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Densities, Refractive Indices, Speeds of Sound, and Surface Tensions for Dilute Aqueous Solutions of 2-Methyl-1-propanol, Cyclopentanone, Cyclohexanone, Cyclohexanol, and Ethyl Acetoacetate at 298.15 K

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ABSTRACT: Densities (ρ), refractive indices (n_D), speeds of sound (u), and surface tensions (σ) for the aqueous binary solutions of partially miscible 2-methyl-1-propanol, cyclopentanone, cyclohexanone, cyclohexanol, and ethyl acetoacetate were determined at the temperature of 298.15 K and atmospheric pressure over the mole fraction range reaching the solubility limit in the aqueous phase. Isentropic compressibilities, determined by means of the Laplace equation, have been calculated from the measured experimental data. To analyze the behavior of surface tensions, the extended Langmuir (EL) model was used.

INTRODUCTION

Studies on the thermodynamic properties of highly dilute aqueous solutions of organic compounds are of considerable theoretical and practical interest. In recent years, there has been an increasing effort to analyze the relationship between surface properties and the activity of a solute in the bulk solution. These studies are focused on dilute solutions due to the importance of the activity coefficients in different practical applications. Our research group has taken a keen interest in the analysis of the surface tension data in the diluted region because, based on these measurements and taking into account the refractive indices and density values, we can precisely assess properties such as standard energy of adsorption, parachor, molar refraction, or the activity coefficient at infinite dilution of the mixture components.

In this work, measurements of refractive index, density, speed of sound, and surface tension for the aqueous binary solutions of partially miscible 2-methyl-1-propanol, cyclopentanone, cyclohexanone, cyclohexanol, and ethyl acetoacetate at the temperature of 298.15 K, including the refractive index of the saturated aqueous phases, are reported.

The refractive index measurement provides important structural information about liquid mixtures, since the refractive index is very sensitive to changes in molecular organization. From the experimental density and speed of sound data, the isentropic compressibility for the binary systems was determined. Special interest was paid in this work to the analysis of the surface tension behavior under the extended Langmuir (EL) model. The EL model is based on a modification of the Langmuir isotherm combined with a mixing rule and includes two adjustable parameters with clear physical meaning.

EXPERIMENTAL SECTION

Chemicals. The pure organic compounds were supplied by Fluka. The maximum water content of the liquids (less than 0.2% in all cases) was determined using a Karl Fisher coulometer. Their mass fraction purities were >99.5% for 2-methyl-1-propanol, cyclopentanone, and cyclohexanone and >99% for cyclohexanol, and ethyl acetoacetate. The ultrapure water was obtained by using an ELIX 3 system from Millipore. The water surface tension value at 298.15 K was 71.78 $\text{mN}\cdot\text{m}^{-1}$, and the values for the density, refractive index, and sound velocity were $\rho = 997.021\ \text{kg}\cdot\text{m}^{-3}$, $n_D = 1.33250$, and $u = 1496.96\ \text{m}\cdot\text{s}^{-1}$ respectively. In Table 1, the experimental refractive index n_D, density ρ, speed of sound u, and surface tension of the pure components σ^0 are compared with literature data.[1−34]

Liquid mixtures were prepared by mass in airtight stoppered bottles, bearing in mind the vapor pressures of the components when establishing the filling sequence. The uncertainty in the mole fraction was estimated to be $<1\cdot10^{-4}$. The saturated aqueous phases were prepared by adding organic solvent to water until the formation of a second phase. The mixtures were constantly stirred and finally left static for at least five hours within a water thermostat to reach equilibrium before taking carefully a sample of the aqueous phase with the aid of a glass Hamilton syringe to measure its refractive index.

The densities and the speed of sound of the pure liquids and mixtures were measured using an Anton Paar DSA-5000 (Austria) digital vibrating tube densimeter. The densimeter is provided with an integrated temperature control; the temperature was stable to ± 0.001 K. The uncertainty in the density measurement is $\pm\ 5\cdot10^{-6}\ \text{g}\cdot\text{cm}^{-3}$, and that for the speed of sound is $\pm\ 0.5\ \text{m}\cdot\text{s}^{-1}$. For refractive index measurements at the wavelength of the D-line of sodium, we have used a Mettler Toledo refractometer (model RA-510M, Germany) having an uncertainty of 10^{-5}, calibrated with ultrapure water. The solutions were prethermostated at 298.15 K

