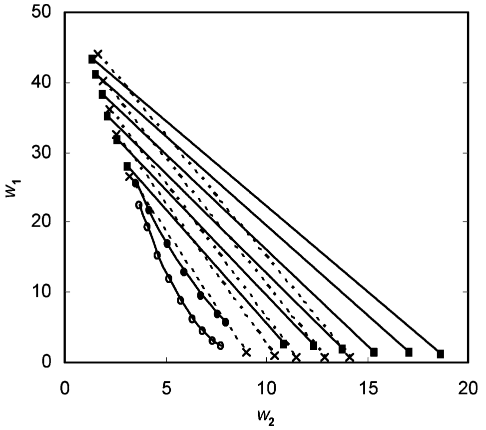

Figure 1. Effect of temperature on the equilibrium phase compositions for the $PEGDME_{2000}$ (1) + sodium carbonate (2) + H_2O (3) system: ●, experimental binodal at 288.15 K; ○, experimental binodal at 318.15 K; —, calculated binodal from eq 2; —■—, tie-lines at 288.15 K; ---×---, tie-lines at 318.15 K. These tie-lines were obtained by connecting the experimental equilibrium phase composition data.

Table 4. Values of Parameters from Least-Squares Linear Fits to the Setschenow-Type Equation (Equation 3) for $PEGDME_{2000}$ (1) + Na_2CO_3 (2) + H_2O (3) at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K

T/K	k_s/(kg·mol^{-1})	intercept	R^2	dev[a]
288.15	1.1648	1.7359	0.9666	0.08
293.15	1.2454	1.5465	0.9724	0.04
298.15	1.2907	1.8181	0.9838	0.03
308.15	1.6772	2.4958	0.9584	0.06
318.15	2.7910	0.9987	0.9555	0.11

[a] $\mathrm{dev} = (\sum_p\sum_l\sum_j(w_{p,l,j}^{\mathrm{cal}} - w_{p,l,j}^{\mathrm{exp}})^2/6N)$, where $w_{p,l,j}$ is the mass fraction of the component j (i.e., polymer, salt, or water) in the phase p for the lth tie-line. N is the number of tie-line data.

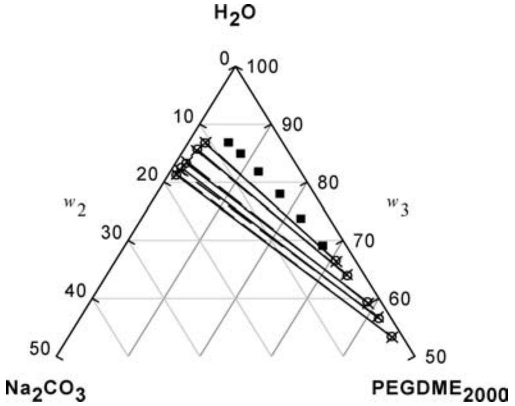

Figure 2. Phase diagram of the $PEGDME_{2000}$ (1) + sodium carbonate (2) + H_2O (3) two-phase system at 298.15 K: ■, experimental binodal; —○—, experimental tie-lines; ---×---, calculated tie-lines using eq 3.

phase-forming ability in the studied system. Additionally, to show the effect of temperature on the equilibrium phase compositions for the investigated system, the experimental tie-lines are compared in Figure 1, for the temperatures (288.15 and 318.15) K, as an example. As shown in Figure 1, the slope and the length of the tie-lines increase with an increase in temperature, similar to the ones observed for the aqueous PEG + trisodium citrate[19] and $PEGDME_{2000}$ + sodium sulfate[10] systems.

For the correlation of LLE data of polymer + salt + water systems, several models have been developed. However, in this work we decided to use a relatively simple two-parameter equation, which can be derived from the binodal theory.[20] This is a Setschenow-type equation and has the following form

$$\ln\left(\frac{C_{\mathrm{P}}^{\mathrm{top}}}{C_{\mathrm{P}}^{\mathrm{bot}}}\right) = k_{\mathrm{p}}(C_{\mathrm{P}}^{\mathrm{bot}} - C_{\mathrm{P}}^{\mathrm{top}}) + k_{\mathrm{s}}(C_{\mathrm{s}}^{\mathrm{bot}} - C_{\mathrm{s}}^{\mathrm{top}}) \tag{3}$$

where k_s is the salting-out coefficient; k_p is the constant related to the activity coefficient; C_P is the concentration of the polymer; and C_s is the concentration of the salt. Superscripts "top" and "bot" stand for the polymer-rich phase and the salt-rich phase, respectively. Recently we successfully used eq 3 for the correlation of tie-line data for the aqueous poly(propylene glycol) + potassium citrate[18] and $PEGDME_{2000}$ + sodium sulfate two-phase systems.[10] The fitting parameters of eq 3 along with the corresponding standard deviations are presented in Table 4 for the investigated system. On the basis of standard deviations reported in Table 4, it is interesting to note that eq 3 with only two parameters represents the experimental LLE data with good accuracy for the studied system at different temperatures. To show the reliability of eq 3, comparison between the experimental and correlated tie-lines is shown in Figure 2 for the $PEGDME_{2000}$ (1) + Na_2CO_3 (2) + H_2O (3) system at 298.15 K, as an example.

