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SPEEDS OF SOUND AND ISENTROPIC COMPRESSIBILITIES OF CYCLOHEXANE+1,3-DIOXOLANE+2-BUTANOL AND *n*-HEXANE+1,3-DIOXOLANE+2-BUTANOL

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Abstract

Speeds of sound of the ternary mixtures cyclohexane+1,3-dioxolane+2-butanol and *n*-hexane+1,3-dioxolane+2-butanol have been measured at the temperatures of 298.15 and 313.15 K. Isentropic compressibilities and excess isentropic compressibilities have been calculated from experimental data. We have also compared the experimental isentropic compressibilities with calculated values from the free length theory and the collision factor theory.

Experimental results show positive values of excess isentropic compressibilities in almost the whole composition range for the ternary mixture containing cyclohexane, meanwhile they are negative for the mixture containing *n*-hexane. Such different behaviour of these systems is related to the large free volume shown by *n*-hexane.

Keywords: butanol, cyclic ether, isentropic compressibilities, speeds of sound, ternary mixture

Introduction

Several papers of many authors [1–6] point out the importance of the study about the thermodynamic behaviour of multicomponent systems involving organic liquids with oxygenated functional groups like alcohols or cyclic polyethers because of industrial applications and the theoretical interest in studying the structure of such solutions. Nevertheless, experimental data for ternary mixtures containing these compounds is scarce. Going on with our research work concerning thermodynamic properties of binary and ternary mixtures containing alkanes, cyclic ethers, and isomers of butanol [7–14] we report here the experimental values of speeds of sound for the ternary mixtures cyclohexane+1,3-dioxolane+2-butanol and *n*-hexane+1,3-dioxolane+2-butanol at the temperatures of 298.15 and 313.15 K.

Isentropic compressibilities and excess isentropic compressibilities have been calculated from experimental data using the equations of Newton–Laplace, and

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Benson and Kiyohara [15], respectively. Excess isentropic compressibilities were correlated with the Redlich–Kister equation [16].

We have also compared experimental isentropic compressibilities with calculated values using the free length theory (FLT) [17, 18] and the collision factor theory (CFT) [19, 20].

Experimental

Materials

The studied compounds were cyclohexane (>99.9 mol%), *n*-hexane (>99 mol%), 1,3-dioxolane (>99 mol%), and 2-butanol (>99 mol%) obtained from Aldrich. These material purities were checked by comparing the measured densities with those reported in the literature and also by means of a chromatographic method. Such studies confirmed the absence of any other significant compounds. No further purification was considered necessary, although 2-butanol was dried with activate molecular sieve type 0.3 nm from Merck.

Properties of the pure compounds at 298.15 and 313.15 K, along with the literature values [2, 21–25], are given in Table 1.

Table 1 Physical properties of the pure components at 298.15 and 313.15 K

Property	T/K	Cyclohexane	n-hexane	1,3-dioxolane	2-butanol
ρ/kg m^{-3}	298.15	773.72 773.89^{a}	655.07 654.84^{a}	1058.62 1058.66^{b}	802.20 802.41^{a}
	313.15	759.47	641.27	1039.97	789.59
u/m s^{-1}	298.15	1254.4	1078.5	1338.8	1211.5
	313.15	1180.8	1008.1	1271.7	1157.1
10^3 α/K^{-1}	298.15	1.220	1.387	1.164	1.059
	313.15	1.257	1.453	1.226	1.105
C_p/J mol^{-1} K^{-1}	298.15	156.2^{c}	195.6^{d}	122.21	196.9^{d}
	313.15	162.0^{c}	200.1^{e}	125.01	212.6^{f}

^{a}Ref [21]; ^{b}Ref [2]; ^{c}Ref [22]; ^{d}Ref [23]; ^{e}Ref [24]; ^{f}Ref [25]

Methods

Speeds of sound, *u*, of the pure compounds and their mixtures were obtained with an Anton Paar DSA-48 vibrating tube densimeter and sound analyzer. The temperature was automatically kept constant within ±0.01 K. The densities needed to obtain the isentropic compressibilities were also measured with the same apparatus. The precision of the speed of sound and density measurements is ± 0.1 m s^{-1} and $\pm 3\cdot 10^{-2}$ kg m^{-3}, respectively. Calibration of the apparatus was carried out with air and deionized twice distilled water. More details of calibration and procedures were reported in a previous paper [26].

