Densities and Viscosities of the Binary Mixtures of Tetrahydrofuran with Isomeric Chlorobutanes at 298.15 K and 313.15 K

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Densities and kinematic viscosities of the binary mixtures tetrahydrofuran + isomeric chlorobutanes have been measured at atmospheric pressure at the temperatures of 298.15 K and 313.15 K. Excess molar volumes, dynamic viscosities, and viscosity deviations have been obtained from experimental data and fitted by the Redlich−Kister equation. Kinematic viscosities have been correlated using the McAllister equation. Excess molar volumes are negative in the whole composition range, while viscosity deviations are small in absolute value and present an inversion of sign for two systems only at one temperature.

Introduction

Our research group has been involved in the last few years in the determination of thermodynamic and transport properties of binary mixtures containing a cyclic ether and a halogenated compound.[1−6] The main objective of this research is to obtain experimental data of relevant properties of these kind of mixtures, which are correlated using different models and equations, and also to understand better the phenomena taking place in the liquid mixtures, especially the role of molecular interactions established between the different components.

In this paper we report densities and viscosities of the binary mixtures tetrahydrofuran + isomers of chlorobutane at atmospheric pressure and the temperatures of 298.15 K and 313.15 K. Kinematic viscosities have been correlated with the McAllister equation. Experimental data have been used to calculate excess molar volumes and viscosity devitions and then fitted with the Redlich−Kister equation. Results have been compared with those previously obtained for the binary mixtures 2-methyltetrahydrofuran + isomers of chlorobutane.

To the best of our knowledge, we only have found a previous reference for the excess molar volumes and viscosity deviations of the binary system tetrahydrofuran + 1-chlorobutane at 298.15 K and 313.15 K.[7]

Experimental Section

Materials. The compounds used were tetrahydrofuran and 1-chlorobutane (> 99.5 %), 2-chlorobutane and 2-methyl-2-chloropropane (> 99 %) obtained from Aldrich, and 2-methyl-1-chloropropane (> 99 %) provided by Fluka. The purities of these compounds were checked by comparing the measured densities with those reported in the literature and also by a cromatographic method, using a semi-capillary methyl silicone column (o.d. 0.530 nm) and a flame ion detector, confirming the absence of other significant compounds, so no further purification was attempted.

Methods. Densities, ρ, of the pure compounds and the binary 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 precision of density measurements is $\pm 5 \times 10^{-3}$ kg·m^{-3}, and the uncertainty of these measurements is $\pm 1 \times 10^{-2}$ kg·m^{-3}.

Kinematic viscosities, ν, were determined using an Ubbelohde viscosimeter (i.d. = 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. 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.[8] After calibration, the estimated uncertainty for dynamic viscosity measurements is ± 0.002 mPa·s.

The compositions of the mixtures are given in mole fraction and were determined by mass using a Sartorius semi-micro balance with a precision of $\pm 10^{-5}$ g. The possible uncertainty in the mole fractions is estimated to be less than 10^{-4}. The pure compounds properties at 298.15 K and 313.15 K, along with literature values at 298.15 K,[9−11] are given in Table 1.

Results and Discussion

The experimental densities, ρ, and calculated excess molar volumes, V^E, of the binary mixtures tetrahydrofuran + isomers of chlorobutane at the temperatures of 298.15 K and 313.15 K are shown in Table 2. Experimental kinematic viscosities, ν, together with calculated dynamic viscosities, η, and viscosity deviations, $\Delta\eta$, of the same binary mixtures at both temperatures are given in Table 3. Viscosity deviations were calculated using the following expression:

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \quad (1)$$

where η is the dynamic viscosity of the mixture, x_i is the mole fraction of component i, and η_i is the dynamic viscosity of pure component i.

