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Surface Properties of Binary Mixtures of Ethylene Glycol with a Series of Aliphatic Alcohols (1-Pentanol, 1-Hexanol, and 1-Heptanol)

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Surface tensions of binary mixtures of ethylene glycol and a homologous series of aliphatic alcohols (1-pentanol, 1-hexanol, and 1-heptanol) were measured as a function of composition using the ring detachment method in the temperature range between (298.15 and 318.15) K. Temperature influence on the behavior of surface tensions and surface properties of binary mixtures has often been used to obtain information about solute structural effects on ethylene glycol. Values of surface tension were correlated with temperature and mole fraction. The experimental data have been used to calculate the surface tension deviation, $\delta\sigma$, as a function of mole fraction, and values obtained are fitted to the Redlich–Kister polynomial equation to obtain the coefficients and the standard errors. The maximum deviation was in both cases always less than 0.5 %. The temperature dependence of σ at fixed composition of solutions was used to estimate surface enthalpy, H^s , and surface entropy, S^s .

Introduction

Among experimental methods of investigating intermolecular interactions and forces in liquids, the surface tension is distinguished because of its simplicity and precision. The surface tension reveals information about the structure and energetics of the surface region between two phases. Variation in surface energy depends on variation in molecular forces and that of the density of patching or molecular size.¹ Thus, the surface tension of solution has an important influence on the transfer of mass and energy across an interface and is one of the most interesting thermophysical properties in many industrial applications such as liquid–liquid extraction, gas absorption, distillation, and condensation. A part of the systematic work on the surface tension of liquids is the determination, correlation, and calculation of surface thermodynamic parameters. Aqueous and nonaqueous solutions of alcohols have been extensively studied due to the importance they have in the understanding of interactions between polar and nonpolar groups with solvent.^{2–11} Also, the surface properties of solution of nonelectrolytes often reflect the interactions of hydrophobic parts of the guest molecules adsorbed at the liquid–vapor interface. For diols, the influence of hydrogen bonds within the diol molecules and intermolecular hydrogen bond with alcohol is reflected in a complex thermodynamic behavior in alcoholic solutions.

Recently, the surface tension of binary mixtures of linear chain alcohols and cyclic alcohols with ethylene glycol was studied.^{12,13} In the present work, we report the surface tension of binary mixtures containing 1-pentanol, 1-hexanol, and 1-heptanol with ethylene glycol at temperatures ranging between (298.15 and 318.15) K. Values of surface tension are correlated with temperature and mole fraction. The experimental data have been used to find surface tension deviation, $\delta\sigma$, and the surface thermodynamic parameters

Table 1. Comparison of Experimental Surface Tension, σ , and Densities, ρ , of Pure Liquids at 298.15 K with Literature Values

substance	$\sigma(\text{exptl})$ $\text{mN}\cdot\text{m}^{-1}$	$\sigma(\text{ref})$ $\text{mN}\cdot\text{m}^{-1}$	$\rho(\text{exptl})$ $\text{g}\cdot\text{cm}^{-3}$	$\rho(\text{ref})$ $\text{g}\cdot\text{cm}^{-3}$
ethylene glycol	47.89	47.99 ¹⁴	1.10971	1.00030 ¹⁴
1-pentanol	24.89	24.74 ¹⁵ 25.36 ¹⁶ 24.97 ¹⁹	0.81110	0.81080 ¹⁵ 0.81083 ¹⁹
1-hexanol	25.69	25.81 ¹⁶ 25.73 ¹⁹	0.81502	0.81515 ¹⁸ 0.81515 ¹⁸
1-heptanol	26.38	26.50 ¹⁷ 26.47 ¹⁹	0.81911	0.81860 ¹⁷ 0.81875 ¹⁹

(surface entropy and surface enthalpy) over the entire concentration range at several temperatures.

Experimental Section

All of the solvents contain ethylene glycol (> 99 %), 1-pentanol (> 99 %), 1-hexanol (> 99 %), and 1-heptanol (> 99 %) purchased from Merck and used without further purification. Their density values were measured by means of an Anton-Paar digital precision densitometer (model DMA 4500) operated in static mode and calibrated with double distilled water. The surface tensions of the pure liquids and their mixtures were determined using a ring-detachment method using a KSV (Sigma 70, Finland) tensiometer. The uncertainty of the surface tension measurement is $\pm 0.1 \text{ mN}\cdot\text{m}^{-1}$ of the final value of the surface tension, and the corresponding reproducibility is $\pm 0.01 \text{ mN}\cdot\text{m}^{-1}$. The platinum ring was thoroughly cleaned and flame-dried before each measurement. All mixtures were prepared by mass in glass stoppered flasks. The balance precision was $\pm 0.0001 \text{ g}$. The temperature was kept constant within $\pm 0.1 \text{ K}$ by a water bath circulator (Pharmacia). The uncertainty of the mole fractions was estimated to be within ± 0.0001 . Surface tension and density values of pure liquids are reported in Table 1 and compared with literature values.