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Table 1. Refractive Index (n_D), Density (ρ), Speed of Sound (u), Isentropic Compressibility (κ_S), and Surface Tension (σ^0) of the Pure Compounds at 298.15 K, Otherwise Indicated

	n_D^{25}		$\rho/\mathrm{g\cdot cm^{-3}}$		$u/\mathrm{m\cdot s^{-1}}$		$\kappa_S/\mathrm{TPa^{-1}}$	$\sigma^0/\mathrm{mN\cdot m^{-1}}$	
	exp	lit.	exp	lit.	exp	lit.	exp	exp	lit.
water	1.33251	1.33248[1]	0.997021	0.99704[2]	1497.0	1497[6]	448	71.78	71.35[10]
		1.3325[2]		0.997045[4]		1497.00[7]			71.8[3]
		1.3325029[3]		0.9970474[3]		1498[8]			71.84[11]
		1.3326[4]		0.997048[5]		1508[9]			71.95[12]
2-methyl-1-propanol	1.39373	1.3937[13]	0.797851	0.79761[13]	1188.1	1185.6[13]	888	22.38	22.11[3,d]
		1.39370[14]		0.7978[3]		1185.63[14]			22.37[18]
		1.393762[15]		0.79784[15]		1188.0[16]			22.47[19]
		1.39389[3]		0.79803[16]		1211.4[17]			22.98[3,a]
cyclopentanone	1.43476	1.43494[21]	0.944107	0.93902[3,d]	1394.1		545	33.78	32.58[3,d]
		1.4354[3]		0.94386[22]					33.87[3,a]
				0.94435[23]					
				0.94865[3,a]					
cyclohexanone	1.44837	1.4480[24]	0.942039	0.93761[3,c]	1408.0	1407.60[26]	535	33.74	34.5[27,a]
		1.4500[3]		0.9410[25]		1417[24]			35.05[3,a]
				0.9424[24]		1430.5[27,a]			
				0.9452[3,a]		1847.7[25]			
cyclohexanol	1.46449	1.46472[29]	0.945077	0.94152[30,d]	1465.3	1443[30,d]	493	33.39	32.82[31]
		1.46477[3]		0.94651[31]					33.91[3,c]
				0.9684[3]					36.2[32,b]
ethyl acetoacetate	1.41690	1.4192[3,a]	1.023479	1.02126[3]	1332.9	1331[34]	550	31.93	32.3[33]
		1.41658[33]		1.0208[34]					
		1.4189[34]		1.02345[33]					

[a] T = 293.15 K. [b] T = 295.65 K. [c] T = 298.65 K. [d] T = 303.15 K.

before the experiments to achieve a quick thermal equilibrium. Experimental densimetric and refractometric techniques have been described in detail in previous works.[35−37]

Surface tensions of the pure liquids and their mixtures σ at the liquid−vapor interface were measured using a Lauda TVT1 automated tensiometer based on the pending drop volume principle. This technique consists of measuring the volume of a drop detaching from a capillary with a circular cross section. The detailed instrumentation design has been described in a previous paper[37] also addressing the procedure and the handling of data. The experimental technique used consists of a hermetically sealed cell allowing for the attainment of a saturated atmosphere and preventing solute loss at the concentrated surface. The temperature of the measurement cell was controlled within ± 0.01 K by a water thermostat. The uncertainty obtained in σ values was less than 0.04 mN·m^{-1}.

■ RESULTS AND DISCUSSION

The density ρ, refractive index n_D, speed of sound u, and isentropic compressibility k_S for the aqueous solutions of (2-methyl-1-propanol, cyclopentanone, cyclohexanone, cyclohexanol, and ethyl acetoacetate) at the temperature of 298.15 K and atmospheric pressure are reported in Table 2. Isentropic compressibility was determined from the density and speed of sound measurements by means of Laplace equation (eq 1).

$$k_S = \rho^{-1}u^{-2} \tag{1}$$

The variation of these properties as a function of the organic compound mole fraction is depicted graphically in Figures 1 to 4. Due to the short range of water solubility, these curves are almost linear in all cases except for the system containing cyclopentanone (the more soluble of the analyzed compounds). Our density results compare well with those reported by Hovorka et al.[38] as can be seen in Figure 1.