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Table 1. Experimental Densities (ρ) and Dynamic Viscosities (η) of Pure Components and Comparison at 298.15 K with Literature Data

	$T = 298.15$ K				$T = 313.15$ K	
	ρ/kg·m^{-3}		η/mPa·s		ρ/kg·m^{-3}	η/mPa·s
component	expt	lit	expt	lit	expt	expt
tetrahydrofuran	881.95	881.97[a]	0.4631	0.461[b]	865.36	0.3979
1-chlorobutane	880.69	880.4[a]	0.4212	0.427[c]	863.82	0.3640
2-chlorobutane	867.37	867.1[a]	0.3938		850.13	0.3371
2-methyl-1-chloropropane	871.13	871.7[a]	0.4295	0.431[c]	853.31	0.3664
2-methyl-2-chloropropane	836.45	836.1[a]	0.4751		818.31	0.3907

[a] Ref 9. [b] Ref 10. [c] Ref 11.

Table 2. Experimental Densities (ρ) and Excess Molar Volumes (V^E) for the Binary Mixtures Tetrahydrofuran (1) + Chlorobutane (2)

$T = 298.15$ K			$T = 313.15$ K		
	ρ	$V^E \times 10^6$		ρ	$V^E \times 10^6$
x_1	kg·m^{-3}	m^3·mol^{-1}	x_1	kg·m^{-3}	m^3·mol^{-1}
Tetrahydrofuran (1) + 1-Chlorobutane (2)					
0.0405	880.75	−0.003	0.0429	864.02	−0.018
0.1083	880.88	−0.010	0.1051	864.26	−0.038
0.2061	881.13	−0.026	0.2057	864.56	−0.057
0.2987	881.35	−0.039	0.3023	864.79	−0.067
0.4018	881.57	−0.049	0.4021	865.00	−0.073
0.4966	881.74	−0.053	0.4974	865.17	−0.075
0.5960	881.86	−0.051	0.5950	865.31	−0.072
0.7018	881.94	−0.044	0.7055	865.43	−0.063
0.7991	881.98	−0.033	0.8024	865.48	−0.050
0.9022	881.98	−0.018	0.9022	865.47	−0.030
0.9489	881.97	−0.010	0.9472	865.44	−0.018
Tetrahydrofuran (1) + 2-Chlorobutane (2)					
0.0348	867.97	−0.026	0.0524	851.13	−0.048
0.0902	868.90	−0.060	0.0985	851.96	−0.082
0.1820	870.40	−0.106	0.1977	853.67	−0.137
0.2532	871.55	−0.135	0.2931	855.24	−0.171
0.3718	873.44	−0.170	0.3996	856.96	−0.195
0.4603	874.85	−0.187	0.5015	858.57	−0.203
0.5819	876.71	−0.191	0.6054	860.16	−0.196
0.6932	878.30	−0.172	0.6994	861.54	−0.175
0.7825	879.43	−0.135	0.8119	863.09	−0.130
0.9161	881.04	−0.061	0.9006	864.24	−0.079
0.9512	881.38	−0.033	0.9563	864.89	−0.038
Tetrahydrofuran (1) + 2-Methyl-1-chloropropane (2)					
0.0571	871.87	−0.032	0.0562	854.45	−0.053
0.1071	872.53	−0.058	0.1267	855.53	−0.102
0.1884	873.57	−0.093	0.2163	856.77	−0.143
0.2881	874.78	−0.122	0.3482	858.48	−0.177
0.4158	876.28	−0.145	0.4419	859.64	−0.187
0.4933	877.18	−0.152	0.5439	860.87	−0.186
0.6022	878.39	−0.150	0.6661	862.25	−0.167
0.7032	879.48	−0.138	0.7494	863.13	−0.142
0.8064	880.49	−0.109	0.8325	863.96	−0.107
0.8983	881.30	−0.070	0.9104	864.68	−0.066
0.9538	881.69	−0.036	0.9613	865.08	−0.030
Tetrahydrofuran (1) + 2-Methyl-2-chloropropane (2)					
0.0526	838.96	−0.093	0.0559	821.08	−0.109
0.1013	841.30	−0.172	0.1219	824.43	−0.233
0.1967	845.97	−0.317	0.2311	829.82	−0.382
0.3170	851.56	−0.416	0.3509	835.82	−0.505
0.4131	856.07	−0.468	0.4172	839.02	−0.536
0.5061	860.43	−0.488	0.5110	843.50	−0.548
0.6033	864.82	−0.462	0.5950	847.38	−0.519
0.7087	869.50	−0.396	0.7018	852.40	−0.460
0.8132	874.03	−0.286	0.8044	856.93	−0.337
0.9031	877.92	−0.168	0.9004	861.05	−0.184
0.9545	880.08	−0.083	0.9545	863.37	−0.085

