Surface Tensions of the Ternary Mixtures Containing an Isomeric Butanol + *n*-Hexane + 1-Chlorobutane at 298.15 K

Montserrat Pérez-Navarro, Isabel Bandrés, Ignacio Giner, Ignacio Gascón, and Carlos Lafuente*

Departamento de Química Orgánica-Química Física, Área de Química Física, Facultad de Ciencias, Universidad de Zaragoza, 50009 Zaragoza, Spain

Surface tensions of the ternary mixtures containing an isomer of butanol (1-butanol, 2-butanol, 2-methyl-1-propanol, or 2-methyl-2-propanol) + *n*-hexane + 1-chlorobutane have been measured at atmospheric pressure and 298.15 K. From these experimental data, surface tension deviations were calculated and fitted by the Cibulka's equation. Experimental results have been compared to the predictions obtained by a group contribution method; furthermore, using this method, the excess surface molar compositions of *n*-hexane in the ternary mixtures have been also estimated.

Introduction

It is well-known that the properties and the behavior of the surface region of a liquid mixture are substantially different from those of the bulk. Consequently, a complete thermodynamic study of a liquid system has to incorporate surface investigations. Moreover, the measurement of surface tension is of outstanding importance in many scientific and technological areas, such as extraction or distillation processes.[1]

Systematic investigations on surface tension of multicomponent systems are rather unusual.[1–6] In the last years our group has been involved in the study of different thermodynamic and transport properties of the ternary mixtures containing an isomer of butanol + *n*-hexane + 1-chlorobutane.[7–10] Following our systematic studies we report here surface tension measurements for these ternary systems at 298.15 K. Surface tension deviations have been calculated from experimental surface tensions and fitted by the Cibulka's equation. Apart from this experimental work we have verified the accuracy in the prediction of surface tensions of a group-contribution method,[11,12] and on the other hand we have calculated using this method the excess surface molar compositions of *n*-hexane in the ternary mixtures. This method combines a model for the description of surface tensions of liquid mixtures[13] with a group contribution method, namely, the universal functional activity coefficient model (UNIFAC), for the calculation of activity coefficients.

Experimental Section

The liquids used were the following: 1-butanol (> 0.998 in mass fraction), 2-methyl-1-propanol and 2-methyl-2-propanol (> 0.995 in mass fraction), and 2-butanol, *n*-hexane, and 1-chlorobutane (≥ 0.99 in mass fraction) provided by Aldrich. The purities of these compounds were checked by measuring their densities. No further purification was considered necessary although the isomeric butanols were dried with activated molecular sieve type 0.3 nm from Merck.

The surface tension, σ, of the pure liquids and their mixtures were determined using a drop volume tensiometer Lauda TVT-2.[14] The tensiometer measures the volume of a drop detaching from a capillary of known diameter. The temperature was kept constant within ± 0.01 K by means of an external Lauda E-200 thermostat. Densities needed to calculate surface tensions from volume drop determinations were measured using an Anton Paar DMA-58 vibrating tube densimeter and can be found in the Supporting Information; the uncertainty of the density measurements is $\pm$ 0.01 $\text{kg}\cdot\text{m}^{-3}$. The uncertainty of the surface tension measurements is $\pm$ 0.01 $\text{mN}\cdot\text{m}^{-1}$.

The compositions of the mixtures are given in mole fraction and were determined by mass using a Sartorius semimicro balance with an uncertainty of $\pm 10^{-5}$ g. The uncertainty in mole fraction is estimated to be less than 10^{-4}.

The pure compound properties at 298.15 K, along with literature values,[15–19] are given in Table 1.

