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Thermodynamic Properties of Water + Ethylene Glycol at 283.15, 293.15, 303.15, and 313.15 K

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Excess volumes (V^E), viscosity deviation ($\Delta\eta$), excess refraction (ΔR), and surface excess (σ^E) of water + ethylene glycol have been determined over the entire composition range at a number of temperatures. The results are fitted to a Redlich−Kister equation, and the corresponding parameters are derived. All the properties have negative values and exhibit a minimum at the same water-rich region of the solution. The results are discussed in terms of molecular interactions.

Introduction

The aim of this work is to provide information on the molecular interactions between water and ethylene glycol in binary mixtures at a series of temperatures. To study the behavior of these mixtures, the density (ρ), the refractive index (n_D), the viscosity (η) and the surface tension (σ) have been measured. A survey in the literature showed that these mixtures have not been studied in sufficient detail. Excess volumes and viscosities at 308.15 K have been reported by Reddy et al. (1994), excess volumes, viscosities, and refractive indices at 298.15 K by Aminabhavi and Gopalakrishna (1995), and surface tension at a series of temperatures by Hoke and Chen (1991). As a part of a program in progress in our laboratory, which concerns the study of thermodynamic properties of binary organic solutions (Palaiologou and Molinou, 1995; Palaiologou, 1996), we report a systematic study of excess volumes (V^E), viscosity deviations ($\Delta\eta$), refractive index deviations (ΔR) and surface tension excess (σ^E) of aqueous ethylene glycol mixtures at (283.15, 293.15, 303.15, and 313.15) K. The data cover the entire range of composition at 10% mass intervals.

Experimental Section

Materials. The ethylene glycol used was Merck p.a. The stated purity was better than 99.5% mass and was used without further purification. The water was doubly distilled, and the conductance was found better than 0.5 μS at 20 °C. The purity of the pure components was assessed by comparing the experimental densities and refractive indices given in Table 1 with literature values: The agreement is very satisfactory. The binary mixtures were prepared by mass (Mettler balance of accuracy 0.01 mg) in flasks with ground glass joints. The mole fractions were known to ±0.0001.

Measurements. Densities (ρ) of pure liquids and binary mixtures were measured with an Anton Paar (model DMA 58), microcomputer-controlled precision densimeter, with built-in solid-state thermostat. The precision in ρ was $\pm 0.000\,01$ g cm^{-3}. The DMA cell was calibrated with dry air and conductivity water. The sample size was 0.7 cm^3, the precision of the internal thermostat was ±0.005 °C, and that of the sample thermostat ±0.01 °C.

The viscosities (η) were measured with a viscosity measuring unit (Schott Gerate AVS 310) equipped with an Ubbelohde capillary viscometer supplied by Canon Instr. Co. The time measurement was fully automatic. A change in light intensity was converted to a digital electric signal and started the internal crystal-controlled clock with readout display. The time measurement tolerance was ±0.005%, and the display accuracy was ±1 digit (0.01 s). The viscometer was immersed in a bath thermostat (model Schott CT 050/2) electronically controlling to the set temperature. The temperature was maintained constant within ±0.03 K, which caused a measuring error up to ±0.1%. The accuracy in the viscosity measurements was ±0.001 mPa·s.

The refractive indices at the sodium D-line were measured with a thermostated Abbe refractometer (model A.Krüss) with a built-in light source for the measuring prism with an accuracy of ±0.0001. The thermostat temperature was constant to ±0.01 °C. The surface tension was measured by a Du Nouy Tensiometer (model 8551) equipped with a platinum−iridium ring of wire diameter 0.37 mm. The tensiometer was calibrated with distilled water at 18 °C, and a correction factor was employed. The platinum−iridium ring was brought to red heat, and the test vessel was cleaned with chromosulfuric acid boiled in distilled water. The accuracy of the surface tension measurement was ±0.1 dyn cm^{-1}. The sample was introduced into a double-walled glass cell, connected to a thermostat where the temperature was constant within ±0.01 °C.

