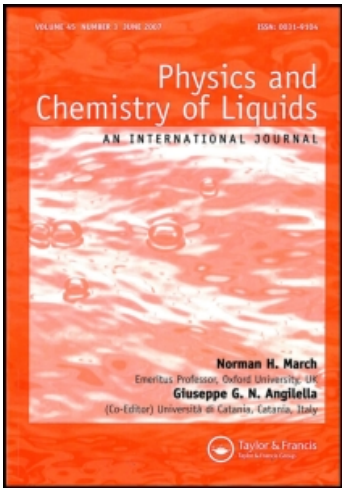

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Densities and Speeds of Sound of the Ternary Mixture 2-Butanol Plus 1-Chlorobutane Plus Tetrahydrofuran

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DENSITIES AND SPEEDS OF SOUND OF THE TERNARY MIXTURE 2-BUTANOL PLUS 1-CHLOROBUTANE PLUS TETRAHYDROFURAN

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Experimental densities (ρ) and speeds of sound (u) were obtained for the ternary system (2-butanol + 1-chlorobutane + tetrahydrofuran) at three temperatures: 283.15, 298.15 and 313.15 K. Excess molar volumes (V^E) and isentropic compressibility deviations ($\Delta\kappa_S$) have been calculated from experimental data. A discussion of the thermodynamic behavior of the ternary system in terms of molecular interactions and structural and packing effects is presented.

Keywords: Excess molar volumes; Isentropic compressibilities; Ternary mixture; 2-Butanol; 1-Chlorobutane; Tetrahydrofuran

INTRODUCTION

Oxygenated compounds such as ethers and alcohols are used as gasoline additives and have been extensively investigated due to their great industrial interest, as it was recently reviewed by Marsh *et al.* [1]. However, references on mixtures containing cyclic ethers are scarce. In our recent studies we have reported new experimental data about thermodynamic properties of binary and ternary mixtures containing cyclic ethers [2–6] together with isomers of butanol, halogenated compounds and alkanes, in order to investigate the influence of the presence of different functional groups in the thermodynamic properties of the liquid mixtures. Continuing our effort, densities and speed of sound for the ternary system 2-butanol + 1-chlorobutane + tetrahydrofuran have been measured at the temperatures of 283.15, 298.15, and 313.15 K, and excess molar volumes and isentropic compressibility deviations have been calculated from experimental data.

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EXPERIMENTAL

The liquids used were 2-butanol (better than 99 mol%), 1-chlorobutane (better than 99 mol %), and tetrahydrofuran (better than 99.5 mol%) obtained from Aldrich. The purity of chemicals was checked by comparing the experimental densities at three temperatures with those reported in the literature [7,8] as they are listed in Table I. We also have employed chromatographic methods to confirm the absence of other significant components. The chemicals were used without additional purification, although the 2-butanol was dried over activated molecular sieve type (0.3 nm) from Merck.

Densities to calculate excess molar volumes were obtained with an Anton Paar DMA-58 vibrating tube densimeter, automatically thermostated at ± 0.01 K. This instrument gives a precision of the density measurements of $\pm 5 \times 10^{-6}$ g cm^{-3}. The accuracy in density measurements is $\pm 1 \times 10^{-5}$ g cm^{-3}.

Isentropic compressibilities have been calculated from densities and speeds of sound. The speeds of sound were determined with an Anton Paar DSA-48 density and sound analyzer automatically thermostated, which gives a precision of ± 0.1 m s^{-1}. The accuracy in speeds of sound measurements is ± 1 m s^{-1}.

The calibrations of both apparatus at each temperature were carried out with deionized doubly distilled water and dry air.

RESULTS

Densities, ρ, speeds of sound, u, isentropic compressibilities, κ_S, excess molar volumes, V^E, and isentropic compressibility deviations, $\Delta\kappa_S$, at 283.15, 298.15, and 313.15 K for the ternary system 2-butanol + 1-chlorobutane + tetrahydrofuran are given in Table II.

