Densities and Viscosities of the Ternary Mixtures 2-Methyl-1-propanol (or 2-Methyl-2-propanol) + *N*-Hexane + 1-Chlorobutane at 298.15 K

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Densities and viscosities of the ternary mixtures 2-methyl-1-propanol (or 2-methyl-2-propanol) + *n*-hexane + 1-chlorobutane have been measured at atmospheric pressure and 298.15 K. Excess molar volumes and viscosity deviations for the ternary mixtures were calculated from experimental data and fitted by the Cibulka equation. These properties have been analyzed taking into account the molecular interactions.

Introduction

Thermodynamic and transport properties of liquid mixtures are of great theoretical and practical interest. In particular, mixtures containing associating and nonassociating molecules are especially relevant from a theoretical point of view for a better understanding of the molecular liquid structure and the intermolecular interactions.

Alcohols are strongly associated with hydrogen bonding. Their binary mixtures with alkanes or chloroalkanes have attracted the attention of the scientific community, and several thermodynamic properties have been obtained; however, scarce information is available about ternary mixtures containing such compounds.

In the last years, the study of thermodynamic and transport properties of ternary mixtures[1–4] containing an isomer of butanol (1-butanol or 2-butanol) + hexane + 1-chlorobutane has been reported. Continuing this systematic study, we present here experimental density and viscosity measurements of the ternary mixtures 2-methyl-1-propanol or 2-methyl-2-propanol + *n*-hexane + 1-chlorobutane at atmospheric pressure at the temperature of 298.15 K. Excess molar volumes and viscosity deviations have been obtained from experimental data. These properties have been correlated with composition using Cibulka's equation,[5] and their values have been analyzed in terms of molecular interactions and compared with previously reported data.

Experimental Section

Materials. The compounds used, 2-methyl-1-propanol, 2-methyl-2-propanol (purity better than 99.5 %), and *n*-hexane and 1-chlorobutane (better than 99 %), were obtained from Aldrich. The purity of the chemicals was checked by comparing the measured densities and viscosities with those reported in the literature. No further purification was attempted. The pure compound properties at 298.15 K, along with literature values, are given in Table 1.

Apparatus and Procedure. Densities, ρ, of the pure compounds and the ternary mixtures were measured with an Anton Paar DMA-58 vibrating-tube densimeter in which the temperature is controlled automatically within ± 0.01 K. The apparatus was calibrated with deionized doubly distilled water and dry air. The uncertainty of density measurements is ± $1 \cdot 10^{-2}$ kg·m^{-3}.

Table 1. Densities, ρ, And Absolute Viscosities, η, of Pure Components at 298.15 K and Comparison with Literature Data

	ρ/kg·m^{-3}		η/mPa·s	
component	exptl	lit.	exptl	lit.
2-methyl-1-propanol	797.77	797.8[8]	3.3528	3.3330[9]
2-methyl-2-propanol	780.78	780.9[8]	4.3839	4.4380[9]
n-hexane	655.07	654.84[9]	0.2949	0.2942[9]
1-chlorobutane	880.69	880.95[9]	0.4235	0.4260[9]

Kinematic viscosities, ν, were determined using an Ubbelohde viscosimeter (inner diameter = 0.63 mm, capillary length = 89.3 mm) with a Schott-Gërate automatic measuring unit model AVS-440, for which the reproducibility of the flow time measurement is ± 0.01 s. At least four time flow measurements were performed for each composition and temperature, and the results were averaged. Kinetic energy corrections were applied to the experimental data. A Schott-Gërate thermostat was used to keep the temperature within ± 0.01 K. Absolute viscosities were obtained from experimental densities and kinematic viscosities ($\eta = \nu\rho$). The calibration of the viscosimeter was carried out with deionized doubly distilled water, and the value used (η = 0.8902 mPa·s at 298.15 K) was taken from Marsh,[6] after calibration of the estimated uncertainty for dynamic viscosity measurements, and is less than 0.002 mPa·s.

