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Experimental Surface Tensions and Derived Surface Properties of Binary Mixtures of Water + Alkoxyethanols (C_1E_m, $m = 1, 2, 3$) and Water + Ethylene Glycol Dimethyl Ether ($C_1E_1C_1$) at (298.15, 308.15, and 318.15) K

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Surface tensions, σ, for binary mixtures of water and ethylene glycol monomethyl ether (2-methoxyethanol, $C_3H_7O_2$; C_1E_1), diethylene glycol monomethyl ether (2-(2-methoxyethoxy)ethanol, $C_5H_{12}O_3$; C_1E_2), triethylene glycol monomethyl ether [2-{2-(2-methoxyethoxy)ethoxy}ethanol, $C_7H_{16}O_4$; C_1E_3), and ethylene glycol dimethyl ether (1,2-dimethoxyethane, $C_4H_{10}O_2$; $C_1E_1C_1$) are measured over the whole composition range. Measurements were made under atmospheric pressure at (298.15, 308.15, and 318.15) K. The experimental data have been used to calculate the surface tension deviations, $\Delta\sigma$, as a function of mole fractions, and values obtained are fitted to the Redlich−Kister polynomial equation to obtain the binary coefficients and the standard errors. The temperature dependence of σ at fixed compositions of solution was used to estimate surface enthalpy, H_σ, and surface entropy, S_σ. Also, the molar surface energies, E_σ, have been deduced at 298.15 K using experimental surface tension data.

Introduction

In the past few years, considerable effort has been spent on the measurement, analysis, and interpretation of basic thermodynamic properties such as density, excess volume, viscosity, and speed of sound of aqueous mixtures of alkyl poly(ethylene glycol) monoethers[1−8] and polyethers.[9−11] The literature reveals that surface tension data of these systems are not available. Surface tension (σ) plays an important role in mass transfer processes such as distillation, extraction, crystallization, and absorption. The key advantage of surface tension data is its ability to reflect changes at the surface caused by very small concentrations of solute. In the case of mixtures of simple liquids, such as water + ethanol,[12] surface changes of 1 mN·m^{-1} can be caused by a solute mole fraction of 1×10^{-3}. Despite the enormous importance of data on surface tension, which is helpful for the solution of a wide range of problems in chemical engineering, not enough attention has been paid to surface tension measurements of these systems. The aim of this work is to provide surface tension data for characterization of the molecular interactions and to relate the surface tensions of mixtures to the bulk-phase properties. Our specific motivation to undertake the present study is to extend the knowledge to brine systems, specifically in order to explore means of influencing crystallization behavior and evaporation pattern.

Experimental Section

Materials. Millipore grade water with conductivity < 0.66×10^{-8} S·cm^{-1} was used throughout this study. Synthesis grade ethylene glycol monomethyl ether (>99.5 mol %), diethylene glycol monomethyl ether (>98 mol %), triethylene glycol monomethyl ether (>97 mol %), and ethylene glycol dimethyl ether (>99 mol %) were obtained from Merck-Schuchardt and were used without further purification. All samples were kept in tightly sealed dark bottles to minimize the absorption of atmospheric moisture and CO_2, and they were dried over 0.4 nm molecular sieves before use. Before measurement, all liquids were partially degassed under vacuum.

Apparatus and Procedure. Binary mixtures were prepared by mass, using an analytical balance with a precision of ±0.0001 g (METTLER AE 200). The mole fraction of each mixture was obtained with an accuracy of 1×10^{-4} from the measured masses of the components. All molar quantities are based on the relative atomic mass table of 1985 issued by IUPAC.[13] Surface tension measurements in both the pure liquids and their mixtures were carried out at different temperatures using a DataPhysics DCAT II automated tensiometer, which employs the Wilhelmy plate method. A wettable platinum blade with an area of 3.98 mm^2 (wetted length 40.20 mm) was immersed in solution and slowly withdrawn to measure the vertical force, which is related to the surface tension by ($\sigma = Fg/W$), where σ is the surface tension, F is the measured vertical force, g is the acceleration of gravity, and W is the perimeter of the blade. The uncertainty obtained in σ values is <0.03 mN·m^{-1}. The temperature of the measurement cell was controlled with a Julabo water thermostat within ±0.1 K.