The expressions which relate κ_S and $\Delta\kappa_S$ with ρ and u are

$$\kappa_S = \frac{1}{\rho u^2} \tag{1}$$

$$\Delta\kappa_S = \kappa_S - \sum_i x_i \kappa_{s,i} \tag{2}$$

TABLE I Densities, ρ, speeds of sound, u, and isentropic compressibilities, $\Delta\kappa_S$, of the pure compounds and comparisons with literature data

Compound	*T*/K	ρ/kg m^{-3}		u/m s^{-1}	$\Delta\kappa_S$/TPa^{-1}
		This paper	*Lit.*	*This paper*	*This paper*
2-Butanol	283.15	814.51	815.2 [7]	1264.8	767.5
	298.15	802.41	802.41 [8]	1212.2	848.1
	313.15	789.63	789.7 [7]	1157.5	945.3
1-Chlorobutane	283.15	897.26	896.2 [7]	1179.4	801.2
	298.15	880.69	880.95 [8]	1118.1	908.3
	313.15	863.87	863.9 [7]	1056.6	1036.9
Tetrahydrofuran	283.15	898.38	895.3 [7]	1350.4	610.4
	298.15	881.96	881.97 [8]	1277.8	694.4
	313.15	865.38	870.5 [7]	1206.0	794.5

TABLE II Densities, ρ, speeds of sound, u, and isentropic compressibilities, κ_S, excess molar volumes, V^E, and isentropic compressibility deviations, $\Delta\kappa_S$, of the ternary system 2-butanol + 1-chlorobutane + tetrahydrofuran at 283.15, 298.15 and 313.15 K

x_1	x_2	ρ/g cm^{-3}	u/m s^{-1}	κ_S/TPa^{-1}	V^E/cm^3 mol^{-1}	$\Delta\kappa_S$/TPa^{-1}
			T = 283.15 K			
0.0454	0.0542	0.89355	1330.4	632.3	0.048	4.4
0.0461	0.0954	0.89356	1320.9	641.4	0.041	5.6
0.1071	0.0545	0.88728	1322.1	644.8	0.106	7.2
0.1085	0.0954	0.88731	1313.0	653.7	0.097	8.1
0.0948	0.1958	0.88895	1293.4	672.4	0.070	9.8
0.0979	0.3018	0.88892	1273.1	694.1	0.059	10.7
0.1001	0.3960	0.88882	1256.1	713.1	0.061	11.4
0.1024	0.4959	0.88865	1238.6	733.5	0.071	12.4
0.1041	0.6012	0.88854	1221.5	754.3	0.081	12.8
0.1067	0.7123	0.88822	1204.0	776.7	0.108	13.6
0.0922	0.8044	0.88943	1191.7	791.7	0.107	13.4
0.0925	0.8425	0.88921	1186.0	799.5	0.134	13.8
0.2056	0.1993	0.87858	1280.4	694.3	0.149	13.5
0.1964	0.3044	0.87989	1261.7	713.9	0.134	14.6