Results and Discussion

The experimental values of density, viscosity, and refractive index are given in Table 2. The excess volumes were calculated according to the equation

$$V^E = V_m - \sum_{i=1}^{2} V_i x_i \tag{1}$$

where V_i represents the molar volume and x_i the mole fraction of the ith component. V_m is the molar volume of the mixture and can be calculated for a binary mixture as $V_m = (x_1M_1 + x_2M_2)/\rho_m$, where ρ_m is the density of the solution and M_1, M_2 are the molecular weights of each

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Table 1. Comparison of Experimental Densities (ρ), Viscosities (η), and Refractive Indices (n_D) of Pure Liquids with Literature Values

T/K	ρ(exp)/g·cm^{-3}	ρ(ref)/g·cm^{-3}	n_D(exp)	n_D(ref)	η(exp)/mPa·s	η(ref)/mPa·s	σ(exp)/mN·m^{-1}	σ(ref)/mN·m^{-1}
Water								
283.15	0.999 69	0.999 70^a	1.3343	1.3334^a	1.2982	1.3070^a	74.27	74.23^a
293.15	0.998 22	0.998 21^a	1.3330	1.3328^a	1.0021	1.0020^a	72.58	72.75^a
298.15	0.997 20^c	0.997 3^e	1.3325^c	1.3314^e	0.9471^c	0.891^e		
303.15	0.995 64	0.995 65^a	1.3320	1.3318^a	0.8075	0.7977^a	71.26	71.20^a
308.15	0.994 20^c	0.994 12^d			0.7381^c	0.7190^d		
313.15	0.992 22	0.992 22^a	1.3306	1.3305^a	0.6660	0.6532^a	69.81	69.60^a
Ethylene Glycol								
283.15	1.120 28		1.4335		30.5126		49.76	
293.15	1.113 23	1.113 50^b	1.4319	1.4318^a	20.8064	21.0^b	49.15	48.40^b
298.15	1.109 50^c	1.000 3^e	1.4310^c	1.4166^e	18.6826^c	16.1^a 9.408^e		47.99^a
303.15	1.105 46		1.4287		13.8678		48.67	
308.15	1.102 50^c	1.105 35^d			11.6956^c	10.473^d		
313.15	1.098 35		1.4254		9.5348		48.07	

[a] *CRC Handbook of Chemistry and Physics*, 74th ed., 1993−1994. [b] Lange, N. A. *Handbook of Chemistry*, 13th ed., 1985. [c] Values from interpolation. [d] Reddy et al. (1994). [e] Aminabhavi and Gopalakrishna (1995).

Table 2. Experimental Densities (ρ), Viscosities (η), and Refractive Indices (n_D) of Ethylene Glycol (1) + Water (2)

x_1	ρ/g·cm^{-3}	η/mPa·s	n_D	x_1	ρ/g·cm^{-3}	η/mPa·s	n_D
T = 283.15 K							
0.0000	0.999 69	1.2982	1.3343	0.2250	1.070 50	5.3013	1.3855
0.0312	1.013 36	1.6961	1.3448	0.3003	1.083 24	7.1250	1.3955
0.0487	1.020 31	1.9711	1.3489	0.4038	1.094 46	9.8892	1.4059
0.0677	1.027 58	2.2643	1.3541	0.5373	1.104 51	14.1111	1.4157
0.1000	1.039 00	2.8498	1.3617	0.7232	1.113 13	20.3512	1.4246
0.1622	1.057 20	3.9470	1.3751	1.0000	1.120 28	30.5126	1.4335
0.2000	1.065 58	4.7651	1.3815				
T = 293.15 K							
0.0000	0.998 22	1.0021	1.3336	0.2250	1.064 79	3.7110	1.3841
0.0312	1.010 93	1.2789	1.3429	0.3003	1.076 57	4.9174	1.3936
0.0487	1.017 70	1.4602	1.3481	0.4038	1.088 06	6.8247	1.4038
0.0677	1.024 52	1.6465	1.3528	0.5373	1.097 84	9.6558	1.4137
0.1000	1.035 08	1.9903	1.3608	0.7232	1.106 16	13.7082	1.4225
0.1622	1.051 78	2.7938	1.3733	1.0000	1.113 23	20.8064	1.4319
0.2000	1.060 00	3.3431	1.3798				
T = 303.15 K							
0.0000	0.995 64	0.8075	1.3326	0.2250	1.058 98	2.7642	1.3820
0.0312	1.008 00	1.0185	1.3425	0.3003	1.070 27	3.5947	1.3921
0.0487	1.014 28	1.1813	1.3468	0.4038	1.081 49	4.8655	1.4017
0.0677	1.020 78	1.3166	1.3518	0.5373	1.090 93	6.6711	1.4110
0.1000	1.028 55	1.5773	1.3594	0.7232	1.099 18	9.4058	1.4197
0.1622	1.045 33	2.1312	1.3722	1.0000	1.103 94	13.8678	1.4287
0.2000	1.054 41	2.4957	1.3796				
T = 313.15 K							
0.0000	0.992 22	0.6660	1.3311	0.2250	1.053 04	2.1155	1.3810
0.0312	1.004 11	0.8238	1.3413	0.3003	1.063 90	2.7887	1.3907
0.0487	1.010 16	0.9179	1.3457	0.4038	1.074 62	3.6131	1.3991
0.0677	1.016 35	1.0342	1.3508	0.5373	1.083 98	4.9055	1.4084
0.1000	1.026 18	1.2440	1.3586	0.7232	1.092 12	6.7370	1.4172
0.1622	1.041 32	1.6344	1.3708	1.0000	1.098 35	9.5348	1.4254
0.2000	1.048	2.0742	1.3803				