Results and Discussion

Experimental results for surface tensions, σ, and deviations in surface tensions, $\Delta\sigma$, at different temperatures are summarized in Table 1. Values of σ were best correlated by means of a polynomial type equation

$$F(x) = \sum_{i=1}^{n} A_i x_1^{\,i-1} \tag{1}$$

where x_1 represents the mole fraction of the organic

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Table 1. Experimental Surface Tension (σ) and Surface Tension Deviations ($\Delta\sigma$) for Binary Mixtures at Various Temperatures

	298.15 K		308.15 K		318.15 K			298.15 K		308.15 K		318.15 K	
	σ	$\Delta\sigma$	σ	$\Delta\sigma$	σ	$\Delta\sigma$		σ	$\Delta\sigma$	σ	$\Delta\sigma$	σ	$\Delta\sigma$
x_1	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$	x_1	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$	$mN \cdot m^{-1}$
Ethylene Glycol Monomethyl Ether (1) + Water (2)													
0	71.99	0	70.38	0	68.82	0	0.3373	36.75	−21.35	35.89	−20.88	34.92	−20.50
0.0509	56.33	−13.56	55.17	−13.16	53.90	−12.90	0.4783	35.02	−17.29	34.18	−16.90	33.22	−16.60
0.0559	55.11	−14.58	54.04	−14.08	52.82	−13.78	0.6258	32.91	−13.32	32.02	−13.04	31.13	−12.83
0.1052	46.86	−20.89	45.88	−20.25	44.80	−19.84	0.7814	31.78	−8.04	30.96	−7.88	30.02	−7.76
0.1494	43.31	−22.53	42.37	−21.98	41.35	−21.54	0.8912	31.02	−4.28	30.21	−4.20	29.28	−4.14
0.2215	39.70	−23.17	38.81	−22.63	37.83	−22.19	1	30.82	0	30.02	0	29.10	0
Diethylene Glycol Monomethyl Ether (1) + Water (2)													
0	71.99	0	70.38	0	68.82	0	0.4427	38.27	−17.23	37.12	−17.11	36.31	−16.63
0.0374	58.57	−12.03	57.12	−11.90	55.97	−11.51	0.4910	37.57	−16.13	36.46	−16.01	35.26	−15.95
0.0558	55.28	−14.63	53.90	−14.46	52.21	−14.61	0.5502	36.87	−14.62	35.92	−14.39	34.53	−14.55
0.1191	45.14	−22.41	43.86	−22.18	43.13	−21.42	0.7494	35.37	−8.70	34.49	−8.56	33.45	−8.49
0.1951	41.05	−23.67	39.78	−23.48	38.96	−22.86	0.8564	35.02	−5.03	34.18	−4.97	33.12	−4.98
0.3021	40.02	−20.71	38.80	−20.56	38.55	−19.43	1	34.73	0	33.91	0	32.95	0
Triethylene Glycol Monomethyl Ether (1) + Water (2)													
0	71.99	0	70.38	0	68.82	0	0.5536	37.76	−14.46	37.04	−14.19	36.12	−14.09
0.0564	54.62	−15.36	53.11	−15.32	51.53	−15.39	0.6606	37.13	−11.27	36.49	−11.04	35.85	−10.77
0.0964	47.43	−21.12	45.98	−21.07	44.95	−20.63	0.7512	36.91	−8.25	36.35	−8.05	35.46	−8.11
0.1673	41.73	−24.29	40.46	−24.13	39.01	−24.19	0.8736	36.69	−4.10	36.18	−3.98	35.29	−4.17
0.3525	40.45	−18.95	39.45	−18.74	38.12	−18.85	1	36.28	0	35.79	0	35.21	0
0.4740	39.10	−15.96	38.29	−15.69	37.42	−15.47							
Ethylene Glycol Dimethyl Ether (1) + Water (2)													
0	71.99	0	70.38	0	68.82	0	0.1942	28.55	−34.08	27.43	−33.73	26.53	−33.17
0.0470	48.35	−21.37	46.83	−21.32	46.47	−20.14	0.2612	27.81	−31.59	26.81	−31.17	25.92	−30.64
0.0660	42.42	−26.39	40.97	−26.28	40.62	−25.10	0.4132	26.08	−25.99	25.12	−25.65	24.78	−24.65
0.0759	40.62	−27.70	39.26	−27.52	37.71	−27.55	0.6313	25.41	−16.15	24.49	−15.92	23.60	−15.59
0.1286	31.96	−33.83	30.69	−33.59	29.26	−33.52	1	23.79	0	22.91	0	21.89	0

