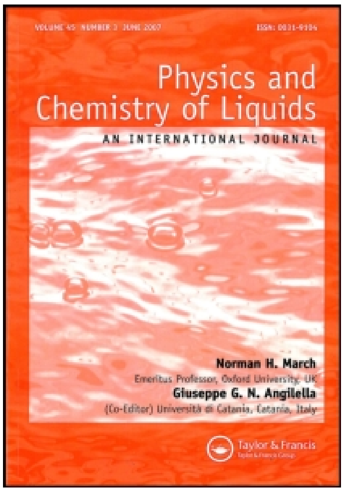

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Densities and Viscosities for the Binary Mixtures (2-Methyl-1-Chloropropane Isomeric Butanol) at 298.15 and 313.15 K

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DENSITIES AND VISCOSITIES FOR THE BINARY MIXTURES (2-METHYL-1-CHLOROPROPANE + ISOMERIC BUTANOL) AT 298.15 AND 313.15 K

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This paper reports excess volumes, V^E, and viscosity deviations, $\Delta\eta$, for binary mixtures of 2-methyl-1-chloropropane with an isomer of butanol at the temperatures 298.15 K and 313.15 K. These properties were obtained from density and viscosity measurements. The results are correlated by means of a Redlich-Kister type equation, and interpreted in terms of molecular interactions. The systems show positive values of V^E except in a short range of compositions for mixtures containing primary butanols (1-butanol at both temperatures and 2-methyl-1-propanol at 298.15 K), whereas $\Delta\eta$ presents negative values at both temperatures over the whole composition range.

Keywords: Binary mixtures; Butanols; 2-Methyl-1-chloropropane; Excess properties

INTRODUCTION

Excess properties of liquid mixtures reveal the existence of specific molecular interactions. The systematic investigations of these excess properties are therefore of great importance in order to get a better knowledge of those interactions. The present work completes a series

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*Corresponding author.

of papers devoted to the study of the volumetric and viscosimetric behaviour of systems formed by an isomer of chlorobutane plus an isomer of butanol [1–4]. This series is comprised into a wider investigation of thermodynamic and transport properties of binary mixtures of butanols with chlorobutanes [5–8], investigation whose objective is to provide experimental information about Cl–OH interaction. As far as we know, there are no literature data for these systems in order to compare our values.

EXPERIMENTAL

The liquids used were: 2-methyl-1-chloropropane, (better than 98.0 mol%), provided by Fluka, and 1-butanol (better than 99.8%), 2-methyl-1-propanol and 2-methyl-2-propanol (better than 99.5%) and 2-butanol (better than 99.0%), all supplied by Aldrich. The purity of chemicals used was checked by measuring their density and was considered sufficient, so no further purification was attempted.

Table I shows the experimental values of density and viscosity for the pure compounds at 298.15 K in comparison with published values [9, 10]. The densities of the pure components and mixtures were measured by means of an Anton Paar DMA-58 vibrating tube densimeter automatically thermostated at 298.15 ± 0.01 K. Calibration was carried out with deionized doubly distilled water and dry air. The precision of the density measurements is estimated to be $\pm 0.00001\ \mathrm{g \cdot cm^{-3}}$.

The viscosity measurements were obtained with an Ubbelhode viscosimeter and a Schott-Geräte automatic measuring unity model

TABLE I Densities and viscosities of pure compounds at $T = 298.15$ K compared with literature data [9]

	ρ / $g \cdot cm^{-3}$		η / $mPa \cdot s$	
Compound	*Exptl.*	*Lit.*	*Exptl.*	*Lit.*
2-Methyl-1-chloropropane	0.87105	0.8717	0.4318	0.431[a]
1-Butanol	0.80575	0.80575	2.5455	2.5710
2-Butanol	0.80241	0.80241	3.0425	2.998
2-Methyl-1-propanol	0.79784	0.7978	3.3996	3.3330
2-Methyl-2-propanol	0.78034	0.7812	4.4059	4.438

[a] Reference [10].

AVS-440. Mole fractions of the mixtures were determined by mass. Details of the procedure have been described previously [11].