0.1997	0.4031	0.87995	1244.0	734.3	0.134	15.7
0.2055	0.5031	0.87973	1226.4	755.8	0.144	17.1
0.1921	0.6077	0.88098	1210.5	774.6	0.157	18.1
0.1960	0.7032	0.88070	1194.3	796.1	0.186	20.7
0.2967	0.0987	0.86977	1290.8	690.0	0.203	14.2
0.3039	0.2019	0.86994	1270.2	712.5	0.189	15.9
0.2956	0.3109	0.87140	1251.5	732.7	0.174	16.5
0.3027	0.4084	0.87132	1233.2	754.7	0.178	18.8
0.2939	0.5104	0.87242	1216.3	774.9	0.191	20.9
0.2993	0.5989	0.87224	1200.2	795.9	0.215	24.2
0.3940	0.1009	0.86136	1281.9	706.5	0.226	15.0
0.3880	0.2030	0.86286	1262.7	726.9	0.208	16.8
0.3975	0.2990	0.86304	1244.8	747.8	0.187	17.9
0.3907	0.3964	0.86424	1227.4	768.1	0.192	20.6
0.4003	0.4961	0.86393	1208.5	792.6	0.222	24.6
0.4953	0.1007	0.85309	1274.3	721.9	0.221	14.5
0.4902	0.2081	0.85483	1254.3	743.6	0.195	16.5
0.4994	0.4027	0.85575	1216.6	789.5	0.217	23.8
0.6000	0.1027	0.84499	1267.3	736.9	0.194	12.7
0.5977	0.2105	0.84666	1246.3	760.4	0.173	16.0
0.5942	0.3078	0.84788	1227.2	783.2	0.190	20.7
0.6877	0.1047	0.83835	1260.7	750.5	0.173	12.1
0.6868	0.1966	0.83981	1242.6	771.2	0.157	15.4
0.8000	0.1047	0.83031	1253.9	766.0	0.113	10.0
0.8420	0.0435	0.82615	1264.1	757.5	0.110	6.6
0.8543	0.1067	0.82661	1250.1	774.2	0.075	9.2
0.8960	0.0444	0.82246	1260.9	764.8	0.070	5.2
			T = 298.15 K			
0.0446	0.0519	0.87746	1261.3	716.3	0.047	4.0
0.0464	0.1079	0.87734	1249.6	730.0	0.041	5.3
0.0556	0.8594	0.87586	1127.7	897.9	0.116	11.1
0.0566	0.9001	0.87557	1122.2	906.9	0.142	11.2
0.1054	0.0536	0.87148	1253.7	730.1	0.115	8.0
0.0900	0.1107	0.87311	1244.2	739.9	0.090	8.0
0.0942	0.2083	0.87297	1225.1	763.2	0.078	9.8
0.0959	0.3161	0.87295	1205.6	788.1	0.075	11.4
0.0992	0.4112	0.87274	1189.2	810.2	0.078	12.6
0.1017	0.5098	0.87253	1173.2	832.6	0.089	13.5
0.1046	0.6176	0.87222	1156.8	856.8	0.111	14.2
0.0887	0.7087	0.87344	1145.0	873.3	0.114	13.6
0.0900	0.8052	0.87304	1131.1	895.3	0.157	14.9