constituent. The results were fitted to the Redlich−Kister equation

$$V^E/\mathrm{cm}^3\ \mathrm{mol}^{-1} = x_1 x_2 \sum_{k=0}^{n} A_k (2x_1 - 1)^k \qquad (2)$$

where A_k are parameters and V^E is the excess volume of the mixture. The optimum number of the coefficients was obtained through examination of the standard deviation according to the equation

$$\sigma_{V^E} = [\sum (V^E_{\mathrm{calc}} - V^E_{\mathrm{exp}})^2/(N - n)]^{1/2} \qquad (3)$$

where N is the number of measurements and n the number of parameters in eq 3. The parameters A_k and the standard deviations are presented in Table 3. The excess volumes are shown in Figure 1, and the values are given in Table 4. The viscosity deviations, $\Delta\eta$, are obtained from the equation

$$\Delta\eta/\mathrm{mPa{\cdot}s} = \eta_m - \sum_{i=1}^{2} \eta_i x_i \qquad (4)$$

where x_i, n_i, n_m refer to the mole fraction and viscosity of the pure components and of the mixture, respectively. $\Delta\eta$ values are listed in Table 4 and are shown in Figure 2. The parameters A_k and the standard deviations σ for $\Delta\eta$ are obtained from equations similar to eqs 2 and 3 and are listed in Table 3.

Table 3. Parameters and Standard Deviations of Excess Functions for Ethylene Glycol (1) + Water (2)

function	A_0	A_1	A_2	A_3	σ
$T = 283.15$ K					
V^E/cm^3·mol^{-1}	−1.5581	0.9190	−0.2178	−0.8563	0.0095
$\Delta\eta$/mPa·s	−12.1077	4.9261	−1.0896	−2.2075	0.0353
ΔR/cm^3·mol^{-1}	−10.8911	−5.4611	−3.2488	−1.8899	0.0074
$T= 293.15$ K					
V^E/cm^3·mol^{-1}	−1.4285	0.6988	0.0133	0.4810	0.0059
$\Delta\eta$/mPa·s	−8.2862	2.5561	−3.1276	−3.0299	0.0252
ΔR/cm^3·mol^{-1}	−10.9746	−5.5662	−3.3823	−1.8933	0.0099
$T = 303.15$ K					
V^E/cm^3·mol^{-1}	−1.4055	0.5598	0.0777	−0.7057	0.0152
$\Delta\eta$/mPa·s	−4.7412	1.5857	−0.6162	−0.8907	0.0094
ΔR/cm^3·mol^{-1}	−10.9831	−5.6351	−3.4791	−1.8828	0.0094
$T=313.15$ K					
V^E/cm^3·mol^{-1}	−1.3040	0.4302	−0.0400	−0.2300	0.0056
$\Delta\eta$/mPa·s	−2.2920	0.8685	0.4768	1.4284	0.0520
ΔR/cm^3·mol^{-1}	−10.9351	−5.6858	−3.5491	−1.7052	0.0120