Table 2. Parameters, A_i, of Eq 1 and Their Standard Deviation, δ

T K	A_1 $mN \cdot m^{-1}$	A_2 $mN \cdot m^{-1}$	A_3 $mN \cdot m^{-1}$	A_4 $mN \cdot m^{-1}$	A_5 $mN \cdot m^{-1}$	A_6 $mN \cdot m^{-1}$	δ $mN \cdot m^{-1}$
Ethylene Glycol Monomethyl Ether (1) + Water (2)							
298.15	71.22	−347.47	1397.31	−2711.81	2451.15	−829.82	±0.97
308.15	69.72	−338.16	1352.61	−2616.16	2358.83	−796.98	±0.90
318.15	68.18	−331.30	1323.00	−2557.88	2306.04	−779.09	±0.88
Diethylene Glycol Monomethyl Ether (1) + Water (2)							
298.15	71.24	−367.28	1614.71	−3321.40	3137.72	−1100.38	±0.97
308.15	69.64	−362.15	1586.57	−3251.28	3063.76	−1072.74	±0.97
318.15	68.07	−358.50	1616.06	−3406.35	3282.49	−1168.95	±1.05
Triethylene Glycol Monomethyl Ether (1) + Water (2)							
298.15	71.87	−388.60	1737.13	−3556.54	3331.84	−1159.62	±0.94
308.15	70.26	−386.74	1737.86	−3560.33	3335.45	−1160.91	±0.94
318.15	68.67	−380.54	1690.27	−3414.68	3158.30	−1087.00	±0.90
Ethylene Glycol Dimethyl Ether (1) + Water (2)							
298.15	71.57	−621.05	3339.44	−8322.74	9423.03	−3866.46	±0.72
308.15	69.95	−618.77	3348.65	−8375.51	9502.45	−3903.85	±0.75
318.15	68.63	−604.98	3204.85	−7827.67	8700.65	−3519.60	±0.44

component. The parameters A_i and the standard deviation δ are given in Table 2.

Measured σ values for the various mixtures have been used to calculate the values of $\Delta\sigma$ from a mole fraction average, using eq 2,

$$\Delta\sigma = \sigma - (x_1\sigma_1 + x_2\sigma_2) \tag{2}$$

where σ is the surface tension of the binary mixture, x_1 and x_2 are the mole fractions, and σ_1 and σ_2 are the surface tension values of components 1 and 2, respectively. The variations of σ and $\Delta\sigma$ with mole fraction of organic solvents at various temperatures are shown in Figures 1 and 2. The $\Delta\sigma$ values over the whole mole fraction range were fitted to the Redlich−Kister[14] polynomial equation:

$$F(x) = x_1x_2\sum_{i=0}^{n}A_i(2x_1 - 1)^i \tag{3}$$

The coefficients A_i of eq 3 were calculated by the method of least squares. The values obtained for the coefficients A_i along with the standard deviations δ are reported in Table 3.

The temperature dependence of σ at fixed compositions of solution allows estimation of surface enthalpy, H_σ, and surface entropy, S_σ. This is obtained at a fixed x_1 using the relation

$$\sigma = H_\sigma + S_\sigma T \tag{4}$$

The definitions[15,16] of these quantities are $H_\sigma = \sigma(\mathrm{d}\sigma/\mathrm{d}T)$ and $S_\sigma = -(\mathrm{d}\sigma/\mathrm{d}T)$. The values of H_σ ($mN \cdot m^{-1} \cdot K^{-1}$) and S_σ ($N \cdot m^{-1}$) thus obtained are reported in Table 4 and graphically represented in Figures 3 and 4. Molar surface energy, E_σ, values have also been deduced at 298.15 K from experimental surface tension data and the density data