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

The pure compound properties at 298.15 K, along with literature values,[7,8] are given in Table 1.

Results and Discussion

The experimental densities of the ternary mixtures 2-methyl-1-propanol or + 2-methyl-2-propanol + *n*-hexane + 1-chlorobutane at 298.15 K are shown in Table 2, along with excess molar volumes, V^E, which were determined using the equation

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$$V^E/m^3 \cdot mol^{-1} = \left(\frac{x_1M_1 + x_2M_2 + x_3M_3}{\rho}\right) - \left(x_1\frac{M_1}{\rho_1} + x_2\frac{M_2}{\rho_2} + x_3\frac{M_3}{\rho_3}\right) \quad (1)$$

where M_i, x_i, and ρ_i are the molecular weights, the mole fractions, and the densities of the pure components, respectively, and ρ is the density of the mixture.

Kinematic viscosities, ν, absolute viscosities, η, and viscosity deviations, $\Delta\eta$, for the same mixtures are collected in Table 3. Viscosity deviations were obtained using:

$$\Delta\eta/mPa \cdot s = \eta - (x_1\eta_1 + x_2\eta_2 + x_3\eta_3) \quad (2)$$

where η is the absolute viscosity of the mixture and η_i is the absolute viscosity of pure component i.

The excess molar volumes and viscosity deviations for the ternary mixtures have been fitted by Cibulka's equation

$$Y = Y_{bin} + x_1x_2(1 - x_1 - x_2)[B_1 + B_2x_1 + B_3x_2] \quad (3)$$

where $Y = V^E$ or $\Delta\eta$ of the ternary mixture and

$$Y_{bin} = x_1x_2\sum_{p=0}^{n} A_p(x_1 - x_2)^p + x_1x_3\sum_{q=0}^{m} A_q(x_1 - x_3)^q + x_2x_3\sum_{r=0}^{o} A_r(x_2 - x_3)^r \quad (4)$$

A_i is the binary parameters of a Redlich–Kister-type equation for the constituent binary mixtures,[1,2,9–11] and x_i is the mole fraction of the component i in the ternary data point.

The coefficients, B_i, and the standard deviations, σ, obtained by the least-squares method are shown in Table 4 along with the parameters of the binary mixtures.

V^E and $\Delta\eta$ surfaces correlated with Cibulka's equation for the ternary systems have been plotted in Figures 1 and 2, respectively. As one can see in Figure 1, excess molar volumes are positive in the whole composition range for both systems, although V^E values are bigger in the system containing 2-methyl-2-propanol and V^E dependence with composition is slightly different in both systems: the ternary system containing 2-methyl-1-propanol behaves like previously reported systems[1,3] containing 1-butanol or 2-butanol. In all these systems, V^E

Table 2. Experimental Densities, ρ, and Calculated Excess Molar Volumes, V^E, of the Ternary Mixtures 2-Methyl-1-propanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3) and 2-Methyl-2-propanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3) at 298.15 K