Mixtures of well-known composition were prepared by mass using a Mettler H20T balance. The uncertainty of the mixtures mole fraction is estimated to be less than ±1·10^{-4}.

Results and discussion

Speeds of sound of the ternary systems cyclohexane+1,3-dioxolane+2-butanol and *n*-hexane+1,3-dioxolane+2-butanol at 298.15 and 313.15 K are given in Table 2 together with calculated isentropic compressibilities and excess isentropic compressibilities.

Table 2 Densities, ρ, speeds of sound, u, isentropic compressibilities, κ_S, and excess isentropic compressibilities, κ_S^E, of the ternary systems cyclohexane(1)+1,3-dioxolane(2)+2-butanol(3) and *n*-hexane(1)+1,3-dioxolane(2)+2-butanol(3) at 298.15 and 313.15 K

x_1	x_2	u/m s^{-1}	$_S$/TPa^{-1}	κ_S^E/TPa^{-1}
		Cyclohexane(1)+1,3-dioxolane(2)+2-butanol(3) at 298.15 K		
0.0535	0.0513	1209.4	845.2	6.7
0.0534	0.1043	1210.8	833.6	7.9
0.0507	0.8481	1301.9	589.2	–3.6
0.0506	0.8977	1312.1	570.9	–1.7
0.0981	0.0579	1207.3	849.4	11.5
0.1029	0.0999	1208.0	840.9	13.1
0.1002	0.2064	1213.4	813.0	12.2
0.0970	0.2979	1219.8	786.7	10.5
0.0995	0.3968	1228.3	757.1	8.9
0.1071	0.4955	1239.1	725.2	6.7
0.1056	0.6016	1253.9	687.5	3.4
0.0989	0.7032	1270.9	649.5	0.9
0.1030	0.7984	1288.7	614.0	0.8
0.1025	0.8477	1298.5	595.5	1.8
0.2078	0.0980	1203.9	852.5	22.9
0.2045	0.2035	1209.1	825.1	22.2
0.2025	0.3100	1216.3	795.0	20.8
0.2032	0.3926	1223.9	769.3	18.7
0.2053	0.4939	1235.4	735.6	15.8
0.2043	0.6006	1250.6	697.3	12.3
0.1986	0.6968	1266.7	661.0	9.6
0.3002	0.1027	1202.1	858.6	30.0
0.3137	0.1895	1205.9	837.7	31.2

Table 2 Continued

x_1	x_2	u/m s^{-1}	$_S$/TPa^{-1}	κ_S^E/TPa^{-1}
Cyclohexane(1)+1,3-dioxolane(2)+2-butanol(3) at 298.15 K				
0.3059	0.2897	1213.3	808.2	28.6
0.3120	0.3843	1222.2	778.7	26.3
0.3108	0.4911	1235.1	742.1	22.3
0.3025	0.5964	1250.3	703.7	18.5
0.4078	0.0993	1201.9	864.2	36.0
0.4014	0.1867	1206.2	841.8	35.9
0.4131	0.2859	1213.9	813.2	34.5
0.4126	0.3958	1225.7	776.9	30.1
0.3979	0.4969	1238.6	741.1	25.5
0.5087	0.0973	1204.1	865.2	39.0
0.5013	0.2002	1210.1	837.9	38.3
0.5154	0.2885	1218.9	810.1	35.5
0.5102	0.3873	1230.6	776.2	30.7
0.6064	0.0978	1208.8	861.6	39.2
0.6069	0.1872	1215.3	836.7	38.0
0.6071	0.2909	1226.9	802.2	32.8
0.7097	0.0966	1216.9	853.2	35.7
0.7055	0.1988	1226.1	822.6	32.7
0.8006	0.0952	1226.7	841.7	29.7
0.8493	0.0498	1229.2	846.7	26.0
0.8455	0.1020	1234.9	830.1	23.2
0.8972	0.0500	1237.9	835.3	18.5
Cyclohexane(1)+1,3-dioxolane(2)+2-butanol(3) at 313.15 K				
0.0561	0.0603	1152.5	945.0	10.4
0.0516	0.0989	1152.5	936.6	12.4
0.0518	0.8433	1231.6	671.4	0.0
0.0523	0.8941	1241.9	649.5	1.1
0.1048	0.0505	1148.8	956.2	16.1
0.1037	0.0957	1149.5	945.6	17.3
0.1099	0.1861	1152.1	922.9	18.5
0.1042	0.2979	1158.6	888.2	16.2
0.0988	0.4066	1169.1	848.5	10.3
0.1078	0.4979	1178.2	816.1	7.5
0.1041	0.6019	1190.2	776.9	5.4