Excess molar volumes and viscosity deviations of the binary mixtures were fitted using a Redlich−Kister equation:

$$Y = x_1 x_2 \sum_{p=0}^{n-1} A_p (x_1 - x_2)^p \qquad (2)$$

Table 3. Experimental Kinematic Viscosities (ν), Dynamic Viscosities (η), and Viscosity Deviations ($\Delta\eta$) for the Binary Mixtures Tetrahydrofuran (1) + Isomeric Chlorobutanes (2)

$T = 298.15$ K				$T = 313.15$ K			
	ν	η	$\Delta\eta$		ν	η	$\Delta\eta$
x_1	mm^2·s^{-1}	mPa·s	mPa·s	x_1	mm^2·s^{-1}	mPa·s	mPa·s
Tetrahydrofuran (1) + 1-Chlorobutane (2)							
0.0524	0.4804	0.4231	−0.0003	0.0524	0.4232	0.3656	−0.0001
0.1080	0.4825	0.4250	−0.0007	0.1080	0.4252	0.3675	−0.0002
0.2063	0.4865	0.4287	−0.0011	0.2063	0.4287	0.3706	−0.0004
0.2991	0.4906	0.4324	−0.0014	0.2991	0.4321	0.3737	−0.0005
0.4088	0.4953	0.4367	−0.0017	0.4088	0.4361	0.3772	−0.0006
0.5070	0.4999	0.4408	−0.0017	0.5070	0.4397	0.3804	−0.0008
0.6074	0.5049	0.4453	−0.0014	0.6074	0.4435	0.3838	−0.0008
0.7001	0.5097	0.4495	−0.0011	0.7001	0.4471	0.3870	−0.0008
0.8054	0.5155	0.4546	−0.0003	0.8054	0.4514	0.3907	−0.0006
0.8910	0.5201	0.4587	0.0001	0.8910	0.4551	0.3939	−0.0003
0.9528	0.5230	0.4613	0.0002	0.9528	0.4577	0.3961	−0.0001
Tetrahydrofuran (1) + 2-Chlorobutane (2)							
0.0544	0.4569	0.3967	−0.0009	0.0544	0.3994	0.3400	−0.0004
0.1110	0.4600	0.3999	−0.0016	0.1110	0.4025	0.3430	−0.0008
0.2078	0.4659	0.4057	−0.0025	0.2078	0.4082	0.3485	−0.0012
0.2947	0.4712	0.4110	−0.0032	0.2947	0.4132	0.3534	−0.0016
0.3996	0.4779	0.4176	−0.0039	0.3996	0.4195	0.3595	−0.0019
0.5075	0.4849	0.4246	−0.0044	0.5075	0.4261	0.3659	−0.0020
0.6041	0.4917	0.4313	−0.0044	0.6041	0.4322	0.3718	−0.0020
0.7055	0.4994	0.4387	−0.0040	0.7055	0.4388	0.3780	−0.0019
0.8076	0.5079	0.4469	−0.0029	0.8076	0.4457	0.3847	−0.0015
0.9065	0.5166	0.4551	−0.0015	0.9065	0.4528	0.3913	−0.0008
0.9544	0.5210	0.4592	−0.0008	0.9544	0.4564	0.3947	−0.0004
Tetrahydrofuran (1) + 2-Methyl-1-chloropropane (2)							
0.0537	0.4944	0.4311	−0.0002	0.0537	0.4307	0.3680	−0.0001
0.1037	0.4957	0.4325	−0.0005	0.1037	0.4320	0.3694	−0.0002
0.2081	0.4981	0.4353	−0.0012	0.2081	0.4346	0.3723	−0.0006
0.3079	0.5005	0.4379	−0.0019	0.3079	0.4372	0.3751	−0.0009
0.4100	0.5031	0.4409	−0.0024	0.4100	0.4400	0.3781	−0.0012
0.5085	0.5063	0.4442	−0.0024	0.5085	0.4428	0.3810	−0.0013
0.5964	0.5091	0.4472	−0.0024	0.5964	0.4454	0.3837	−0.0015
0.7050	0.5130	0.4512	−0.0020	0.7050	0.4487	0.3871	−0.0014
0.8051	0.5168	0.4550	−0.0015	0.8051	0.4520	0.3904	−0.0013
0.8989	0.5207	0.4589	−0.0009	0.8989	0.4554	0.3938	−0.0009
0.9510	0.5229	0.4610	−0.0005	0.9510	0.4575	0.3957	−0.0006
Tetrahydrofuran (1) + 2-Methyl-2-chloropropane (2)							
0.0634	0.5649	0.4743	−0.0001	0.0557	0.4766	0.3913	0.0002
0.1288	0.5617	0.4733	−0.0002	0.0959	0.4759	0.3917	0.0003
0.2423	0.5563	0.4718	−0.0004	0.1963	0.4743	0.3928	0.0006
0.3198	0.5524	0.4705	−0.0008	0.2960	0.4724	0.3935	0.0007
0.4555	0.5459	0.4684	−0.0012	0.3892	0.4704	0.3940	0.0005
0.5341	0.5423	0.4673	−0.0015	0.5210	0.4674	0.3945	0.0000
0.6598	0.5368	0.4656	−0.0016	0.5951	0.4656	0.3946	−0.0004
0.7441	0.5335	0.4647	−0.0015	0.6368	0.4647	0.3947	−0.0006
0.8407	0.5300	0.4639	−0.0011	0.8089	0.4610	0.3951	−0.0014
0.8916	0.5284	0.4636	−0.0008	0.9009	0.4598	0.3959	−0.0012
0.9610	0.5263	0.4633	−0.0003	0.9526	0.4596	0.3968	−0.0007