Table 1. Densities, ρ, and Surface Tensions, σ, of Pure Compounds at 298.15 K and Comparison with Literature Data

	$\rho/\text{kg}\cdot\text{m}^{-3}$		$\sigma/\text{mN}\cdot\text{m}^{-1}$	
component	exptl	lit.	exptl	lit.
1-butanol	805.80	805.75[a]	24.25	24.20[b]
2-butanol	802.44	802.41[a]	23.08	23.1[c]
2-methyl-1-propanol	797.77	797.8[a]	22.47	22.37[c]
2-methyl-2-propanol	780.78	780.63[d]	20.30	20.1[c]
n-hexane	655.07	654.84[a]	18.06	17.90[b]
1-chlorobutane	880.69	880.95[a]	23.29	23.1[e]

[a] From ref 13. [b] From ref 15. [c] From ref 16. [d] From ref 14. [e] From ref 17.

Results and Discussion

Surface tension deviations, $\Delta\sigma$, have been calculated from experimental measurements of surface tension according to the following equation:

$$\Delta\sigma = \sigma - x_1\sigma_1 - x_2\sigma_2 - x_3\sigma_3 \qquad (1)$$

where σ is the surface tension of the mixture and x_i and σ_i are the mole fraction and surface tension of component i, respectively. The values of surface tensions and surface tension deviations are given in Table 2.

The surface tension deviations for the ternary mixtures have been fitted by Cibulka's equation:[20]

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* To whom correspondence should be addressed. Tel.: +34976762295. Fax: +34976761202; e-mail: celadi@unizar.es.

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Table 2. Experimental Surface Tensions, σ, and Surface Tension Deviations, $\Delta\sigma$, of the Ternary Mixtures of Isomeric Butanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3) at 298.15 K