The solubility of the organic solutes in water were obtained from experimental refractive index measurements. Refractive indices were fitted to a second-order polynomial as a function of x, as exemplified in Figure 2 for cyclopentanone (inset); having n_D of the aqueous saturated phases, the aqueous solubility ($x_{\mathrm{aq.sol}}$) of the solutes was calculated from this polynomial. These values are reported in Table 3 together with literature data.

All systems show positive refractive index deviations with respect to ideality in a volume fraction basis ($\Delta_\phi n_D$) as depicted in Figure 5. This is in fairly agreement with the negative excess molar volumes v^E (Figure 5B) since n_D is inversely proportional to the molar free volume of one mole of hard-core spheres[39,40] (the molar free volume is the unoccupied volume of the hard-core spheres); the systems with the smallest $\Delta_\phi n_D$ show the smallest v^E, except in the case of 2-methyl-1-propanol and ethyl acetoacetate. The difference is ascribed to stronger structural changes in the system {water + 2-methyl-1-propanol} than in the system {water + ethyl acetoacetate}.

The system showing the lowest compressibility is cyclohexanol since the speed of sound in its mixtures is the highest. This system has the smallest $\Delta_\phi n_D$ and v^E. Cyclohexanone and cyclopentanone aqueous mixtures follow in $\Delta_\phi n_D$ and v^E, but u of cyclohexanone system is higher, so it has lower compressibility.

Table 2. Refractive Index (n_D), Density (ρ), Speed of Sound (u), and Isentropic Compressibilities (κ_S) of the Aqueous Binary Systems at 298.15 K

		ρ	u	κ_S
x	n_D	$g \cdot cm^{-3}$	$m \cdot s^{-1}$	TPa^{-1}
(x) 2-methyl-1-propanol + (1 − x) Water				
0.0002	1.33255	0.996913	1497.4	447
0.0005	1.33270	0.996679	1498.7	447
0.0008	1.33282	0.996486	1499.8	446
0.0014	1.33308	0.996069	1502.0	445
0.0017	1.33321	0.995867	1503.2	444
0.0032	1.33378	0.994931	1508.4	442
0.0046	1.33437	0.994035	1513.5	439
0.0067	1.33521	0.992809	1520.4	436
0.0077	1.33564	0.992181	1524.0	434
0.0099	1.33655	0.990950	1531.1	430
0.0124	1.33754	0.989513	1539.1	427
0.0140	1.33823	0.988700	1543.5	425
(x) Cyclopentanone + (1 − x) Water				
0.0005	1.33279	0.997025	1498.5	447
0.0012	1.33314	0.997000	1500.5	445
0.0022	1.33368	0.996984	1503.3	444
0.0027	1.33401	0.996974	1505.0	443
0.0038	1.33456	0.996959	1508.0	441
0.0044	1.33524	0.996949	1509.9	440
0.0059	1.33587	0.996936	1515.1	437
0.0079	1.33686	0.996929	1519.3	435
0.0092	1.33744	0.996932	1522.4	433
0.0144	1.34015	0.996953	1534.8	426
0.0182	1.34216	0.996988	1543.1	421
0.0232	1.34478	0.997022	1552.0	416
0.0305	1.34803	0.997028	1563.1	411
0.0394	1.35161	0.996922	1572.4	406
0.0501	1.35641	0.996560	1578.7	403
0.0564	1.35922	0.996241	1579.9	402
0.0683	1.36333	0.995391	1578.3	403
(x) Cyclohexanone + (1 − x) Water				
0.0004	1.33276	0.997006	1498.2	447
0.0007	1.33302	0.996984	1499.5	446
0.0008	1.33308	0.996979	1499.8	446
0.0011	1.33329	0.996963	1500.9	445
0.0016	1.33363	0.996940	1502.6	444
0.0022	1.33407	0.996915	1504.7	443
0.0024	1.33421	0.996903	1505.5	443
0.0028	1.33453	0.996881	1507.0	442
0.0031	1.33472	0.996874	1508.0	441
0.0039	1.33536	0.996842	1510.9	439
0.0046	1.33575	0.996817	1513.0	438
0.0068	1.33735	0.996753	1520.3	434
0.0082	1.33835	0.996720	1524.5	432
0.0082		0.996722	1524.4	432
0.0104	1.33985	0.996669	1530.7	428
(x) Cyclohexanol + (1 − x) Water				
0.00013	1.33259	0.997006	1497.4	447
0.00013	1.33260	0.997001	1497.4	447
0.00026	1.33269	0.996986	1498.0	447
0.00026	1.33269	0.996982	1498.0	447
0.00031	1.33272	0.996974	1498.2	447
0.00031	1.33272	0.996974	1498.2	447
0.00033	1.33275	0.996971	1498.3	447
0.0005	1.33292	0.996935	1499.4	446
0.0009	1.33323	0.996872	1501.2	445
0.0015	1.33368	0.996784	1503.9	444
0.0022	1.33419	0.996692	1506.9	442
0.0032	1.33497	0.996566	1511.4	439
0.0042	1.33572	0.996457	1515.5	437
0.0055	1.33679	0.996321	1521.3	434
0.0064	1.33736	0.996243	1524.6	432
(x) Ethyl Acetoacetate + (1 − x) Water				
0.0004	1.33275	0.997260	1498.1	447
0.0007	1.33302	0.997485	1499.4	446
0.0015	1.33355	0.997958	1502.0	444
0.0021	1.33397	0.998313	1503.9	443
0.0033	1.33485	0.999086	1508.0	440
0.0042	1.33543	0.999587	1510.5	438
0.0053	1.33623	1.000266	1513.9	436
0.0069	1.33725	1.001165	1518.2	433
0.0082	1.33815	1.001912	1521.7	431
0.0102	1.33943	1.003029	1526.5	428
0.0136	1.34157	1.004795	1533.5	423
0.0169	1.34351	1.006385	1538.8	420