where Y is V^E or $\Delta\eta$ of the binary mixture, A_p are adjustable parameters, obtained by the least-squares method, and n is the number of parameters. The values of these parameters are given in Table 4 together with the corresponding standard deviations, σ.

Table 4. Coefficients of the Redlich−Kister Equation and the Corresponding Standard Deviations (σ) for Excess Molar Volumes and Viscosity Deviations

property	T/K	A_0	A_1	A_2	A_3	$\sigma(Y)$
		Tetrahydrofuran (1) + 1-Chlorobutane (2)				
$V^E \times 10^6$/m^3·mol^{-1}	298.15	−0.212	−0.018	0.089	−0.070	0.000
	313.15	−0.296	0.014	−0.111	0.042	0.001
$\Delta\eta$/mPa·s	298.15	−0.0068	0.0015	0.0065	0.0059	0.0001
	313.15	−0.0031	−0.0022	0.0004	0.0025	0.0000
		Tetrahydrofuran (1) + 2-Chlorobutane (2)				
$V^E \times 10^6$/m^3·mol^{-1}	298.15	−0.763	−0.138	0.019	0.174	0.001
	313.15	−0.809	−0.020	−0.138	0.073	0.001
$\Delta\eta$/mPa·s	298.15	−0.0174	−0.0057	0.0005	0.0073	0.0000
	313.15	−0.0081	−0.0020	−0.0011	0.0012	0.0000
		Tetrahydrofuran (1) + 2-Methyl-1-chloropropane (2)				
$V^E \times 10^6$/m^3·mol^{-1}	298.15	−0.607	−0.066	−0.122	−0.050	0.001
	313.15	−0.749	0.048	−0.181	0.077	0.001
$\Delta\eta$/mPa·s	298.15	−0.0099	−0.0003	0.0037	−0.0043	0.0001
	313.15	−0.0054	−0.0027	−0.0017	−0.0037	0.0000
		Tetrahydrofuran (1) + 2-Methyl-2-chloropropane (2)				
$V^E \times 10^6$/m^3·mol^{-1}	298.15	−1.937	0.044	0.034	−0.035	0.005
	313.15	−2.199	0.023	0.142	0.026	0.006
$\Delta\eta$/mPa·s	298.15	−0.0055	−0.0059	0.0010	0.0028	0.0000
	313.15	0.0006	−0.0096	−0.0084	−0.0019	0.0000