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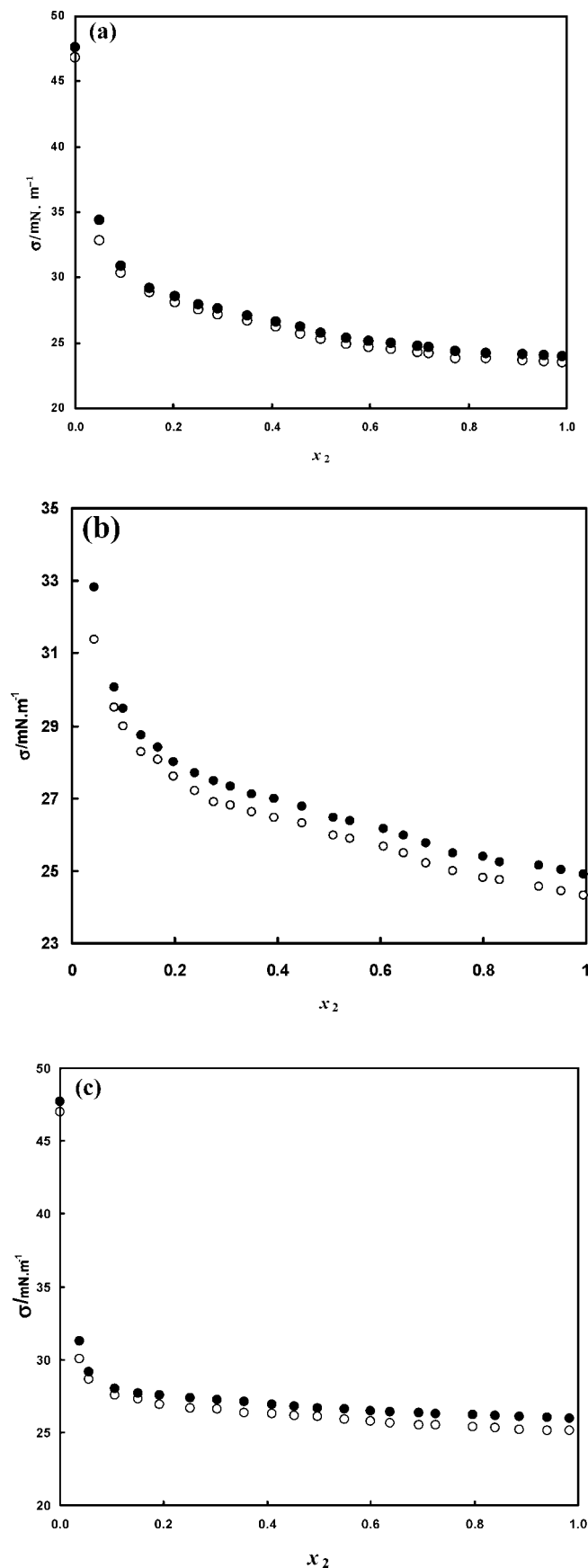


Figure 1. (a) Plot of surface tension ($\sigma/\text{mN}\cdot\text{m}^{-1}$) vs mole fraction of 1-pentanol (x_2) at \bullet , 303.15 K and \circ , 313.15 K. (b) Plot of surface tension ($\sigma/\text{mN}\cdot\text{m}^{-1}$) vs mole fraction of 1-hexanol (x_2) at \bullet , 303.15 K and \circ , 313.15 K. (c) Plot of surface tension ($\sigma/\text{mN}\cdot\text{m}^{-1}$) vs mole fraction of 1-heptanol (x_2) at \bullet , 303.15 K and \circ , 313.15 K.