x_1	x_2	σ / $\mathrm{mN \cdot m^{-1}}$	$\Delta\sigma$ / $\mathrm{mN \cdot m^{-1}}$	x_1	x_2	σ / $\mathrm{mN \cdot m^{-1}}$	$\Delta\sigma$ / $\mathrm{mN \cdot m^{-1}}$
1-Butanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3)				2-Butanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3)			
0.0610	0.8385	18.45	−0.51	0.0507	0.0506	22.72	−0.29
0.0544	0.8825	18.34	−0.39	0.0499	0.0996	22.33	−0.43
0.1013	0.1103	22.22	−0.59	0.0494	0.8511	18.48	−0.35
0.1029	0.2001	21.48	−0.86	0.0478	0.8511	18.48	−0.35
0.1070	0.2967	20.80	−1.04	0.1033	0.0453	22.66	−0.37
0.1064	0.3976	20.23	−1.08	0.0938	0.1000	22.20	−0.55
0.1027	0.5004	19.72	−1.05	0.0955	0.2017	21.45	−0.77
0.1008	0.5934	19.30	−0.98	0.1003	0.2970	20.85	−0.87
0.1118	0.6834	18.92	−0.90	0.1013	0.3996	20.36	−0.82
0.1131	0.7856	18.59	−0.70	0.0930	0.5062	19.84	−0.78
0.1082	0.8380	18.41	−0.60	0.0960	0.6002	19.48	−0.65
0.2064	0.1976	21.43	−1.02	0.0963	0.7002	19.08	−0.53
0.1992	0.3017	20.64	−1.26	0.0975	0.8012	18.59	−0.49
0.1955	0.4013	20.04	−1.34	0.1016	0.8468	18.40	−0.44
0.2022	0.4943	19.59	−1.31	0.1979	0.1001	22.00	−0.72
0.2994	0.2032	21.26	−1.25	0.1952	0.1965	21.27	−0.95
0.2978	0.3007	20.59	−1.41	0.1976	0.3022	20.69	−0.98
0.3007	0.3962	20.06	−1.45	0.1906	0.4030	20.23	−0.91
0.2991	0.4992	19.57	−1.40	0.1959	0.4987	19.79	−0.85
0.3975	0.3998	20.00	−1.58	0.1996	0.6010	19.30	−0.80
0.3945	0.5028	19.47	−1.57	0.1952	0.7051	18.84	−0.72
0.4908	0.1046	22.19	−1.02	0.2955	0.0997	21.87	−0.84
0.4948	0.2019	21.30	−1.41	0.2947	0.2037	21.09	−1.07
0.4892	0.3061	20.53	−1.63	0.2949	0.3049	20.51	−1.12
0.4947	0.4000	19.91	−1.76	0.2106	0.4519	20.01	−0.87
0.5932	0.1028	22.33	−0.99	0.2964	0.5036	19.60	−0.99
0.5911	0.2022	21.35	−1.45	0.2917	0.6070	19.08	−0.97
0.5939	0.3020	20.53	−1.75	0.3929	0.0969	21.80	−0.90
0.6883	0.2020	21.38	−1.51	0.3964	0.2002	20.98	−1.18
0.7934	0.1003	22.54	−0.99	0.4153	0.2923	20.39	−1.28
0.8459	0.0516	23.23	−0.60	0.3959	0.4013	19.87	−1.24
0.8394	0.1004	22.62	−0.95	0.3960	0.5037	19.32	−1.25
0.2067	0.5932	19.21	−1.18	0.4907	0.1023	21.65	−1.00
0.2975	0.2017	21.28	−1.24	0.4949	0.1980	20.86	−1.29
0.2985	0.6004	19.16	−1.28	0.4957	0.3024	20.24	−1.36
0.3985	0.1990	21.28	−1.35	0.4910	0.4048	19.70	−1.37
0.3985	0.3028	20.49	−1.60	0.5940	0.1013	21.65	−0.99
0.2009	0.0957	22.29	−0.69	0.8005	0.1002	21.83	−0.77
0.2971	0.0986	22.22	−0.84	0.5913	0.2029	20.85	−1.25
0.3997	0.0970	22.24	−0.93	0.5967	0.3014	20.21	−1.38
0.0585	0.0520	22.80	−0.27	0.6916	0.1007	21.77	−0.85
0.0611	0.1010	22.36	−0.46	0.6921	0.2010	20.89	−1.20
0.0970	0.0535	22.77	−0.33	0.8529	0.0472	22.45	−0.41
0.2037	0.6971	18.87	−0.97	0.8477	0.0989	21.89	−0.70
0.6885	0.1019	22.40	−1.02	0.9003	0.0477	22.49	−0.36
2-Methyl-1-propanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3)				2-Methyl-2-propanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3)			
0.0512	0.0457	22.70	−0.31	0.0507	0.0505	22.44	−0.43
0.0520	0.1032	22.18	−0.53	0.0625	0.0990	21.97	−0.62
0.0588	0.8475	18.51	−0.30	0.0665	0.8392	18.40	−0.30
0.0520	0.8900	18.38	−0.21	0.0615	0.8854	18.26	−0.22
0.1064	0.0506	22.59	−0.35	0.1175	0.0963	21.73	−0.71
0.0973	0.0987	22.14	−0.55	0.1010	0.1991	21.05	−0.90
0.1016	0.2005	21.36	−0.80	0.0991	0.3002	20.46	−0.96
0.1049	0.2950	20.76	−0.90	0.1128	0.3938	19.92	−0.97
0.1041	0.3969	20.18	−0.95	0.1017	0.4987	19.49	−0.89
0.0987	0.5006	19.67	−0.92	0.1047	0.6016	19.05	−0.78
0.0989	0.5963	19.26	−0.83	0.1027	0.6989	18.73	−0.60
0.0990	0.6987	18.89	−0.66	0.1202	0.7866	18.38	−0.44
0.1001	0.8025	18.61	−0.40	0.1015	0.8488	18.25	−0.30
0.1045	0.8432	18.44	−0.35	0.2078	0.0980	21.36	−0.80
0.1986	0.1000	22.00	−0.60	0.2016	0.1997	20.66	−0.98
0.2026	0.2015	21.22	−0.85	0.1849	0.3571	19.84	−1.03
0.2014	0.3023	20.54	−1.00	0.2117	0.4959	19.12	−0.94
0.2038	0.4007	19.97	−1.06	0.1925	0.6063	18.74	−0.80
0.2022	0.4965	19.50	−1.03	0.2062	0.6934	18.42	−0.63
0.1998	0.6018	19.05	−0.93	0.2955	0.1049	21.00	−0.86
0.1981	0.7050	18.69	−0.75	0.2939	0.2018	20.31	−1.05
0.2963	0.2016	21.05	−0.94	0.2968	0.3039	19.70	−1.11
0.2571	0.2556	20.74	−1.00	0.2967	0.4034	19.23	−1.06
0.2999	0.3973	19.79	−1.18	0.3034	0.4998	18.82	−0.95
0.2969	0.5036	19.27	−1.14	0.2986	0.6032	18.44	−0.80
0.2923	0.6050	18.89	−1.00	0.3991	0.1013	20.68	−0.89
0.3954	0.1012	21.77	−0.67	0.3976	0.2042	19.94	−1.09