		ρ	$V^E \cdot 10^6$			ρ	$V^E \cdot 10^6$
x_1	x_2	kg·m^{-3}	m^3·mol^{-1}	x_1	x_2	kg·m^{-3}	m^3·mol^{-1}
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	863.32	0.099	0.0490	0.0506	860.51	0.200
0.0520	0.1032	847.47	0.116	0.0625	0.0990	845.74	0.251
0.0588	0.8475	677.96	0.146	0.0665	0.8392	677.54	0.272
0.0520	0.8900	670.58	0.118	0.1137	0.0485	853.72	0.376
0.1064	0.0506	857.37	0.153	0.1175	0.0963	840.41	0.376
0.0973	0.0987	844.80	0.171	0.1010	0.1991	815.20	0.360
0.1016	0.2005	817.47	0.206	0.0991	0.3002	790.15	0.372
0.1049	0.2950	793.40	0.227	0.1128	0.3938	766.48	0.397
0.1041	0.3969	768.94	0.253	0.1017	0.4987	743.86	0.374
0.0987	0.5006	745.68	0.264	0.1047	0.6016	721.19	0.393
0.0989	0.5963	724.85	0.261	0.1202	0.7866	682.01	0.419
0.0990	0.6987	703.62	0.240	0.1015	0.8488	672.04	0.361
0.1001	0.8025	683.04	0.202	0.1027	0.6989	701.36	0.368
0.1045	0.8432	674.90	0.185	0.2078	0.0980	830.29	0.533
0.1986	0.1000	836.19	0.227	0.2016	0.1997	804.37	0.534
0.2026	0.2015	808.98	0.248	0.1985	0.3054	778.31	0.558
0.2014	0.3023	783.58	0.282	0.1849	0.3571	767.49	0.523
0.2038	0.4007	759.77	0.299	0.2117	0.4959	732.99	0.568
0.2022	0.4965	737.98	0.308	0.1925	0.6063	711.10	0.550
0.1998	0.6018	715.31	0.294	0.2062	0.6934	691.91	0.537
0.1981	0.7050	694.12	0.268	0.2955	0.1049	819.34	0.632
0.3007	0.0999	828.20	0.220	0.2939	0.2018	794.17	0.655
0.2963	0.2016	801.48	0.244	0.2968	0.3039	768.51	0.675
0.2571	0.2556	790.74	0.270	0.2967	0.4034	745.13	0.668
0.2999	0.3973	752.90	0.271	0.3034	0.4998	722.83	0.663
0.2969	0.5036	728.82	0.275	0.2986	0.6032	701.12	0.652
0.2923	0.6050	707.18	0.263	0.3991	0.1013	809.76	0.698
0.3954	0.1012	820.49	0.181	0.3976	0.2042	783.05	0.719
0.3944	0.2042	793.14	0.195	0.4048	0.2928	760.23	0.747
0.3958	0.3031	767.97	0.211	0.3963	0.4018	735.36	0.747
0.3971	0.4012	744.27	0.213	0.3961	0.5071	711.91	0.724
0.4512	0.4576	726.76	0.188	0.4963	0.1012	800.00	0.724
0.4973	0.0994	812.90	0.137	0.4956	0.2032	773.42	0.748
0.4957	0.2035	785.24	0.141	0.4968	0.3046	748.01	0.790
0.4953	0.3007	760.64	0.141	0.4960	0.4080	723.77	0.802
0.4980	0.4053	735.11	0.152	0.5958	0.1031	789.55	0.715
0.5946	0.1032	804.04	0.082	0.5995	0.2013	763.28	0.771
0.5975	0.2049	776.66	0.071	0.5927	0.3065	737.86	0.800
0.5959	0.3059	751.02	0.088	0.6990	0.0987	780.54	0.662
0.7002	0.1011	795.83	0.034	0.6988	0.2025	753.41	0.686
0.7024	0.2009	768.74	0.037	0.7980	0.1028	770.02	0.539
0.7978	0.1016	787.12	0.018	0.8487	0.0503	779.82	0.414
0.8494	0.0514	796.65	0.010	0.8512	0.0978	766.46	0.439
0.8507	0.0993	782.97	0.016	0.8926	0.0532	774.97	0.330

Table 3. Experimental Kinematic Viscosities, ν, and Calculated Absolute Viscosities, η, and Viscosity Deviations, $\Delta\eta$, of the Ternary Mixtures 2-Methyl-1-propanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3) and 2-Methyl-2-propanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3) at 298.15 K