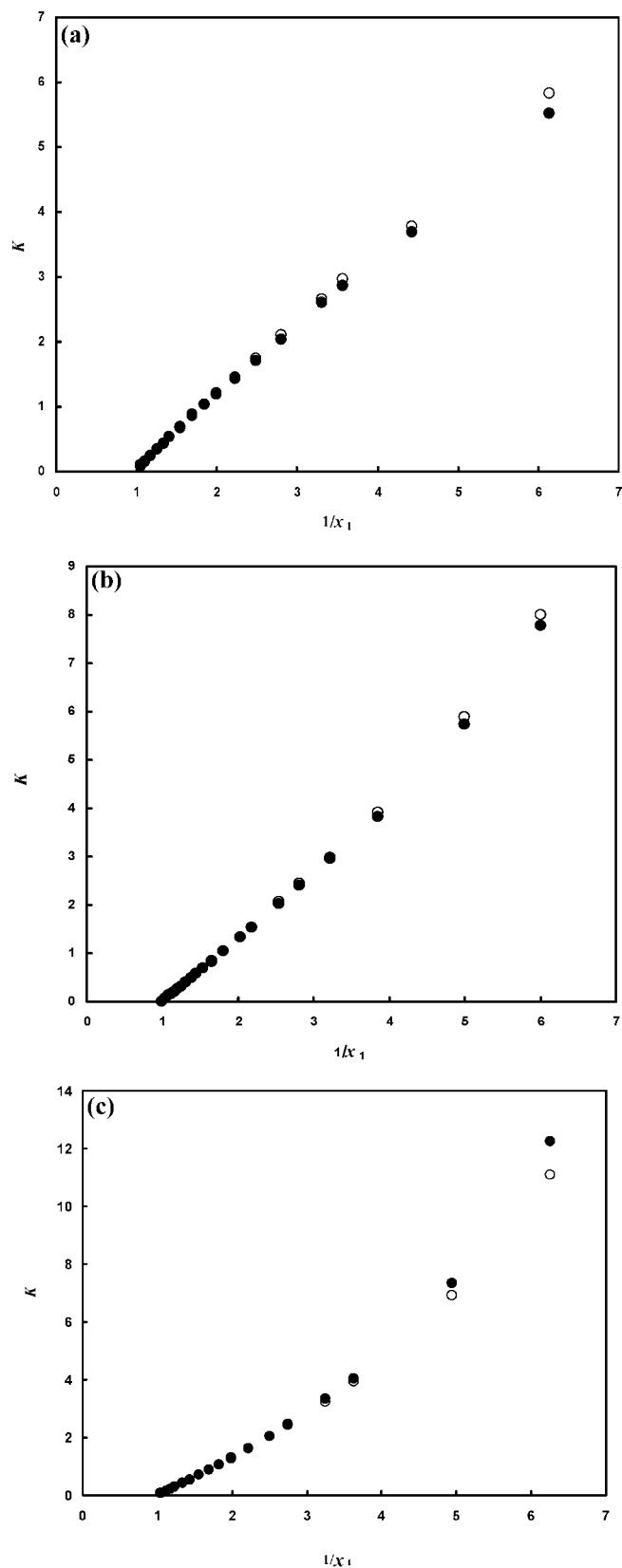


Figure 2. (a) Plot of $K = x_2/(\sigma^* - x_2)$ vs $1/x_1$ for ethylene glycol + 1-pentanol mixtures at \bullet , 303.15 K and \circ , 318.15 K [ethylene glycol is component 1 and 1-pentanol is component 2]. (b) Plot of $K = x_2/(\sigma^* - x_2)$ vs $1/x_1$ for ethylene glycol + 1-hexanol mixtures at \bullet , 303.15 K and \circ , 318.15 K [ethylene glycol is component 1 and 1-hexanol is component 2]. (c) Plot of $K = x_2/(\sigma^* - x_2)$ vs $1/x_1$ for ethylene glycol + 1-heptanol mixtures at \bullet , 303.15 K and \circ , 318.15 K [ethylene glycol is component 1 and 1-heptanol is component 2].

