Static Relative Permittivities of Water + Propane-1,2-diol and Water + Propane-1,3-diol under Pressures up to 300 MPa at 298.15 K

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Static relative permittivities (ϵ_r) of water + propane-1,2-diol and water + propane-1,3-diol were measured at four mole fractions under pressures up to 300 MPa at 298.15 K. A Tait-type equation was used to represent ϵ_r with pressure (P) for each composition. Values of $(\partial \ln \epsilon_r/\partial P)_T$ and $\epsilon_r^{-2}(\partial\epsilon_r/\partial P)_T$ at 0.1 MPa for both aqueous propanediol systems were determined by use of the static relative permittivity at 0.1 MPa, ϵ_r(0.1 MPa), and the Tait-type parameters, A and B. Values of $(\partial \ln \epsilon_r/\partial P)_T$ and the isothermal compressibility (κ_T) at 0.1 MPa over the entire composition range are compared for both aqueous propanediol systems. A relationship between $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T at 0.1 MPa was derived from the density dependence of ϵ_r. Furthermore, composition dependence of $\epsilon_r^{-2}(\partial\epsilon_r/\partial P)_T$ at 0.1 MPa was examined for the present systems.

Introduction

The static relative permittivities (ϵ_r) of many mixtures of water + organic solvents have been extensively studied as a function of composition around room temperature at atmospheric pressure. Information on pressure dependence of ϵ_r is useful to analyze some properties of electrolytes in polar solvents, but limited ϵ_r values have been measured for aqueous organic mixtures under high pressure. We have so far reported ϵ_r results for some aqueous organic mixtures at selected mole fractions under high pressures up to 300 MPa at 298.15 K and examined composition dependence of $(\partial \ln \epsilon_r/\partial P)_T$ and $(\partial\epsilon_r/\partial P)_T$ values at 0.1 MPa for each aqueous organic mixture.[1−4]

To extend our understanding of dielectric properties for water + organic solvents, this paper reports ϵ_r results of water + propane-1,2-diol and water + propane-1,3-diol at selected mole fractions under pressures up to 300 MPa at 298.15 K. Since the isothermal compressibilities (κ_T) for both aqueous propanediol systems were determined previously as a function of composition at 0.1 MPa and 298.15 K,[5] the composition dependence of $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T at 0.1 MPa is compared for both water + propanediol mixtures. A relationship between $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T at 0.1 MPa is derived, and the condition that those values are approximately equal each other is described. A comparison of $\epsilon_r^{-2}(\partial\epsilon_r/\partial P)_T$ values at 0.1 MPa with composition is also made among water + propane-1,2-diol, water + propane-1,3-diol, water + butane-1,2-diol, and water + butane-1,3-diol systems.

Experimental Section

Propane-1,2-diol (>99.0 %) and propane-1,3-diol (98 %) were obtained from Kanto Chemical Co., Inc. and Aldrich Chemical Co., Inc. Each propanediol was refluxed over CaH_2 for several hours and then distilled under N_2 stream at reduced pressure at least twice. Water was distilled from an alkaline solution of $KMnO_4$ and then distilled. The refractive index (n_D) for each substance was measured with an Abbe refractometer at 298.15 K; these agreed to within ± 0.0001 with those reported previously.[1,6] The purity was checked by GC analysis and found to be >99.9 % for propane-1,2-diol and >99.8 % for propane-1,3-diol. All the aqueous propanediol mixtures were prepared gravimetrically with a precision of 0.00001 g. The compositions were accurate to ± 0.0001 in the mole fraction of propanediol at the nominal values.

The ϵ_r measurements were carried out by means of a transformer ratio-arm bridge with a three-terminal dielectric cell in the frequency range of (10 to 50) kHz. A high-pressure vessel equipped with the dielectric cell was immersed in a thermostated bath where the temperature was controlled to ± 0.01 K. Details of the apparatus and experimental procedure for the measurements have been described elsewhere.[1] Pressure was accurate to ± 0.07 MPa. Uncertainty in ϵ_r is estimated to be within ± 0.1 %. Measurements of ϵ_r were carried at least twice for each aqueous mixture, and the ϵ_r values thus obtained were reproduced to within ± 0.09 %.