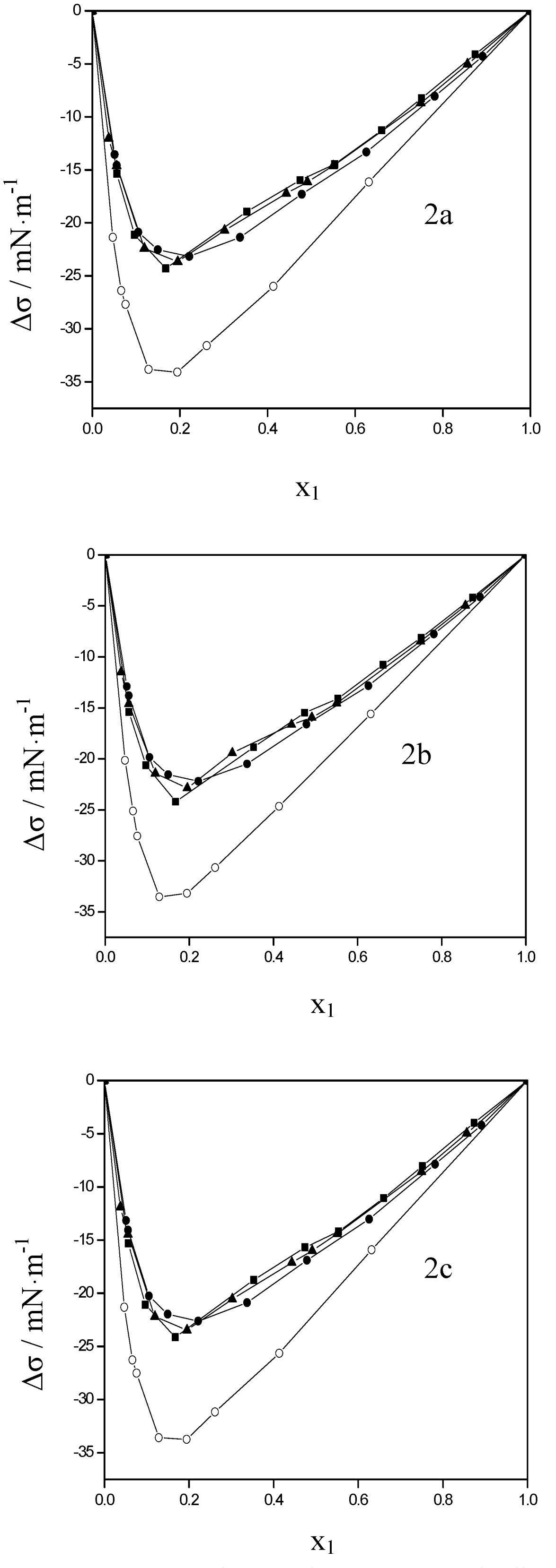

Figure 1. Comparison of the surface tension, σ, of different mixtures at (a) 298.15 K, (b) 308.15 K, and (c) 318.1K: (●) ethylene glycol monomethyl ether; (▲) diethylene glycol monomethyl ether; (■) triethylene glycol monomethyl ether; (○) ethylene glycol dimethyl ether.