Table 2. Continued

x_1	x_2	σ / $\text{mN}\cdot\text{m}^{-1}$	$\Delta\sigma$ / $\text{mN}\cdot\text{m}^{-1}$	x_1	x_2	σ / $\text{mN}\cdot\text{m}^{-1}$	$\Delta\sigma$ / $\text{mN}\cdot\text{m}^{-1}$
0.3944	0.2042	20.90	−1.00	0.4048	0.2928	19.42	−1.13
0.3958	0.3031	20.20	−1.18	0.3963	0.4018	18.95	−1.05
0.3971	0.4012	19.64	−1.23	0.3961	0.5071	18.53	−0.92
0.4512	0.4576	19.28	−1.25	0.4963	0.1012	20.41	−0.87
0.4973	0.0994	21.71	−0.65	0.4956	0.2032	19.71	−1.04
0.4957	0.2035	20.81	−1.01	0.4968	0.3046	19.16	−1.05
0.4953	0.3007	20.12	−1.19	0.4960	0.4080	18.75	−0.92
0.4980	0.4053	19.51	−1.25	0.5958	0.1031	20.13	−0.84
0.5946	0.1032	21.63	−0.63	0.5995	0.2013	19.47	−0.97
0.5975	0.2049	20.73	−1.00	0.5927	0.3065	18.96	−0.95
0.5959	0.3059	20.07	−1.13	0.6990	0.0987	19.98	−0.70
0.7002	0.1011	21.69	−0.50	0.6988	0.2025	19.34	−0.80
0.7024	0.2009	20.81	−0.85	0.7980	0.1028	19.87	−0.50
0.7978	0.1016	21.70	−0.40	0.8487	0.0503	20.17	−0.32
0.8494	0.0514	22.24	−0.08	0.8512	0.0978	19.80	−0.43
0.8980	0.0489	22.19	−0.11	0.8926	0.0532	20.07	−0.27

Table 3. Coefficients of the Redlich−Kister Equation, A_p, for the Constituent Binary Mixtures, Coefficients of the Cibulka's Equation, B_p, for the Ternary Mixtures, and the Corresponding Standard Deviations, *s*, for Surface Tension Deviations

system	A_0	A_1	A_2	A_3	A_4	s ($\Delta\sigma$)
1-butanol + *n*-hexane[19]	−6.78	−4.82	−0.91	2.81		0.01
2-butanol + *n*-hexane[19]	−5.88	−0.20	−0.33	−2.28		0.01
2-methyl-1-propanol + *n*-hexane[20,21]	−4.57	−0.79	1.10	−0.93		0.00
2-methyl-2-propanol + *n*-hexane[20,21]	−3.03	0.02	−0.06	−0.84		0.00
1-butanol + 1-chlorobutane[18]	−1.00	−0.41	−0.25	0.43		0.00
2-butanol + 1-chlorobutane[18]	−1.16	0.53	0.31	1.36		0.01
2-methyl-1-propanol + 1-chlorobutane[18]	0.02	0.97	1.42	2.40	1.86	0.00
2-methyl-2-propanol + 1-chlorobutane[18]	−1.35	0.48	−0.34	1.07		0.02
n-hexane + 1-chlorobutane[22]	−3.14	0.57	−0.63	2.81		0.00