		ν	η	$\Delta\eta$			ν	η	$\Delta\eta$
x_1	x_2	$mm^2 \cdot s^{-1}$	mPa·s	mPa·s	x_1	x_2	$mm^2 \cdot s^{-1}$	mPa·s	mPa·s
2-Methyl-1-propanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3)					2-Methyl-2-propanol (1) + *n*-Hexane (2) + 1-Chlorobutane (3)				
0.0518	0.0469	0.4853	0.4188	−0.1504	0.0402	0.0610	0.4862	0.4175	−0.1574
0.0513	0.0997	0.4816	0.4087	−0.1523	0.0505	0.0980	0.4793	0.4062	−0.2047
0.0509	0.8589	0.4594	0.3107	−0.1514	0.0480	0.8632	0.4553	0.3073	−0.1953
0.0535	0.8917	0.4596	0.3080	−0.1575	0.0503	0.8963	0.4557	0.3046	−0.2028
0.0931	0.0474	0.5015	0.4310	−0.2591	0.0944	0.0462	0.4942	0.4233	−0.3681
0.0948	0.0995	0.4984	0.4211	−0.2673	0.0922	0.0976	0.4899	0.4129	−0.3632
0.0935	0.2002	0.4912	0.4019	−0.2697	0.0979	0.1982	0.4839	0.3948	−0.3909
0.0903	0.3040	0.4982	0.3948	−0.2541	0.0921	0.3025	0.4781	0.3779	−0.3715
0.0956	0.4024	0.4988	0.3832	−0.2686	0.0918	0.4055	0.4467	0.3422	−0.3927
0.0938	0.5067	0.4949	0.3686	−0.2645	0.0946	0.5022	0.4175	0.3105	−0.4231
0.0915	0.6114	0.4990	0.3604	−0.2525	0.0935	0.6078	0.4264	0.3075	−0.4081
0.0917	0.7099	0.4741	0.3327	−0.2681	0.0901	0.7145	0.4373	0.3059	−0.3825
0.0905	0.8169	0.4705	0.3204	−0.2631	0.0920	0.8116	0.4660	0.3169	−0.3666
0.0906	0.8564	0.4710	0.3172	−0.2616	0.0907	0.8616	0.4657	0.3123	−0.3596
0.1958	0.0982	0.5592	0.4681	−0.5163	0.1930	0.1018	0.5318	0.4418	−0.7330
0.1930	0.2001	0.5683	0.4604	−0.5027	0.1928	0.2012	0.5231	0.4210	−0.7402
0.1969	0.3016	0.5491	0.4306	−0.5309	0.1952	0.3027	0.5164	0.4025	−0.7551
0.1950	0.4032	0.5332	0.4052	−0.5377	0.1955	0.4087	0.5079	0.3831	−0.7621
0.1981	0.5045	0.5298	0.3903	−0.5486	0.1915	0.5093	0.5024	0.3678	−0.7486
0.1955	0.6088	0.5248	0.3748	−0.5431	0.1920	0.6060	0.5029	0.3577	−0.7483
0.1940	0.7097	0.5249	0.3640	−0.5365	0.1942	0.7048	0.4608	0.3183	−0.7837
0.2978	0.0957	0.6440	0.5342	−0.7493	0.2936	0.0988	0.5917	0.4858	−1.0878
0.2974	0.1973	0.6299	0.5056	−0.7637	0.2955	0.2050	0.5842	0.4634	−1.1040
0.2987	0.3033	0.6208	0.4815	−0.7780	0.2532	0.2680	0.5621	0.4395	−0.9523
0.2969	0.4027	0.6119	0.4601	−0.7813	0.2899	0.4065	0.5628	0.4194	−1.0999
0.2988	0.5058	0.6076	0.4425	−0.7912	0.2880	0.5135	0.5530	0.3989	−1.0992
0.2973	0.6105	0.6033	0.4256	−0.7903	0.2951	0.6141	0.5520	0.3859	−1.1273
0.4026	0.0951	0.7612	0.6254	−0.9652	0.3963	0.0960	0.6718	0.5451	−1.4356
0.3970	0.2009	0.7381	0.5858	−0.9748	0.3920	0.2048	0.7172	0.5618	−1.3878
0.4006	0.3014	0.7318	0.5621	−0.9961	0.4352	0.2871	0.7948	0.6029	−1.5072
0.4009	0.4035	0.7446	0.5536	−0.9924	0.3892	0.4095	0.7017	0.5153	−1.3969
0.3972	0.5070	0.7169	0.5162	−1.0056	0.3924	0.5080	0.6524	0.4645	−1.4477
0.4996	0.0986	0.9150	0.7438	−1.1305	0.4955	0.1009	0.7786	0.6229	−1.7500
0.5005	0.2003	0.9008	0.7078	−1.1561	0.4970	0.2116	0.7991	0.6161	−1.7485
0.4998	0.3032	0.8903	0.6764	−1.1722	0.4957	0.3169	0.8433	0.6284	−1.7175
0.4999	0.4053	0.8853	0.6506	−1.1851	0.4942	0.4128	0.7748	0.5601	−1.7675
0.6000	0.0989	1.1464	0.9225	−1.2459	0.5871	0.0986	0.9281	0.7346	−2.0014
0.6006	0.1998	1.1359	0.8834	−1.2737	0.5970	0.2042	0.9067	0.6917	−2.0699
0.5989	0.3045	1.1257	0.8454	−1.2933	0.5922	0.3062	0.8754	0.6461	−2.0834
0.7022	0.0979	1.5005	1.1951	−1.2728	0.7065	0.0980	1.0386	0.8101	−2.3988
0.7045	0.1988	1.4947	1.1495	−1.3121	0.6768	0.2331	0.9147	0.6839	−2.3900
0.7591	0.0928	1.7826	1.4136	−1.2216	0.8047	0.1097	1.4319	1.0991	−2.4972
0.8534	0.0467	2.4817	1.9795	−0.9379	0.8585	0.0588	2.0265	1.5736	−2.2423
0.8507	0.0953	2.4425	1.9152	−0.9880	0.8556	0.1002	1.9672	1.5058	−2.2933
0.9007	0.0475	2.9193	2.3154	−0.7404	0.9056	0.0459	2.7711	2.1502	−1.8539

