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Isothermal Vapor−Liquid Equilibria for the 2-Propanol + Water System Containing Poly(ethylene glycol) at 298.15 K

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Isothermal vapor−liquid equilibrium measurements were performed for the 2-propanol + water system containing poly(ethylene glycol) (PEG) at 298.15 K by a flow type apparatus. Three different PEGs whose molecular weights are 200, 1000, and 20 000 were employed, and their mass fractions in the liquid phase were up to 0.35. The volatility of water was reduced by the addition of PEG, and the azeotropic point of the 2-propanol + water system was changed to a higher mole fraction of 2-propanol. Activity coefficients of solvent were calculated by assuming the monodispersity of PEG. It was found that the activity coefficients of water were selectively decreased by the addition of PEG. These phenomena did not depend on the molecular weight of PEG.

Introduction

Although distillation has been widely utilized as a separation process in chemical industries, it is difficult to separate volatile components in an azeotropic mixture. It is known that the azeotropic point for some alcohol + water systems disappears by the addition of inorganic electrolyte, because the volatility of water is reduced by hydration of the inorganic electrolyte. The phenomena is the so-called "salt effect" on the azeotrope. In our previous study (Mishima et al., 1986, 1987; Hongo and Hibino, 1989; Kumagae et al., 1992), vapor−liquid equilibria for alcohol + water systems containing calcium chloride were reported. Our successive study is whether the addition of a nonionic substance would induce such phenomena as the salt effect. The substance should have a nonvolatile nature and a strong interaction with a water molecule. Nonionic polymer is preferable for the usage. Especially, PEG has a peculiar interaction with water molecules (Kjellander and Florin, 1981) because the molecule structure of the repeating unit of an open ring of crown ether (Frensdolff, 1971).

Vapor−liquid equilibria for the 2-propanol + water system containing three different molecular weights of PEG were measured at 298.15 K using of a flow type apparatus. The effects of PEG were investigated by varying the concentration and molecular weight of PEG for the azeotropic mixture of 2-propanol + water.

Experimental Section

Materials. 2-Propanol used in this work was of guaranteed reagent grade, and its mass fraction purity reported by the supplier, Wako Pure Chemical Industries Co. Ltd., was higher than 0.995. No significant impurity could be detected with gas chromatography. PEG was of chemical reagent grade (Wako Pure Chemical Industries Co. Ltd.) with average molecular weights of 200, 1000, and 20 000. The polydispersity of PEG was measured by a gel permeation chromatograph equipped with a differential reflactometer (GL Science Inc. RI 504), with the column of Asahipak GF-510HQ (Asahi Chemical Industry Co. Ltd.). The molecular weight and polydispersity of PEG are listed in Table 1. As shown in the table, the polydispersity is no larger than 1.200. No significant impurity could be detected in the ^{1}H-NMR (JOEL JNM-Ex90) spectrum of PEG. All reagents were used as received.

Table 1. Molecular Weight and Polydispersity of PEG Used in This Study

polymer	av mol wt[a] M_w	polydispersity[b] M_w/M_n
PEG200	200	1.085
PEG1000	1000	1.176
PEG20000	20000	1.108

[a] Reported by supplier. [b] Measured with a gel permeation chromatograph.

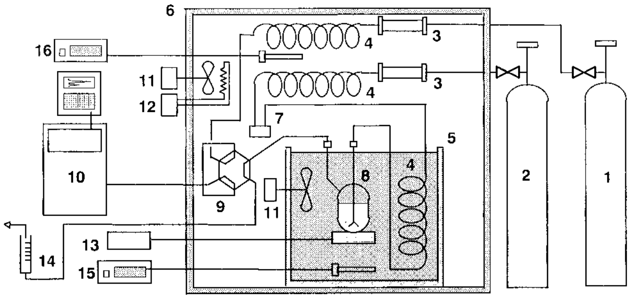

Figure 1. Schematic diagram of flow type apparatus: (1) carrier gas for gas chromatograph; (2) carrier gas for sample; (3) gas dryer tube; (4) heat exchanger; (5) water bath; (6) air chamber; (7) mass flow controller; (8) sample cell; (9) six-way valve; (10) gas chromatography; (11) impeller; (12) heater; (13) waterproofing magnetic stirrer; (14) flow meter; (15) thermistor thermometer.

