

# Viscosities of Binary Mixtures of Isomeric Butanols or Isomeric Chlorobutanes with 2-Methyltetrahydrofuran

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Viscosities for the binary mixtures of isomeric butanols or isomeric chlorobutanes with 2-methyltetrahydrofuran were determined at the temperatures 298.15 K and 313.15 K; the viscosities of the mixture of 2-methyl-2-chloropropane with 2-methyltetrahydrofuran were measured only at 298.15 K. The viscosity data were correlated by the McAllister equation. Viscosity deviations for the mixtures containing isomeric butanols are negative over the whole composition range. On the other hand, for the mixtures involving isomeric chlorobutanes, viscosity deviations are slightly negative, except for that for 1-chlorobutane; in this case viscosity deviations present an inversion of sign.

## 1. Introduction

Viscosity data of liquid mixtures are of great interest from both practical and theoretical points of view. Following our systematic study<sup>1–8</sup> on viscosity of liquid mixtures containing isomeric butanols or isomeric chlorobutanes, we present in this paper viscosity measurements of binary mixtures containing isomeric butanols or isomeric chlorobutanes and 2-methyltetrahydrofuran at 298.15 K and 313.15 K, except those for 2-chloro-2-methylpropane are measured only at 298.15 K due to its low normal boiling temperature. Viscosity data have been used to calculate the viscosity deviations. As far as we know there are no viscosity measurements on these systems.

## 2. Experimental Section

**Materials.** The liquids used were 1-butanol (>99.8 mol %), 2-methyl-1-propanol, 2-methyl-2-propanol, and 1-chlorobutane (>99.5 mol %), and 2-butanol, 2-chlorobutane, 2-methyl-2-chloropropane, and 2-methyltetrahydrofuran (>99 mol %) obtained from Aldrich, and 2-methyl-1-chloropropane (>99 mol %) provided by Fluka. All the substances were used without further purification, although before measurement butanols were dried over activated molecular sieve type 0.3 nm from Merck.

**Methods.** Kinematic viscosities,  $\nu$ , were determined using an Ubbelohde viscosimeter with a Schott-Geräte automatic measuring unit model AVS-440. The temperature was kept constant within  $\pm 0.01$  K by means of a Schott-Geräte thermostat. The viscosimeter was calibrated with deionized doubly distilled water. The uncertainty of flow time measurements was  $\pm 0.01$  s, and the corresponding uncertainty in the kinematic viscosity was  $\pm 1 \times 10^{-4}$  mm<sup>2</sup>·s<sup>-1</sup>. Kinetic energy corrections were applied to the experimental data.

Densities,  $\rho$ , required for converting kinematic viscosities to absolute viscosities ( $\eta = \rho\nu$ ), were measured using an Anton Paar DMA-58 vibrating tube densimeter in which temperature is controlled automatically within  $\pm 0.01$  K. The apparatus was calibrated with deionized twice distilled water and dry air. The precision of density measurements

**Table 1. Densities and Viscosities of Pure Compounds at 298.15 K and Comparison with Literature Data**

compound	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\eta/\text{mPa}\cdot\text{s}$	
	exptl	lit.	exptl	lit.
1-butanol	0.805 56	0.8060 <sup>a</sup>	2.5720	2.5710 <sup>a</sup>
2-butanol	0.802 25	0.8026 <sup>a</sup>	2.9960	2.998 <sup>a</sup>
2-methyl-1-propanol	0.797 72	0.7978 <sup>a</sup>	3.3890	3.333 <sup>a</sup>
2-methyl-2-propanol	0.780 68	0.7812 <sup>a</sup>	4.3660	4.438 <sup>a</sup>
1-chlorobutane	0.880 69	0.88095 <sup>a</sup>	0.4212	0.427 <sup>c</sup>
2-chlorobutane	0.867 37	0.8671 <sup>b</sup>	0.3935	
2-methyl-1-chloropropane	0.870 89	0.8719 <sup>b</sup>	0.4285	0.431 <sup>c</sup>
2-methyl-2-chloropropane	0.835 67	0.8362 <sup>b</sup>	0.4727	
2-methyltetrahydrofuran	0.849 27	0.84882 <sup>d</sup>	0.4750	0.4573 <sup>d</sup>

<sup>a</sup> Reference 9. <sup>b</sup> Reference 10. <sup>c</sup> Reference 11. <sup>d</sup> Reference 12.

is  $\pm 5 \times 10^{-6}$  g·cm<sup>-3</sup>, and the accuracy of these measurements after proper calibration is  $\pm 10^{-5}$  g·cm<sup>-3</sup>.