Table 5. Parameters and Root Mean Square Deviations (RMSD) for the McAllister Equation

T = 298.15 K			T = 313.15 K		
ν_{12} mm^2·s^{-1}	ν_{21} mm^2·s^{-1}	RMSD	ν_{12} mm^2·s^{-1}	ν_{21} mm^2·s^{-1}	RMSD
Tetrahydrofuran (1) + 1-Chlorobutane (2)					
0.5108	0.4917	0.0003	0.4470	0.4345	<0.0001
Tetrahydrofuran (1) + 2-Chlorobutane (2)					
0.4961	0.4739	0.0001	0.4370	0.4165	0.0001
Tetrahydrofuran (1) + 2-Methyl-1-chloropropane (2)					
0.5118	0.5020	0.0001	0.4473	0.4396	0.0001
Tetrahydrofuran (1) + 2-Methyl-2-chloropropane (2)					
0.5362	0.5539	0.0001	0.4623	0.4756	0.0003

The kinematic viscosities of the binary mixtures were correlated using the McAllister equation:[12]

$$\ln(\nu/\text{mm}^2\cdot\text{s}^{-1}) = x_1^3 \ln \nu_1 + x_2^3 \ln \nu_2 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} - \ln[x_1 + x_2 M_2/M_1] + 3x_1^2 x_2 \ln[(2 + M_2/M_1)/3] + 3x_1 x_2^2 \ln[(1 + 2M_2/M_1)/3] + x_2^3 \ln(M_2/M_1) \quad (4)$$

where ν is the kinematic viscosity of the binary mixture, ν_1 and ν_2 are the kinematic viscosities of the pure components, M_1 and M_2 are the molecular masses of the components, and ν_{12} and ν_{21} are adjustable parameters characteristic of the system.

The estimated parameters of the McAllister equation and the corresponding root mean square deviations (RMSD) for all the systems are shown in Table 5. As one can see in this table, the McAllister equation correlates very well the viscosity data of the binary systems.

Excess molar volumes of the binary mixtures have been plotted in Figures 1 and 2. V^E values are negative for all the mixtures in the whole composition range at both temperatures, 298.15 K and 313.15 K. The more negative V^E values correspond to the binary mixture containing 2-methyl-2-chloropropane, and V^E increases in the sequence 2-chlorobutane < 2-methyl-1-chloropropane < 1-chlorobutane. The minimum V^E values for all the mixtures appear near the equimolecular composition. A comparison of both Figures 1 and 2 allow the observation that V^E values become slightly more negative (bigger absolute values) when temperature increases from 298.15 K to 313.15 K. It is also remarkable that V^E values are very close for the binary mixtures containing 2-chlorobutane or 2-methyl-1-chloropropane. V^E values reported here are very similar to those previously obtained for the binary mixtures 2-methyl-tetrahydrofuran + isomers of chlorobutane[4] and follow the same sequence. We also observe a good agreement between our values and those previously reported by Postigo et al.[7] (the average absolute deviation in densities is 0.44 kg·m^{-3}).