Equipment and Procedures. A schematic diagram of the flow type apparatus employed is shown in Figure 1. The apparatus was partly improved in this study, and other equipment and operating procedures have been described elsewhere (Kumagae et al., 1992; Uchizono et al., 1983; Kuratsu et al., 1984; Hongo et al., 1994). The sample solution, volume about 100 cm^3, was prepared by use of a precise direct-reading balance (Exact Inc. AV 1581). The weight capacity of the balance is from 0.1 mg to 1 kg. The solution was loaded into a glass cell (part 8 in Figure 1) of inner volume about 200 cm^3. A stirrer tip was in the cell to agitate the viscous polymer solution. The cell was held in a water bath (5) at 298.15 K, which was measured by a thermistor thermometer (15) (Takara Thermistor Co. Ltd. D641) with an accuracy of ±0.01 K. A waterproofing magnetic stirrer (13) was located in the water bath. Carrier gas (2) (helium) passed through a gas dryer tube

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Table 2. Vapor−Liquid Equilibrium for 2-Propanol (1) + Water (2) + PEG200 (3) at 298.15 K[a]

w_3	x'_1	P_1/kPa	P_2/kPa	y'_1	$x_1{}^b$	$\gamma_1{}^b$	$\gamma_2{}^b$	w_3	x'_1	P_1/kPa	P_2/kPa	y'_1	$x_1{}^b$	$\gamma_1{}^b$	$\gamma_2{}^b$
0.00	0.069 5	2.339	2.942	0.442 9	0.069 5	5.819	1.001	0.20	0.000 0	0.000	3.078	0.000 0	0.000 0		0.997
0.00	0.120 0	3.008	2.879	0.510 9	0.120 0	4.333	1.036	0.20	0.100 8	2.784	2.792	0.449 2	0.098 0	4.913	1.012
0.00	0.200 9	3.327	2.844	0.539 1	0.200 9	2.862	1.127	0.20	0.200 0	3.318	2.680	0.553 2	0.193 4	2.964	1.097
0.00	0.299 5	3.550	2.771	0.561 6	0.299 5	2.049	1.253	0.20	0.299 6	3.554	2.592	0.578 3	0.288 3	2.131	1.230
0.00	0.399 9	3.673	2.681	0.578 1	0.399 9	1.588	1.414	0.20	0.399 8	3.749	2.463	0.603 5	0.382 6	1.693	1.358
0.00	0.500 3	3.867	2.555	0.602 1	0.500 3	1.336	1.619	0.20	0.503 6	4.002	2.291	0.636 0	0.479 4	1.443	1.536
0.00	0.601 8	4.132	2.343	0.638 1	0.601 8	1.187	1.864	0.20	0.601 3	4.247	2.019	0.677 8	0.569 5	1.289	1.693
0.00	0.698 5	4.418	2.104	0.677 4	0.698 5	1.093	2.209	0.20	0.691 8	4.439	1.678	0.725 7	0.652 2	1.177	1.829
0.00	0.797 6	4.759	1.665	0.740 8	0.797 6	1.031	2.605	0.20	0.803 2	4.764	1.194	0.799 6	0.753 0	1.094	2.048
0.00	0.900 2	5.214	0.964	0.843 9	0.900 2	1.001	3.060	0.20	0.884 1	5.018	0.736	0.872 0	0.825 5	1.051	2.156
								0.20	1.000 0	5.493	0.000	1.000 0	0.928 2	1.023	
0.05	0.000 0	0.000	3.140	0.000 0	0.000 0		0.999								
0.05	0.113 2	2.858	2.786	0.506 3	0.112 5	4.391	1.001	0.35	0.000 0	0.000	2.974	0.000 0	0.000 0		0.989
0.05	0.204 9	3.236	2.762	0.539 5	0.203 5	2.749	1.108	0.35	0.099 5	2.589	2.706	0.488 9	0.093 8	4.774	1.010
0.05	0.297 5	3.418	2.680	0.560 6	0.295 1	2.002	1.218	0.35	0.196 2	3.197	2.615	0.550 1	0.182 9	3.022	1.105
0.05	0.400 7	3.646	2.620	0.581 9	0.397 0	1.587	1.397	0.35	0.293 7	3.497	2.438	0.589 3	0.271 1	2.231	1.184
0.05	0.504 6	3.856	2.480	0.608 6	0.499 3	1.335	1.603	0.35	0.405 6	3.759	2.217	0.629 1	0.369 6	1.758	1.296
0.05	0.653 4	4.245	2.163	0.662 5	0.645 4	1.137	2.000	0.35	0.499 3	3.985	1.992	0.666 8	0.450 5	1.529	1.396
0.05	0.698 4	4.367	1.990	0.687 0	0.689 5	1.095	2.116	0.35	0.523 3	4.040	1.931	0.676 6	0.470 9	1.483	1.426
0.05	0.806 3	4.651	1.463	0.760 7	0.795 1	1.011	2.425	0.35	0.608 0	4.211	1.671	0.715 9	0.542 4	1.342	1.514
0.05	0.899 8	5.083	0.867	0.854 3	0.886 3	0.992	2.780	0.35	0.742 0	4.520	1.165	0.795 1	0.652 9	1.197	1.625
0.05	1.000 0	5.719	0.000	1.000 0	0.984 0	1.005		0.35	0.852 1	4.816	0.687	0.875 2	0.741 4	1.123	1.690
								0.35	1.000 0	5.184	0.000	1.000 0	0.857 2	1.045	