RESULTS AND DISCUSSION

The excess properties were calculated from our measurements according to the following equations:

$$V^E = \sum_{i=1}^{2} x_i M_i \left(\frac{1}{\rho} - \frac{1}{\rho_i} \right) \tag{1}$$

$$\Delta\eta = \eta - \sum_{i=1}^{2} x_i \eta_i \tag{2}$$

where x_i, M_i, ρ_i and η_i designate mole fraction, molecular weight ($\mathrm{g \cdot mol^{-1}}$), density ($\mathrm{g \cdot cm^{-3}}$) and absolute viscosity ($\mathrm{mPa \cdot s}$) of pure component i, respectively. Symbols without subscript refer to the mixture, subscript 1 is used for 2-methyl-1-chloropropane and 2 for isomeric butanol.

The excess properties for binary mixtures were fitted to a Redlich-Kister polynomial equation [12]:

$$Y_{ij}^E = x_i x_j \sum_{p=0}^{p} A_p (x_i - x_j)^p \tag{3}$$

where Y_{ij}^E is V^E or $\Delta\eta$, x_i denotes the mole fraction of component i in the i, j mixture with $x_j = 1 - x_i$, and A_p are adjustable parameters.

The calculated values of the excess functions for the binary mixtures are presented in Tables II and III, and graphically shown in Figures 1–4. The coefficients A_p (Eq. (3)) and the standard deviations σ obtained by the least squares method are given in Table IV.

The behaviour exhibited by the binary mixtures can be interpreted through the interactions present in the pure liquids and in the mixtures.

In this respect and first of all, the main characteristics of the pure compounds to be taken into account are the hydrogen bond associated structure of the alcohols, and the dipole–dipole interactions existing

TABLE II Densities and excess volumes for 2-methyl-1-chloropropane (1) with an isomeric butanol (2) at indicated temperature

x_1	ρ $g \cdot cm^{-3}$	V^E $cm^3 \cdot mol^{-1}$	x_1	ρ $g \cdot cm^{-3}$	V^E $cm^3 \cdot mol^{-1}$	x_1	ρ $g \cdot cm^{-3}$	V^E $cm^3 \cdot mol^{-1}$
2-Methyl-1-chloropropane (1)+1-Butanol (2) at 298.15 K								
0.0963	0.81310	−0.022	0.4024	0.83404	0.033	0.7038	0.85254	0.129
0.2008	0.82058	−0.016	0.4986	0.84013	0.063	0.8085	0.85873	0.146
0.3020	0.82744	0.008	0.6037	0.84659	0.095	0.8864	0.86346	0.130
2-Methyl-1-chloropropane (1)+2-Butanol (2) at 298.15 K								
0.1040	0.80955	0.110	0.5031	0.83587	0.413	0.8931	0.86210	0.302
0.2086	0.81656	0.213	0.6066	0.84260	0.443	0.9455	0.86625	0.186
0.3067	0.82303	0.297	0.7105	0.84944	0.440			
0.4072	0.82960	0.367	0.8172	0.85662	0.394			
2-Methyl-1-chloropropane (1)+2-Methyl-1-propanol (2) at 298.15 K								
0.1050	0.80650	−0.021	0.5054	0.83626	0.112	0.8927	0.86276	0.159
0.2093	0.81462	0.009	0.6114	0.84361	0.146	0.9432	0.86650	0.107
0.3095	0.82212	0.047	0.7121	0.85043	0.174			
0.4092	0.82940	0.081	0.8062	0.85674	0.189			
2-Methyl-1-chloropropane (1)+2-Methyl-2-propanol (2) at 298.15 K								
0.0925	0.78746	0.264	0.4119	0.81474	0.667	0.7136	0.84204	0.619
0.2054	0.79660	0.500	0.5110	0.82370	0.677	0.8180	0.85180	0.514
0.3181	0.80639	0.618	0.6120	0.83281	0.664	0.8909	0.85885	0.398
2-Methyl-1-chloropropane (1)+1-Butanol (2) at 313.15 K								
0.0990	0.80101	−0.015	0.4101	0.82044	0.044	0.7030	0.83670	0.151
0.2018	0.80775	−0.009	0.5088	0.82607	0.082	0.7994	0.84193	0.164
0.3152	0.81481	0.014	0.6102	0.83168	0.120	0.9101	0.84822	0.120