Even though u of 2-methyl-1-propanol mixtures is bigger than that of ethyl acetoacetate mixtures, the heteromolecular interactions of 2-methyl-1-propanol with water are the responsible of a higher compressibility in comparison to ethyl acetoacetate. No certain trend of k_S with respect to $\Delta_\phi n_D$, v^E, u, or π^* can be recognized.

Table 4 contains the experimental surface tension measurements at 298.15 K. The surface tension of all mixtures studied decreases as the concentration increases. This trend was nonlinear, the change in surface tension caused by a change in the organic compound concentration being larger at low concentrations than at high. Figure 6 shows the plot of the reduced surface pressures, $\pi^* = \pi/\pi^0 = (\sigma_{water} - \sigma)/(\sigma_{water} - \sigma^0)$, versus the organic compound volume fraction, where σ^0 is the surface tension of the pure solute, and thus, π^0 is the maximum surface pressure that the system can attain. The σ values obtained by Habrdová et al.[12] for the system {cyclopentanone + water} coincide with our measurements as shown in Figure 6. For the other systems treated in this work no experimental data were found.

The EL[41] model was applied to these mixtures to interpret the results obtained. The EL model allows for the behavior analysis of binary systems that deviate either positively or negatively from ideality.[42−44] Briefly, it considers the surface of a binary liquid mixture as a thin but finite layer and starts by developing the following expression for the relationship between the volume fractions of component B in the surface and the bulk, ϕ_B^S and ϕ_B,