The refractive index deviations, ΔR, are calculated from the Lorentz−Lorentz (Lorentz, 1909) equation

$$\Delta R/\text{cm}^3\ \text{mol}^{-1} = R_m - \sum_{i=1}^{2} R_i \Phi_i \qquad (5)$$

where R_i and R_m are the molar refractions of the pure components and the mixture, respectively, and Φ_i is the volume fraction of the ith component, given as

$$\Phi_i = x_i V_i / \sum_{i=1}^{2} x_i V_i \qquad (6)$$

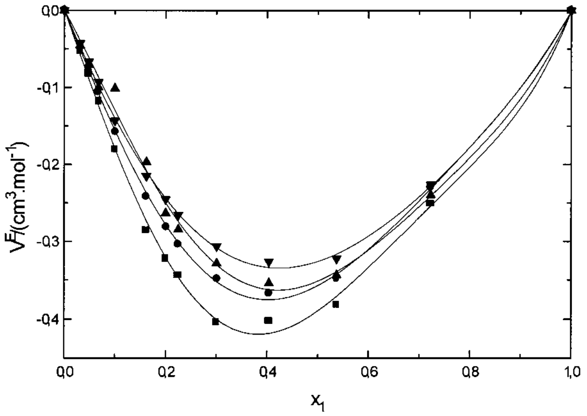

Figure 1. Excess molar volumes for ethylene glycol (1) + water (2) mixtures at 283.15 K (■), 293.15 K (●), 303.15 K (▲), and 313.15 K (▼).

The molar refraction R_i was obtained from the Lorentz−Lorentz equation

$$R_i = \frac{n_D^2 - 1}{n_D^2 + 2} V_i \qquad (7)$$

where n_D is the refractive index for a pure liquid. The equation for a binary mixture becomes

$$R_m = \frac{n_{D(m)}^2 - 1}{n_{D(m)}^2 + 2} V_m \qquad (8)$$

where $n_{D(m)}$ is the refractive index of the mixture and R_m is the molar refraction of the mixture. The deviations in refractive indices (ΔR) are given in Table 4 and are shown

Table 4. Excess Volumes (V^E), Viscosity Deviations ($\Delta\eta$), and Refractive Index Deviations (ΔR) of Ethylene Glycol (1) + Water (2)

x_1	V^E/cm^3·mol^{-1}	$\Delta\eta$/mPa·s	ΔR	x_1	V^E/cm^3·mol^{-1}	$\Delta\eta$/mPa·s	ΔR
$T = 283.15$ K							
0.0000	0	0	0	0.2250	−0.3444	−2.5701	−2.6423
0.0312	−0.0532	−0.5136	−0.6215	0.3003	−0.4039	−2.9463	−2.8797
0.0487	−0.0822	−0.7498	−0.9346	0.4038	−0.4028	−3.2058	−2.9097
0.0677	−0.1177	−1.0117	−1.2282	0.5373	−0.3815	−2.884	−2.609
0.1000	−0.1804	−1.3698	−1.6598	0.7232	−0.2511	−2.0748	−1.7826
0.1622	−0.2853	−2.0898	−2.2604	1.0000	0	0	0
0.2000	−0.3229	−2.376	−2.5144				
$T = 293.15$ K							
0.0000	0	0	0	0.2250	−0.3037	−1.7471	−2.6667
0.0312	−0.0438	−0.3411	−0.6389	0.3003	−0.3491	−2.0319	−2.9041
0.0487	−0.0738	−0.5064	−0.9425	0.4038	−0.3678	−2.1744	−2.9408
0.0677	−0.1056	−0.6964	−1.2430	0.5373	−0.3487	−1.9872	−2.6319
0.1000	−0.1578	−0.9922	−1.6724	0.7232	−0.2299	−1.6164	−1.8053
0.1622	−0.2418	−1.4206	−2.2832	1.0000	0	0	0
0.2000	−0.2817	−1.6199	−2.5382				
$T = 303.15$ K							
0.0000	0	0	0	0.2250	−0.2845	−0.9819	−2.6788
0.0312	−0.0456	−0.1965	−0.6360	0.3003	−0.3286	−1.1348	−2.911
0.0487	−0.0707	−0.2622	−0.9514	0.4038	−0.3541	−1.2157	−2.9494
0.0677	−0.1002	−0.3751	−1.2482	0.5373	−0.3436	−1.1537	−2.6452
0.1000	−0.1013	−0.5362	−1.6818	0.7232	−0.2399	−0.8469	−1.8084
0.1622	−0.1969	−0.7947	−2.2908	1.0000	0	0	0
0.2000	−0.2633	−0.9239	−2.5278				
$T = 313.15$ K							
0.0000	0	0	0	0.2250	−0.2658	−0.546	−2.6643
0.0312	−0.0429	−0.1189	−0.6382	0.3003	−0.3066	−0.5406	−2.9033
0.0487	−0.0666	−0.18	−0.9524	0.4038	−0.3263	−0.6341	−2.9488
0.0677	−0.0933	−0.2322	−1.2501	0.5373	−0.3225	−0.5257	−2.639
0.1000	−0.1435	−0.3089	−1.6863	0.7232	−0.2267	−0.3429	−1.7953
0.1622	−0.2154	−0.4701	−2.2898	1.0000	0	0	0
0.2000	−0.2457	−0.3656	−2.5041				