Results and Discussion

The ϵ_r values measured for water + propane-1,2-diol and water + propane-1,3-diol at 298.15 K are listed as a function of pressure in Table 1, in which x_2 is the mole fraction of propanediol. Data for pure propanediols are also included in Table 1.[6] Figure 1 illustrates the composition dependence of ϵ_r(0.1 MPa) values at 298.15 K for water + propane-1,2-diol and water + propane-1,3-diol. Available ϵ_r(0.1 MPa) data from the literature[7−11] for each aqueous propanediol mixture are also plotted for comparison in each figure. Our ϵ_r(0.1 MPa) data as a function of x_2 for each system are represented by the following equation:

$$\epsilon_r(0.1\ \mathrm{MPa}) = \sum_{i=0}^{N} A_i x_2^{\,i} \tag{1}$$

Values of the coefficients A_i for each system obtained by the method of nonweighted least-squares are given in Table

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Table 1. Static Relative Permittivities (ϵ_r) for Water + Propanediol Mixtures as a Function of Pressure at a Temperature of 298.15 K

	ϵ_r at P/MPa						
x_2	0.1	50	100	150	200	250	300
	Water (1) + Propane-1,2-diol (2)						
0.2000	56.45	57.87	59.19	60.41	61.53	62.60	63.61
0.4000	44.23	45.53	46.73	47.80	48.80	49.75	50.64
0.6000	37.09	38.28	39.34	40.29	41.19	42.02	42.81
0.8000	32.25	33.34	34.30	35.15	35.95	36.71	37.42
1.0000[a]	28.67	29.67	30.56	31.34	32.08	32.76	33.40
	Water (1) + Propane-1,3-diol (2)						
0.2000	59.05	60.30	61.43	62.49	63.47	64.40	65.30
0.4000	49.10	50.18	51.17	52.07	52.89	53.68	54.42
0.6000	42.85	43.80	44.67	45.45	46.19	46.88	47.53
0.8000	38.54	39.39	40.15	40.83	41.47	42.06	42.63
1.0000[a]	34.69	35.42	36.11	36.72	37.30	37.83	38.34

[a] The ϵ_r data for pure propanediols are cited from ref 6.

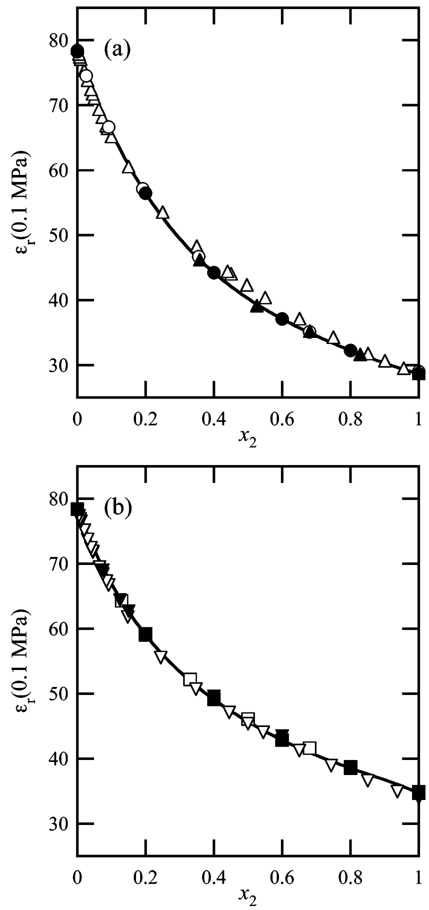

Figure 1. Static relative permittivities at 0.1 MPa and 298.15 K, ϵ_r(0.1 MPa), against mole fraction x_2 for (a) water (1) + propane-1,2-diol (2) (●, this work; ○, Verbeeck et al.;[7] ▲, Tombari et al.;[8] △, George and Sastry[9]) and (b) water (1) + propane-1,3-diol (2) (■, this work; □, Wang et al.;[10] ▼, Safonova et al.;[11] ▽, George and Sastry[9]). The smoothed curves are based on the values of A_i listed in Table 2.