(*continued*)

TABLE II Continued

x_1	x_2	ρ/g cm^{-3}	u/m s^{-1}	κ_S/TPa^{-1}	V^E/cm^3 mol^{-1}	$\Delta\kappa_S$/TPa^{-1}
0.0920	0.8448	0.87268	1125.2	905.0	0.186	15.8
0.1992	0.1112	0.86303	1233.1	762.0	0.183	13.2
0.1886	0.2130	0.86446	1215.0	783.6	0.160	14.7
0.2008	0.4182	0.86407	1178.8	832.9	0.167	18.2
0.1868	0.5013	0.86532	1166.3	849.6	0.172	19.3
0.1907	0.6089	0.86500	1149.0	875.7	0.207	21.7
0.1941	0.7043	0.86456	1133.5	900.2	0.259	25.3
0.2917	0.0991	0.85497	1227.7	776.0	0.229	15.6
0.2857	0.2014	0.85613	1209.0	799.1	0.216	17.7
0.2951	0.3090	0.85593	1189.3	826.0	0.219	20.1
0.2849	0.4055	0.85713	1173.5	847.1	0.225	22.2
0.2916	0.5101	0.85685	1155.4	874.2	0.255	25.9
0.2799	0.6181	0.85778	1138.3	899.7	0.301	30.0
0.3929	0.0986	0.84660	1219.9	793.7	0.259	17.8
0.3860	0.2045	0.84805	1201.4	817.0	0.247	19.5
0.3955	0.3152	0.84808	1180.8	845.6	0.251	23.0
0.3904	0.4143	0.84896	1163.6	869.9	0.271	26.9
0.3818	0.5174	0.84981	1146.0	896.0	0.321	32.3
0.4923	0.1033	0.83880	1213.1	810.1	0.271	18.0
0.4895	0.2065	0.84007	1193.8	835.3	0.262	21.5
0.4862	0.3193	0.84123	1173.6	863.1	0.272	25.7
0.4823	0.4160	0.84198	1155.7	889.2	0.311	31.7
0.5982	0.1006	0.83087	1207.9	824.9	0.255	17.0
0.5972	0.2114	0.83229	1186.8	853.0	0.246	21.6
0.5938	0.3078	0.83328	1168.1	879.5	0.279	28.0
0.6858	0.1055	0.82472	1202.7	838.3	0.223	15.9
0.6881	0.2138	0.82587	1180.9	868.3	0.231	22.4
0.7987	0.1057	0.81708	1197.3	853.7	0.161	13.9
0.8408	0.0604	0.81357	1205.0	846.5	0.137	9.9
0.8357	0.1061	0.81464	1195.3	859.2	0.138	13.6
0.9017	0.0598	0.80962	1202.4	854.3	0.091	8.5
			T = 313.15 K			
0.0449	0.0549	0.86112	1191.1	818.5	0.040	4.0
0.0465	0.0931	0.86107	1183.1	829.7	0.029	5.6
0.0550	0.8634	0.85920	1066.4	1023.4	0.134	11.4
0.0560	0.9026	0.85888	1061.4	1033.5	0.164	11.8
0.1055	0.0547	0.85532	1184.6	833.2	0.121	9.5
0.0918	0.1074	0.85673	1176.6	843.2	0.095	8.8
0.0938	0.1955	0.85675	1160.9	866.1	0.081	10.0
0.0967	0.3157	0.85655	1139.9	898.5	0.085	12.9
0.0989	0.4113	0.85637	1124.8	922.9	0.092	13.8
0.1006	0.4949	0.85621	1112.2	944.3	0.101	14.6
0.1032	0.5997	0.85583	1096.9	971.2	0.131	15.8
0.0881	0.7096	0.85683	1082.9	995.2	0.144	15.5
0.0913	0.8063	0.85623	1069.8	1020.6	0.195	16.9
0.0909	0.8465	0.85603	1064.4	1031.1	0.226	17.7
0.2007	0.0953	0.84703	1169.0	863.9	0.197	16.1
0.2025	0.1987	0.84730	1151.7	889.8	0.187	16.6
0.1935	0.3051	0.84825	1134.3	916.3	0.190	18.6
0.2050	0.4139	0.84750	1116.0	947.4	0.213	21.7
0.1853	0.5012	0.84912	1104.1	966.1	0.215	22.2
0.1934	0.6073	0.84841	1087.7	996.3	0.260	25.4
0.1943	0.7047	0.84803	1073.2	1023.8	0.327	29.2
0.2953	0.0969	0.83918	1162.9	881.2	0.250	18.7
0.3030	0.2016	0.83917	1144.0	910.6	0.246	21.6
0.2965	0.3073	0.84014	1126.9	937.2	0.249	23.5
0.3025	0.4078	0.83991	1110.0	966.3	0.279	27.3
0.2919	0.5099	0.84081	1094.4	993.0	0.313	30.9

(*continued*)

TABLE II Continued

x_1	x_2	ρ/g cm^{-3}	u/m s^{-1}	κ_S/TPa^{-1}	V^E/cm^3 mol^{-1}	$\Delta\kappa_S$/TPa^{-1}
0.2808	0.6159	0.84142	1078.3	1022.1	0.384	36.0
0.3866	0.3971	0.83359	1106.0	980.7	0.319	31.7
0.3976	0.5017	0.83281	1087.5	1015.4	0.398	39.3
0.4924	0.1016	0.82407	1152.0	914.4	0.290	21.0
0.4872	0.2090	0.82535	1132.7	944.3	0.294	25.7
0.4856	0.3197	0.82606	1113.3	976.7	0.330	31.5
0.4968	0.4039	0.82543	1097.2	1006.4	0.393	39.1
0.5937	0.2135	0.81787	1126.9	962.9	0.296	27.1
0.5931	0.3072	0.81837	1109.2	993.3	0.352	34.9
0.6896	0.1017	0.81042	1143.8	943.3	0.237	20.1
0.6857	0.1977	0.81146	1125.5	972.8	0.282	27.0
0.8002	0.1045	0.80322	1138.9	959.8	0.189	19.4

where x_i and κ_S, i are, respectively, the mole fraction and the isentropic compressibility of component i.