reported elsewhere[1,3,6,9] using the relation

$$E_\sigma = \sigma(M/\rho)^{2/3} \tag{5}$$

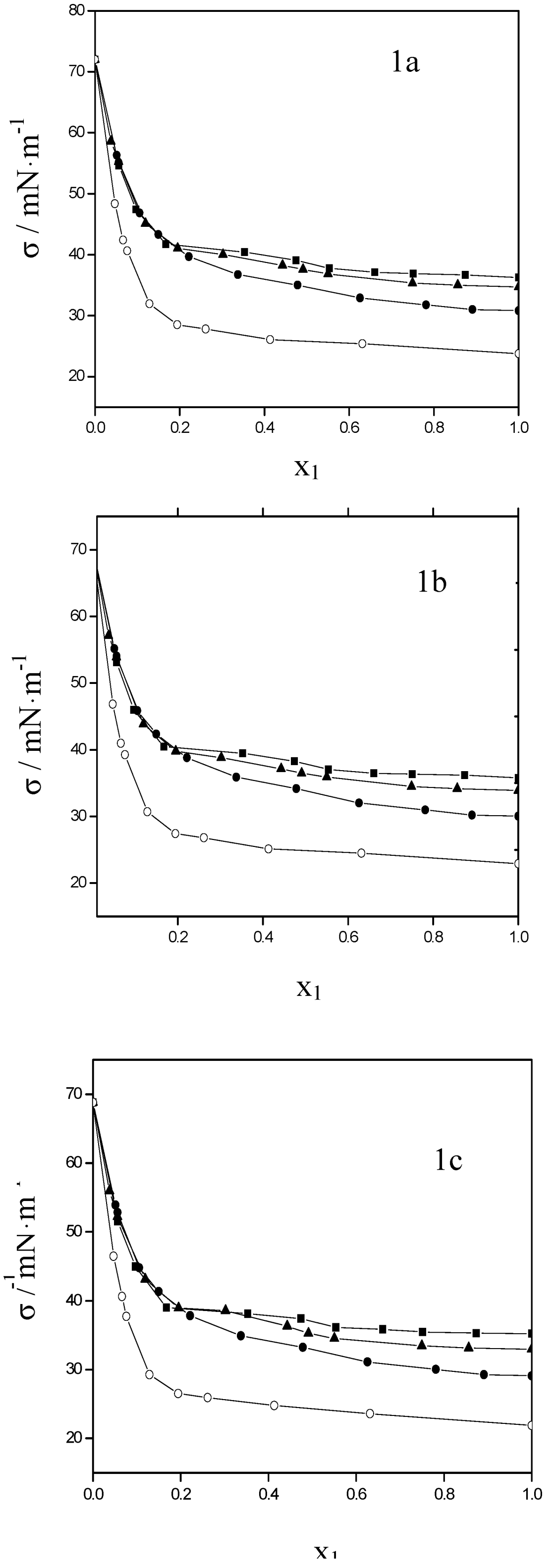

Figure 2. Comparison of the deviations in surface tension, $\Delta\sigma$, of different mixtures at (a) 298.15 K, (b) 308.15 K, and (c) 318.15 K: (●) ethylene glycol monomethyl ether; (▲) diethylene glycol monomethyl ether; (■) triethylene glycol monomethyl ether; (○) ethylene glycol dimethyl ether.

where ρ is the density and M is the molar mass of the compound. The values of E_σ are reported in Table 5 and graphically represented in Figure 5.

Plots of σ versus mole fraction of cosolvents, at different temperatures shown in Figure 1, indicate that the values

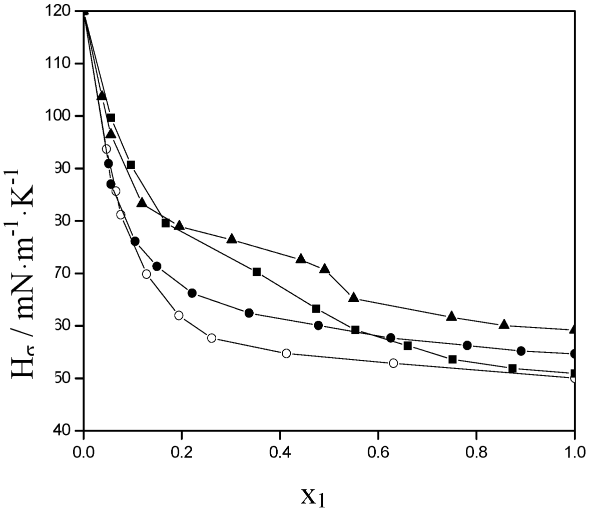

Figure 3. Plots of surface enthalpy, H_σ, as a function of mole fraction of different mixtures: (●) ethylene glycol monomethyl ether; (▲) diethylene glycol monomethyl ether; (■) triethylene glycol monomethyl ether; (○) ethylene glycol dimethyl ether.

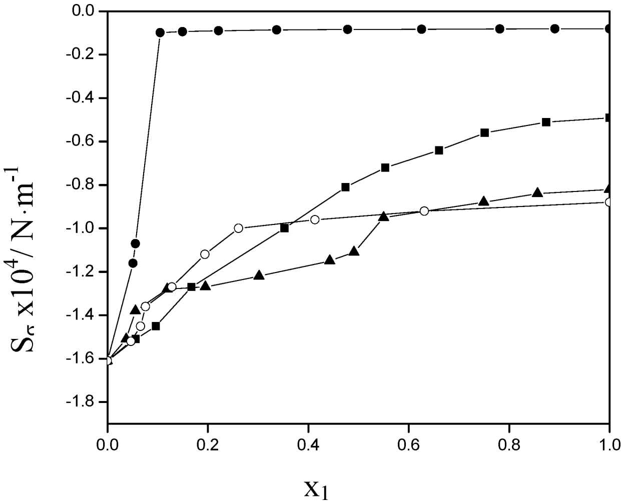

Figure 4. Plots of surface entropy, S_σ, as a function of mole fraction of different mixtures: (●) ethylene glycol monomethyl ether; (▲) diethylene glycol monomethyl ether; (■) triethylene glycol monomethyl ether; (○) ethylene glycol dimethyl ether.