system	B_1	B_2	B_3	s ($\Delta\sigma$)
1-butanol + *n*-hexane + 1-chlorobutane	−16.74	10.95	14.70	0.02
2-butanol + *n*-hexane + 1-chlorobutane	−11.31	−16.92	46.17	0.02
2-methyl-1-propanol + *n*-hexane + 1-chlorobutane	−7.82	−15.76	11.31	0.02
2-methyl-2-propanol + *n*-hexane + 1-chlorobutane	−11.85	−9.64	20.43	0.02

$$\Delta\sigma = \Delta\sigma_{\text{bin}} + x_1x_2(1 - x_1 - x_2)[B_1 + B_2x_1 + B_3x_2] \tag{2}$$

where

$$\Delta\sigma_{\text{bin}} = x_1x_2\sum_{p=0}^{n}A_p(x_1 - x_2)^p + x_1x_3\sum_{q=0}^{m}A_q(x_1 - x_3)^q + x_2x_3\sum_{r=0}^{o}A_r(x_2 - x_3)^r \tag{3}$$

in which A_i is the binary parameters of a Redlich−Kister type equation for the constituent binary mixtures,[21–25] and x_i the mole fraction of the component *i* in the ternary data point. The coefficients, B_i, and the standard deviations, *s*, obtained by the least-squares method are shown in Table 3 along with the parameters of the binary mixtures.

$\Delta\sigma$ three-dimensional surfaces correlated with Cibulka's equation for the ternary systems have been graphically represented against the composition of the liquid mixtures in Figures 1 to 4. As one can see in these figures, $\Delta\sigma$ values are negative in the whole composition range for the ternary systems containing 1-butanol and 2-methyl-2-propanol, while $\Delta\sigma$ values exhibit an inversion of sign in the region very rich in the butanol isomer for the ternary systems containing 2-butanol and 2-methyl-1-propanol with very small or small $\Delta\sigma$ positive values, respectively. A similar behavior was reported for the constituent binary mixture isomer of butanol + chlorobutane.[21] The most negative $\Delta\sigma$ values are reached in the ternary system containing 1-butanol, while the less negative $\Delta\sigma$ values are obtained for the mixture containing 2-methyl-2-propanol.

Because of the Gibbs adsorption phenomenon, the components of a mixture with lower surface tension have a great tendency to migrate to the surface, while the other components tend to stay in the bulk. So, when a mixture is formed, $\Delta\sigma$ negative values are expected, and $\Delta\sigma$ values are more negative when the difference between the surface tension of the pure components is higher. Nevertheless, favorable interactions among the components of the mixture can counterbalance the migration to the surface of the more active components. If we take into account the bulk behavior of the four ternary systems,

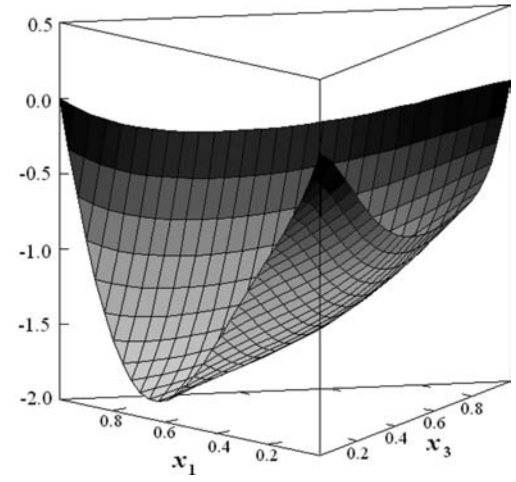

Figure 1. Surface tension deviation, $\Delta\sigma$, correlated using the Cibulka's equation, for the ternary system 1-butanol (1) + *n*-hexane (2) + 1-chlorobutane (3) at 298.15 K.