2-Methyl-1-chloropropane (1) + 2-Butanol (2) at 313.15 K								
0.1081	0.79616	0.167	0.4014	0.81408	0.441	0.6684	0.83040	0.520
0.2018	0.80190	0.275	0.4984	0.81995	0.495	0.7627	0.83650	0.468
0.3054	0.80819	0.377	0.6007	0.82621	0.521	0.8672	0.84349	0.359
2-Methyl-1-chloropropane (1) + 2-Methyl-1-propanol (2) at 313.15 K								
0.1023	0.79369	0.023	0.4044	0.81452	0.152	0.7049	0.83378	0.246
0.2042	0.80099	0.058	0.5018	0.82085	0.194	0.7977	0.83972	0.235
0.3083	0.80815	0.104	0.6029	0.82732	0.227	0.8975	0.84640	0.167
2-Methyl-1-chloropropane (1) + 2-Methyl-2-propanol (2) at 313.15 K								
0.1043	0.77266	0.319	0.4095	0.79809	0.724	0.7035	0.82386	0.721
0.2073	0.78084	0.535	0.5065	0.80651	0.757	0.8037	0.83322	0.598
0.3018	0.78878	0.653	0.6105	0.81568	0.748	0.9009	0.84275	0.399

TABLE III Absolute viscosities and viscosity deviations for 2-methyl-1-chloropropane (1) with an isomeric butanol (2) at indicated temperature

x_1	η $mPa \cdot s$	$\Delta\eta$ $mPa \cdot s$	x_1	η $mPa \cdot s$	$\Delta\eta$ $mPa \cdot s$	x_1	η $mPa \cdot s$	$\Delta\eta$ $mPa \cdot s$
2-Methyl-1-chloropropane (1) + 1-Butanol (2) at 298.15 K								
0.1061	1.9812	−0.3400	0.4036	0.9932	−0.6992	0.7007	0.5857	−0.4787
0.2070	1.5541	−0.5538	0.4999	0.8229	−0.6659	0.7972	0.5150	−0.3455
0.3058	1.2330	−0.6661	0.6059	0.6801	−0.5847	0.8971	0.4612	−0.1881
2-Methyl-1-chloropropane (1) + 2-Butanol (2) at 298.15 K								
0.1077	1.9458	−0.8155	0.4066	0.8335	−1.1475	0.7018	0.5343	−0.6760
0.2064	1.3806	−1.1230	0.5036	0.7002	−1.0276	0.8050	0.4815	−0.4594
0.3079	1.0463	−1.1924	0.6480	0.5685	−0.7822	0.8972	0.4478	−0.2524
2-Methyl-1-chloropropane (1) + 2-Methyl-1-propanol (2) at 298.15 K								
0.1154	2.3639	−0.6932	0.4017	1.0186	−1.1888	0.7057	0.5775	−0.7277
0.2046	1.7278	−1.0645	0.4943	0.8243	−1.1083	0.8002	0.5104	−0.5144
0.3086	1.2873	−1.1964	0.6102	0.6718	−0.9168	0.8978	0.4593	−0.2758
2-Methyl-1-chloropropane (1) + 2-Methyl-2-propanol (2) at 298.15 K								
0.0982	2.3447	−1.6709	0.4029	0.8424	−1.9624	0.6979	0.5388	−1.0936
0.2011	1.4771	−2.1296	0.5031	0.6988	−1.7078	0.7997	0.4863	−0.7415
0.3047	1.0652	−2.1298	0.6023	0.6040	−1.4083	0.8948	0.3827	−0.4672
2-Methyl-1-chloropropane (1) + 1-Butanol (2) at 313.15 K								
0.1061	1.3955	−0.2131	0.4036	0.7548	−0.4342	0.7007	0.4768	−0.2932
0.2070	1.1208	−0.3454	0.4999	0.6405	−0.4127	0.7972	0.4261	−0.2077
0.3058	0.9144	−0.4125	0.6059	0.5441	−0.3596	0.8971	0.3888	−0.1042
2-Methyl-1-chloropropane (1) + 2-Butanol (2) at 313.15 K								
0.1077	1.2539	−0.3821	0.4066	0.6299	−0.5746	0.7018	0.4357	−0.3427
0.2064	0.9644	−0.5291	0.5036	0.5461	−0.5184	0.8050	0.4015	−0.2279
0.3079	0.7565	−0.5905	0.6480	0.4591	−0.3969	0.8972	0.3778	−0.1185