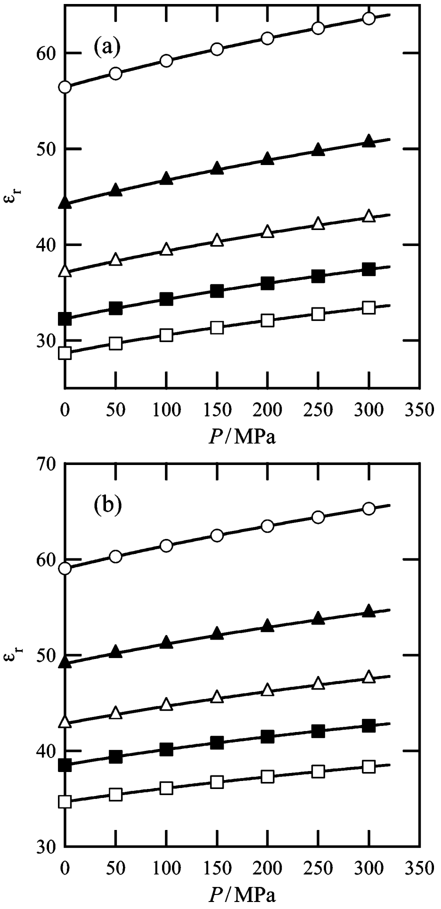

Figure 2. Pressure dependence of the static relative permittivities ϵ_r at 298.15 K for (a) water (1) + propane-1,2-diol (2) and (b) water (1) + propane-1,3-diol (2): ○, x_2 = 0.2000; ▲, x_2 = 0.4000; △, x_2 = 0.6000; ■, x_2 = 0.8000; □, x_2 = 1.0000. The smoothed curves are based on the values of ϵ_r(0.1 MPa), A and B listed in Table 3.

Table 2. Coefficients A_i and Standard Deviation $\sigma(\epsilon_r$(0.1 MPa)) for Least-Squares Representations of ϵ_r(0.1 MPa) by Eq 1 at a Temperature of 298.15 K

A_0	A_1	A_2	A_3	A_4	A_5	$\sigma(\epsilon_r$(0.1 MPa))
		Water (1) + Propane-1,2-diol (2)				
78.41	−144.08	200.20	−153.00	47.14		0.00
		Water (1) + Propane-1,3-diol (2)				
78.41	−138.45	272.49	−376.20	294.01	−95.58	0.00

2, along with the standard deviation of the fit $\sigma(\epsilon_r$(0.1 MPa)). A marked decrease in ϵ_r(0.1 MPa) is observed at lower x_2 in aqueous propanediol as well as in other aqueous organic mixtures so far studied in our laboratory[1,2,4] except in water + dimethyl sulfoxide.[3] The literature ϵ_r(0.1 MPa) values for water + propane-1,2-diol by Verbeeck et al.[7] and Tombari et al.[8] agree with the present ϵ_r(0.1 MPa) ones within ± 0.7 % and ± 1.4 %, respectively. The values in the mole fraction range of 0.35 to 0.65 by George and Sastry[9] are about 4 % larger than ours, but their results in other composition ranges are in agreement with the present values within ± 1.9 %. For water + propane-1,3-diol, the ϵ_r(0.1 MPa) values by Wang et al.,[10] Safonova et al.,[11] and George and Sastry[9] coincide with ours within ± 1.7 %, ± 2.3 %, and ± 2.2 %, respectively.