The mixtures were prepared using a Mettler H20T balance. The maximum estimated error in the mole fractions is  $\pm 1 \times 10^{-4}$ .

The pure compound properties (densities and absolute viscosities) at 298.15 K are shown together with literature data<sup>9–12</sup> in Table 1.

## 3. Results and Discussion

The kinematic,  $\nu$ , and absolute,  $\eta$ , viscosities together with the densities,  $\rho$ , for the binary mixtures are given in Table 2.

The kinematic viscosities were correlated by using the McAllister equation:<sup>13</sup>

$$\ln \nu = x_1^3 \ln \nu_1 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 - \ln[x_1 + x_2 M_2/M_1] + 3x_1^2 x_2 \ln[(2 + M_2/M_1)/3] + 3x_1 x_2^2 \ln[(1 + 2M_2/M_1)/3] + x_2^3 \ln(M_2/M_1) \quad (1)$$

where  $\nu$  refers to the kinematic viscosity of the mixture,  $\nu_1$  and  $\nu_2$  are the kinematic viscosities of components 1 and 2,  $M_1$  and  $M_2$  are the molecular masses of the components, and  $\nu_{12}$  and  $\nu_{21}$  are adjustable parameters characteristic of the system.

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**Table 2. Experimental Kinematic Viscosities,  $\nu$ , Densities,  $\rho$ , and Absolute Viscosities,  $\eta$ , for the Studied Mixtures**