Table 4. Coefficients of the Redlich−Kister Equation, A_p, for the Constituent Binary Mixtures and Coefficients of Cibulka's Equation, B_p, for the Ternary Mixtures and the Corresponding Standard Deviations, σ, for Excess Molar Volumes and Viscosity Deviations

system	property	A_0	A_1	A_2	A_3	A_4	$\sigma(Y)$
2-methyl-1-propanol + *n*-hexane[9]	$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	0.762	−0.990	0.423	0.153	–	0.003
	$\Delta\eta$/mPa·s	−4.830	−3.176	−1.699	−0.352	–	0.004
2-methyl-2-propanol + *n*-hexane[a]	$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	3.238	0.641	1.023	−1.694	–	0.007
	$\Delta\eta$/mPa·s	−7.1791	−6.0220	−8.1441	−7.2062	–	0.0228
2-methyl-1-propanol + 1-chlorobutane[10,11]	$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	0.611	−0.584	0.285	−0.441	–	0.002
	$\Delta\eta$/mPa·s	−4.3746	−2.7251	−1.5136	−0.4556	–	0.0016
2-methyl-2-propanol + 1-chlorobutane[10,11]	$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	2.788	0.039	1.005	−0.376	–	0.004
	$\Delta\eta$/mPa·s	−6.9662	−5.4959	−4.5994	−6.2236	−4.8128	0.0110
n-hexane + 1-chlorobutane[1,2]	$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	0.505	0.255	−0.108	−0.202	–	0.003
	$\Delta\eta$/mPa·s	−0.0755	0.0115	−0.0015	0.0093	–	0.0005

system	property	B_1	B_2	B_3	$\sigma(Y)$
2-methyl-1-propanol + *n*-hexane + 1-chlorobutane	$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	5.303	−11.705	−1.333	0.004
	$\Delta\eta$/mPa·s	1.7000	9.0802	0.6370	0.0077
2-methyl-2-propanol + *n*-hexane + 1-chlorobutane	$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	2.056	−4.772	−3.651	0.011
	$\Delta\eta$/mPa·s	−0.1688	33.9644	−0.4394	0.0245