[a] P_1 and P_2 are the partial pressures of 2-propanol and water. [b] Calculated form hypothetical molecular weight of PEG200.

Table 3. Vapor−Liquid Equilibrium for 2-Propanol (1) + Water (2) + PEG1000 (3) at 298.15 K[a]

w_3	x'_1	P_1/kPa	P_2/kPa	y'_1	$x_1{}^b$	$\gamma_1{}^b$	$\gamma_2{}^b$	w_3	x'_1	P_1/kPa	P_2/kPa	y'_1	$x_1{}^b$	$\gamma_1{}^b$	$\gamma_2{}^b$
0.05	0.000 0	0.000	3.155	0.000 0	0.000 0		1.000	0.20	0.000 0	0.000	3.140	0.000 0	0.000 0		0.999
0.05	0.107 4	3.097	2.919	0.514 8	0.107 3	4.990	1.037	0.20	0.203 1	3.408	2.794	0.549 5	0.201 8	2.920	1.118
0.05	0.206 4	3.401	2.837	0.545 2	0.206 1	2.852	1.134	0.20	0.299 8	3.656	2.685	0.576 6	0.297 5	2.125	1.224
0.05	0.302 9	3.612	2.821	0.561 5	0.302 4	2.067	1.283	0.20	0.395 4	3.886	2.549	0.603 8	0.391 9	1.714	1.347
0.05	0.400 7	3.829	2.710	0.585 6	0.400 0	1.655	1.435	0.20	0.594 8	4.368	2.110	0.674 3	0.588 5	1.283	1.666
0.05	0.502 9	4.057	2.540	0.615 0	0.501 8	1.398	1.622	0.20	0.505 7	4.140	2.346	0.638 3	0.500 7	1.429	1.518
0.05	0.601 5	4.219	2.374	0.640 0	0.600 1	1.215	1.891	0.20	0.693 2	4.638	1.762	0.724 7	0.685 1	1.170	1.841
0.05	0.705 0	4.558	2.053	0.689 5	0.703 2	1.120	2.209	0.20	0.796 0	4.969	1.309	0.791 5	0.785 7	1.093	2.058
0.05	0.752 1	4.772	1.831	0.722 7	0.750 1	1.100	2.345	0.20	0.898 1	5.350	0.703	0.883 8	0.885 5	1.044	2.217
0.05	0.799 7	4.885	1.586	0.754 8	0.797 5	1.059	2.514								
0.05	0.894 9	5.183	0.932	0.847 6	0.892 3	1.004	2.816								

[a] P_1 and P_2 are the partial pressures of 2-propanol and water. [b] Calculated form hypothetical molecular weight of PEG1000.

