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Density and Surface Tension of Binary Mixtures of Acetonitrile + 1-Alkanol at 293.15 K

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Surface tension and density of {acetonitrile + methanol, ethanol, 1-propanol, 1-butanol, or 1-pentanol} at the temperature 293.15 K and normal atmospheric pressure have been measured as a function of mole fraction. The Redlich−Kister polynomial equation was fitted to each of the experimental surface tension deviation and excess molar volume results. Using the resulting polynomial equations, the surface tension deviations and the excess molar volumes ($\Delta\sigma$, V_m^E) were calculated over the entire composition range. The results obtained are discussed and interpreted in terms of the type and nature of the specific intermolecular interaction between the components.

Introduction

The thermodynamic and transport properties of liquids and liquid mixtures[1] have been used to understand the molecular interactions between the components of the mixture and also for engineering applications concerning heat transfer, mass transfer, and fluid flow. Surface tension and density data of binary liquid mixtures are important from practical and theoretical points of view to understand liquid theory. Acetonitrile, alkanols, and their binary mixtures are used as solvents in chemistry and modern chemical technology.[2] Acetonitrile and alkanol mixtures are also used as very powerful cosolvents of polymers.[3] Molar excess volumes (V_m^E)[4−13] and molar excess enthalpies (H_m^E)[14,15] for (1-alkanol + acetonitrile) systems have been measured previously. Excess molar functions for (butanenitrile + ethanol, 1-butanol, 1-propanol, 2-propanol, 1-hexanol, and 1-octanol) systems have been reported for 298.15 K.[16−18] The experimental data of the excess molar volumes of 1-alkanol + nitrile mixtures (1-octanol, 1-nonanol, 1-decanol, or 1-undecanol + propanenitrile) and (1-undecanol + butanenitrile) at 298.15 K and atmospheric pressure have also been measured.[19] Dielectric constants, viscosities, and densities for acetonitrile + methanol mixtures have been presented at 298.15 K.[20] Density and viscosity of (butanenitrile + butanole isomers) have been measured.[21] Excess Gibbs free energies, excess enthalpies, and volumes were also measured for (butanenitrile + 2-butanol, 2-methyl-1-propanol, and 2-methyl-2-propanol).[22,23] Excess functions for binary mixtures of (butanenitrile + methanol, 1-pentanol, 1-heptanol, 1-nonanol, or 1-decanol) have been measured at 298.15 K.[24] Excess molar enthalpies and excess molar volumes of (alkanol + nitrile compounds) were also measured.[25] Densities and viscosities of binary mixtures of 1-propanol and 2-propanol with acetonitrile have been measured.[26] A survey of the literature, however, shows that there are no reports on the measurement of surface tension and density for acetonitrile + alkanols at 293.15 K. We present here results of the surface tension (σ_m) and density (ρ_m) for binary mixtures of acetonitrile with alkanols (C_1−C_5) at 293.15 K.

Experimental Section

The source and purity of the chemical compounds used in this study are shown in Table 1 together with the experimental surface tension and density measurements. Mixtures were prepared by mass with a Mettler AT201 balance (repeatability $\pm 1 \times 10^{-5}$ g) in airtight stoppered bottles, and the error in the mole fraction was estimated to be $< 10^{-4}$.

Surface Tension Measurements. The surface tension of pure components and mixtures at the liquid−vapor interface was measured using the pendant drop method.[40] The temperature of the measurement cell was controlled at (293.15 ± 0.1) K by a thermostatic water bath (Grant, model W14) via an external circulating loop. Each value reported was an average of three measurements with an uncertainty of $\pm$ 0.1 mN·m^{-1}.

Density Measurements. The measurement of the densities of the pure components and the binary mixtures was carried out using an Anton-Paar densitometer (model DMA 55) operated in static mode with an uncertainty of $\pm 10^{-5}$ g·cm^{-3}. Each value reported was an average of three measurements. Prior to measuring the unknown densities, the instrument was calibrated with double distilled water and air at 293.15 K. The temperature of the measuring cell was maintained at (293.15 ± 0.1) K using the Grant thermostatic water bath.