$x_1$	$\nu/\text{mm}^2\cdot\text{s}$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$	$x_1$	$\nu/\text{mm}^2\cdot\text{s}$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$
1-Butanol (1) + 2-Methyltetrahydrofuran (2) at 298.15 K				1-Chlorobutane (1) + 2-Methyltetrahydrofuran (2) at 298.15 K			
0.0000	0.5593	0.849 28	0.4750	0.6036	1.2082	0.825 28	0.9971
0.0488	0.5913	0.847 62	0.5012	0.7034	1.4661	0.820 68	1.2032
0.1039	0.6216	0.845 56	0.5256	0.8042	1.8409	0.815 80	1.5018
0.2050	0.6927	0.841 78	0.5831	0.9060	2.4024	0.810 56	1.9473
0.3006	0.7699	0.838 03	0.6452	0.9582	2.7999	0.807 81	2.2618
0.4051	0.8806	0.833 86	0.7343	1.0000	3.1928	0.805 56	2.5720
0.5099	1.0508	0.829 37	0.8715				
2-Butanol (1) + 2-Methyltetrahydrofuran (2) at 298.15 K				2-Chlorobutane (1) + 2-Methyltetrahydrofuran (2) at 298.15 K			
0.0000	0.5593	0.849 28	0.4750	0.6018	1.1120	0.820 77	0.9127
0.0437	0.5664	0.847 10	0.4798	0.6999	1.3928	0.816 27	1.1369
0.0915	0.6070	0.844 81	0.5128	0.8077	1.8597	0.811 31	1.5088
0.1968	0.6452	0.839 74	0.5418	0.8708	2.2624	0.808 35	1.8288
0.3024	0.7162	0.834 82	0.5979	0.9361	2.8980	0.805 28	2.3337
0.3993	0.8061	0.830 17	0.6692	1.0000	3.7345	0.802 25	2.9960
0.5051	0.9497	0.825 31	0.7838				
2-Methyl-1-propanol (1) + 2-Methyltetrahydrofuran (2) at 298.15 K				2-Methyl-1-chloropropane (1) + 2-Methyltetrahydrofuran (2) at 298.15 K			
0.0000	0.5593	0.849 28	0.4750	0.5992	1.2569	0.820 51	1.0313
0.0397	0.5850	0.847 52	0.4958	0.7013	1.5836	0.815 04	1.2907
0.0951	0.6173	0.844 97	0.5216	0.7995	2.0619	0.809 54	1.6692
0.1999	0.6872	0.840 08	0.5773	0.9048	2.9259	0.803 45	2.3508
0.3008	0.7773	0.835 46	0.6494	0.9557	3.5460	0.800 39	2.8382
0.3982	0.8961	0.830 71	0.7444	1.0000	4.2484	0.797 71	3.3890
0.4988	1.0403	0.825 72	0.8590				
2-Methyl-2-propanol (1) + 2-Methyltetrahydrofuran (2) at 298.15 K				2-Methyl-2-chloropropane (1) + 2-Methyltetrahydrofuran (2) at 298.15 K			
0.0000	0.5593	0.849 28	0.4750	0.5918	1.2469	0.808 40	1.0080
0.0380	0.5808	0.846 42	0.4916	0.6972	1.6660	0.801 38	1.3351
0.0906	0.6072	0.842 39	0.5115	0.7952	2.1969	0.795 17	1.7469
0.1910	0.6720	0.835 27	0.5613	0.8989	3.3357	0.788 08	2.6288
0.3192	0.7785	0.826 46	0.6434	0.9532	4.3728	0.784 19	3.4291
0.3935	0.8656	0.821 40	0.7110	1.0000	5.5926	0.780 67	4.3660
0.4981	1.0312	0.814 49	0.8399				
1-Butanol (1) + 2-Methyltetrahydrofuran (2) at 313.15 K				1-Chlorobutane (1) + 2-Methyltetrahydrofuran (2) at 313.15 K			
0.0000	0.4916	0.834 01	0.4100	0.6036	0.9695	0.812 07	0.7873
0.0488	0.5106	0.832 35	0.4250	0.7034	1.1555	0.807 96	0.9336
0.1039	0.5345	0.830 50	0.4439	0.8042	1.3925	0.803 45	1.1188
0.2050	0.5881	0.827 07	0.4864	0.9060	1.7561	0.798 76	1.4027
0.3006	0.6515	0.823 64	0.5366	0.9562	1.9934	0.796 23	1.5872
0.4051	0.7339	0.820 00	0.6018	1.0000	2.2249	0.794 19	1.7670
0.5099	0.8412	0.815 98	0.6864				
2-Butanol (1) + 2-Methyltetrahydrofuran (2) at 313.15 K				2-Chlorobutane (1) + 2-Methyltetrahydrofuran (2) at 313.15 K			
0.0000	0.4916	0.834 01	0.4100	0.6018	0.8777	0.806 88	0.7082
0.0437	0.4909	0.831 74	0.4083	0.6999	1.0525	0.802 66	0.8448
0.0915	0.5061	0.829 48	0.4198	0.8077	1.3404	0.797 97	1.0696
0.1968	0.5543	0.824 64	0.4571	0.8708	1.5651	0.795 28	1.2447
0.3024	0.6045	0.820 02	0.4957	0.9361	1.8563	0.792 38	1.4709
0.3993	0.6705	0.815 81	0.5470	1.0000	2.2383	0.789 44	1.7670
0.5051	0.7712	0.811 20	0.6256				
2-Methyl-1-propanol (1) + 2-Methyltetrahydrofuran (2) at 313.15 K				2-Methyl-1-chloropropane (1) + 2-Methyltetrahydrofuran (2) at 313.15 K			
0.0000	0.4916	0.834 01	0.4100	0.5992	0.9964	0.807 01	0.8041
0.0397	0.5064	0.832 15	0.4214	0.7013	1.2155	0.801 97	0.9748
0.0951	0.5312	0.829 82	0.4408	0.7995	1.5132	0.796 99	1.2060
0.1999	0.5877	0.825 25	0.4850	0.9048	2.0220	0.791 39	1.6002
0.3008	0.6543	0.820 88	0.5371	0.9557	2.3621	0.788 49	1.8625
0.3982	0.7473	0.816 54	0.6102	1.0000	2.7383	0.785 89	2.1520
0.4988	0.8466	0.811 84	0.6873				
2-Methyl-2-propanol (1) + 2-Methyltetrahydrofuran (2) at 313.15 K				2-Methyl-2-chloropropane (1) + 2-Methyltetrahydrofuran (2) at 313.15 K			
0.0000	0.4916	0.834 01	0.4100	0.5918	0.9492	0.793 51	0.7532
0.0380	0.5036	0.831 02	0.4185	0.6972	1.1675	0.786 72	0.9185
0.0906	0.5271	0.826 98	0.4359	0.7952	1.4695	0.780 27	1.1466
0.1910	0.5726	0.819 77	0.4694	0.8989	1.9695	0.773 14	1.5227
0.3192	0.6489	0.811 22	0.5264	0.9532	2.3493	0.769 08	1.8068
0.3935	0.7058	0.806 46	0.5692	1.0000	2.7387	0.765 33	2.0960
0.4981	0.8153	0.799 58	0.6519				

The estimated parameters of the McAllister equation and the corresponding standard deviations for all the systems are shown in Table 3. As can be seen in this table, the equation of McAllister correlates very well the viscosity data; the poorest correlation is presented for the mixture 2-methyl-2-propanol with 2-methyltetrahydrofuran.