The correlation equations[21] given by Othmer−Tobias (eq 4a) and Bancroft (eq 4b) have also been used to correlate the tie-line compositions.

$$\left(\frac{1 - w_1^{\mathrm{top}}}{w_1^{\mathrm{top}}}\right) = k\left(\frac{1 - w_2^{\mathrm{bot}}}{w_2^{\mathrm{bot}}}\right)^n \tag{4a}$$

$$\left(\frac{w_3^{\mathrm{bot}}}{w_2^{\mathrm{bot}}}\right) = k_1\left(\frac{w_3^{\mathrm{top}}}{w_1^{\mathrm{top}}}\right)^r \tag{4b}$$

where k, n, k_1, and r represent fit parameters. These equations have also been used to assess the reliability of LLE data. A linear dependency of the plots $\log((1 - w_1^{\mathrm{top}})/w_1^{\mathrm{top}})$ against $\log((1 - w_2^{\mathrm{top}})/w_2^{\mathrm{bot}})$ and $\log(w_3^{\mathrm{bot}}/w_2^{\mathrm{bot}})$ against $\log(w_3^{\mathrm{top}}/w_1^{\mathrm{top}})$ indicated an acceptable consistency of the results. The corresponding correlation coefficient values, R, and the values of the fitted parameters together with the standard deviations are given in Table 5. On the basis of the obtained standard deviations, we conclude that eqs 4a and 4b can also be satisfactorily used to correlate the tie-line data of the investigated system.

The location of the plait point for the studied system at each working temperature was also estimated by extrapolation from the auxiliary curve satisfactorily fitted with the following linear equation

$$w_1 = f + gw_2 \tag{5}$$

where f and g represent fit parameters. For the studied system, the estimated values for plait points along with the obtained

Table 5. Values of Parameters of Equation 4a and Equation 4b for PEGDME (1) + Na_2CO_3 (2) + H_2O (3) at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K

T/K	k	n	R^2	k_1	r	R^2	dev[a]
288.15	0.24700	1.10807	0.994	3.56158	0.90276	0.993	0.17
293.15	0.16718	1.28472	0.992	4.06333	0.76246	0.992	0.31
398.15	0.25815	1.03683	0.985	3.77079	0.94894	0.985	0.40
308.15	0.13395	1.28756	0.981	4.87835	0.75175	0.983	0.61
318.15	0.07972	1.52493	0.997	5.29461	0.66374	0.997	0.19

[a] Definition is given in Table 4.

Table 6. Values of Parameters of Equation 5 and the Plait Point for PEGDME (1) + Na_2CO_3 (2) + H_2O (3) at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K

T/K	f	g	R^2	plait point (100 w_1, 100 w_2, 100 w_3)
288.15	0.0740	1.9669	0.9866	(17.28, 5.02, 77.70)
293.15	0.1029	1.9011	0.9928	(16.89, 5.92, 77.19)
298.15	0.0767	2.0854	0.9913	(17.63, 4.78, 77.59)
308.15	0.0251	2.8086	0.9845	(16.01, 4.81, 79.18)
318.15	–0.0258	3.3212	0.9942	(13.75, 4.92, 81.33)

fitting parameters for eq 5 and the corresponding correlation coefficients are listed in Table 6.

The crystal structure of the obtained powder was determined through powder X-ray diffractometry (XRD). The (XRD) spectrum has been kept in the Supporting Information for further study and shows that the powder is composed completely of monohydrate sodium carbonate.

Figure 3 shows the complete phase diagram for the studied system plotted on triangular coordinates. The letters L and S denote liquid and solid phases, respectively. Most of the phase diagram consists of either a solid−liquid−liquid or solid−solid−liquid region.

In Figure 3, region 1L represents unsaturated solutions. The biphasic zone is labeled 2L. The two liquid regions occupy a small portion of the total phase diagram, although the coexisting phases are generally quite different in composition. There are two single liquid−liquid regions, one with a liquid rich in water and another with a liquid rich in $PEGDME_{2000}$. Any addition of sodium carbonate would alter the compositions of the two immiscible phases.