Results and Discussion

Experimental results for the surface tension (σ_m) and density (ρ_m) of mixtures at the temperature 293.15 K are summarized in Tables 2 and 3.

The surface tension deviation from linearity $\Delta\sigma$ was defined by

$$\Delta\sigma/(\text{mN}\cdot\text{m}^{-1}) = \sigma_m - x_1\sigma_1 - x_2\sigma_2 \qquad (1)$$

where σ_m is the surface tension of the mixture and σ_1 and σ_2 are that of components 1 and 2, respectively.

The excess molar volume (V_m^E) was defined by

$$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1}) = \frac{M_1x_1 + M_2x_2}{\rho_m} - \frac{M_1x_1}{\rho_1} - \frac{M_2x_2}{\rho_2} \qquad (2)$$

where ρ_m is the density of the mixture and M_1, M_2, x_1, x_2, ρ_1,

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Table 1. Source, Purity, Density ρ, and Surface Tension σ of the Pure Compounds at 293.15 K

		ρ/g·cm^{-3}		σ/mN·m^{-1}	
compound	source and purity	this work	lit	this work	lit
acetonitrile	Sigma-Aldrich >99.9 %	0.77830	0.7820[a] 0.7821[b] 0.783[c]	29.25	29.29[k]
methanol	Fluka >99.5 %	0.78783	0.7914[d] 0.79108[e] 0.792[c]	22.78	22.95[l] 22.50[k]
ethanol	Aldrich >99.8 %	0.78644	0.7893[d] 0.78824[e] 0.789[c]	22.31	22.3[m] 22.31[l] 22.39[k]
1-propanol	BDH >99.5 %	0.8013	0.8035[d] 0.804[c] 0.8031[f] 0.80364[e]	23.78	23.69[l] 23.73[f] 23.7[n] 23.71[k]
1-butanol	BDH >99.5 %	0.80733	0.8089[d] 0.810[c] 0.8104[f]	24.503	24.16[f] 24.53[n]
1-pentanol	Aldrich 99 %	0.81511	0.8144[d] 0.81412[g] 0.81453[h] 0.814[c] 0.8144[i] 0.8151[j] 0.8153[f]	25.457	25.69[f] 25.79[n] 25.79[k]

[a] Ref 27. [b] Ref 26. [c] Ref 28. [d] Ref 29. [e] Ref 30. [f] Ref 31. [g] Ref 32. [h] Ref 33. [i] Ref 34. [j] Ref 35. [k] Ref 36. [l] Ref 37. [m] Ref 38. [n] Ref 39.

Table 2. Experimental Surface Tension (σ_m) at 293.15 K for the Mixtures of $C_2H_3N + C_pH_{2p+1}OH$ (p = 1, 2, 3, 4, and 5)

x_1	σ_m/(mN·m^{-1})	x_1	σ_m/(mN·m^{-1})
Acetonitrile (1) + Methanol (2)		Acetonitrile (1) + 1-Butanol (2)	
0.1001	23.80	0.1026	24.65
0.2011	24.73	0.2021	24.78
0.2996	25.53	0.3020	24.94
0.4997	26.85	0.5024	25.52
0.7012	27.94	0.7014	26.53
0.8009	28.42	0.8007	27.25
0.8956	28.85	0.8995	28.14
Acetonitrile (1) + Ethanol (2)		Acetonitrile (1) + 1-Pentanol (2)	
0.0999	22.81	0.1003	25.49
0.2007	23.30	0.2031	25.54
0.3039	23.85	0.3014	25.57
0.5001	25.06	0.5003	25.82
0.6994	26.51	0.7000	26.64
0.8013	27.36	0.8001	27.29
0.8996	28.26	0.8993	28.11
Acetonitrile (1) + 1-Propanol (2)			
0.1004	24.02		
0.2018	24.23		
0.3013	24.52		
0.5004	25.32		
0.7003	26.50		
0.8002	27.27		
0.8997	28.19		