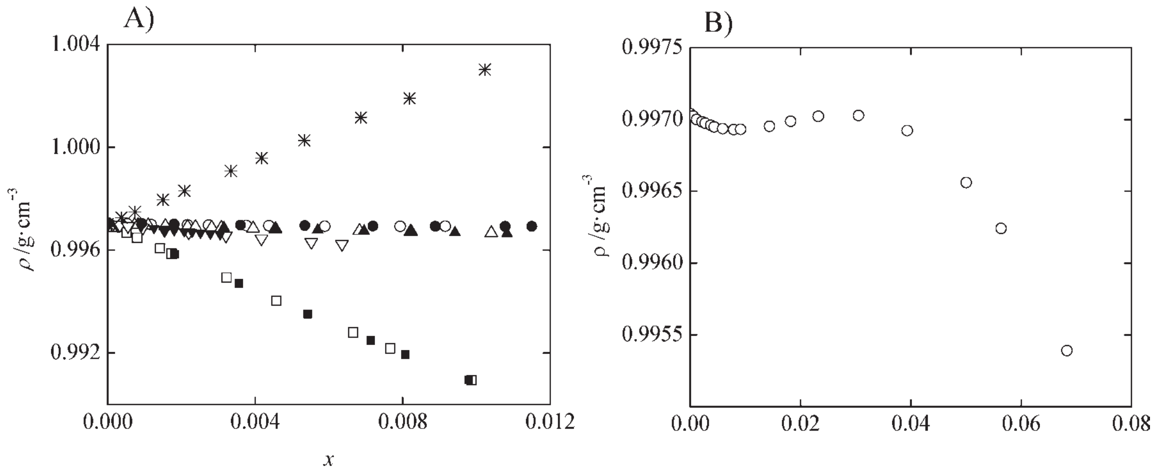

Figure 1. Experimental density ρ versus molar fraction x of the organic compound of the aqueous binary systems at T = 298.15 K. (A) □, 2-Methyl 1-propanol; ○, cyclopentanone; △, cyclohexanone; ▽, cyclohexanol; ∗, ethyl acetoacetate. Full symbols are data from ref 38: ■, 2-methyl 1-propanol; ●, cyclopentanone; ▲, cyclohexanone; ▼, cyclohexanol. (B) Behavior of the cyclopentanol system in the whole achieved composition interval.

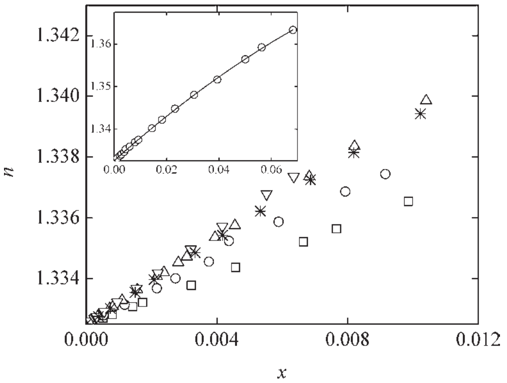

Figure 2. Experimental refractive index n_D versus molar fraction x of the organic compound of the aqueous binary systems at T = 298.15 K: □, 2-methyl 1-propanol; ○, cyclopentanone; △, cyclohexanone; ▽, cyclohexanol; ∗, ethyl acetoacetate. The inset shows the behavior of the cyclopentanol system in the whole achieved composition interval. The line stands for the fitting of the experimental data to a second-order polynomial, $n_D = 1.33257 + 0.55075x - 1.45931x^2$, for the estimation of the aqueous solubility $x_{aq.sol}$ from the experimental n_D of the aqueous saturated phase.

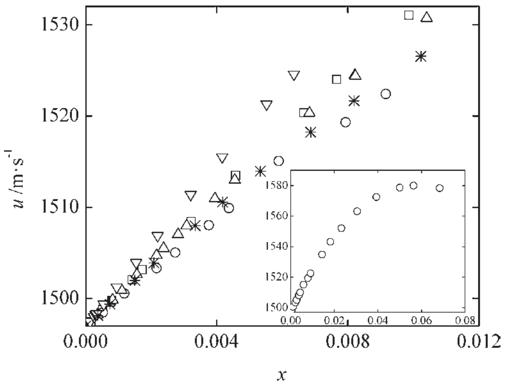

Figure 3. Experimental speed of sound u versus molar fraction x of the organic compound of the aqueous binary systems at T = 298.15 K: □, 2-methyl 1-propanol; ○, cyclopentanone; △, cyclohexanone; ▽, cyclohexanol; ∗, ethyl acetoacetate. The inset shows the behavior of the cyclopentanol system in the whole achieved composition interval.

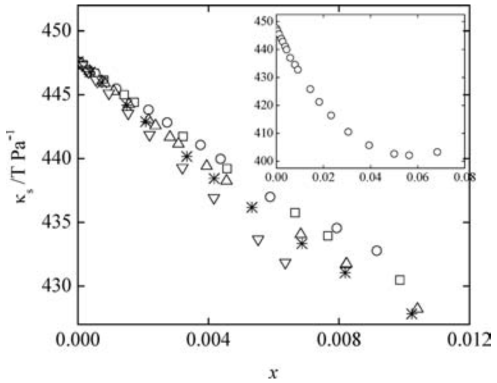

Figure 4. Calculated isentropic compressibility k_S versus molar fraction x of the organic compound of the aqueous binary systems at T = 298.15 K: □, 2-methyl 1-propanol; ○, cyclopentanone; △, cyclohexanone; ▽, cyclohexanol; ∗, ethyl acetoacetate. The inset shows the behavior of the cyclopentanol system in the whole achieved composition interval.