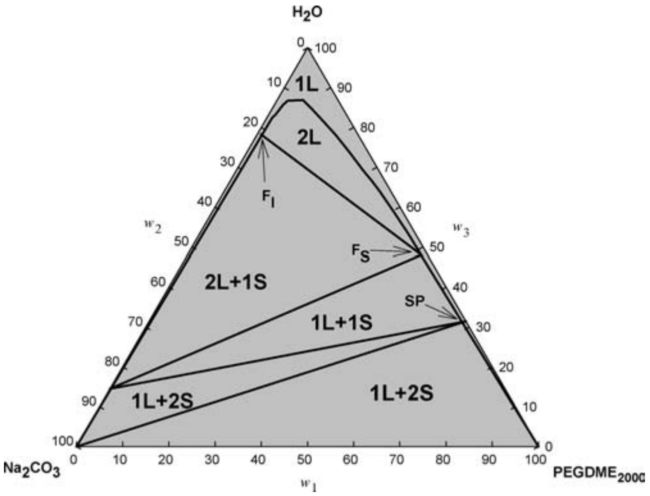

Figure 3. Complete phase diagram of the $PEGDME_{2000}$ (1) + sodium carbonate (2) + H_2O (3) two-phase system at 298.15 K in triangular coordinates: 1L, liquid phase; 2L, two-liquid phase; 2L+1S, a solid phase ($Na_2CO_3 \cdot H_2O$) and two-liquid phase; 1L+1S, solid phase ($Na_2CO_3 \cdot H_2O$) and liquid saturated; first 1L+2S, two-solid phase (Na_2CO_3 and $Na_2CO_3 \cdot H_2O$) and a liquid phase; second zone of 1L+2S, two-solid phase (Na_2CO_3 and $PEGDME_{2000}$) and a liquid phase; SP, the solubility point of $PEGDME_{2000}$ in water; F_I and F_S, eutectic points in the bottom and top phases, respectively.

The 2L+1S region represents two immiscible liquid phases in equilibrium with a solid phase. In this case, the system is saturated with Na_2CO_3, and the composition of each liquid phase will remain constant even though the relative amount of the two liquids will change, as predicted by the phase rule.

The 1L+1S region contains the monohydrate Na_2CO_3 in equilibrium with $PEGDME_{2000}$ saturated solutions. Finally, according to solubility data obtained for $PEGDME_{2000}$ and Na_2CO_3 in the system studied, it seems that the first 1L+2S region consists of two solid phases (monohydrate and anhydrous sodium carbonate) and a liquid phase and the second one consists of two solid phases (anhydrous sodium carbonate and $PEGDME_{2000}$) and a liquid phase.

It was experimentally determined that the solubility of $PEGDME_{2000}$ in water at 298.15 K was 68.6 % by mass. The eutectic points F_I and F_S were 1.3 % ($PEGDME_{2000}$), 20.9 % (Na_2CO_3), and 77.8 % (H_2O) in the bottom phase and 50.6 % ($PEGDME_{2000}$), 0.8 % (Na_2CO_3), and 48.6 % (H_2O) in the top phase. There exists a zone of insignificant size, which is not visible in the diagram, as it is practically superimposed by the ordinate. This zone varies between solubility of the Na_2CO_3 in water (23.17 % by mass)[22] and the invariant F_I (1.3 % $PEGDME_{2000}$, 20.9 % Na_2CO_3, and 77.8 % H_2O) that corresponds to crystals in equilibrium with a saturated Na_2CO_3 solution. These observations are very similar to those obtained for the phase diagram of the PEG + Na_2CO_3 + H_2O system, given by Taboada et al.[9]

The phase diagram of the system investigated is also very similar to the poly(ethylene glycol) dimethyl ether 2000 ($PEGDME_{2000}$) + Na_2SO_4 + H_2O system, studied previously.[10] However, in the phase diagram of the system containing sodium sulfate,[10] there is one kind of salt and five zones, but for the system containing sodium carbonate, we observe two kinds of salt and six zones.

Conclusions

Liquid−liquid equilibrium data for the $PEGDME_{2000}$ (1) + Na_2CO_3 (2) + H_2O (3) system were obtained at T = (288.15, 293.15, 298.15, 308.15, and 318.15) K. The experimental binodal data were satisfactorily correlated with the Merchuk equation. The effect of temperature on the phase-forming ability for the studied system was also studied, and it was observed that the area of the biphasic region increased slightly with an increase of temperature. Additionally, the tie lines were satisfactorily correlated with the Setschenow and Othmer−Tobias equations. Also, liquid−liquid−solid equilibria were determined for this system, and it was observed that the solid phase in equilibrium with the biphasic region was $Na_2CO_3 \cdot H_2O$. Furthermore, the complete phase diagram was determined for the $PEGDME_{2000}$ (1) + Na_2CO_3 (2) + H_2O (3) system at 298.15 K.

Supporting Information Available:

The (XRD) spectrum has been presented in Figure S1. The peaks were indexed accurately according to the JCPDS card No. 08-0037 which is attributed to the monohydrate sodium carbonate crystal structure. This material is available free of charge via the Internet at http://pubs.acs.org.

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