2-Methyl-1-chloropropane (1) + 2-Methyl-1-propanol (2) at 313.15 K								
0.1154	1.5599	−0.3770	0.4017	0.7582	−0.6645	0.7057	0.4679	−0.4086
0.2046	1.1909	−0.5858	0.4943	0.6325	−0.6238	0.8002	0.4215	−0.2852
0.3086	0.9221	−0.6678	0.6102	0.5324	−0.5157	0.8978	0.3866	−0.1449
2-Methyl-1-chloropropane (1) + 2-Methyl-2-propanol (2) at 313.15 K								
0.0982	1.3590	−0.5376	0.4029	0.6303	−0.7430	0.6979	0.4370	−0.4297
0.2011	0.9661	−0.7538	0.5031	0.5423	−0.6590	0.7997	0.4053	−0.2865
0.3047	0.7571	−0.7849	0.6023	0.4810	−0.5499	0.8948	0.3751	−0.1534

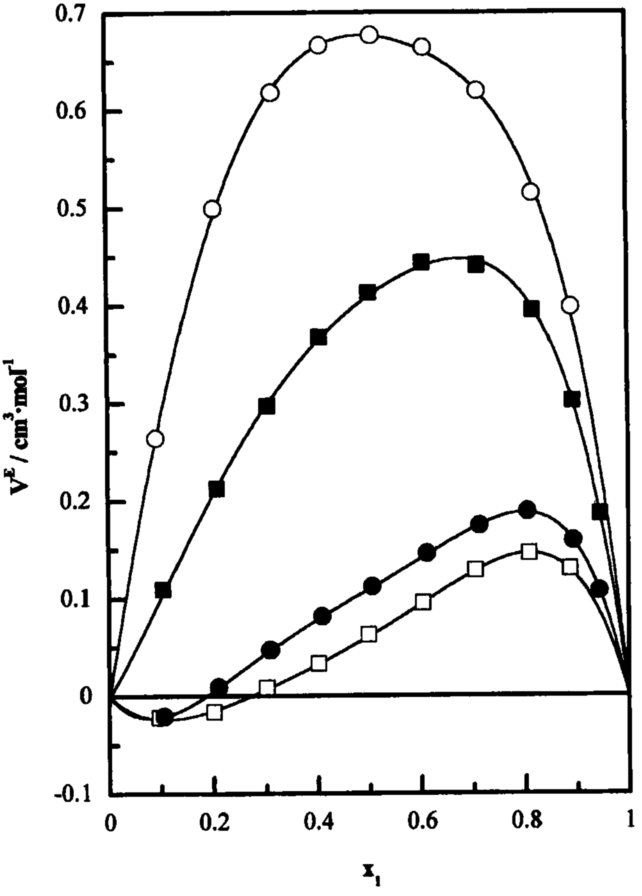

FIGURE 1 Excess volumes, V^E, of 2-methyl-1-chloropropane (1)+a butanol (2) at 298.15 K as a function of mole fraction x_1: 1-butanol (□); 2-butanol (■); 2-methyl-1-propanol (•); 2-methyl-2-propanol (○).

between alcohol monomers and also between the polar molecules of 2-methyl-1-chloropropane. In the mixture appear the specific interaction Cl–OH and also structural factors. The contribution of these effects is described in detail in Ref. [2].