[a] Unpublished results.

maximum values appear inside the triangular diagram and correspond to ternary mixtures with small mole fractions of the butanol isomer. However, in the ternary system containing 2-methyl-2-propanol, the maximum V^E values belong to the binary system *n*-hexane + tertiary alcohol. V^E values increase in the sequence 1-butanol < 2-methyl-1-propanol < 2-butanol

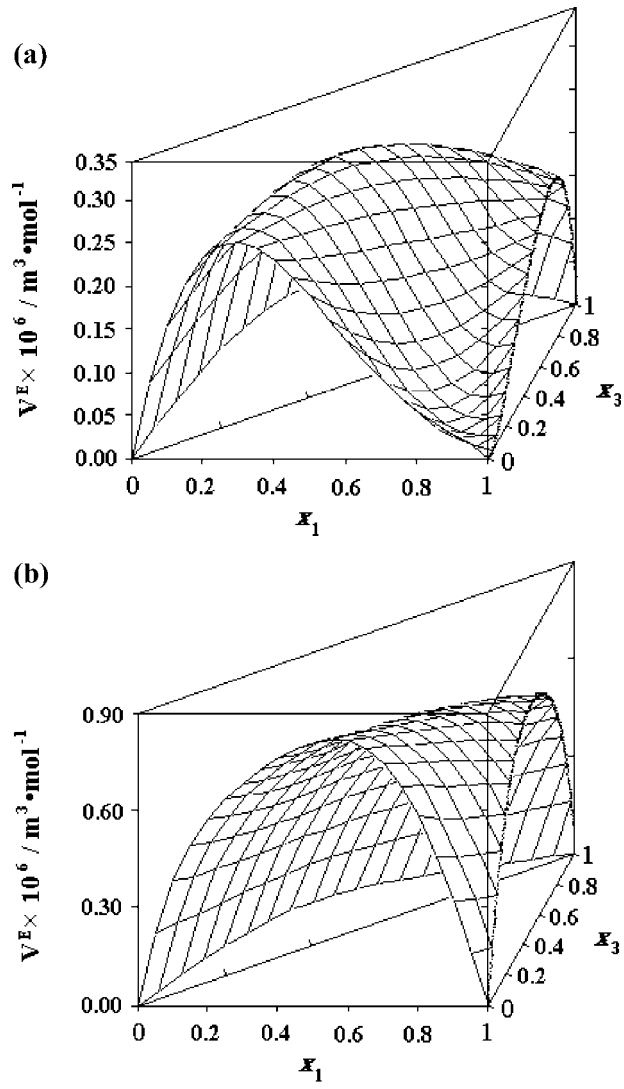

Figure 1. V^E surface at 298.15 K according to Cibulka′s equation for the ternary systems: (a) 2-methyl-1-propanol (1) + *n*-hexane (2) + 1-chlorobutane (3); (b) 2-methyl-2-propanol (1) + *n*-hexane (2) + 1-chlorobutane (3).