Table 3. Parameters, A_i, of Eq 3 and Their Standard Deviation, δ

T / K	A_1 / mN·m^{-1}	A_2 / mN·m^{-1}	A_3 / mN·m^{-1}	A_4 / mN·m^{-1}	A_5 / mN·m^{-1}	δ / mN·m^{-1}
Ethylene Glycol Monomethyl Ether (1) + Water (2)						
298.15	−68.02	50.45	−44.17	95.00	−94.12	±0.35
308.15	−66.48	49.54	−44.67	91.33	−88.21	±0.33
318.15	−65.33	48.47	−43.79	89.32	−86.20	±0.32
Diethylene Glycol Monomethyl Ether (1) + Water (2)						
298.15	−63.56	43.56	−51.42	119.33	−100.52	±0.41
308.15	−62.92	44.35	−50.84	116.18	−99.40	±0.42
318.15	−62.47	36.71	−38.86	124.02	−112.89	±0.32
Triethylene Glycol Monomethyl Ether (1) + Water (2)						
298.15	−61.67	43.41	−61.44	129.58	−87.08	±0.22
308.15	−60.63	43.47	−61.35	129.68	−87.45	±0.21
318.15	−59.99	46.15	−66.38	121.44	−82.03	±0.26
Ethylene Glycol Dimethyl Ether (1) + Water (2)						
298.15	−89.29	93.53	−41.62	3.34	−388.53	±0.33
308.15	−88.29	91.80	−37.18	10.24	−387.54	±0.36
318.15	−91.74	51.36	41.50	465.84		±0.26

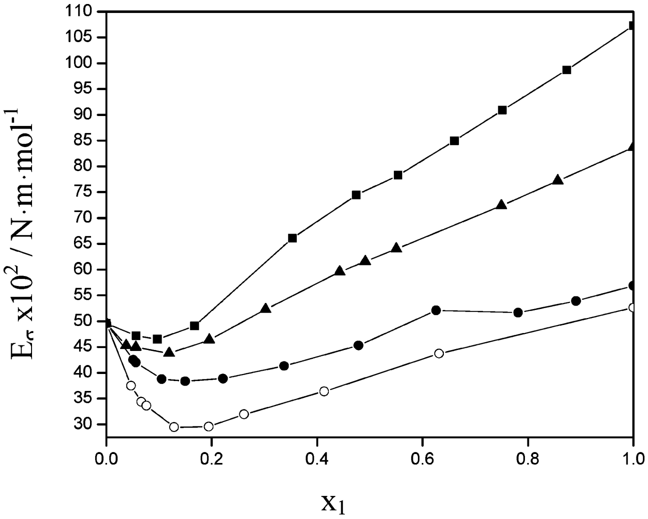

Figure 5. Plots of surface energy, E_σ, as a function of mole fraction of different mixtures at 298.15 K: (●) ethylene glycol monomethyl ether; (▲) diethylene glycol monomethyl ether; (■) triethylene glycol monomethyl ether; (○) ethylene glycol dimethyl ether.