Table 2. Surface Tension, σ , of 1-Pentanol, 1-Hexanol, and 1-Heptanol Mixed with Ethylene Glycol at Various Temperatures and Mole Fractions of Alcohols (x_2)

	x_2	$\sigma/\text{mN}\cdot\text{m}^{-1}$					x_2	$\sigma/\text{mN}\cdot\text{m}^{-1}$				
		T/K						T/K				
		298.15	303.15	308.15	313.15	318.15		298.15	303.15	308.15	313.15	318.15
1-pentanol	0.0488	35.01	34.40	33.65	33.18	32.85	0.5520	26.53	25.39	25.21	25.02	24.91
	0.0931	32.10	30.89	30.71	30.51	30.37	0.5982	25.71	25.15	24.98	24.81	24.68
	0.1523	29.93	29.14	29.25	29.05	28.87	0.6425	26.32	24.96	24.79	24.67	24.53
	0.2043	29.26	28.56	28.41	28.27	28.09	0.6979	26.21	24.75	24.57	24.41	24.25
	0.2502	28.35	27.94	27.81	27.67	27.51	0.7197	26.12	24.63	24.51	24.37	24.19
	0.2912	28.04	27.65	27.51	27.35	27.17	0.7739	25.98	24.35	24.18	23.98	23.82
	0.3503	27.01	27.05	26.91	26.80	26.68	0.8370	24.89	24.23	24.15	23.92	23.78
	0.4094	27.39	26.62	26.48	26.31	26.18	0.9113	25.45	24.13	24.00	23.81	23.65
	0.4586	26.71	26.19	25.98	25.82	25.69	0.9536	25.25	24.02	23.89	23.75	23.61
	0.5003	26.75	25.77	25.61	25.43	25.27						
1-hexanol	0.0000	47.79	47.57	47.25	47.10	46.40	0.5405	26.82	26.38	26.25	26.07	25.88
	0.0429	32.50	32.81	32.68	31.55	31.38	0.6061	26.71	26.17	26.01	25.85	25.67
	0.1009	29.07	29.49	29.34	29.27	28.97	0.6452	26.62	25.97	25.78	25.65	25.48
	0.2761	28.00	27.47	27.31	27.15	26.91	0.6897	26.51	25.75	25.58	25.38	25.21
	0.3098	27.90	27.32	27.19	27.02	26.80	0.7407	26.43	25.48	25.30	25.16	24.99
	0.3500	27.38	27.12	26.98	26.81	26.63	0.8000	26.34	25.38	25.19	25.02	24.81
	0.3942	27.32	26.98	26.83	26.68	26.47	0.8333	26.28	25.25	25.17	24.98	24.75
	0.4468	27.24	26.78	26.63	26.47	26.31	0.9091	26.22	25.16	24.97	24.78	24.56
	0.5079	27.03	26.48	26.35	26.17	25.97	0.9524	26.16	25.02	24.81	24.61	24.45
	0.0559	29.24	29.16	28.96	28.78	28.64	0.5502	26.25	26.60	26.24	26.07	25.91
1-heptanol	0.1059	27.73	28.02	27.88	27.71	27.55	0.5999	26.17	26.48	26.12	25.92	25.78
	0.1508	27.18	27.66	27.55	27.42	27.28	0.6369	26.00	26.39	26.09	25.83	25.65
	0.1914	27.05	27.54	27.21	27.06	26.91	0.6928	26.08	26.31	26.01	25.68	25.52
	0.2512	26.93	27.36	26.98	26.81	26.67	0.7246	26.01	26.25	25.91	25.62	25.48
	0.3027	26.78	27.20	26.84	26.71	26.57	0.7978	25.92	26.18	25.85	25.52	25.37
	0.3559	26.67	27.07	26.69	26.53	26.35	0.8403	25.83	26.12	25.78	25.51	25.31
	0.4085	26.57	26.89	26.65	26.42	26.28	0.8875	25.71	26.07	25.68	25.45	25.21
	0.4532	26.41	26.77	26.57	26.32	26.16	0.9404	25.62	26.02	25.62	25.45	25.14
	0.4966	26.32	26.67	26.42	26.23	26.07						

Table 3. Surface Tension Parameters a and b (Equations 3 and 4) for 1-Pentanol/Ethylene Glycol, 1-Hexanol/Ethylene Glycol, and 1-Heptanol/Ethylene Glycol

system		T/K				
		298.15	303.15	308.15	313.15	318.15
ethylene glycol + 1-pentanol	b	0.791	0.266	0.893	0.794	0.752
	a	1.020	1.165	0.967	0.112	1.173
	SD	0.061	0.035	0.033	0.083	0.093
ethylene glycol + 1-hexanol	b	0.952	0.691	0.673	0.655	0.661
	a	0.969	1.067	1.081	1.089	1.827
	SD	0.019	0.066	0.051	0.083	0.093
ethylene glycol + 1-heptanol	b	0.894	0.576	0.602	0.614	0.607
	a	1.024	1.148	1.133	1.122	1.126
	SD	0.027	0.007	0.024	0.054	0.037