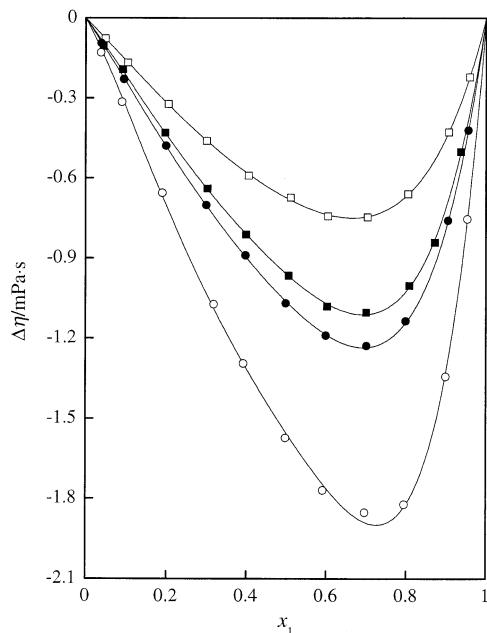
Viscosity deviations can be calculated from our measurements according to the following equation:

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \quad (2)$$

These viscosity deviations are graphically represented in Figures 1–4. The values of viscosity deviations have been fitted to a Redlich–Kister equation:

$$\Delta\eta = x_1(1 - x_1) \sum_{i=0}^{i=p-1} A_i(2x_1 - 1)^i \quad (3)$$

where  $A_i$  are adjustable parameters, obtained by the least-squares method. The values of these parameters are



**Figure 1.** Viscosity deviations,  $\Delta\eta$ , of an isomeric butanol + 2-methyltetrahydrofuran mixture at 298.15 K as a function of mole fraction of isomeric butanol: (□) 1-butanol; (■) 2-butanol; (●) 2-methyl-1-propanol; (○) 2-methyl-2-propanol; (---) Redlich–Kister equation.

**Table 3. Parameters and Standard Deviations for the McAllister Equation**

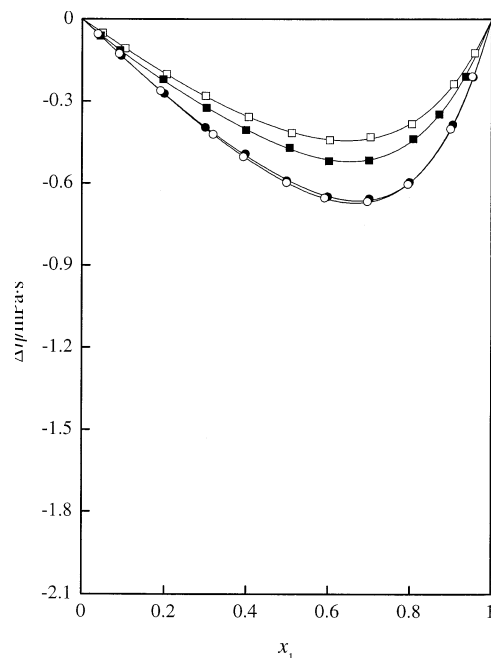
$T = 298.15 \text{ K}$			$T = 313.15 \text{ K}$		
$\nu_{12}$	$\nu_{21}$	$\sigma$	$\nu_{12}$	$\nu_{21}$	$\sigma$
1-Butanol + 2-Methyltetrahydrofuran					
1.0995	0.7868	0.0070	0.9178	0.6454	0.0036
2-Butanol + 2-Methyltetrahydrofuran					
0.9084	0.7122	0.0103	0.8141	0.5693	0.0072
2-Methyl-1-propanol + 2-Methyltetrahydrofuran					
1.0431	0.8021	0.0054	0.8817	0.6628	0.0055
2-Methyl-2-propanol + 2-Methyltetrahydrofuran					
0.9221	0.7958	0.0273	0.8458	0.6276	0.0053
1-Chlorobutane + 2-Methyltetrahydrofuran					
0.5019	0.5350	0.0001	0.4428	0.4709	0.0002
2-Chlorobutane + 2-Methyltetrahydrofuran					
0.4815	0.5194	0.0003	0.4269	0.4579	0.0001
2-Methyl-1-chloropropane + 2-Methyltetrahydrofuran					
0.5082	0.5337	0.0002	0.4480	0.4688	0.0001
2-Methyl-2-chloropropane + 2-Methyltetrahydrofuran					
0.5527	0.5512	0.0003			