Table 3. Refractive Index (n_D) of the Saturated Aqueous Phase and the Estimated Aqueous Solubility ($x_{aq.sol}$) at 298.15 K, Otherwise Indicated

		$x_{aq.sol}$	
	n_D^{25}	exp.	lit.
2-methyl-1-propanol	1.34105	0.0209	0.021[20] 0.0263[3]
cyclopentanone	1.36406	0.0703	
cyclohexanone	1.34429	0.0166	0.0043[3,a] 0.0158[28]
cyclohexanol	1.33780	0.0069	0.0070[3,b]
ethyl acetoacetate	1.34511	0.0190	0.0185[3]

[a] T = 293.15 K. [b] T = 297.75 K.

respectively,

$$\phi_B^S = \beta\phi_B/(1 - \phi_B + \beta\phi_B) \qquad (2)$$

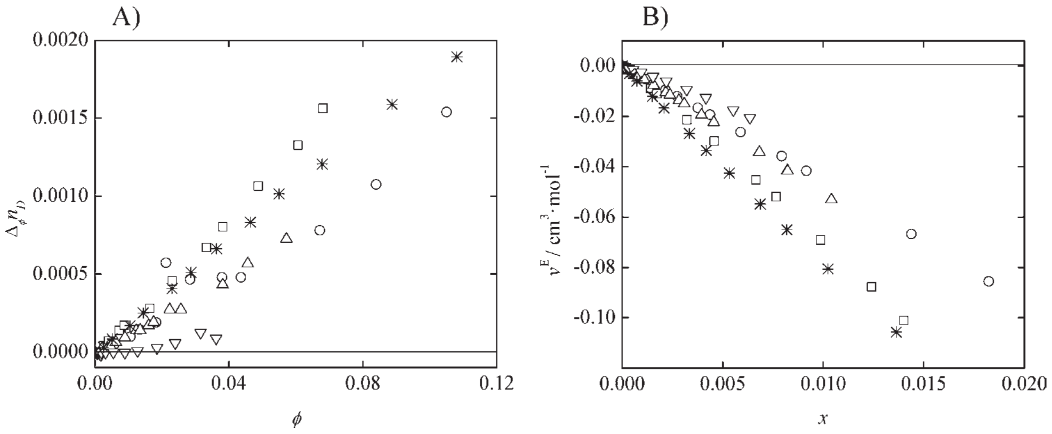

Figure 5. Plot of (A) refractive index deviations, $\Delta_\phi n_D$, and (B) excess molar volumes, v^E, for the aqueous binary systems at T = 298.15 K: □, 2-methyl 1-propanol; ○, cyclopentanone; △, cyclohexanone; ▽, cyclohexanol; ∗, ethyl acetoacetate.

Table 4. Surface Tension (σ) of the Aqueous Binary Mixtures at 298.15 K

x	σ / mN·m^{-1}	x	σ / mN·m^{-1}	x	σ / mN·m^{-1}
2-Methyl-1-propanol		Cyclopentanone		Cyclohexanone	
0.0002	69.11	0.0009	68.12	0.0004	68.22
0.0005	64.78	0.0017	65.38	0.0007	65.55
0.0008	61.89	0.0033	62.09	0.0008	64.98
0.0014	56.85	0.0062	57.71	0.0011	63.44
0.0017	54.91	0.0142	50.93	0.0016	61.18
0.0032	48.38	0.0302	43.76	0.0022	59.02
0.0046	44.28	0.0537	38.49	0.0024	58.19
0.0067	39.61	0.0680	36.57	0.0028	56.82
0.0077	37.67			0.0031	56.07
0.0099	34.59			0.0039	53.97
0.0124	31.40			0.0046	52.59
0.0140	30.13			0.0068	48.96
				0.0082	47.23
Cyclohexanol		Ethyl Acetoacetate		0.0082	47.28
0.00013	68.81	0.0004	67.63	0.0104	44.89
0.00013	69.60	0.0007	65.01		
0.00026	66.67	0.0015	61.08		
0.00026	67.07	0.0021	58.81		
0.00031	66.07	0.0033	55.27		
0.00031	65.81	0.0042	53.47		
0.00033	65.63	0.0053	51.18		
0.0005	60.94	0.0069	48.58		
0.0009	55.98	0.0082	46.97		
0.0015	51.24	0.0102	44.62		
0.0022	47.45	0.0136	41.46		
0.0032	43.15	0.0169	38.81		
0.0042	40.14				
0.0055	37.01				
0.0064	35.45				

where the parameter $\beta = (\phi_B^S/\phi_B)/(\phi_A^S/\phi_A)$ is a measure of the lyophobicity of B relative to A. The second ingredient of the EL model is a mixing rule with a Margules-type term,