Figure 2 shows the experimental ϵ_r results for both water + propanediol systems as a function of pressure. In any aqueous mixture, $(\partial\epsilon_r/\partial P)_T$ becomes smaller with increasing pressure. To represent the pressure dependence of the ϵ_r data for each solution, the following Tait-type equation, usually referred to as the Owen−Brinkley equation,[12] is usually applied:

$$1 - \frac{\epsilon_r(0.1\ \text{MPa})}{\epsilon_r(P)} = A \ln \frac{B + P}{B + 0.1\ \text{MPa}} \qquad (2)$$

where $\epsilon_r(P)$ is the static relative permittivity at the pressure P. The Tait-type parameters, A and B, for each solution obtained by a nonweighted least-squares method

Table 3. Static Relative Permittivity at 0.1 MPa, Parameters of the Tait-Type Equation, A and B, Standard Deviations of Fit of Eq 2, $\sigma(\epsilon_r)$, and $(\partial \ln \epsilon_r/\partial P)_T$, κ_T, and $\epsilon_r^{-2}(\partial \epsilon_r/\partial P)_T$ at 0.1 MPa and at a Temperature of 298.15 K

x_2	ϵ_r(0.1 MPa)	A	B/MPa	$\sigma(\epsilon_r)$	$(\partial \ln \epsilon_r/\partial P)_T$/TPa^{-1}	κ_T[a]/TPa^{-1}	$\epsilon_r^{-2}(\partial \epsilon_r/\partial P)_T$/TPa^{-1}
Water (1) + Propane-1,2-diol (2)							
0.0000[b]	78.41	0.2185	463.6	0.02	471.2	451.2	6.01
0.2000	56.45	0.1698	318.9	0.00	532.3	364.9	9.43
0.4000	44.23	0.1704	272.3	0.01	625.6	386.6	14.14
0.6000	37.09	0.1682	247.4	0.01	679.6	423.7	18.32
0.8000	32.25	0.1674	234.3	0.01	714.2	454.2	22.14
1.0000[b]	28.67	0.1650	220.8	0.01	746.9	476.7	26.05
Water (1) + Propane-1,3-diol (2)							
0.2000	59.05	0.1553	352.6	0.01	440.3	355.9	7.46
0.4000	49.10	0.1440	308.9	0.00	466.0	356.1	9.49
0.6000	42.85	0.1462	312.1	0.00	468.3	371.8	10.93
0.8000	38.54	0.1347	289.4	0.01	465.3	385.4	12.07
1.0000[b]	34.69	0.1436	318.7	0.01	450.4	399.5	12.98

[a] The values are calculated from the Tait parameters reported in ref 5. [b] The data for pure water and propanediols are, respectively, cited from ref 1 and ref 6.

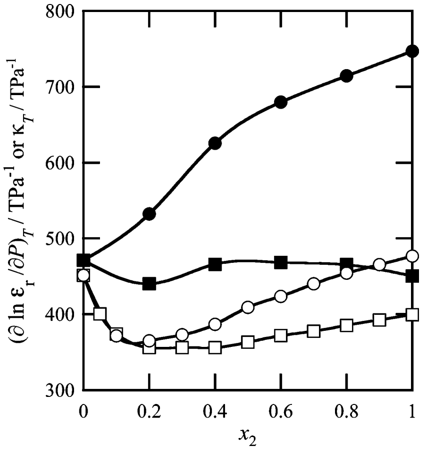

Figure 3. Composition dependence of $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T at 0.1 MPa and 298.15 K for water (1) + propane-1,2-diol (2) (●, ○) and water (1) + propane-1,3-diol (2) (■, □). Closed and open symbols represent $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T, respectively. The curves by a cubic spline fit to data points are to guide to the eye.

along with the standard deviations $\sigma(\epsilon_r)$ of the fit are summarized in Table 3, in which the results for pure water and propanediols reported in the previous papers[1,6] are also tabulated. The $\sigma(\epsilon_r)$ values for each aqueous propanediol mixtures are less than 0.01, so the Tait-type equation is also applicable for the present aqueous solutions to represent the pressure dependence of ϵ_r very accurately.