Table 3. Experimental Density (ρ_m) at 293.15 K for the Mixtures of $C_2H_3N + C_pH_{2p+1}OH$ (p = 1, 2, 3, 4, and 5)

x_1	ρ_m/(g·cm^{-3})	x_1	ρ_m/(g·cm^{-3})
Acetonitrile (1) + Methanol (2)		Acetonitrile (1) + 1-Butanol (2)	
0.1001	0.78846	0.1026	0.80536
0.2011	0.78804	0.2021	0.80318
0.2996	0.78713	0.3020	0.80080
0.4997	0.78482	0.5024	0.79542
0.7012	0.78213	0.7014	0.78923
0.8009	0.78075	0.8007	0.78570
0.8956	0.77947	0.8995	0.78201
Acetonitrile (1) + Ethanol (2)		Acetonitrile (1) + 1-Pentanol (2)	
0.0999	0.78657	0.1003	0.81220
0.2007	0.78599	0.2031	0.80906
0.3039	0.78505	0.3014	0.80603
0.5001	0.78308	0.5003	0.79964
0.6994	0.78089	0.7000	0.79242
0.8013	0.7798	0.8001	0.78811
0.8996	0.77891	0.8993	0.78338
Acetonitrile (1) + 1-Propanol (2)			
0.1004	0.79954		
0.2018	0.79763		
0.3013	0.79562		
0.5004	0.79105		
0.7003	0.78597		
0.8002	0.78329		
0.8997	0.78068		

and ρ_2 are the molecular weight, mole fraction, and density of pure components 1 and 2, respectively.

For each mixture, the surface tension deviation ($\Delta\sigma$) and excess molar volume (V_m^E) were fitted to the Redlich−Kister polynomial equation[41] in the form

$$y = x_1(1 - x_1)\sum_{j=1}^{n} a_j(2x_1 - 1)^{j-1} \tag{3}$$

where $y = \Delta\sigma$ or V_m^E and x_1 denotes the mole fraction of acetonitrile. In each case, the optimum number of coefficients is ascertained from an examination of the variation in standard deviation (S) as given by

$$S = [\sum[(y_{obs} - y_{cal})_i^2/(n - m)]]^{1/2} \tag{4}$$

where n is the total number of data points and m is the number of coefficients considered. The coefficients a_j listed in Table 4 were estimated by the least-squares method. These parameters were used to obtain the calculated curves, which have been shown with solid line, in Figures 1 and 2.

Surface Tension. The experimental σ_m for the binary mixtures {acetonitrile + 1-alkanol} at the temperature 293.15 K is reported in Table 2 for the various mole fractions of acetonitrile. Figure 1 shows $\Delta\sigma$ as a function of mole fraction of acetonitrile for the binary mixtures of {acetonitrile + 1-alkanol}. As it can be seen from Figure 1, $\Delta\sigma$ is positive for (acetonitrile + methanol) while $\Delta\sigma$ is negative for all the other mixtures studied in this work over the whole range of mole fraction. In all systems, the experimental data show a negative deviation from