Table 4. Vapor−Liquid Equilibrium for 2-Propanol (1) + Water (2) + PEG20000 (3) at 298.15 K[a]

w_3	x'_1	P_1/kPa	P_2/kPa	y'_1	$x_1{}^b$	$\gamma_1{}^b$	$\gamma_2{}^b$	w_3	x'_1	P_1/kPa	P_2/kPa	y'_1	$x_1{}^b$	$\gamma_1{}^b$	$\gamma_2{}^b$
0.05	0.083 6	2.767	2.903	0.488 0	0.083 6	5.722	1.003	0.05	0.742 6	4.666	1.877	0.713 1	0.742 6	1.086	2.310
0.05	0.123 9	3.160	2.897	0.521 8	0.123 9	4.408	1.047	0.05	0.777 9	4.738	1.708	0.735 0	0.777 9	1.053	2.436
0.05	0.193 4	3.469	2.859	0.548 2	0.193 5	3.099	1.122								
0.05	0.247 1	3.598	2.828	0.559 9	0.247 1	2.517	1.190	0.20	0.084 2	2.687	2.923	0.479 0	0.084 2	5.519	1.011
0.05	0.291 5	3.664	2.783	0.568 3	0.291 5	2.173	1.244	0.20	0.175 5	3.368	2.812	0.545 0	0.175 5	3.318	1.081
0.05	0.356 3	3.780	2.749	0.579 0	0.356 3	1.834	1.353	0.20	0.299 9	3.732	2.700	0.580 2	0.299 8	2.152	1.222
0.05	0.440 2	3.915	2.643	0.596 9	0.440 2	1.537	1.496	0.20	0.408 2	3.983	2.550	0.609 7	0.408 0	1.688	1.365
0.05	0.520 0	4.118	2.549	0.617 6	0.519 9	1.369	1.682	0.20	0.488 9	4.130	2.389	0.633 6	0.488 7	1.461	1.481
0.05	0.594 2	4.309	2.390	0.643 2	0.594 2	1.254	1.865	0.20	0.603 3	4.431	2.105	0.678 0	0.603 0	1.270	1.681
0.05	0.629 1	4.348	2.277	0.656 3	0.629 1	1.195	1.944	0.20	0.691 8	4.666	1.799	0.721 7	0.691 4	1.167	1.850
0.05	0.691 3	4.517	2.097	0.682 9	0.691 3	1.130	2.152								

[a] P_1 and P_2 are the partial pressures of 2-propanol and water. [b] Calculated form hypothetical molecular weight of PEG20000.

(3) which contained molecular sieves having a pore diameter of 0.5 nm, and the flow rate of helium was set to be 10 cm^3/min by a mass-flow controller (7) (STEC Inc. SEC-400mk2). Then, the carrier gas was introduced into the cell and saturated with the solvent vapor. Successively, a known volume of the carrier gas was isolated by a six-way valve (9), and introduced into a gas chromatograph (10) equipped with a thermal conductivity detector (GL Science Inc. GC-380TCD). The packing in the precolumn (glass tube, i.d. 2.0 mm × 1.83 m long) was 20% PEG1000 (poly(ethylene glycol)) on Flusin T 80/100 mesh and that in the main column (glass tube, i.d. 2.0 mm × 1.83 m long) was Gasukuropak 54 (polystyrene closs-linked by divinylbenzene) 60/80 mesh. The temperature in the air chamber (6) was maintained at 313 K to avoid condensation of the saturated vapor. Teflon pipe (i.d. 4.0 mm × 300 mm long) was used as a connector between the cell and the six-way valve, and the temperature of the six-way valve was also maintained at 333 K for the same purpose.

We measured the peak area of each substance supplied from pure solvent or mixtures at several temperatures close to 298.15 K. First, the peak area of each solvent at 298.15 K was determined by interpolation. Next, the corresponding temperature t_i^* which represented the same peak area, was obtained for each pure solvent. The partial pressure P_i was regarded as the following equation:

$$P_i = P_i^\circ(t_i^*) \qquad i = 1, 2 \tag{1}$$

where subscripts 1 and 2 indicate 2-propanol and water,

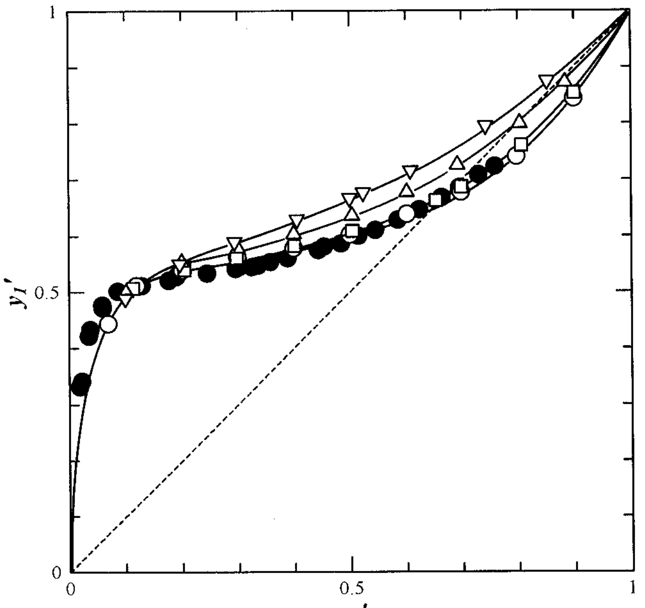

Figure 2. Vapor–liquid equilibrium of the 2-propanol (1) + water (2) system containing PEG200 (3) at 298.15 K: (○) $w_3 = 0.00$, (□) 0.05, (△) 0.20 and (▽) 0.35, this work; (●) $w_3 = 0.00$, Schumacher et al. (1948). x' and y' are calculated on a PEG200 free basis.

Table 5. Results of Thermodynamic Consistency Tests for the 2-Propanol (1) + Water (2) System at 298.15 K

test	criterion of consistency (charater; +)	result of this work
method 1 (Van Ness et al. and Fredenslund)	$\overline{\Delta y} < 0.01$	0.07
method 2 (Redlich–Kister and Herington)	$D < 10\%$	1.754%

$P_i^\circ(t_i^*)$ denotes the saturated vapor pressure of the pure solvent at the corresponding temperature t_i^*, which can be calculated from the equation for the vapor pressure such as the Antoine equation (Gmehling et al., 1981a). Assuming that the vapor phase is ideal, the PEG free basis mole fraction of solvent in the vapor phase y_i' was calculated by the following equation:

$$y_i' = \frac{P_i}{P_1 + P_2} \qquad i = 1, 2 \qquad (2)$$

The peak area of PEG is too small to be detected by a gas chromatograph, so the vapor pressure of PEG is neglected. Then, the mole fraction of each solvent in the vapor phase was regarded as the same as the PEG free basis mole fraction in the vapor phase.

Results and Discussion

Prior to the measurement, the miscibility of PEG for pure solvent was investigated. PEG200, -1000, and -20000 are soluble in water up to higher concentrations, but the solubility of PEG1000 and -20000 is much less in 2-propanol. After taking into account the expected solubility in the mixture, measurements of vapor–liquid equilibria were performed at mass fraction of PEG (component 3) w_3 to the mixture of solvent set to be 0.00, 0.05, 0.20, and 0.35. In this study, precise values of the activity coefficient could not be calculated, because we did not have detailed information about the molecular weight of PEG. However, the various PEG's used are almost monodispersed, as shown in Table 1, so a hypothetical molecular weight was defined under the following assumptions:

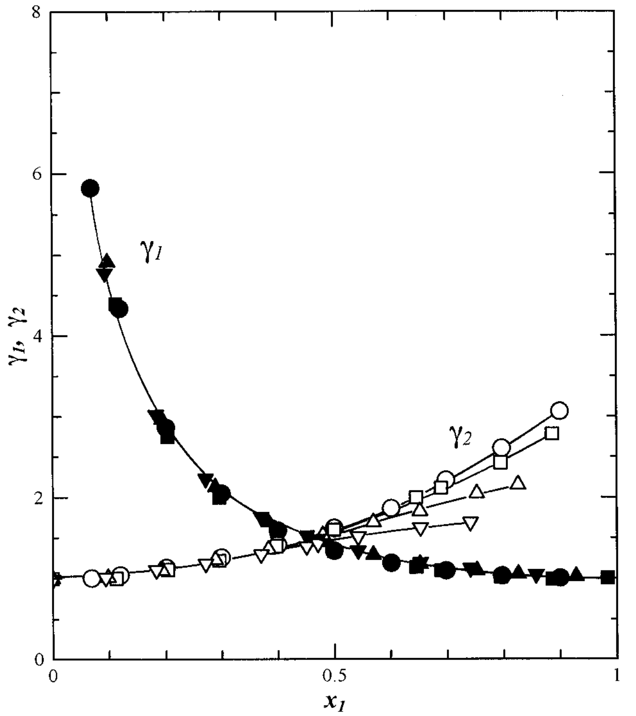

Figure 3. Hypothetical activity coefficients for the 2-propanol (1) + water (2) system containing PEG200 (3) at 298.15 K: (●) γ_1 and (○) γ_2 at $w_3 = 0.00$; (■) γ_1 and (□) γ_2 at $w_3 = 0.05$; (▲) γ_1 and (△) γ_2 at $w_3 = 0.20$; (▼) γ_1 and (▽) γ_2 at $w_3 = 0.35$.

(1) The degree of polymerization is a natural number, and PEG is perfectly monodispersed.

(2) The hypothetical molecular weight is the closest to that reported by the supplier. According to the assumptions, the hypothetical molecular weights of PEG200, -1000, -20000 are assumed to be 194.23, 987.23, and 20018.15, respectively. Therefore, the hypothetical mole fraction of solvent in liquid phase x_i is calculated from the following equation:

$$x_i = x_i'\left\{1 - \frac{w_3/M_3}{(1 - w_3)/(x_1'M_1 + x_2'M_2) + w_3/M_3}\right\} \qquad i = 1, 2 \quad (3)$$

where x_1' and x_2' are the PEG free basis mole fractions of 2-propanol (1) and water (2) in the liquid phase, M_1 and M_2 are the molecular weights of 2-propanol and water, and M_3 is the hypothetical molecular weight of PEG.

Then, the hypothetical activity coefficient of component i, γ_i, was evaluated by the following equation:

$$\gamma_i = P_i/x_iP_i^\circ \qquad i = 1, 2 \qquad (4)$$

where P_i° is the vapor pressure of the pure solvent at 298.15 K. In eq 4, it was assumed that x_i would be equal to that at the preparation, because the volatility of each component is very small.

Experimental results of the vapor–liquid equilibria of three ternary systems for 2-propanol + water containing PEG200, -1000, and -20000 were presented in Tables 2–4. The homogeneous liquid phase was maintained in the whole range of concentrations shown in the tables, but two liquid phases coexisted at mole fractions of 2-propanol larger than that shown in Tables 3 and 4. Though a small amount of helium could be soluble in the solution, the reliability of the present experimental procedure was ascertained in previous work (Mishima et al., 1986, 1987; Hongo and Hibino, 1989; Kumagae et al., 1992; Hongo et

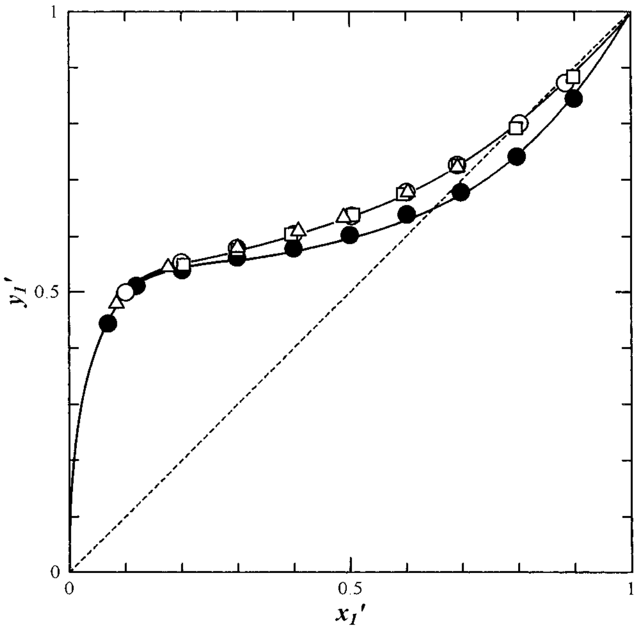

Figure 4. Vapor−liquid equilibrium of the 2-propanol (1) + water (2) system containing various molecular weights of PEG (3) at 298.15K: (●) $w_3 = 0.00$; (○) with PEG200 at $w_3 = 0.20$; (□) with PEG1000 at $w_3 = 0.20$; (△) with PEG20000 at $w_3 = 0.20$. x' and y' are calculated on a PEG free basis.