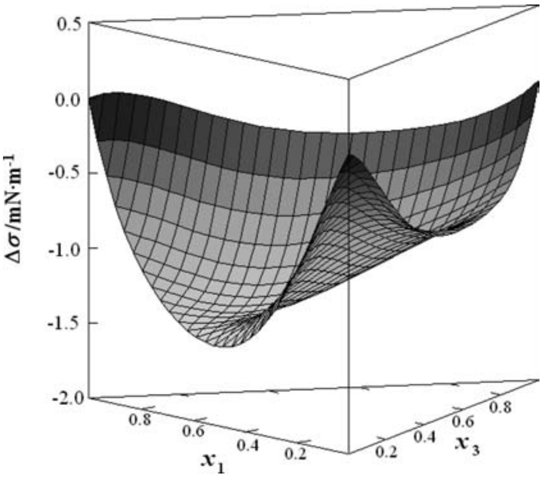

Figure 2. Surface tension deviation, $\Delta\sigma$, correlated using the Cibulka's equation, for the ternary system 2-butanol (1) + *n*-hexane (2) + 1-chlorobutane (3) at 298.15 K.

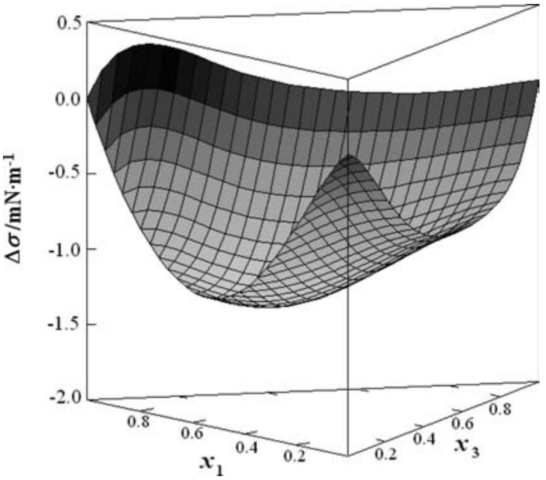

Figure 3. Surface tension deviation, $\Delta\sigma$, correlated using the Cibulka's equation, for the ternary system 2-methyl-1-propanol (1) + *n*-hexane (2) + 1-chlorobutane (3) at 298.15 K.

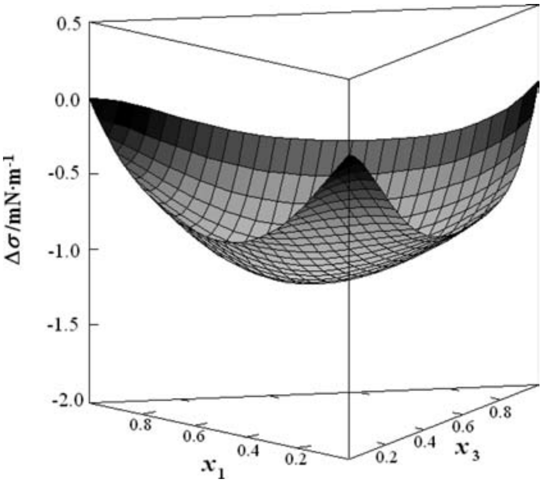

Figure 4. Surface tension deviation, $\Delta\sigma$, correlated using the Cibulka's equation, for the ternary system 2-methyl-2-propanol (1) + *n*-hexane (2) + 1-chlorobutane (3) at 298.15 K.

which all show G^E positive values,[9,10] we can infer that the new interactions, formed in the mixing process between molecules of butanol and chlorobutane, cannot compensate the weakening of the self-interactions; therefore the above-mentioned surface migration is only partially prevented.