The values of both excess properties of the ternary system have been fitted to the Cibulka equation [9]:

$$Y^E = Y^E_{\text{bin}} + (C_1 + C_2 x_1 + C_3 x_2) \tag{3}$$

where Y^E is the excess molar volume or the isentropic compressibility deviation, x_i is the mole fraction of component i in the ternary mixture, C_i are adjustable parameters obtained by the least squares method, and Y^E_{bin} is the binary contribution, which is given by

$$Y^E_{\text{bin}} = \sum_{i \neq j} Y^E_{ij} = Y^E_{12} + Y^E_{13} + Y^E_{23} \tag{4}$$

where Y^E_{ij} is a Redlich–Kister polynomial equation,

$$Y^E_{ij} = x_i x_j \sum_{p=0}^{n} A_p (x_i - x_j)^p \tag{5}$$

where x_i and x_j are the mole fractions in the binary mixture and A_p are binary adjustable parameters obtained by least squares.

The Redlich–Kister parameters for all the constituent binary mixtures have been taken from literature values [5,6,10], and they are presented in Table III along with standard deviations in the properties correlation. Parameters of the Cibulka equation are reported in Table IV along with the corresponding standard deviations.

Isolines at constant values of excess molar volumes for the ternary system have been graphically represented in Fig. 1, while isentropic compressibility deviations are plotted in Fig. 2.

Excess molar volumes of the ternary system are positive in almost the whole composition range and they increase when temperature rises, as can be seen in Fig. 1.

Isentropic compressibility deviations are positive throughout the entire composition range, and they also increase with increasing temperature, as shown in Fig. 2.

TABLE III Parameters of the Redlic–Kister equation, A_p, for the constituent binary mixtures and standard deviations, $\sigma(Y^E)$

Property	*T*/K	A_0	A_1	A_2	A_3	$\sigma(Y^E)$
2-Butanol + 1-chlorobutane						
	283.15	1.113	−0.689	0.185	−0.389	0.003
V^E	298.15	1.639	−0.550	0.374	−0.631	0.004
	313.15	2.105	−0.500	0.620	−0.890	0.005
	283.15	124.0	−44.2	23.9	−29.3	0.1
$\Delta\kappa_S$	298.15	163.6	−41.8	19.4	−25.3	0.1
	313.15	206.0	−35.2	39.9	18.0	0.5
2-Butanol + tetrahydrofuran						
	283.15	1.014	−0.119	0.063	−0.062	0.002
V^E	298.15	1.191	−0.098	0.060	−0.179	0.003
	313.15	1.228	−0.121	−0.007	−0.136	0.002
	283.15	50.1	−17.7	−4.0	2.1	0.1
$\Delta\kappa_S$	298.15	61.0	−17.5	−10.6	5.6	0.1
	313.15	80.9	−29.8	11.7	48.3	0.2
1-Chlorobutane + tetrahydrofuran						
	283.15	−0.221	0.010	0.016	−0.060	0.001
V^E	298.15	−0.235	0.000	0.025	−0.023	0.001
	313.15	−0.281	0.006	−0.064	−0.045	0.001
	283.15	26.2	−1.4	0.7	−2.4	0.1
$\Delta\kappa_S$	298.15	24.3	−2.8	−4.2	9.8	0.1
	313.15	22.6	−0.6	−8.0	7.6	0.1

TABLE IV Parameters of the Cibulka equation, C_i, for the ternary mixture 2-butanol + 1-chlorobutane + tetrahydrofuran and standard deviations, $\sigma(Y^E)$

Property	*T*/K	C_1	C_2	C_3	$\sigma(Y^E)$
	283.15	1.495	−3.528	−3.416	0.006
V^E	298.15	0.747	−4.228	−2.647	0.006
	313.15	1.420	−6.653	−3.093	0.006
	283.15	48.5	−190.3	−325.5	0.3
$\Delta\kappa_S$	298.15	−1.4	−153.3	−321.5	0.4
	313.15	−62.3	−314.5	−257.9	0.9

The largest values of both properties, V^E and $\Delta\kappa_S$, correspond to the binary system 2-butanol + 1-chlorobutane and the smallest values to the binary mixture 1-chlorobutane + tetrahydrofuran.