Results and Discussion

The surface tensions of the studied binary mixtures at the indicated temperatures are given in Table 2. In all the systems at specified temperatures, the surface tension, σ , decreased with an increase of the alcohol mole fraction. This trend is nonlinear, and the change in surface tension caused by a given change in alcohol concentration is larger at low concentration than at high concentration (see Figure 1). Connors and Wright⁹ proposed an equation for surface tension of mixtures that has been used successfully in the chemical literature as follows^{4,9,12}

$$\sigma = \sigma_1 - \left(1 + \frac{bx_1}{1 - ax_1}\right)x_2(\sigma_1 - \sigma_2) \quad (1)$$

where σ_1 and σ_2 are the surface tensions of pure ethylene glycol and pure alcohol, respectively, and x_1 and x_2 are the corresponding mole fractions. The σ is the surface tension of the

mixed solvent. By definition of reduced surface tension (or dimensionless surface tension) as

$$\sigma^* = \frac{\sigma_1 - \sigma}{\sigma_1 - \sigma_2} \quad (2)$$

and by linearization of eq 1, we obtain

$$\frac{x_2}{(\sigma^* - x_2)} = \frac{1}{ax_1} - \frac{b}{a} \quad (3)$$

So the fitted parameters b and a were determined at each

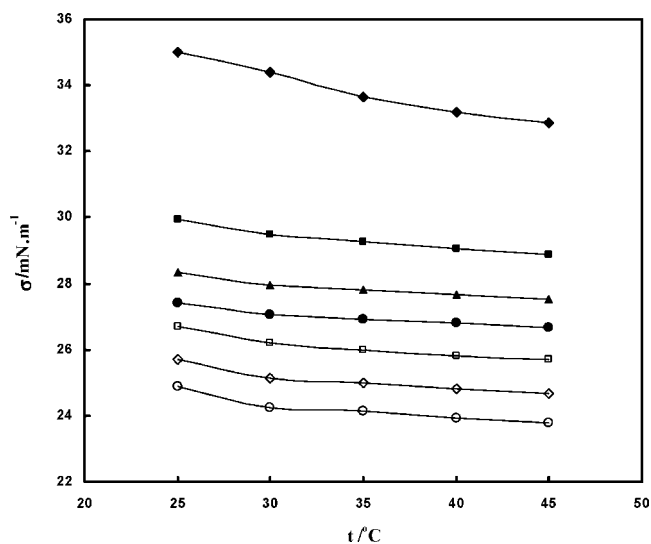
**Figure 3.** Surface tension ($\sigma/\text{mN}\cdot\text{m}^{-1}$) of pentanol (2) + ethylene glycol (1) as a function of temperature at \blacklozenge , $x_2 = 0.0488$; \blacksquare , $x_2 = 0.1523$; \blacktriangle , $x_2 = 0.2502$; \bullet , $x_2 = 0.3503$; \square , $x_2 = 0.4586$; \diamond , $x_2 = 0.5982$; \circ , $x_2 = 0.8320$.

Table 4. Surface Tension Parameters of Equation 4 for the 1-Alkanol/Ethylene Glycol Systems