Table 2 Continued

x_1	x_2	u/m s^{-1}	${}_{S}$/TPa^{-1}	κ_S^ETPa^{-1}
Cyclohexane(1)+1,3-dioxolane(2)+2-butanol(3) at 313.15 K				
0.0982	0.7170	1206.5	730.5	3.5
0.0871	0.8098	1223.1	690.5	2.7
0.1001	0.8460	1228.0	678.2	5.8
0.1994	0.1091	1143.8	958.0	28.3
0.2111	0.1910	1146.5	937.4	29.4
0.2091	0.2926	1151.8	907.0	28.4
0.2081	0.3910	1160.1	872.6	24.5
0.2044	0.4991	1170.9	832.8	20.7
0.2051	0.6023	1184.2	791.9	16.7
0.1987	0.7022	1198.6	751.0	14.7
0.3038	0.0991	1139.3	974.2	38.2
0.3119	0.1887	1141.7	951.8	40.1
0.3084	0.2938	1148.9	917.4	36.4
0.3120	0.3916	1156.5	884.6	34.1
0.3038	0.4924	1167.5	845.5	29.1
0.3002	0.5925	1180.9	804.5	24.5
0.4033	0.0979	1137.2	983.2	44.9
0.3986	0.2090	1142.9	950.3	43.0
0.4125	0.2960	1148.9	922.8	41.5
0.4103	0.3988	1158.1	885.9	37.6
0.3989	0.4907	1168.0	850.6	33.8
0.5036	0.0970	1137.5	987.7	48.8
0.5001	0.2028	1142.8	956.8	47.9
0.5046	0.2946	1151.7	923.0	42.2
0.5012	0.3979	1162.4	884.0	37.1
0.6034	0.0987	1141.7	984.1	47.1
0.6015	0.2066	1148.8	950.2	44.7
0.6047	0.2975	1157.9	916.7	39.9
0.7086	0.1014	1147.8	976.5	43.7
0.7035	0.1895	1153.9	948.6	42.0
0.7954	0.1039	1155.4	965.3	37.5
0.8482	0.0513	1157.3	973.1	32.8
0.8391	0.1071	1162.8	953.0	28.8
0.8808	0.0642	1164.3	959.3	25.0