Table 5. Surface Tension of Ethylene Glycol (1) + Water (2)

	σ/mN·m^{-1}					
T/K	$x_1 = 0.0000$	$x_1 = 0.0312$	$x_1 = 0.0487$	$x_1 = 0.0677$	$x_1 = 0.1000$	$x_1 = 0.1622$
283.15	74.27	69.84	68.17	66.16	63.56	61.96
293.15	72.58	68.45	66.87	64.77	62.29	60.69
303.15	71.26	67.16	65.51	63.51	61.18	59.51
313.15	69.81	65.92	64.25	62.11	59.92	58.09
323.15	67.88	64.38	62.91	60.72	58.65	56.94

T/K	$x_1 = 0.2000$	$x_1 = 0.2250$	$x_1 = 0.3003$	$x_1 = 0.4038$	$x_1 = 0.5313$	$x_1 = 0.7232$	$x_1 = 1.0000$
283.15	59.76	59.29	58.10	55.99	53.85	51.80	49.76
293.15	58.61	58.16	56.96	54.97	53.04	50.76	49.15
303.15	57.45	57.18	56.00	53.97	51.68	50.13	48.67
313.15	56.34	55.96	54.90	52.87	50.75	48.92	48.07
323.15	55.14	54.92	53.71	51.93	50.06	48.05	47.46

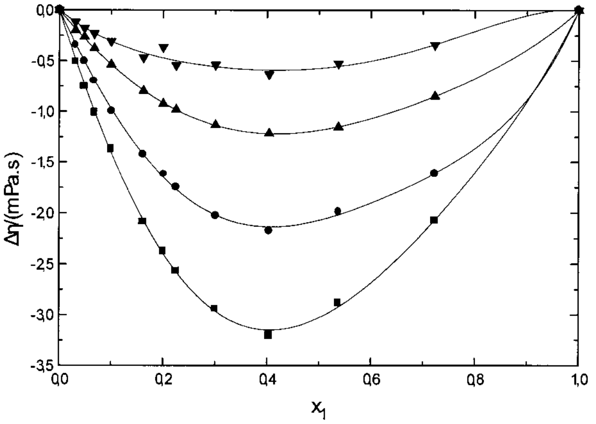

Figure 2. Viscosity deviations of ethylene glycol (1) + water (2) mixtures at 283.15 K (■), 293.15 K (●), 303.15 K (▲), and 313.15 K (▼).