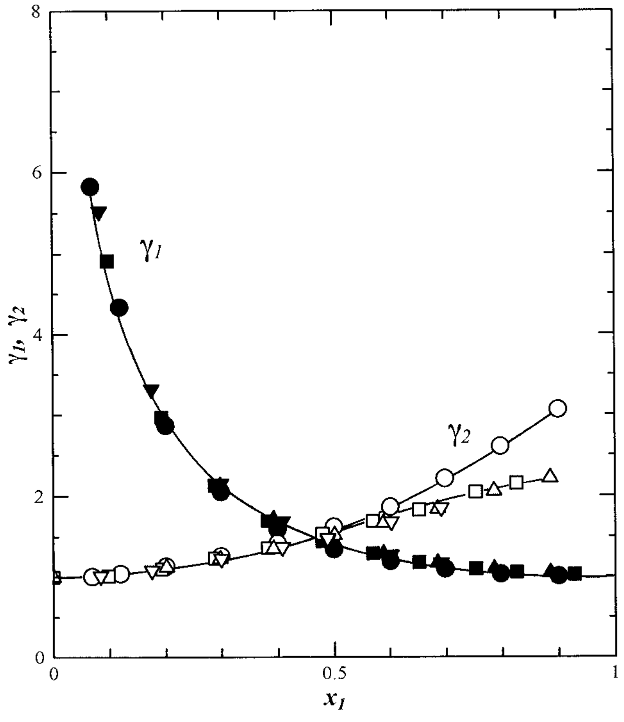

Figure 5. Hypothetical activity coefficients for the 2-propanol + water system containing various molecular weights of PEG at 298.15 K: (●) γ_1 and (○) γ_2 at $w_3 = 0.00$; (■) γ_1 and (□) γ_2 with PEG200 at $w_3 = 0.20$; (▲) γ_1 and (△) $\gamma/32$ with PEG1000 at $w_3 = 0.20$; (▼) γ_1 and (▽) γ_2 with PEG20000 at $w_3 = 0.20$.

al., 1991, 1994). The accuracy of the result in the partial pressure is considered to be within ±2%. Further, the results were compared with those of the literature. The x_1, y_1 relation of the binary system, 2-propanol + water, has been reported by Schumacher et al. (1948) at the same temperature. A typical illustration is shown in Figure 2, where some discrepancies can be seen in the mole fraction range from $x_1 = 0.1$ to 0.5.

The experimental data were tested for thermodynamic consistency by using the point test of Fredenslund et al. (1977) and Van Ness et al. (1973) and the area test of Herington et al. (1951) and of Redlich and Kister (1948) as described by Gmehling et al. (1981b).

Table 5 shows the results of the tests. Both tests indicate that the experimental vapor−liquid equilibrium data are thermodynamically consistent.

From the values shown in the tables, the extrapolated activity coefficient of water at $x_1' = 0.000$ would be smaller than unity. Though more detailed discussion will be required, these tendencies have been often observed in some systems where molecular interaction in a mixture is more stable than that in pure solvent (Gmehling and Kolbe, 1988).

Figure 2 shows the x_1', y_1' relation of the 2-propanol (1) + water (2) system containing various mass fractions of PEG200 (3) at 298.15 K. As shown in the figure, a significant change of the azeotropic point cannot be observed in the system at $w_3 = 0.05$. However, the azeotropic point shifts to the larger mole fraction of 2-propanol of the system at $w_3 = 0.20$ and seems to disappear at $w_3 = 0.35$. The effect of PEG200 on the azeotropic mixture of the alcohol + water system is similar to that of calcium chloride. Comparing the selectivity of solvent, the hypothetical activity coefficient of each solvent was also shown in Figure 3. In the figure, the activity coefficient of water was reduced at the larger concentration of PEG200, while that of 2-propanol could not be changed at any concentration of PEG200. Then, the selective hydration of PEG200 would cause a shift of the azeotropic point.

The vapor−liquid equilibrium data for the 2-propanol + water system containing various molecular weights of PEG at $w_3 = 0.20$ were shown in Figure 4. The shift of the azeotropic point can be observed.

Figure 5 shows the hypothetical activity coefficients of solvent for the 2-propanol + water system containing various molecular weights of PEG at $w_3 = 0.20$. In the figure, the hypothetical activity coefficient of each solvent shows the same tendency as that shown in Figure 3 and does not depend on the molecular weight of PEG.