Table 2 Continued

x_1	x_2	u/m s^{-1}	$_S$/TPa^{-1}	κ_S^E/TPa^{-1}
		n-hexane(1)+1,3-dioxolane(2)+2-butanol(3) at 298.15 K		
0.0466	0.0609	1199.0	866.3	–4.4
0.0507	0.0951	1199.0	860.6	–5.3
0.0513	0.8474	1286.1	611.5	–30.8
0.0519	0.8926	1294.9	594.7	–30.7
0.1005	0.0549	1184.1	902.0	–9.3
0.1032	0.0895	1183.9	895.9	–9.7
0.1026	0.1892	1188.0	870.1	–12.2
0.0959	0.2958	1196.4	835.1	–16.3
0.1009	0.3865	1202.0	810.1	–21.6
0.0961	0.4900	1214.2	771.7	–27.7
0.1024	0.6752	1237.0	708.0	–40.3
0.0916	0.7161	1247.2	686.0	–39.8
0.0956	0.8075	1262.4	652.3	–45.3
0.0974	0.8523	1271.0	634.7	–48.2
0.1935	0.0952	1161.6	951.4	–13.8
0.1877	0.2078	1166.5	919.2	–18.0
0.2054	0.2916	1166.6	905.9	–23.9
0.1978	0.4020	1176.8	865.4	–32.8
0.1858	0.5092	1190.5	820.2	–41.7
0.1991	0.5952	1197.0	796.3	–51.3
0.1971	0.6977	1211.5	755.3	–60.4
0.2887	0.1014	1140.0	1009.7	–11.9
0.2946	0.2001	1141.7	987.2	–17.9
0.2928	0.3003	1147.8	955.3	–27.4
0.2798	0.3388	1152.8	935.2	–31.0
0.2937	0.4883	1162.7	890.7	–48.3
0.2976	0.5981	1173.4	851.4	–61.4
0.3856	0.1048	1121.8	1065.6	–7.7
0.4001	0.1970	1122.4	1048.2	–15.0
0.4006	0.2991	1128.1	1015.5	–27.3
0.4056	0.3992	1134.4	983.3	–40.5
0.4031	0.5031	1144.2	942.5	–55.7
0.4953	0.1046	1105.3	1124.2	–1.8
0.4906	0.2058	1110.2	1090.9	–14.4

Table 2 Continued

x_1	x_2	u/m s^{-1}	$_S$/TPa^{-1}	κ_S^E/TPa^{-1}
		n-hexane(1)+1,3-dioxolane(2)+2-butanol(3) at 298.15 K		
0.4990	0.2956	1114.3	1065.2	–26.4
0.5129	0.3878	1119.8	1036.9	–41.8
0.5882	0.1100	1095.3	1165.3	0.7
0.5952	0.2027	1098.8	1139.1	–11.7
0.6013	0.2927	1104.3	1108.9	–27.2
0.6896	0.1109	1087.2	1205.7	2.8
0.6823	0.2121	1093.1	1168.7	–13.6
0.7874	0.1061	1082.2	1239.1	2.7
0.8594	0.0491	1077.8	1276.6	9.1
0.8424	0.1071	1081.1	1252.7	–0.3
0.9030	0.0378	1076.8	1290.5	9.0
		n-hexane(1)+1,3-dioxolane(2)+2-butanol(3) at 313.15 K		
0.0496	0.0518	1140.0	976.7	–5.1
0.0497	0.1077	1140.2	965.0	–3.1
0.0504	0.8487	1214.6	697.7	–30.2
0.0491	0.8936	1225.5	675.3	–31.6
0.1010	0.0535	1124.4	1017.7	–11.4
0.1024	0.0965	1123.7	1010.1	–10.2
0.1035	0.1995	1125.8	984.3	–11.9
0.1005	0.2889	1131.6	953.2	–16.9
0.0999	0.3936	1139.1	916.7	–23.6
0.0956	0.4973	1150.2	874.1	–31.0
0.0925	0.6045	1163.5	828.9	–38.2
0.0913	0.7078	1177.2	786.2	–43.2
0.0975	0.8054	1190.4	748.3	–50.6
0.0976	0.8443	1197.5	730.3	–53.1
0.1910	0.0968	1098.4	1082.2	–14.6
0.1881	0.2094	1102.3	1048.4	–20.2
0.2044	0.2952	1102.3	1032.4	–29.1
0.1981	0.4063	1111.9	986.6	–40.6
0.1915	0.5043	1122.1	943.8	–50.1
0.1963	0.6021	1130.9	906.9	–61.6
0.1956	0.6969	1142.0	866.4	–70.3
0.2956	0.0940	1073.4	1163.6	–14.3