Table 4. Root-Mean-Square Deviations for the Prediction of Surface Tensions, rmsd, Using a Group-Contribution Method

system	rmsd/%
1-butanol + *n*-hexane + 1-chlorobutane	1.79
2-butanol + *n*-hexane + 1-chlorobutane	2.03
2-methyl-1-propanol + *n*-hexane + 1-chlorobutane	1.72
2-methyl-2-propanol + *n*-hexane + 1-chlorobutane	2.93

Surface Tension Predictions

The assumptions that the bulk and surface phases are in equilibrium and the partial molar area of component i is the same as its molar area lead to the following equations:[13]

$$\sigma = \sigma_i + \frac{RT}{A_i}\ln\frac{x_i^{\sigma}\gamma_i^{\sigma}}{x_i^{b}\gamma_i^{b}} \qquad (i = 1 \text{ to } 3) \tag{4}$$

$$\sum_i x_i^{\sigma} = 1 \tag{5}$$

where σ and σ_i are the surface tensions for the mixture and the pure compound i, A_i is the molar surface area of pure component i (this magnitude can be calculated from the molar volume, $A_i = V_i^{2/3}N_0^{1/3}$), T is the temperature, x_i^{σ} and x_i^{b} are the mole fractions in the surface and bulk liquid phases, and γ_i^{σ} and γ_i^{b} are the corresponding activity coefficients.

Suarez et al.[11] and Zhibao et al.[12] have used the UNIFAC method[26,27] to calculate the surface and bulk phase activity coefficients; nevertheless, in this work we have employed the modified UNIFAC method by Gmehling et al.[28] In this modification the introduction of separate structure groups for primary, secondary, and tertiary alcohols allows us to discriminate the behavior of a series of isomeric alcohols.

To evaluate the agreement between experimental and predicted results, the mean relative square deviation (MRSD) expressed in percentage has been calculated using the following equation:

$$\mathrm{MRSD} = 100\left(\frac{1}{m}\sum_{i=1}^{m}\left(\frac{\sigma_{i,\mathrm{exp}} - \sigma_{i,\mathrm{pred}}}{\sigma_{i,\mathrm{exp}}}\right)^2\right)^{1/2} \tag{6}$$

where m is the number of experimental data.

The values for the MRSD expressed in percentage are given in Table 4. These values indicate that the group contribution method gives an excellent prediction of surface tension, with the overall MRSD average being 2.11 %.

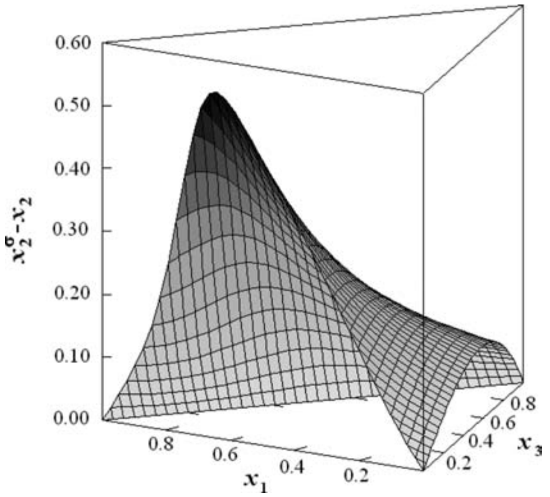

Figure 5. Excess surface mole fractions of *n*-hexane, $x_2^{\sigma} - x_2$, for the ternary system 1-butanol (1) + *n*-hexane (2) + 1-chlorobutane (3) at 298.15 K.

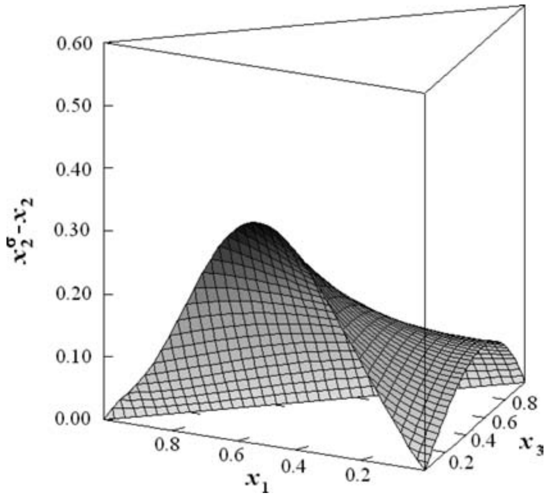

Figure 6. Excess surface mole fractions of *n*-hexane, $x_2^\sigma - x_2$, for the ternary system 2-butanol (1) + *n*-hexane (2) + 1-chlorobutane (3) at 298.15 K.