	x_2	K_1 $\text{mN} \cdot \text{m}^{-1}$	K_2 $\text{mN} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$	SD	x_2	K_1 $\text{mN} \cdot \text{m}^{-1}$	K_2 $\text{mN} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$	SD
1-pentanol	0.0488	38.014	0.120	0.26	0.5003	28.076	0.066	0.29
	0.0931	33.604	0.077	0.38	0.5520	27.939	0.072	0.36
	0.1523	31.270	0.056	0.34	0.5982	27.866	0.076	0.41
	0.2043	30.359	0.056	0.19	0.6425	27.763	0.077	0.44
	0.2502	29.381	0.043	0.13	0.6979	27.820	0.085	0.47
	0.2912	28.972	0.041	0.08	0.7739	27.969	0.099	0.58
	0.3503	28.270	0.037	0.11	0.8370	27.709	0.094	0.57
	0.4094	28.507	0.055	0.22	0.9113	27.320	0.088	0.50
	0.4586	28.325	0.062	0.26	0.9536	26.909	0.071	0.47
	0.0429	34.63	0.070	0.43	0.5405	27.81	0.044	0.09
1-hexanol	0.1009	30.89	0.043	0.11	0.6061	27.76	0.048	0.14
	0.1680	29.99	0.047	0.23	0.6452	27.72	0.052	0.18
	0.2761	29.11	0.050	0.12	0.6897	27.76	0.059	0.21
	0.3098	28.99	0.050	0.14	0.7407	27.71	0.064	0.29
	0.3500	28.25	0.036	0.03	0.8000	27.47	0.068	0.28
	0.3942	28.25	0.040	0.06	0.8333	27.62	0.067	0.30
	0.4468	28.20	0.043	0.10	0.9091	27.73	0.074	0.31
	0.5079	28.10	0.049	0.13	0.9524	27.69	0.077	0.35
	0.0559	30.862	0.052	0.15	0.5502	27.861	0.044	0.06
	0.1059	29.047	0.033	0.02	0.5999	27.832	0.047	0.07
1-heptanol	0.1508	28.406	0.025	0.01	0.6369	27.674	0.046	0.05
	0.1914	28.646	0.039	0.06	0.6928	27.945	0.055	0.05
	0.2512	28.659	0.045	0.07	0.7246	27.837	0.054	0.06
	0.3027	28.417	0.042	0.07	0.7978	27.800	0.055	0.06
	0.3559	28.448	0.048	0.07	0.8403	27.665	0.053	0.04
	0.4085	28.257	0.045	0.05	0.8875	27.558	0.052	0.06
	0.4532	28.071	0.043	0.02	0.9404	27.441	0.051	0.07
	0.4966	27.800	0.039	0.03				

Table 5. Surface Entropy, S^s , and Surface Enthalpy, H^s , for the Systems under Investigation

	x_2	$S^s/\text{mN} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$	$H^s/\text{mN} \cdot \text{m}^{-1}$	x_2	$S^s/\text{mN} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$	$H^s/\text{mN} \cdot \text{m}^{-1}$
1-pentanol	0.0488	0.120	70.871	0.5003	0.066	46.418
	0.0931	0.077	54.986	0.5520	0.072	48.046
	0.1523	0.056	46.978	0.5982	0.076	49.058
	0.2043	0.056	46.008	0.6425	0.077	49.385
	0.2502	0.043	41.264	0.6979	0.085	51.600
	0.2912	0.041	40.198	0.7739	0.099	55.741
	0.3503	0.037	38.506	0.8370	0.094	54.052
	0.4094	0.055	43.661	0.9113	0.088	51.785
	0.4586	0.062	45.596	0.9536	0.071	46.578
	0.0429	0.070	53.360	0.5405	0.044	39.872
1-hexanol	0.1009	0.043	42.684	0.6061	0.048	41.014
	0.1680	0.047	42.957	0.6452	0.052	42.116
	0.2761	0.050	42.900	0.6897	0.059	44.092
	0.3098	0.050	42.800	0.7407	0.064	45.502
	0.3500	0.036	38.168	0.8000	0.068	46.723
	0.3942	0.040	39.240	0.8333	0.067	46.127
	0.4468	0.043	40.173	0.9091	0.074	48.272
	0.5079	0.049	41.513	0.9524	0.077	48.987
	0.0559	0.052	45.117	0.5502	0.044	39.922
	0.1059	0.033	38.183	0.5999	0.047	40.616
1-heptanol	0.1508	0.025	35.170	0.6369	0.046	40.065
	0.1914	0.039	39.332	0.6928	0.055	42.970
	0.2512	0.045	41.059	0.7246	0.054	42.542
	0.3027	0.042	39.956	0.7978	0.055	42.870
	0.3559	0.048	41.455	0.8403	0.053	42.124
	0.4085	0.045	40.580	0.8875	0.052	41.825
	0.4532	0.043	39.824	0.9404	0.051	41.199
	0.4966	0.039	38.382			

temperature from the slope and intercept, respectively. These plots for (1-pentanol + ethylene glycol), (1-hexanol + ethylene glycol), and (1-heptanol + ethylene glycol) at (303.15 and 318.15) K are represented in Figure 2, and obtained a and b values for these systems are listed in Table 3.

Temperature influence on the behavior of volumetric and surface properties of aqueous solutions has often been used to obtain information about solute structural effects on solvent structure. For most binary aqueous solutions at a given concentration, surface tension shows a linear dependency with temperature, decreasing as temperature increases. However, several authors have reported nonlinear behavior and thermal

anomalies in aqueous mixtures.^{20,21} In our selected systems, surface tension decreased with increasing temperature for any given concentration of alcohols. Figure 3 shows the variation of surface tension with the temperature for the 1-pentanol + ethylene glycol solutions.