The sign and magnitude of excess molar volumes depend on the strength of the interaction between the molecules, on the structural effects and packing phenomena, both related with differences in shape and size [11]. Isentropic compressibility deviations also depend on all these factors but packing phenomena are better evidenced through $\Delta\kappa_S$ values [12].

In our ternary mixture there are many different effects to consider: for the pure compounds 2-butanol is self-associated with hydrogen bonds, while 1-chlorobutane and tetrahydrofuran present permanent dipolar moments, which reveals the existence of dipole–dipole interactions in the pure liquids. We must also take into account the packing of the molecules in the pure compounds. As we can see in Table I the smallest values of isentropic compressibility correspond to tetrahydrofuran, which

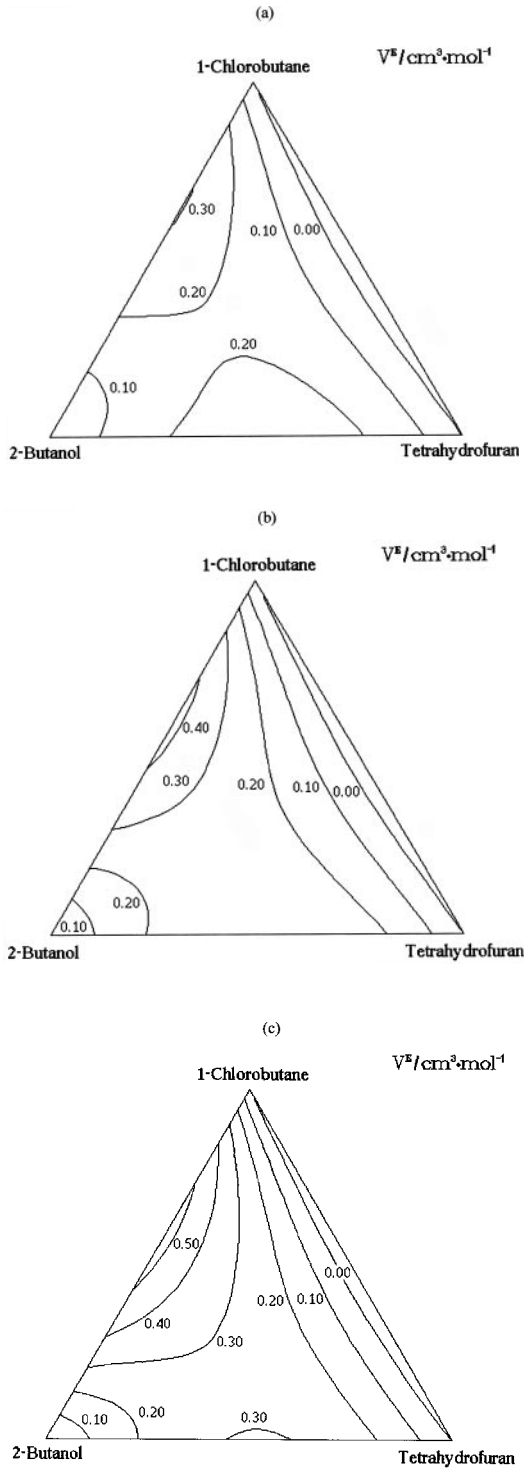

FIGURE 1 Isolines of constant excess molar volumes, V^E, of the ternary system 2-butanol + 1-chlorobutane + tetrahydrofuran: (a) at 283.15 K; (b) at 298.15 K; (c) at 313.15 K.