Consequently, the "polymer effect", which is similar to the "salt effect", can be observed in the azeotropic 2-propanol + water system containing various molecular weights of PEG.

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Literature Cited

Fredenslund, A.; Gmehling, J.; Rasmunssen, P. *Vapor-Liquid Equilibria Using UNIFAC*; Elsevier: Amsterdam, 1977.

Frensdorff, H. K. Stability Constants of Cyclic Polyether Complexes with Univalent Cations. *J. Am. Chem. Soc.* **1971**, *93*, 600−606.

Gmehling, J.; Kolbe, B. *Thermodynamik-Chemieingenieurweszsen/Verfahrenstechnik*; Georg Thieme: Stuttgart, 1988.

Gmehling, J.; Onken, U.; Arlt, W. *Vapor-Liquid Equilibrium Data Collection*; DECHEMA: Frankfurt am Main, 1981a; Vol. I, Part 1a, p 248.

Gmehling, J.; Onken, U.; Arlt, W. *Vapor-Liquid Equilibrium Data Collection*; DECHEMA: Frankfurt am Main, 1981b; Vol. I, Part 1a, p XXV.

Herington, E. F. G. Tests for consistency of experimental isobaric vapor liquid equilibrium data. *J. Inst. Petrol.* **1951**, *37*, 457−470.

Hongo, M.; Hibino, T. Measurement of Vapor-Liquid Equilibria of Binary and Ternary Systems Containing 1-Propanol, Water and Calciumu Chloride at 298.15 K. *Kagaku Kogaku Ronbunshu* **1989**, *15*, 863−867.

Hongo, M.; Kusunoki, M. (Vapor-Liquid) Equilibria of (butane-2-ol + butane-1-ol) at the temperature 298.15 K. *J. Chem. Thermodyn.* **1991**, *23*, 1181−1182.

Hongo, M.; Tsuji, T.; Fukuchi, K.; Arai, Y. Vapor-Liquid Equilibria of Methanol + Hexane, Methanol + Heptane, Ethanol + Hexane, Ethanol + Heptane and Ethanol + Octane at 298.15 K. *J. Chem. Eng. Data* **1994**, *39*, 688−691.

Kjellander, R.; Florin, E. Water Structure and Changes in the Thermal Stability of the System Poly(ethylene oxide). *J. Chem. Soc., Faraday Trans. 1* **1981**, *77*, 2053−2077.

Kumagae, Y.; Mishima, K.; Hongo, M.; Kusunoki, M.; Arai, Y. Calcium Chloride on Vapor-Liquid Equilibria of Alcohol-Alcohol and Alcohol-Water Binary Systems. *Can. J. Chem. Eng.* **1992**, *70*, 1180−1185.

Kuratsu, M.; Tomori, Y.; Hongo, M.; Arai, Y. *Bunrigijyutsu* **1984**, *14*, 160 (in Japanese).

Mishima, K.; Iwai, Y.; Yamaguchi, S.; Isonaga, H.; Arai, Y.; Hongo, M. Correlation of Vapor-Liquid Equilibria of Ethanol-Water-Calcium Chloride and Methanol-Ethanol-Calcium Chloride Systems at 25 °C. *Memories of Facalty of Engineering, Kyushu Univ.* **1986**, *46*, 407−433.

Mishima, K.; Matsubara, K.; Arai, Y.; Hongo, M. *Kagaku Kogaku Ronbunshu* **1987**, *13*, 850−853 (in Japanese).

Redlich, O.; Kister, A. T. Algebraic representitation of thermodynamic properties and classification of solutions. *Ind. Eng. Chem.* **1948**, *40*, 345−348.

Schumacher, J. E.; Hunt, M. Nitromethane-Isopropyl Alcohol − Water System. *Ind. Eng. Chem.* **1948**, *34*, 701−704.

Uchizono, Y.; Kai, M.; Tashima, Y.; Arai, Y. *Kagaku Kogaku Ronbunshu* **1983**, *9*, 485−489 (in Japanese).

Van Ness, H. C.; Byer, S. M.; Gibbs, R. E. Vapor-Liquid equilibrium: Part I. An appraisal of data reduction methods. *AIChE J.* **1973**, *19*, 238−244.

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