The volumetric behaviour of the systems is notably different depending on the isomer of butanol involved. So, at 298.15 K, those systems containing secondary or tertiary butanol (2-butanol and 2-methyl-2-propanol, respectively) show positive V^E values in the whole

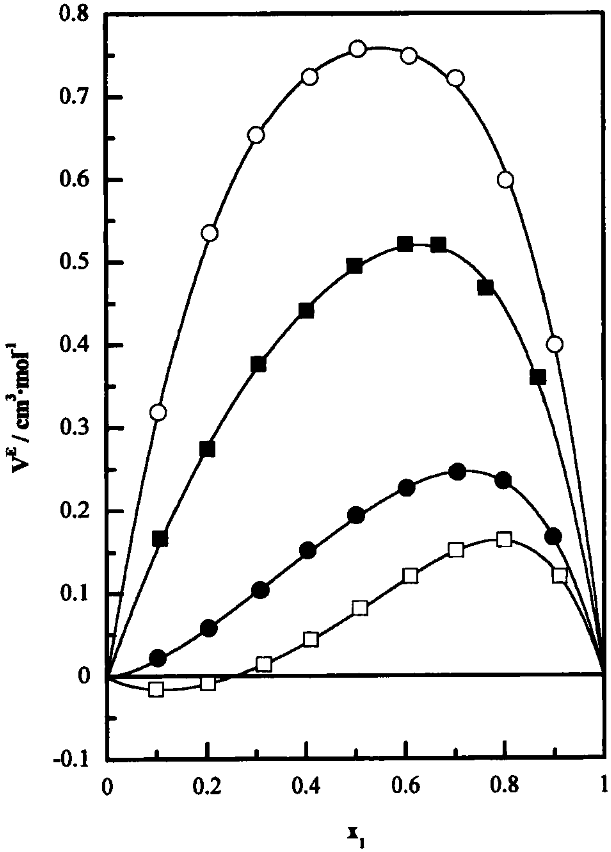

FIGURE 2 Excess volumes, V^E, of 2-methyl-1-chloropropane (1)+a butanol (2) at 313.15 K as a function of mole fraction x_1: 1-butanol (□); 2-butanol (■); 2-methyl-1-propanol (•); 2-methyl-2-propanol (o).

composition range, being the greatest the values corresponding to mixtures with 2-methyl-2-propanol. When the butanol involved is the primary one, sigmoidal shaped curves are obtained. The region of negative V^E is that of poor compositions in halogenated compound (up to $x = 0.2 - 0.3$). When temperature is 313.15 K all of the systems present higher values of the excess volume in such a way that the curve for the mixture 2-methyl-1-chloropropane+2-methyl-1-propanol becomes positive over the entire composition range. At both temperatures, the systems with the linear primary isomer show lower V^E than those with the branched one.

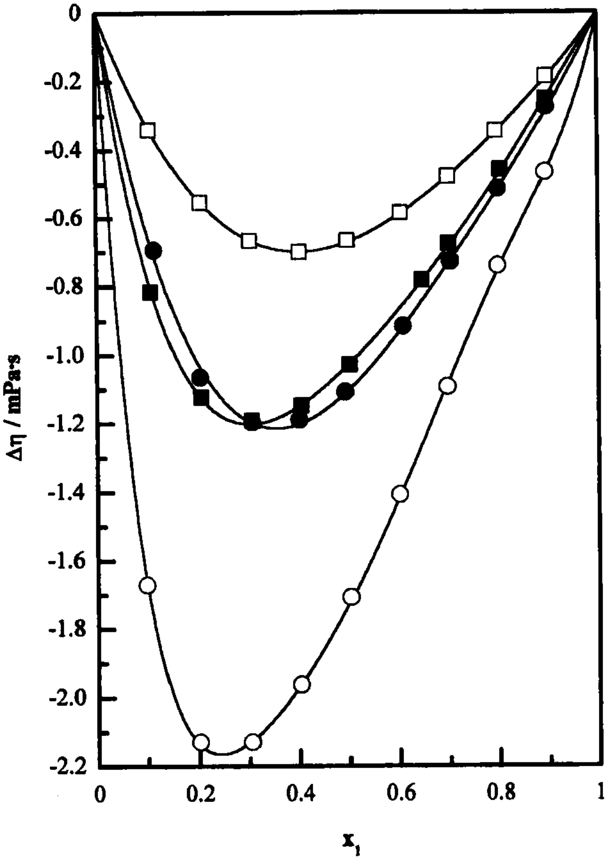

FIGURE 3 Viscosity deviations, $\Delta\eta$, of 2-methyl-1-chloropropane (1) + a butanol (2) at 298.15 K as a function of mole fraction x_1: 1-butanol (□); 2-butanol (■); 2-methyl-1-propanol (•); 2-methyl-2-propanol (o).