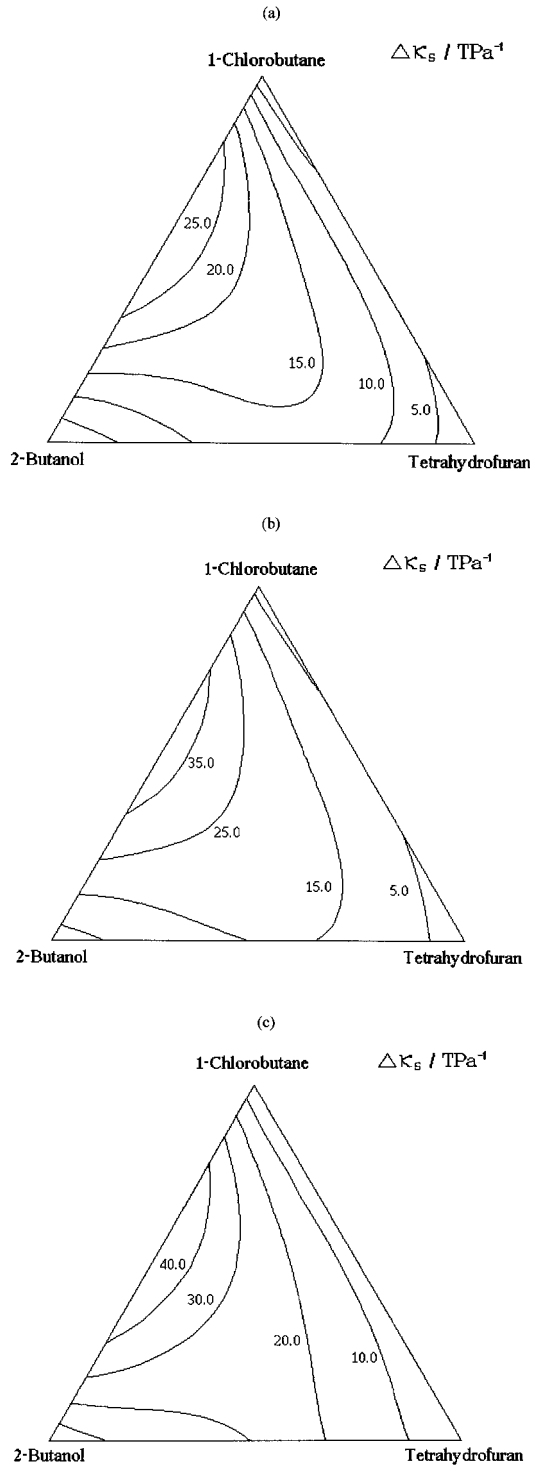

FIGURE 2 Isolines of constant isentropic compressibility deviations, $\Delta\kappa_S$, of the ternary system 2-butanol + 1-chlorobutane + tetrahydrofuran: (a) at 283.15 K; (b) at 298.15 K; (c) at 313.15 K.

indicate that the cyclic ether presents the most compact structure of the three compounds.

When the mixing process occurs, like interactions weaken but specific interactions between the different components become more significant. The breaking or weakness of self-association and dipole–dipole interactions lead to positive V^E and $\Delta\kappa_S$ values, while specific interactions between the different functional groups, O–OH, Cl–OH, and O–Cl result in negative values.

Positive values of V^E indicate that the weakness of the pure compounds interactions is more important than forming cross-associates between the different compounds. While positive isentropic compressibility deviations reveal that the packing of molecules in the mixture is less compact than in the pure compounds.

Values of V^E and $\Delta\kappa_S$ are largest in the binary systems 2-butanol + 1-chlorobutane and 2-butanol + trtrahydrofuran than in the mixture 1-chlorobutane +tetrahydrofuran, showing that the associated structure of 2-butanol is quite disturbed when it is mixed with other polar molecules. This effect is more relevant in 2-butanol than in 1-butanol, as we can confirm comparing our results with those previously reported for the ternary system 1-butanol + 1-chlorobutane + tetrahydrofuran [6]. We have observed that V^E and $\Delta\kappa_S$ values are significantly largest in the ternary mixture containing 2-butanol than in the system with 1-butanol. Although sterically hindered alcohols are considerably less associated [13] and they have smaller hydrogen-bonds energies [14], the breaking of more bonds in the mixtures of 2-butanol outweighs the smaller contribution per bond to the investigated properties [15].

The smallest values of V^E and $\Delta\kappa_S$ appear at high mole fractions of tetrahydrofuran, which presents the most compact structure of the three compounds, as we have outlined.

Concerning the increase of both properties with temperature, it can be explained by the increase of thermal energy, which contributes to weaken like interactions.

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