The surface tensions of the binary mixtures were correlated with temperature by the following expression, proposed for the pure components introduced by Jasper¹⁶

$$\sigma / \text{mN} \cdot \text{m}^{-1} = K_1 - K_2(t / ^\circ \text{C}) \quad (4)$$

where K_1 and K_2 are fitting coefficients and t is the temperature in $^\circ \text{C}$. Equation 4 also fitted the data of Table 2 for each

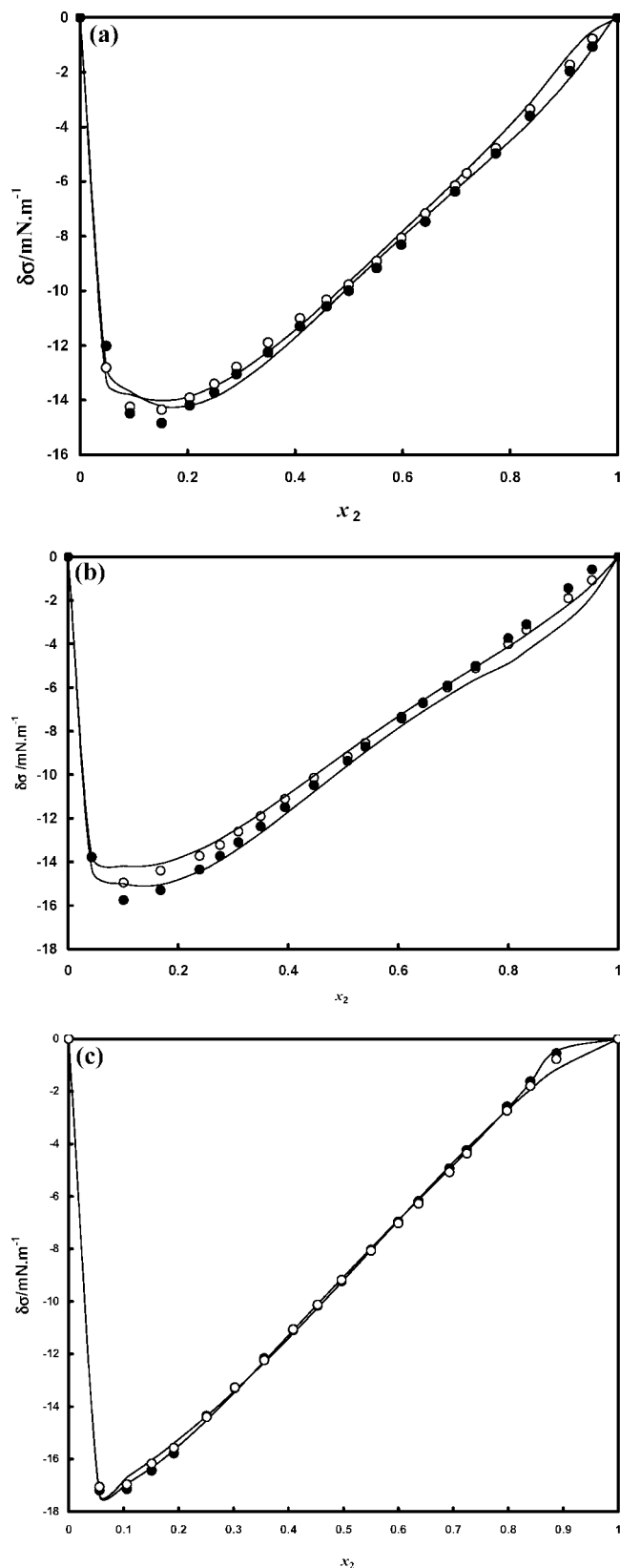


Figure 4. (a) Plot of surface tension deviation ($\delta\sigma/\text{mN}\cdot\text{m}^{-1}$) vs mole fraction of 1-pentanol (x_2) at \bullet , 303.15 K and \circ , 318.15 K. (b) Plot of surface tension deviation ($\delta\sigma/\text{mN}\cdot\text{m}^{-1}$) vs mole fraction of 1-hexanol (x_2) at \bullet , 303.15 K and \circ , 318.15 K. (c) Plot of surface tension deviation ($\delta\sigma/\text{mN}\cdot\text{m}^{-1}$) vs mole fraction of 1-heptanol (x_2) at \bullet , 303.15 K and \circ , 318.15 K.

concentration, with deviates less than 0.6 %. The fitted values of K_1 and K_2 are listed in Table 4.