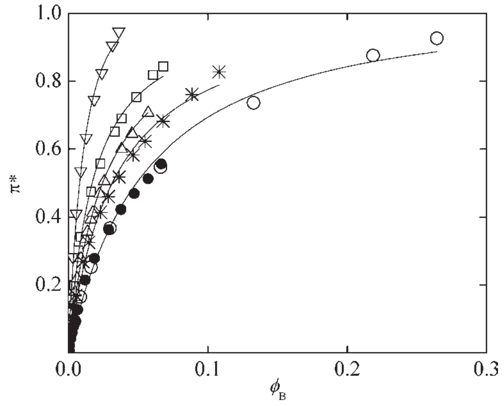

Figure 6. Experimental reduced surface pressure π^* versus molar volume fraction ϕ_2 of the organic compound of the aqueous binary systems at T = 298.15 K: □, 2-methyl-1-propanol; ○, cyclopentanone; △, cyclohexanone; ▽, cyclohexanol; ∗, ethyl acetoacetate. ●, Cyclopentanone of ref 12 volume fraction was calculated using the densities of pure compounds reported in this work. Solid lines stand for the fitting to EL model (eq 5); in the case of cyclopentanone only the experimental data reported in this work were fitted.

$$\sigma = \phi_A^S\sigma_A + \phi_B^S\sigma_B - \lambda\phi_A^S\phi_B^S\pi^0 \qquad (3)$$

where σ_A and σ_B are the surface tensions of the pure components A and B, π^0 is the positive difference between them (A is the component of higher surface tension), and λ is a parameter that represents the effect of unlike-pair interactions on the surface tension of the mixture and is zero when this effect is negligible. From eq 3 the surface pressure π can be written as:

$$\pi = \pi^0\phi_B^S(\alpha\phi_A^S + \phi_B^S) \qquad (4)$$

where $\alpha = \lambda + 1$; and substituting for ϕ_B^S and $\phi_A^S(= 1 - \phi_B^S)$ from eq 2 yields

$$\pi = \pi^0\beta[\beta + \alpha(\phi_A/\phi_B)]/[\beta + (\phi_A/\phi_B)]^2 \qquad (5)$$

In Figure 6 the fitting curves to eq 5 corresponding to the systems analyzed in this work show the ability of the EL model to describe the surface tension behavior. Table 5 reports the values of parameters α and β obtained after adjusting the experimental

Table 5. Parameters α and β and Standard Deviation s of the EL Model Obtained by Fitting Equation 5 to the Surface Tension Experimental Data at 298.15 K

	α	β	s
2-methyl-1-propanol	1.3 ± 0.2	41 ± 8	0.017
cyclopentanone	1.5 ± 0.3	12 ± 3	0.028
cyclohexanone	1.0	40 ± 1	0.013
cyclohexanol	2.3 ± 0.1	41 ± 2	0.015
ethyl acetoacetate	1.0	31 ± 1	0.023

values of surface tension to the above equation. The values of $\beta > 1$ indicate that the concentration of the organic compound is higher at the surface, in regard to the bulk concentration. The values of β are consistent with the solubility of the solutes; the less soluble in water, the higher value of β and the more hydrophobic organic compounds. An exception occurs with ethyl acetoacetate and 2-methyl-1-propanol; β for the slightly less soluble ethyl acetoacetate is smaller than that for 2-methyl-1-propanol. For the aqueous systems containing ethyl acetoacetate and cyclohexanone, $\alpha = 1$ (or $\lambda = 0$), which implies that the heteromolecular interactions do not affect significantly the surface tension of the solution. On the contrary, for the mixtures {2-methyl-1-propanol and cyclopentanone + water} λ is clearly greater than 0. The heteromolecular interactions of 2-methyl-1-propanol with water are more significant than those of ethyl acetoacetate, conferring 2-methyl-1-propanol a more hydrophobic character. When comparing 2-methyl-1-propanol with cyclohexanone, the solubility of 2-methyl-1-propanol is also higher; that the values of β for both solutes are the same is attributed likewise to the higher value of α for 2-methyl-1-propanol. The cyclohexanol aqueous mixture shows the largest α, what clearly suggests structural changes in the mixing process which explain, for example, the fact that even though the density of the pure cyclohexanol is higher than that of the cyclohexanone, the density of the cyclohexanol aqueous mixtures is lower than that of the cyclohexanone system.

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