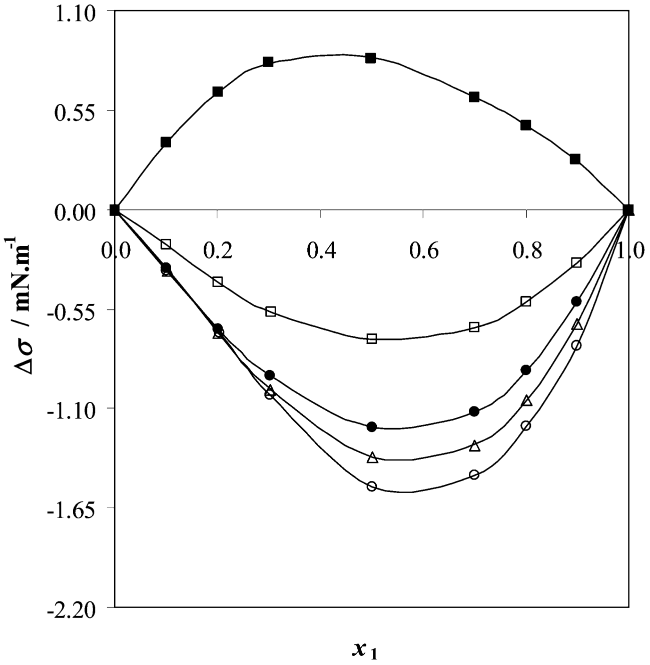

Figure 1. Plot of $\Delta\sigma$ against mole fraction of acetonitrile at 293.15 K for the mixtures of acetonitrile with ■, methanol; □, ethanol; ●, 1-propanol; △, 1-butanol; and ○. 1-pentanol. The points are measured, and the lines are calculated from eqs 1 and 3 using the parameters in Table 4.

Table 4. Redlich−Kister Coefficients and Standard Deviations (S) of Equations 3 and 4 for σ_m and V_m^E for the Investigated Systems

system		a_1	a_2	a_3	a_4	a_5	S
acetonitrile + methanol	$\Delta\sigma/(mN \cdot m^{-1})$	3.3645	−1.2363	0.2809	0.7830		0.0045
	$V_m^E/(cm^3 \cdot mol^{-1})$	−0.5581	0.2992	−0.1777	0.3283		0.0013
acetonitrile + ethanol	$\Delta\sigma/(mN \cdot m^{-1})$	−2.8849	−0.4780	−0.1924	−0.3034	0.7221	0.0013
	$V_m^E/(cm^3 \cdot mol^{-1})$	−0.1275	0.3629	−0.1787	0.3443		0.0040
acetonitrile + 1-propanol	$\Delta\sigma/(mN \cdot m^{-1})$	−4.8231	−1.1571	−0.3318	−0.2152	0.9316	0.0030
	$V_m^E/(cm^3 \cdot mol^{-1})$	0.2303	0.2879	0.1728	0.0495		0.0027
acetonitrile + 1-butanol	$\Delta\sigma/(mN \cdot m^{-1})$	−5.4836	−1.8291	0.2414	−0.4166		0.0077
	$V_m^E/(cm^3 \cdot mol^{-1})$	0.4437	0.2082	0.1063	0.2671		0.0028
acetonitrile + 1-pentanol	$\Delta\sigma/(mN \cdot m^{-1})$	−6.1256	−2.6155	1.6298	−0.3936	−2.4178	0.0016
	$V_m^E/(cm^3 \cdot mol^{-1})$	1.3534	−0.2727	0.1422	0.4855		0.0032

linearity, indicating an enrichment of one component (the component with a lower surface tension, which in this work is linear alcohols) at the liquid−vapor interface. The strong interactions between acetonitrile and methanol give rise to an increase in surface tension, which is in turn responsible for the positive $\Delta\sigma$ for the (acetonitrile + methanol) system. The strong interactions between acetonitrile and methanol decrease the tendency of methanol to adsorb at the liquid−vapor interface. This figure also shows that, at equimolar concentrations of acetonitrile from ethanol up to 1-pentanol, $\Delta\sigma$ becomes more negative as the length of the alkanol chain increases. The minima in the $\Delta\sigma - x_1$ curves are slightly shifted toward acetonitrile rich mole fractions, moving from $x = 0.544$ for ethanol up to 0.581 for 1-pentanol mixtures as shown in Figure 1.