Viscosity deviations of the binary mixtures have been plotted in Figures 3 and 4. It could be pointed out that $\Delta\eta$ values are very small in absolute value, which is good agreement with previously reported $\Delta\eta$ values for the binary mixtures 2-methyl-tetrahydrofuran + isomers of chlorobutane[1]. $\Delta\eta$ curves at 298.15 K are sigmoid for the binary systems containing 1-chlorobutane or 2-methyl-2-chloropropane, and there is an inversion in sign

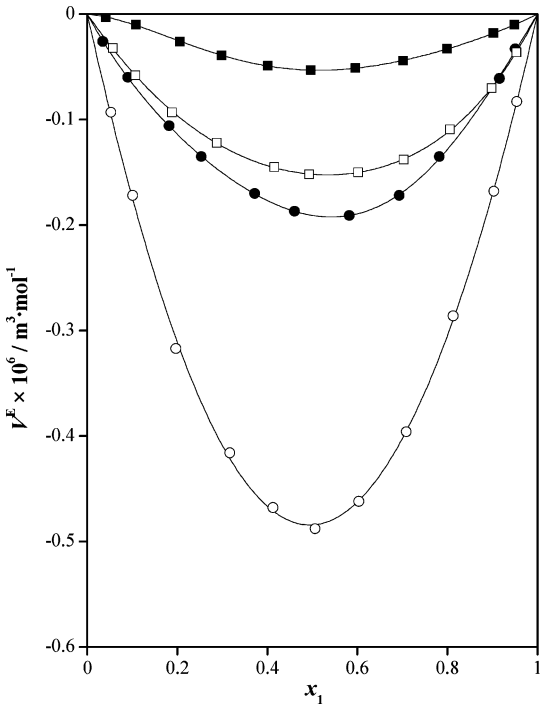

Figure 1. Excess molar volumes, V^E, for the binary mixtures tetrahydrofuran (1) + isomers of chlorobutane (2) at 298.15 K: ■, 1-chlorobutane; ●, 2-chlorobutane; □, 2-methyl-1-chloropropane; ○, 2-methyl-2-chloropropane.

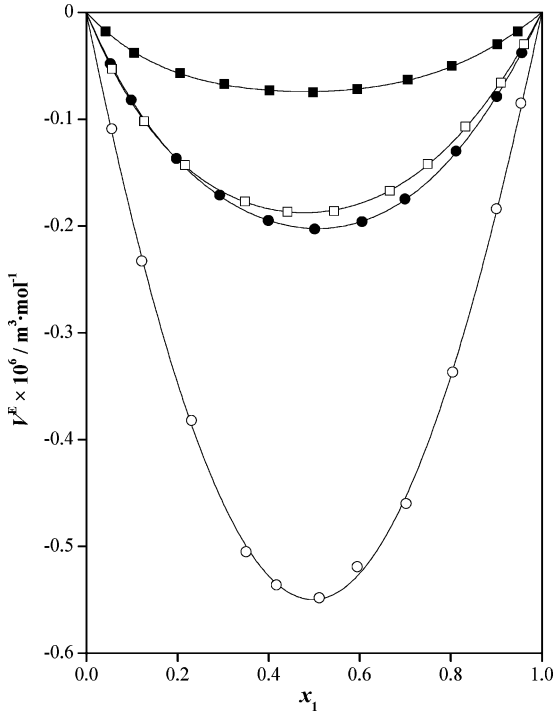

Figure 2. Excess molar volumes, V^E, for the binary mixtures tetrahydrofuran (1) + isomers of chlorobutane (2) at 313.15 K: ■, 1-chlorobutane; ●, 2-chlorobutane; □, 2-methyl-1-chloropropane; ○, 2-methyl-2-chloropropane.