Values of $(\partial \ln \epsilon_r/\partial P)_T$ at 0.1 MPa for polar liquids are required to evaluate the limiting slope of the curves of apparent molar volumes of strong electrolytes against the square root of concentration. The values at 0.1 MPa can be evaluated from the Tait-type parameters as follows:

$$\left(\frac{\partial \ln \epsilon_r}{\partial P}\right)_T = \frac{A}{B + 0.1\ \text{MPa}} \tag{3}$$

Table 3 lists also the values of $(\partial \ln \epsilon_r/\partial P)_T$ at 0.1 MPa thus calculated.

It has been reported that there was a good agreement between $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T at 0.1 MPa for a few pure solvents.[13] The κ_T values at 0.1 MPa determined from the Tait parameters previously reported[5] are listed in Table 3. Figure 3 depicts the composition dependence of $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T at 0.1 MPa and 298.15 K for each aqueous propanediol mixture. For each water + propanediol, the $(\partial \ln \epsilon_r/\partial P)_T$ values are higher than κ_T values over the entire composition range. For water + propane-1,2-diol, the $(\partial \ln \epsilon_r/\partial P)_T$ values increase with x_2, while κ_T values show a minimum at $x_2 = 0.2$ and an increase above that composition. Furthermore, there is a large difference in magnitude between these values; the largest difference is found at $x_2 = 0.4$, and its value is as high as about 63 %. For water + propane-1,3-diol, a shallow minimum was observed in $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T around $x_2 = 0.2$. Above that composition, κ_T values increase slightly to 399.5 TPa^{-1} at $x_2 = 1$, while $(\partial \ln \epsilon_r/\partial P)_T$ values increase but almost keep constant. Therefore our results suggest that the use of κ_T value to estimate $(\partial \ln \epsilon_r/\partial P)_T$ value for any aqueous propanediol mixture except for water brings about unreliable results.

The disagreement between $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T values at 0.1 MPa can be explained as follows. It is expected that there is a relationship between ϵ_r and density (ρ) for each solution because pressure dependence of density can be usually represented by the following Tait equation:

$$1 - \frac{\rho(0.1\ \text{MPa})}{\rho(P)} = A' \ln \frac{B' + P}{B' + 0.1\ \text{MPa}} \tag{4}$$

Here $\rho(P)$ and $\rho(0.1\ \text{MPa})$ are respectively the density at pressure P and 0.1 MPa, and A' and B' are the Tait parameters. The form of eq 4 is similar to eq 2, but the values of the Tait parameters, A and B, are different from those of the Tait-type parameters, A' and B', for the solutions of the same composition. The parameters A' and B' for water + propanediol mixtures have been determined from the compression measurements at pressures up to 200 MPa,[5] so the densities at high pressures up to 200 MPa can be calculated from eq 4 in combination with available $\rho(0.1\ \text{MPa})$ data for water + propane-1,2-diol[9,14,15] and water + propane-1,3-diol.[9] Plots of ϵ_r against ρ thus determined at various compositions for each aqueous propanediol up to 200 MPa are illustrated in Figure 4, where ϵ_r values for water were plotted against density up to 300 MPa because densities up to 300 MPa at 298.15 K are available.[16] The following equation was applied to correlate the ϵ_r data with density by a least-squares method:

$$\epsilon_r(P) = \epsilon_r(0.1\ \text{MPa}) + a[\rho(P) - \rho(0.1\ \text{MPa})] + b[\rho(P) - \rho(0.1\ \text{MPa})]^2 \tag{5}$$

This equation means that every curve passes through the data at 0.1 MPa (i.e., $\rho(0.1\ \text{MPa})$ and $\epsilon_r(0.1\ \text{MPa})$), and the coefficients a and b are independent of pressure but depend