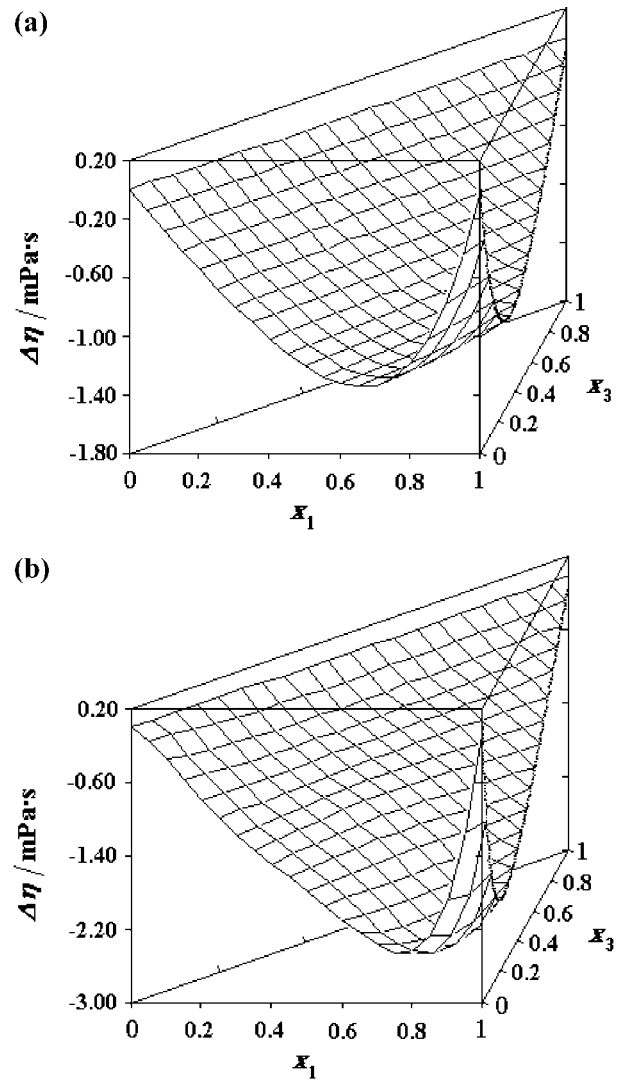

Figure 2. $\Delta\eta$ surface at 298.15 K according to Cibulka's equation for the ternary systems: (a) 2-methyl-1-propanol (1) + *n*-hexane (2) + 1-chlorobutane (3); (b) 2-methyl-2-propanol (1) + *n*-hexane (2) + 1-chlorobutane (3).

< 2-methyl-2-propanol. This sequence is directly related to the self-association of butanol isomers: 1-butanol and 2-methyl-1-propanol are primary alcohols, strongly associated in the pure state, while 2-butanol and 2-methyl-2-propanol are secondary and tertiary alcohols, respectively, and hydrogen bonding is weaker.

Figure 2 shows that viscosity deviations are negative for both systems in the complete composition range. $\Delta\eta$ dependence with composition is quite similar for both systems, although values are more negative in the ternary system containing 2-methyl-2-propanol. Previously reported[2,4] $\Delta\eta$ for the mixtures containing 1-butanol or 2-butanol also show a similar behavior, with minimum viscosity deviations appearing in mixtures containing high concentrations of butanol isomer and small quantities of chlorobutane. $\Delta\eta$ values are less negative in the system with 1-butanol, while they are very similar for the ternary systems containing 2-butanol or 2-methyl-1-propanol. Negative viscosity deviations indicate that our systems have easier flow than the pure liquids, which is characteristic of liquid mixtures with positive deviations of Raoult's law.[12]

Positive excess molar volumes and negative viscosity deviations indicate that the main effect in the mixtures is the breaking of the self-interactions in the compounds during the mixing process: in these systems the rupture of hydrogen bonds of butanol isomers and the breaking of dipole–dipole interactions of 1-chlorobutane predominate over the formation of new interactions OH−Cl.

V^E and $\Delta\eta$ values are bigger in absolute value in the ternary system containing 2-methyl-2-propanol, which indicates that the breaking of hydrogen bonding of the alcohol molecules is more effective when the alcohol is weakly self-associated. In line with this, the smaller absolute values of both properties correspond to the ternary system of 1-butanol, which is the strongest self-associated component in the mixtures.

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