collected in Table 4 together with the standard deviations,  $\sigma$ :

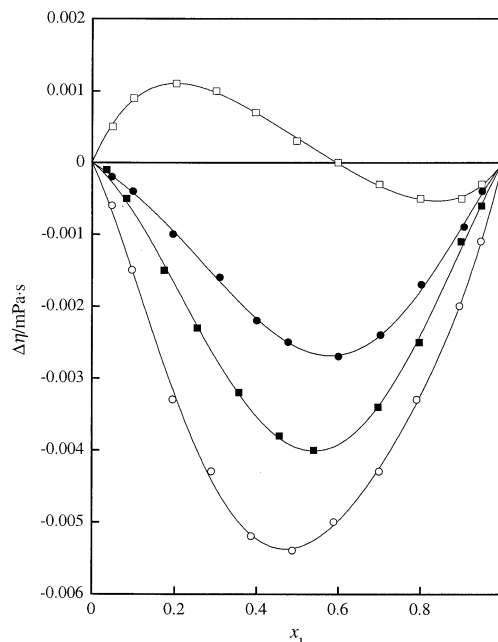
$$\sigma = \left[ \sum_{i=1}^{i=n} (\Delta\eta^{\text{exptl}} - \Delta\eta^{\text{cal}})^2 / (n - p) \right]^{1/2} \quad (4)$$

where  $n$  is the number of experimental data points and  $p$  is the number of adjustable parameters.

Viscosity deviations for the mixtures containing isomeric butanols present negative values at both the temperatures 298.15 K and 313.15 K.  $\Delta\eta$  becomes less negative following the sequence 2-methyl-2-propanol < 2-methyl-1-propanol < 2-butanol < 1-butanol. The minimum  $\Delta\eta$  values for all the mixtures appear in the alcohol-rich region, around  $x_1 = 0.7$ . A comparison of viscosity behavior with temperature



**Figure 2.** Viscosity deviations,  $\Delta\eta$ , of an isomeric butanol + 2-methyltetrahydrofuran mixture at 313.15 K as a function of mole fraction of isomeric butanol: (□) 1-butanol; (■) 2-butanol; (●) 2-methyl-1-propanol; (○) 2-methyl-2-propanol; (---) Redlich–Kister equation.



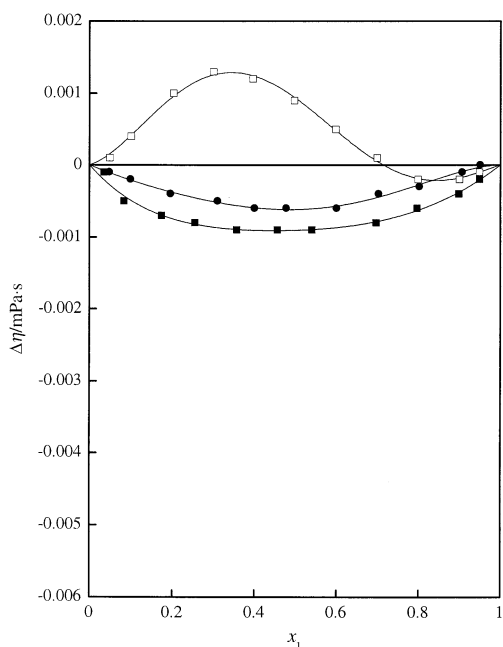
**Figure 3.** Viscosity deviations,  $\Delta\eta$ , of an isomeric chlorobutane + 2-methyltetrahydrofuran mixture at 298.15 K as a function of mole fraction of isomeric chlorobutane: (□) 1-chlorobutane; (■) 2-chlorobutane; (●) 2-methyl-1-chloropropane; (○) 2-methyl-2-chloropropane; (---) Redlich–Kister equation.

indicates that  $\Delta\eta$  values decrease in absolute value with temperature; this effect is more remarkable for the mixture containing 2-methyl-2-propanol.