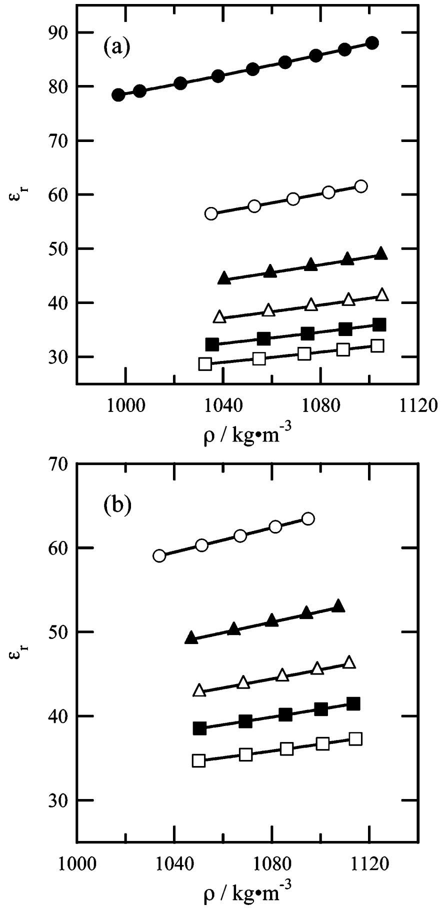

Figure 4. Density dependence of ϵ_r at 298.15 K for (a) water (1) + propane-1,2-diol (2) and (b) water (1) + propane-1,3-diol (2): ●, $x_2 = 0.0000$; ○, $x_2 = 0.2000$; ▲, $x_2 = 0.4000$; △, $x_2 = 0.6000$; ■, $x_2 = 0.8000$; □, $x_2 = 1.0000$. The smoothed curves are based on the values of ϵ_r(0.1 MPa) in Table 3 and ρ(0.1 MPa), a and b in Table 4.

Table 4. Density at 0.1 MPa, Coefficients a and b in Eq 5, y, and $(\partial \ln \epsilon_r/\partial P)_T/\kappa_T$ Values at 0.1 MPa and 298.15 K

x_2	ρ(0.1 MPa) / kg·m^{-3}	a / kg^{-1}·m^3	$10^5 b$ / kg^{-2}·m^6	y^a	$(\partial \ln \epsilon_r/\partial P)_T$ / κ_T
		Water (1) + Propane-1,2-diol (2)			
0.0000	997.0	0.08114	10.57	1.03	1.04
0.2000	1035.1	0.07956	5.021	1.46	1.46
0.4000	1040.5	0.06908	3.134	1.63	1.62
0.6000	1038.5	0.05765	5.536	1.61	1.60
0.8000	1035.5	0.04989	5.696	1.60	1.57
1.0000	1032.6	0.04384	6.105	1.58	1.57
		Water (1) + Propane-1,3-diol (2)			
0.2000	1033.9	0.07122	2.083	1.25	1.24
0.4000	1046.9	0.06182	1.711	1.32	1.31
0.6000	1050.2	0.05170	4.224	1.27	1.26
0.8000	1050.4	0.04479	2.602	1.22	1.21
1.0000	1050.0	0.03720	5.279	1.13	1.13

[a] y is equal to $a\rho$(0.1 MPa)/ϵ_r(0.1 MPa).

on x_2 and temperature. Table 4 lists the values of a and b for both water + propanediol mixtures. It is found that a value decreases with an increase in x_2. Differentiation of eq 5 with pressure and rearrangement leads the following equation at 0.1 MPa:

$$\left(\frac{\partial \ln \epsilon_r}{\partial P}\right)_T / \kappa_T = \frac{a\rho(0.1\ \text{MPa})}{\epsilon_r(0.1\ \text{MPa})} = y \tag{6}$$