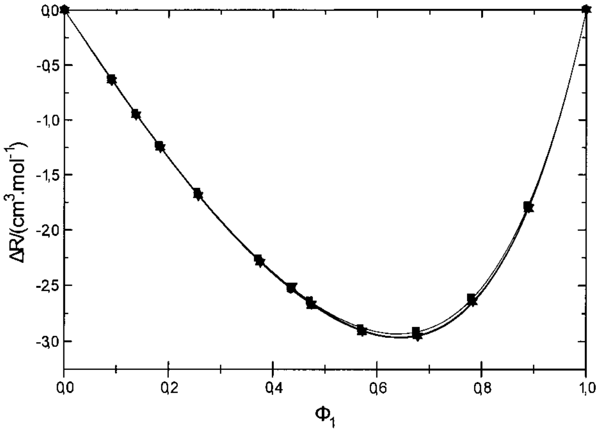

Figure 3. Molar refraction deviations versus the volume fraction of ethylene glycol (1) + water (2) mixtures independent of temperature.

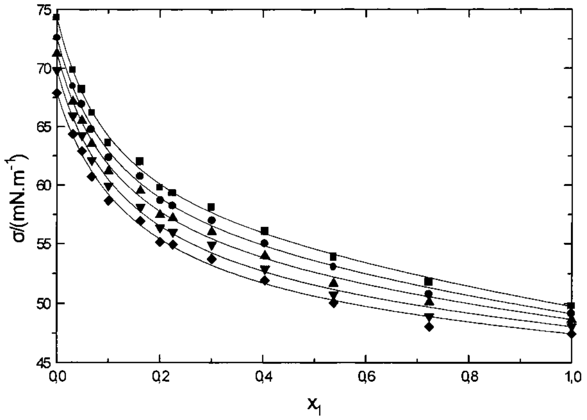

Figure 4. Surface tension of ethylene glycol (1) + water (2) mixtures at 283.15 K (■), 293.15 K (●), 303.15 K (▲), 313.15 K (▼), and 323.15 K (◆).

Table 6. Surface Tension Parameters α, *β* (Eq 10) for Ethylene Glycol (1) + Water (2)

t/°C	α	β
10	0.6721	0.8957
20	0.6912	0.8810
30	0.7253	0.8859
40	0.7623	0.8800
50	0.7792	0.8714

Table 7. Surface Tension Parameters k_1, k_2 (Eq 9) for Ethylene Glycol (1) + Water (2)

x_1	k_1	k_2	x_1	k_1	k_2
0.0312	71.185	0.134	0.2250	60.384	0.109
0.0487	69.484	0.131	0.3003	59.186	0.108
0.0677	67.516	0.135	0.4038	57.012	0.102
0.1000	64.777	0.122	0.5373	54.837	0.099
0.1622	63.230	0.126	0.7232	52.734	0.093
0.2000	60.913	0.115	1.0000	50.326	0.057

in Figure 3. The results were fitted to similar equations to eqs 2 and 3, and the parameters A_k and the standard deviations σ, are listed in Table 3.

The surface tension values (σ) are tabulated in Table 5 and are shown versus the mole fraction of ethylene glycol in Figure 4. The curves were obtained by fitting the data to the concentration according to the equation (Connors et al., 1989)

$$\sigma = \sigma_2 - \left(1 + \frac{\alpha x_2}{1 - \beta x_2}\right) x_1(\sigma_2 - \sigma_1) \qquad (9)$$

where σ_2, σ_1, and σ are the surface tension of water, ethylene glycol, and the mixture. The parameters α, β are given in Table 6. The surface tension of the mixtures was correlated with the temperature by fitting the following expression (Jasper, 1972)

$$\sigma = k_1 - k_2 t \qquad (10)$$

The coefficients k_1 and k_2 are given in Table 7, and the deviations were less than 0.5%. There is a decrease of the surface tension with increase of the temperature. In Figure 5 results are compared with literature values at different concentrations, together with those of Hoke and Chen (1991). The agreement is good, although a comparison cannot be complete, as the temperature and the concentrations differ from our own. Deviations from the ideal (excess