Table 6. A_k Parameters and Their Standard Deviations (SD) from Equation 9 for Alkanol/Ethylene Glycol Systems

	T/K	A_0	A_1	A_2	A_3	A_4	SD
1-pentanol	298.15	37.84	-49.28	185.06	-212.95	89.20	0.24
	303.15	36.36	-63.27	156.25	-181.04	75.97	0.41
	308.15	35.15	-50.94	113.08	-123.01	49.85	0.30
	313.15	34.80	-49.30	108.67	-118.34	48.13	0.29
	318.15	34.52	-49.04	110.98	-124.84	52.11	0.25
1-hexanol	298.15	25.61	-21.80	109.48	-125.88	52.56	0.20
	303.15	34.52	-57.36	162.03	-201.79	88.35	0.27
	308.15	34.37	-57.37	162.00	-201.64	88.18	0.28
	313.15	33.03	-45.04	121.14	-148.19	64.14	0.15
	318.15	32.84	-45.05	121.19	-148.34	64.23	0.15
1-heptanol	298.15	29.98	-4.79	73.20	-90.26	39.28	0.19
	303.15	29.89	-20.49	53.31	-63.62	27.02	0.13
	308.15	29.81	-21.38	54.07	-62.84	25.99	0.10
	313.15	29.63	-21.31	54.85	-66.32	28.69	0.10
	318.15	29.48	-21.15	53.74	-63.91	27.07	0.10

With a particular choice of dividing surface, the temperature derivative of surface tension corresponds to excess surface entropy per unit area^{22,23}

$$S^s = -\left(\frac{\partial\sigma}{\partial T}\right) \quad (5)$$

More precisely, it was shown^{23,24} that S^s represents the variation of entropy per unit area due to an interface formation. This equation (eq 5) is valid at constant surface concentration of a solute, not the bulk one, but changing the temperature in a rather narrow range should not change the surface composition too much.

In a similar manner, and with similar limitations, the surface enthalpy per unit area can be derived as

$$H^s = \sigma - T\left(\frac{\partial\sigma}{\partial T}\right) \quad (6)$$

The calculated surface entropies and enthalpies are tabulated in Table 5. When organic solutes adsorb at the liquid–air interface, their hydrophilic groups remain immersed in solution, and the hydrophobic ones are oriented toward air. Recently, comparing some systems shows that concentration dependences of surface entropy and enthalpy are almost independent of the hydrophilic part of the solute molecule.^{26,27}

Deviations from surface tension, $\delta\sigma$, were calculated from our measurements according to the following equation

$$\delta\sigma = \sigma - \sum_{i=1}^2 x_i \sigma_i \quad (7)$$

where x_i and σ_i are the mole fraction and surface tension of component i , respectively. The values of excess surface tension are also graphically represented in Figure 4. The excess surface tensions were correlated by means of the Redlich–Kister²⁸ equation as follows

$$\delta\sigma = x_1 x_2 \sum A_k (x_1 - x_2)^k \quad k = 0, 1, 2, \dots, n \quad (8)$$

where A_k is the adjustable parameter determined by the method of least-squares. The values of these parameters are given in Table 6 together with the standard deviations. For these mixtures, the values are negative over the whole composition range. The negative values are reached in the mixture containing the pure compounds showing the greatest surface tension difference, that is, alcohols with ethylene glycol. This equation provides a simple tool for interpolation of surface tension in these mixtures with an accuracy that is within the experimental uncertainty.

Conclusions

Surface tension measurements for mixed systems containing some aliphatic alcohols and ethylene glycol have been carried out and reported at various temperatures in this paper. The obtained data indicate that the surface tensions of all three systems in various compositions of alcohol/ethylene glycol show a linear decrease with an increase of temperature. On the other hand, experimental plots illustrate a nonlinear dependency between surface tension and bulk composition of species. Thermodynamic properties of the surface for three systems such as surface entropy and surface enthalpy show a complicated behavior. Experimentally obtained surface tensions were correlated by means of the Redlich–Kister equation and provided a series of adjustable equation parameters for prediction of surface tension of such mixed systems.

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