With respect to the mixtures containing isomeric chlorobutanes, it could be pointed out that  $\Delta\eta$  values are very small. Viscosity deviations for the mixture 1-chlorobutane + 2-methyltetrahydrofuran display sigmoid curves, and there is an inversion of sign from positive to negative values at  $x_1 = 0.6$  at 298.15 K and at  $x_1 = 0.7$  at 313.15 K. For the

Table 4. Parameters and Standard Deviations for Equation 3

system	$A_0$	$A_1$	$A_2$	$A_3$	$\sigma$
$T = 298.15 \text{ K}$					
1-butanol	-2.7000	-1.6147	-1.0483	-0.5923	0.0051
2-butanol	-3.8386	-2.6348	-2.0158	-1.1657	0.0088
2-methyl-1-propanol	-4.2549	-2.9506	-2.2512	-1.3061	0.0057
2-methyl-2-propanol	-6.2244	-4.5454	-4.6077	-3.5284	0.0230
1-chlorobutane	0.0013	-0.0072	0.0015	-0.0034	0.0000
2-chlorobutane	-0.0159	-0.0040	0.0086	0.0011	0.0001
2-methyl-1-chloropropane	-0.0103	-0.0054	0.0046	0.0033	0.0000
2-methyl-2-chloropropane + 2-methyltetrahydrofuran	-0.0214	0.0030	0.0034	-0.0085	0.0001
$T = 313.15 \text{ K}$					
1-butanol	-1.6343	-0.8839	-0.4977	-0.2221	0.0029
2-butanol	-1.8851	-1.1487	-0.6454	-0.0264	0.0047
2-methyl-1-propanol	-2.3506	-1.5163	-1.0264	-0.4519	0.0037
2-methyl-2-propanol	-2.3962	-1.5373	-0.9166	-0.3774	0.0026
1-chlorobutane	0.0037	-0.0082	-0.0038	0.0064	0.0000
2-chlorobutane	-0.0036	0.0004	-0.0018	0.0007	0.0001
2-methyl-1-chloropropane + 2-methyltetrahydrofuran	-0.0025	0.0002	0.0011	0.0009	0.0000



**Figure 4.** Viscosity deviations,  $\Delta\eta$ , of an isomeric chlorobutane + 2-methyltetrahydrofuran mixture at 313.15 K as a function of mole fraction of isomeric chlorobutane: (□) 1-chlorobutane; (■) 2-chlorobutane; (●) 2-methyl-1-chloropropane; (—) Redlich–Kister equation.

rest of the isomeric chlorobutanes,  $\Delta\eta$  values are negative over the whole composition range. At 298.15 K,  $\Delta\eta$  decreases in the sequence 1-chlorobutane > 2-methyl-1-chloropropane > 2-chlorobutane > 2-methyl-2-chloropropane; at 313.15 K, the sequence is 1-chlorobutane > 2-methyl-1-chloropropane > 2-chlorobutane. On the other hand, the  $\Delta\eta$  change, in absolute value, with temperature is small.

For the mixtures containing isomeric butanols, the values are clearly negative, and it is known that the viscosity behavior depends, among other factors, on the strength of the interactions between like and unlike molecules. So, in this case, we could conclude that the interaction between the oxygen atom of the ether and the hydroxyl group of the butanols is not enough to compensate the weakening of dipole–dipole interactions in the 2-methyltetrahydrofuran and the breaking of hydrogen bonds in the butanols. The less negative  $\Delta\eta$  values are presented

by the mixture 1-butanol + 2-methyltetrahydrofuran, and the more negative ones, by the mixture 2-methyl-2-propanol + 2-methyltetrahydrofuran; this fact indicates that the  $\Delta\eta$  values for these mixtures are closely related with the associated structure of the considered butanol.

For the mixtures containing isomeric chlorobutanes, the weakening of the dipole–dipole interactions in the pure compounds after the mixing process is outweighed by the Cl–O interaction, leading to small viscosity deviations. Comparing the results for both kinds of mixtures and taking into account previous works,<sup>1–8</sup> we can conclude that the viscosimetric behavior is less influenced by the size and shape of the mixing compounds than by the existence of strong interactions between the compounds.

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