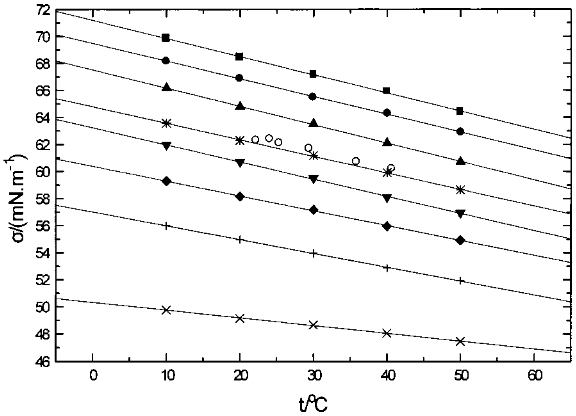

Figure 5. Surface tension of ethylene glycol (1) + water (2) mixtures as a function of temperature at different mole fractions of ethylene glycol: $x_2 = 0.0312$ (■), $x_2 = 0.0487$ (●), $x_2 = 0.0677$ (▲), $x_2 = 0.1000$ (∗), $x_2 = 0.1622$ (▼), $x_2 = 0.2250$ (◆), $x_2 = 0.4038$ (+), $x_2 = 1.0000$ (×), and $x_2 = 0.1000$; Hoke and Chen (1991) (○).

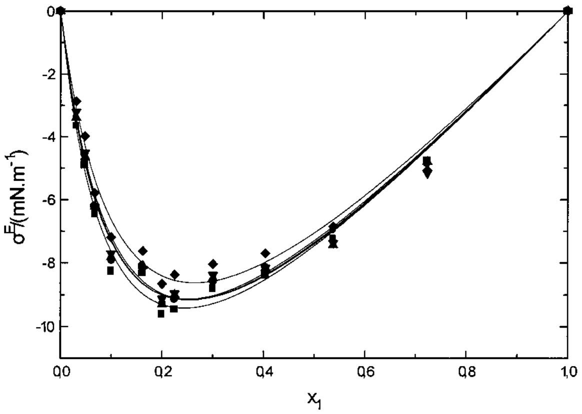

Figure 6. Excess surface tension as a function of the mole fraction of ethylene glycol at 283.15 K (■), 293.15 K (●), 303.15 K (▲), 313.15 K (▼), and 323.15 (◆).

surface tensions) were calculated from

$$\sigma^E = \sigma - (\sigma_2 x_2 + \sigma_1 x_1) \tag{11}$$

The experimental points were fitted to eqs 9 and 11, and the curves obtained are shown in Figure 6. The deviations show a decrease as the temperature increases.

The excess volumes, V^E, are negative for all mixtures, indicating interactions between water and ethylene glycol mixtures and disruption of the intramolecular interactions in water and ethylene glycol. The minima of V^E observed occur in the low mole fraction of ethylene glycol, $x_1 = 0.4$, in agreement with the published works (Reddy et al., 1994; Aminabhavi and Gopalakrisha, 1995).

The temperature coefficient of V^E is positive as shown in Figure 7.

The viscosity deviations ($\Delta\eta$) versus the mole fraction of ethylene glycol are listed in Table 4 and are plotted in Figure 2. The $\Delta\eta$ values are negative through all the composition range and for all temperatures investigated, and they increase with a rise in temperature. The minimum values shift toward the same water-rich region as in the case of the excess volume.

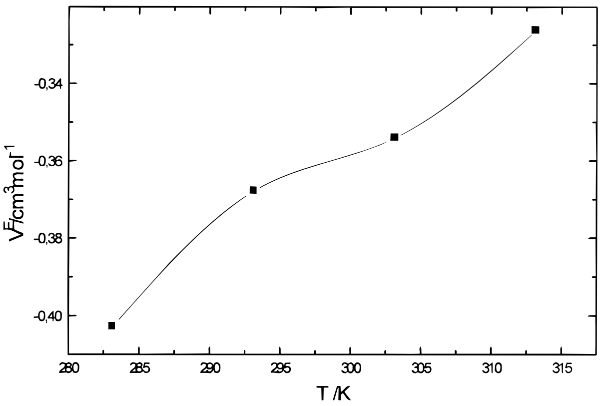

Figure 7. Temperature dependence of the equimolar excess volumes for water + ethylene glycol.

Figure 3 shows the refractive index deviations (ΔR) versus the volume fraction of ethylene glycol. The values are negative at the whole composition range for all mixtures. The minimum values occur at the same mole fractions as previously. The values are independent of temperature as predicted by the theory, the molar refraction depending only on the wavelength of the light used for measurement (Atkins, 1983; Glasstone, 1965).

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