As to the $\Delta\eta$, they are negative over the whole composition range at both temperatures. The most negative values correspond to the system of 2-methyl-1-chloropropane with 2-methyl-2-propanol, and the least negative to those with 1-butanol. The mixtures with 2-butanol and 2-methyl-1-propanol at 298.15 K show very similar values of $\Delta\eta$, being the corresponding curves almost superposed. At 313.15 K the viscosity deviations are rather more negative for the branched alcoholic isomer. Anyway, an increase in the working temperature makes $\Delta\eta$ less negative, especially for the mixtures containing 2-methyl-2-propanol.

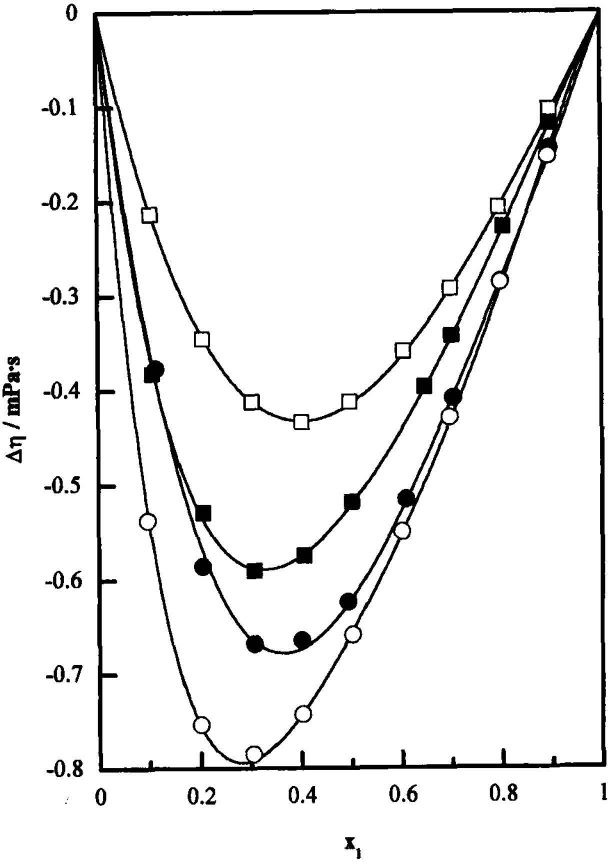

FIGURE 4 Viscosity deviations, $\Delta\eta$, of 2-methyl-1-chloropropane (1) + a butanol (2) at 313.15 K as a function of mole fraction x_1: 1-butanol (□); 2-butanol (■); 2-methyl-1-propanol (●); 2-methyl-2-propanol (○).

The volumetric behaviour of the systems here studied shows great similarity with the systems consisting of the same alcohols and 1-chlorobutane [1], not only in the sigmoidal shape of the curves of primary butanols but also in the values and positions of both maxima and minima. These values and positions are practically identical independently of the chlorinated isomer and the working temperature. When the halogenated derivative is 2-chlorobutane or 2-methyl-2-chloropropane [2, 3] the results obtained were lower, particularly for the tertiary halogenated isomer. In any case the V^E values for every

TABLE IV Coefficients, A_p, and standard deviations, $\sigma(Y^E)$, for Eq. (3) at indicated temperature