Table 4. Derived Surface Enthalpy, H_σ, and Surface Entropy, S_σ, for Various Binary Mixtures

x_1	H_σ / mN·m^{-1}·K^{-1}	10^4S_σ / N·m^{-1}	x_1	H_σ / mN·m^{-1}·K^{-1}	10^4S_σ / N·m^{-1}
Ethylene Glycol Monomethyl Ether (1) + Water (2)					
0	119.99	−1.61	0.3373	62.39	−0.086
0.0509	90.92	−1.16	0.4783	60.06	−0.084
0.0559	87.01	−1.07	0.6258	57.66	−0.083
0.1052	76.08	−0.098	0.7814	56.23	−0.082
0.1494	71.34	−0.094	0.8912	55.17	−0.081
0.2215	66.24	−0.089	1	54.67	−0.080
Diethylene Glycol Monomethyl Ether (1) + Water (2)					
0	119.99	−1.61	0.4427	72.56	−1.15
0.0374	103.65	−1.51	0.4910	70.66	−1.11
0.0558	96.42	−1.38	0.5502	65.19	−0.95
0.1191	83.30	−1.28	0.7494	61.61	−0.88
0.1951	78.92	−1.27	0.8564	60.06	−0.84
0.3021	76.39	−1.22	1	59.17	−0.82
Triethylene Glycol Monomethyl Ether (1) + Water (2)					
0	119.99	−1.61	0.5536	59.23	−0.72
0.0564	99.64	−1.51	0.6606	56.21	−0.64
0.0964	90.66	−1.45	0.7512	53.61	−0.56
0.1673	79.60	−1.27	0.8736	51.90	−0.51
0.3525	70.27	−1.00	1	50.89	−0.49
0.4740	63.25	−0.81			
Ethylene Glycol Dimethyl Ether (1) + Water (2)					
0	119.99	−1.61	0.1942	61.94	−1.12
0.0470	93.67	−1.52	0.2612	57.63	−1.00
0.0660	85.65	−1.45	0.4132	54.70	−0.96
0.0759	81.17	−1.36	0.6313	52.84	−0.92
0.1286	69.82	−1.27	1	50.03	−0.88

of σ of all the binary mixtures investigated here decrease with an increase of the cosolvent mole fraction and follow the sequence ethylene glycol dimethyl ether < ethylene glycol monomethyl ether < diethylene glycol monomethyl ether < triethylene glycol monomethyl ether, indicating an increase in surface tension with the addition of an oxyethylene group in an alkoxyethanol molecule. There is a systematic decrease in σ with a rise in temperature for all mixtures. A comparison of data at different temperatures reveals that the temperature coefficient $(\partial\sigma/\partial T)_P$ decreases for all mixtures with increasing temperature. Further, Figure 2 shows that $\Delta\sigma$ is negative, with a sharp minimum in the water rich region, for all the binary mixtures studied, and the deviations are in the order ethylene glycol dimethyl ether < triethylene glycol monomethyl ether < diethylene glycol monomethyl ether < ethylene glycol monomethyl ether. It is worth noting that the dependence of $\Delta\sigma$ upon

Table 5. Derived Molar Surface Energies, E_σ, for Various Binary Mixtures at 298.15 K

x_1	$\frac{10^2E_\sigma}{N\cdot m\cdot mol^{-1}}$	x_1	$\frac{10^2E_\sigma}{N\cdot m\cdot mol^{-1}}$	x_1	$\frac{10^2E_\sigma}{N\cdot m\cdot mol^{-1}}$
Ethylene Glycol Monomethyl Ether (1) + Water (2)					
0	49.57	0.1494	38.36	0.6258	52.10
0.0509	42.55	0.2215	38.88	0.7814	51.69
0.0559	42.01	0.3373	41.34	0.8912	53.93
0.1052	38.78	0.4783	45.29	1	56.87
Diethylene Glycol Monomethyl Ether (1) + Water (2)					
0	49.57	0.1951	46.36	0.5502	64.01
0.0374	45.29	0.3021	52.31	0.7494	72.41
0.0558	44.99	0.4427	59.58	0.8564	77.23
0.1191	43.76	0.4910	61.56	1	83.66
Triethylene Glycol Monomethyl Ether (1) + Water (2)					
0	49.57	0.3525	66.07	0.7512	90.90
0.0564	47.20	0.4740	74.46	0.8736	98.70
0.0964	46.52	0.5536	78.25	1	107.31
0.1673	49.06	0.6606	84.95		
Ethylene Glycol Dimethyl Ether (1) + Water (2)					
0	49.57	0.1286	29.46	0.4132	36.40
0.0470	37.51	0.1942	29.57	0.6313	43.70
0.0660	34.38	0.2612	31.97	1	52.63
0.0759	33.64				

composition tends to resemble that of bulk phase properties such as excess molar volumes and excess molar isentropic compressibilities.

Acknowledgment

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