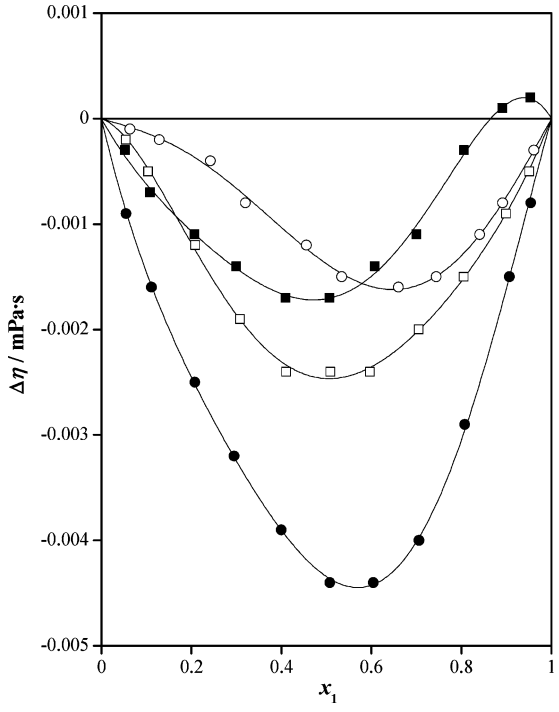

Figure 3. Viscosity deviations, $\Delta\eta$, for the binary mixtures tetrahydrofuran (1) + isomers of chlorobutane (2) at 298.15 K: ■, 1-chlorobutane; ●, 2-chlorobutane; □, 2-methyl-1-chloropropane; ○, 2-methyl-2-chloropropane.

in the system containing 1-chlorobutane from negative to positive values at $x_1 = 0.85$. However, only the binary system with 2-methyl-2-chloropropane shows a sigmoid curve at 313.15 K. In this system the inversion in sign takes place from positive to negative values at $x_1 = 0.52$. For the rest of binary mixtures, $\Delta\eta$ values are negative in the whole composition range. At both temperatures, $\Delta\eta$ values decrease in the sequence: 1-chlorobutane > 2-methyl-1-chloropropane > 2-chlorobutane. A comparison of viscosity behavior with temperature indicates that $\Delta\eta$ values decrease in absolute value with temperature, except for the mixture containing 2-methyl-2-chloropropane. Previously reported data for the binary system tetrahydrofuran + 1-chlorobutane[7] also show small viscosity deviations (the average absolute deviation in dynamic viscosities is 0.01 mPa·s).

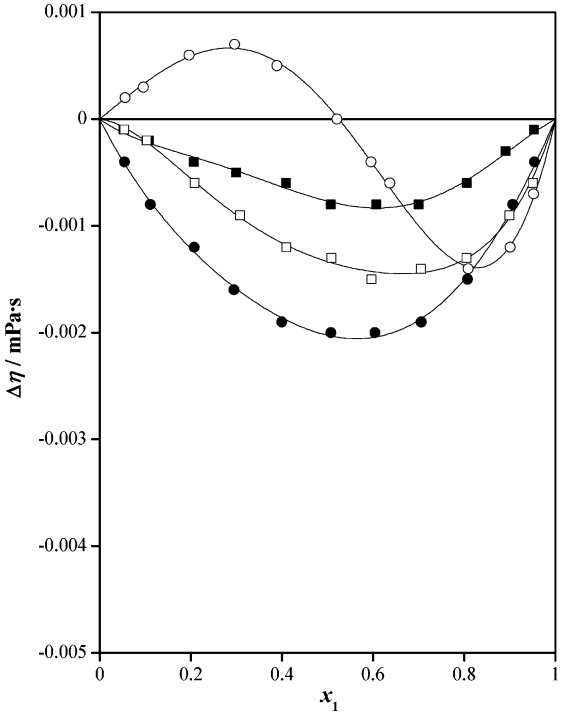

Figure 4. Viscosity deviations, $\Delta\eta$, for the binary mixtures tetrahydrofuran (1) + isomers of chlorobutane (2) at 313.15 K: ■, 1-chlorobutane; ●, 2-chlorobutane; □, 2-methyl-1-chloropropane; ○, 2-methyl-2-chloropropane.

Negative excess molar volumes reveal the existence of a specific interaction between unlike molecules.[13] This interaction takes place between a lone pair of electrons of the oxygen atom of the cyclic ether and the Cl atom of the halogenated compound.[14] It is also known that viscosity deviations depend, among other factors, on the strength of the interactions between like and unlike molecules. Small viscosity deviations obtained for the binary systems reveal that unlike interactions could compensate the breaking of the dipole−dipole interactions existing in the pure compounds.

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