Here $a\rho$(0.1 MPa)/ϵ_r(0.1 MPa) is replaced by y. Validity of this equation can be judged from a comparison of y values with $(\partial \ln \epsilon_r/\partial P)_T/\kappa_T$ values at 0.1 MPa listed in Table 4, where y values were calculated from a, ρ(0.1 MPa), and ϵ_r(0.1 MPa) values summarized in Tables 3 and 4, and $(\partial \ln \epsilon_r/\partial P)_T/\kappa_T$ values were obtained from $(\partial \ln \epsilon_r/\partial P)_T$ and κ_T values at 0.1 MPa tabulated in Table 3. Agreement between y and $(\partial \ln \epsilon_r/\partial P)_T/\kappa_T$ values at 0.1 MPa is quite satisfactory; most of y values coincide with $(\partial \ln \epsilon_r/\partial P)_T/\kappa_T$ values at 0.1 MPa within ± 0.01. If a linear correlation between ϵ_r and ρ is used (i.e., b is fixed as zero), a maximum deviation between y and $(\partial \ln \epsilon_r/\partial P)_T/\kappa_T$ values is as high as about 9 %. So it is reasonable to use eq 5 to represent density dependence of ϵ_r for the present systems. Furthermore, it is clarified that the $(\partial \ln \epsilon_r/\partial P)_T$ value becomes close to the κ_T value only in the case that a value of y is almost unity. Since y value for water is 1.03 at 298.15 K, it can be considered that the $(\partial \ln \epsilon_r/\partial P)_T$ value is nearly equal to the κ_T value for water, as suggested by Kawaizumi and Zana.[13] But y values at other compositions are in the ranges of 1.46 to 1.63 for water + propane-1,2-diol and 1.13 to 1.32 for water + propane-1,2-diol at 298.15 K, as tabulated in Table 4.

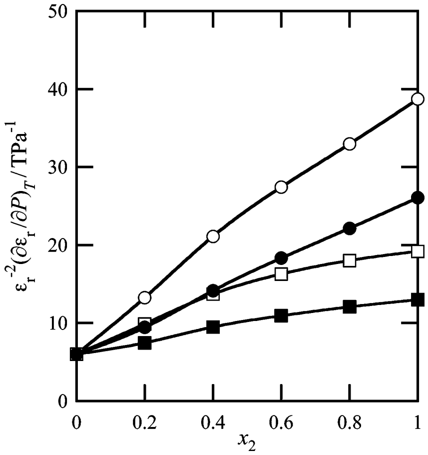

Figure 5. Composition dependence of $\epsilon_r^{-2}(\partial \epsilon_r/\partial P)_T$ at 0.1 MPa and 298.15 K for water (1) + propane-1,2-diol (2) (●), water (1) + propane-1,3-diol (2) (■), water (1) + butane-1,2-diol (2)[4] (○), and water (1) + butan-1,3-diol (2)[4] (□). The curves by a cubic spline fit to data points are to guide to the eye.

Values of $\epsilon_r^{-2}(\partial \epsilon_r/\partial P)_T$ at 0.1 MPa are needed to estimate a volume contribution due to electrostriction in the partial molar volume of an ion at infinite dilution by use of the Born equation. These values at 0.1 MPa can be evaluated also ϵ_r(0.1 MPa) and the Tait-type parameters as follows:

$$\epsilon_r^{-2}\left(\frac{\partial \epsilon_r}{\partial P}\right)_T = \frac{A}{\epsilon_r(0.1\ \text{MPa})(B + 0.1\ \text{MPa})} \tag{7}$$

The calculated $\epsilon_r^{-2}(\partial \epsilon_r/\partial P)_T$ values at 0.1 MPa for both aqueous propanediol mixtures are shown as a function of x_2 in Figure 5, where the values[4] for water + butane-1,2-diol and water + butane-1,3-diol are also plotted for comparison. The $\epsilon_r^{-2}(\partial \epsilon_r/\partial P)_T$ values at 0.1 MPa for each system increase with an increase in the composition of organic component. The value for water + alkane-1,2-diol is higher than that for water + alkane-1,3-diol at the same composition. Moreover, the $\epsilon_r^{-2}(\partial \epsilon_r/\partial P)_T$ values at 0.1 MPa at higher mole fraction ($x_2 > 0.6$) for water + butane-1,n-diol are about twice those for water + propane-1,n-diol (n = 2 or 3). From these results, it is found that the $\epsilon_r^{-2}(\partial \epsilon_r/\partial P)_T$ value at 0.1 MPa for water + alkanediol is strongly influenced by the relative position of the OH groups and a size of alkyl end group in alkanediol.

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