Function	A_0	A_1	A_2	A_3	A_4	σ
2-Methyl-1-chloropropane (1)+1-Butanol (2) at 298.15 K						
$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	0.247	0.620	0.460	0.604	–	0.001
$\Delta\eta/\mathrm{mPa \cdot s}$	−2.6745	1.1304	−0.2386	−0.2402	–	0.0017
2-Methyl-1-chloropropane (1)+2-Butanol (2) at 298.15 K						
$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	1.637	0.767	0.808	0.815	–	0.004
$\Delta\eta/\mathrm{mPa \cdot s}$	−4.1094	2.8750	−2.3573	1.2699	–	0.0064
2-Methyl-1-chloropropane (1)+2-Methyl-1-propanol (2) at 298.15 K						
$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	0.440	0.616	0.461	0.933	–	0.001
$\Delta\eta/\mathrm{mPa \cdot s}$	−4.4077	2.7921	−1.0956	−0.3905	–	0.0184
2-Methyl-1-chloropropane (1)+2-Methyl-2-propanol (2) at 298.15 K						
$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	2.708	−0.043	1.435	0.974	–	0.005
$\Delta\eta/\mathrm{mPa \cdot s}$	−6.8950	5.4973	−3.5526	4.8114	−6.5123	0.0100
2-Methyl-1-chloropropane (1)+1-Butanol (2) at 313.15 K						
$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	0.309	0.780	0.486	0.333	–	0.001
$\Delta\eta/\mathrm{mPa \cdot s}$	−1.6582	0.7028	−0.0845	−0.0061	–	0.0023
2-Methyl-1-chloropropane (1)+2-Butanol (2) at 313.15 K						
$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	1.974	0.779	0.768	0.282	–	0.005
$\Delta\eta/\mathrm{mPa \cdot s}$	−2.0854	1.3570	−0.8394	0.5221	–	0.0045
2-Methyl-1-chloropropane (1)+2-Methyl-1-propanol (2) at 313.15 K						
$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	0.765	0.812	0.403	0.290	–	0.002
$\Delta\eta/\mathrm{mPa \cdot s}$	−2.4833	1.5239	−0.4428	−0.1839	–	0.0110
2-Methyl-1-chloropropane (1)+2-Methyl-2-propanol (2) at 313.15 K						
$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	3.018	0.307	1.438	0.475	–	0.006
$\Delta\eta/\mathrm{mPa \cdot s}$	−2.6261	1.9419	−1.7797	1.3084	–	0.0061

isomer of chlorobutane follow the increasing sequence: 1-butanol < 2-methyl-1-propanol < 2-butanol < 2-methyl-2-propanol.

From the viewpoint of a defined alcoholic isomer, the situation is not so regular: whereas the mixtures containing 2-methyl-2-propanol show similar V^E curves with all the chlorinated isomers (at 298.15 K symmetric curves with a maximum in $0{,}68\,\mathrm{cm^3 \cdot mol^{-1}}$), the systems involving the remaining butanols present the greatest excess volumes when the chlorine atom is placed on a primary carbon and the lowest ones with the tertiary chlorinated isomer.

From all these data it can be concluded that the volumetric behaviour is governed by the nature of the carbon atom that carries the functional group (Cl, OH). This is mainly true for the butanol isomers because the more important effect in the mixing process is the breaking of their association. The remaining effects become prominent only when the values here reported are compared with those of systems butanol isomer + 1-bromobutane or 2-methyl-1-bromopropane [13, 14], in which the presence of bromine atom instead of chlorine one leads to a similar behaviour but reaching greater excess volumes (always positive).

On the other hand, the viscosimetric behaviour is very similar to that observed for the mixtures of the isomers of butanol with the remaining isomers of chlorobutane [2–4], and even with 2-methyl-1-bromobutane [14]. The most remarkable feature in all these systems is the similarity in the values for the mixtures containing 2-butanol and 2-methyl-1-propanol, especially at 298.15 K. In all these cases and according to the expectancy, the characteristic that determines the $\Delta\eta$ values is the nature of the alcoholic isomer: the breaking of the association in butanols allows an easier flow of the mixture, then $\Delta\eta$ shows always negative values. However, it seems that this property is not so influenced by the atom carrying the functional group as was V^E. Instead, the specific interactions and the interstitial accommodation produced in the mixing process acquire a major role, yielding both effects to a positive contribution to the viscosity deviations.

From the results it could be concluded that the specific interactions Cl–OH are easier with primary alcohols while the structural accommodation is favoured in the case of linear molecules. In this way it could be explained the fact that the most negative values were always obtained for the mixtures with 2-methyl-2-propanol and the least negative for the mixtures with 1-butanol. For the other two isomers of butanol, the effects are opposite and the system containing them shows very similar values. In the case of 2-butanol the major contribution would be the accommodation whereas the specific interaction Cl–OH would be the prevailing effect in the case of 2-methyl-1-propanol.

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