Excess Molar Volume. The experimental ρ_m for the binary mixtures {acetonitrile + 1-alkanol} at the temperature 293.15 K is reported in Table 3 for various mole fractions of acetonitrile. The calculated V_m^E for the binary mixtures {acetonitrile + methanol, ethanol, 1-propanol, 1-butanol, and 1-pentanol} are plotted as a function of the mole fraction of acetonitrile in Figure 2. As can be seen from Figure 2, V_m^E is negative for (acetonitrile + methanol) mixture while for (acetonitrile + ethanol) mixture it takes both negative and positive values. Excess molar volumes are positive for mixtures of acetonitrile with 1-propanol, 1-butanol, and 1-pentanol over the whole range of mole fractions. This figure also shows that, at equimolar concentrations of acetonitrile from 1-pentanol up to1-propanol, V_m^E becomes less positive as the length of the alkanol chain decreases.

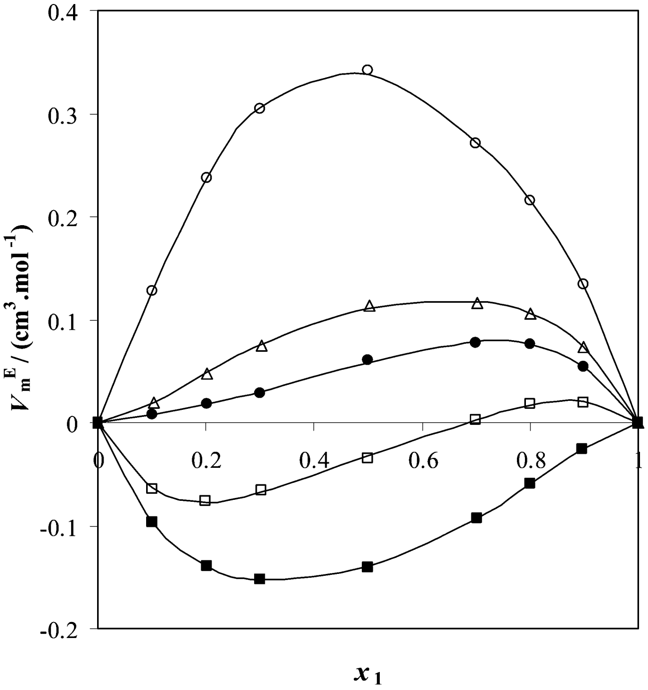

Figure 2. Plot of V_m^E against mole fraction of acetonitrile at 293.15 K for the mixtures of acetonitrile with ■, methanol; □, ethanol; ●, 1-propanol; △, 1-butanol; and ○, 1-pentanol. The points are measured, and the lines are calculated from eqs 2 and 3 using the parameters in Table 4.

The negative V_m^E values of (acetonitrile + methanol) mixtures can be attributed to interstitial accommodation of acetonitrile in the H-bonded network of methanol which outweighs the positive V_m^E owing to physical interactions and the breaking up of methanol clusters in the presence of acetonitrile molecules. Acetonitrile favors dipole−dipole interactions due to its high dipole moment (3.92 D). The positive V_m^E values for mixtures of acetonitrile with 1-propanol, 1-butanol, and 1-pentanol can be ascribed to dominance of disruption of H-bonds between alkanols over dipole−dipole interactions between alkanol (C_3−C_5) and acetonitrile molecules. The increase in V_m^E with the increase in chain length of 1-alkanols implies that dipole−dipole interaction is weaker in higher alkanols owing to their decreased polarizabilities[42] with increase of chain length.

Conclusions

Surface tensions and densities for {acetonitrile + alkanol} binary mixtures were determined at 293.15 K. The deviation of surface tension from linearity ($\Delta\sigma$) is negative in all of the systems analyzed in this work with the exception of {acetonitrile + methanol}. However, the excess molar volume (V_m^E) is positive for the {acetonitrile + 1-propanol, 1-butanol, and 1-pentanol} mixtures but negative for the mixture with methanol, while for {acetonitrile + ethanol}, V_m^E takes both positive and negative values. These differences have been accounted for in terms of molecular interactions.

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