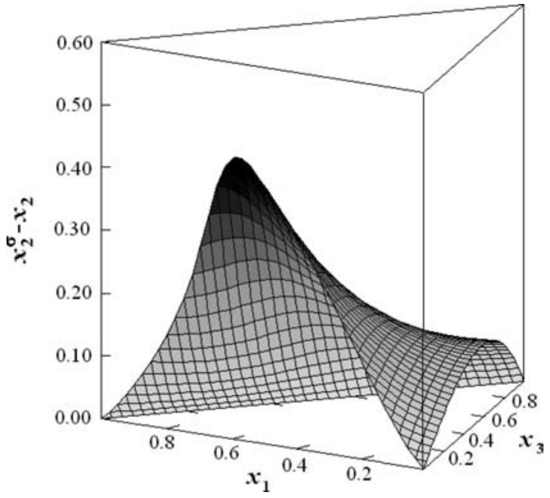

Figure 7. Excess surface mole fractions of *n*-hexane, $x_2^\sigma - x_2$, for the ternary system 2-methyl-1-propanol (1) + *n*-hexane (2) + 1-chlorobutane (3) at 298.15 K.

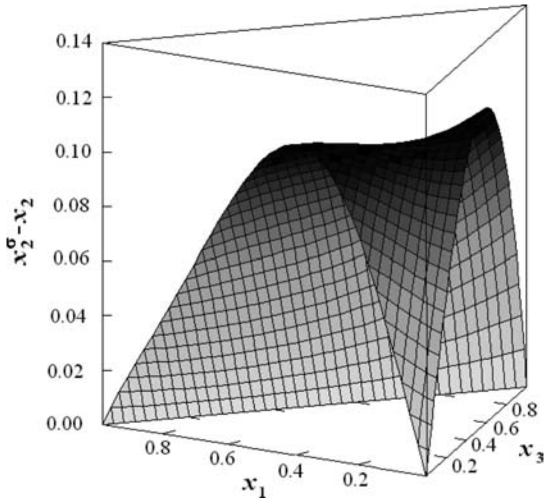

Figure 8. Excess surface mole fractions of *n*-hexane, $x_2^\sigma - x_2$, for the ternary system 2-methyl-2-propanol (1) + *n*-hexane (2) + 1-chlorobutane (3) at 298.15 K.

The better agreement between experimental and estimated values has been obtained for 1-butanol + *n*-hexane + 1-chlorobutane and 2-methyl-1-propanol + *n*-hexane + 1-chlorobutane mixtures, while the greater differences have been found for the system containing 2-methyl-2-propanol.

We have mentioned that the surface of a mixture is enriched usually in the component of lower surface tension by means of a mechanism designated as Gibbs adsorption. Quantitative information about this enrichment is given by the surface mole fractions, x_i^σ; this magnitude can be estimated by means of eq 4. A representation of $x_2^\sigma - x_2$, that is, excess surface mole fraction of *n*-hexane in the ternary mixtures, is shown in Figures 5 to 8. In all of the cases, there is an amount of hexane that migrates to the surface by the mechanism of adsorption abovementioned especially for the mixture containing 1-butanol, with the $x_2^\sigma - x_2$ values being higher in the rich butanol regions. It is also remarkable that for the mixture containing 2-methyl-2-propanol the excess surface mole fraction of *n*-hexane is lower than for the rest of the ternary mixtures, probably due to a competition with 2-methyl-2-propanol molecules for the surface.

Supporting Information Available:

Table S1: densities of the ternary mixtures at 298.15 K. This material is available free of charge via the Internet at http://pubs.acs.org.

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