Table 2 Continued

x_1	x_2	u/m s^{-1}	$_{\mathrm{s}}$/TPa^{-1}	κ_S^E/TPa^{-1}
		n-hexane(1)+1,3-dioxolane(2)+2-butanol(3) at 313.15 K		
0.2964	0.1976	1076.2	1134.0	–22.5
0.2932	0.3011	1081.2	1097.1	–33.3
0.3072	0.3873	1084.7	1073.0	–46.8
0.2957	0.4970	1095.9	1020.9	–61.2
0.2799	0.6135	1110.9	960.2	–75.6
0.3890	0.0969	1056.0	1229.1	–12.1
0.4023	0.1963	1055.9	1208.5	–21.1
0.4086	0.2761	1059.0	1183.5	–32.8
0.4037	0.4066	1068.0	1129.3	–53.2
0.3986	0.4979	1076.8	1086.2	–69.4
0.4956	0.1048	1039.0	1297.9	–6.4
0.4794	0.2108	1045.0	1252.4	–21.5
0.5042	0.2843	1045.5	1239.1	–33.3
0.5103	0.3893	1051.7	1199.1	–52.7
0.5968	0.0951	1027.3	1358.1	–2.0
0.6006	0.1963	1030.0	1326.5	–15.9
0.5987	0.2930	1035.9	1285.7	–35.2
0.7017	0.0957	1018.0	1411.6	2.5
0.6881	0.2033	1023.9	1364.2	–17.9
0.8069	0.1042	1012.7	1451.4	1.0
0.8408	0.0575	1010.0	1479.5	7.7
0.8381	0.1046	1011.9	1461.1	–0.8
0.8905	0.0499	1008.4	1498.0	7.3

Isentropic compressibilities have been calculated from the Newton–Laplace equation:

$$\kappa_S = \frac{1}{\rho u^2} \qquad (1)$$

and excess isentropic compressibility, κ_S^E, is defined as:

$$\kappa_S^{id} = \kappa_S - \kappa_S^{id} \qquad (2)$$

where the isentropic compressibility for the ideal mixture, κ_S^{id}, is given by an expression proposed by Benson and Kiyohara [15]:

$$\kappa_{S}^{id}=\sum_{i=1}^{3}\phi_{i}\left[\kappa_{S,i}+\frac{TV_{i}\alpha_{i}^{2}}{C_{p,i}}\right]-T\left(\sum_{i}x_{i}V_{i}\right)\frac{\left(\sum_{i}\phi_{i}\alpha_{i}\right)^{2}}{\left(\sum_{i}x_{i}C_{p,i}\right)} \tag{3}$$

being ϕ_i the volume fraction of component *i* in the mixture, x_i the corresponding mole fraction, T the absolute temperature, and $\kappa_{S,i}$, V_i, α_i and $C_{p,i}$ the isentropic compressibility, the molar volume, the cubic expansion coefficient, and the molar heat capacity of the pure components, respectively. The cubic expansion coefficients were obtained from density measurements at several temperatures and the heat capacity of 1,3-dioxolane was determined with a programmable differential scanning microcalorimeter Setaram DSC II.

The excess isentropic compressibilities of the ternary systems were fitted to the Redlich–Kister equation for ternary mixtures [16]:

$$\kappa_{S}^{E}=\kappa_{S,bin}^{E}+x_{1}x_{2}(1-x_{1}-x_{2})[C_{1}+C_{2}x_{1}+C_{2}x_{1}+C_{3}x_{2}] \tag{4}$$

where x_i is the mole fraction of component *i* in the mixture, C_i are adjustable parameters obtained by the least square method and $\kappa_{S,bin}^{E}$ is the contribution to the excess property of the constituent binary mixtures, which is given by:

$$\kappa_{S,bin}^{E}=\kappa_{S,12}^{E}+\kappa_{S,13}^{E}+\kappa_{S,23}^{E} \tag{5}$$

and each $\kappa_{S,ij}^{E}$ is obtained by means of a Redlich–Kister polynomial equation:

$$\kappa_{S,ij}^{E}=x_{i}x_{j}\sum_{p=0}^{n}A_{p,ij}(x_{i}-x_{j})^{p} \tag{6}$$

where x_i is the mole fraction of component *i* in the ternary mixture and $A_{p,ij}$ are adjustable parameters obtained by the least square method. Parameters of the constituent binary mixtures, obtained from experimental measurements either performed in our laboratory or taken from previous papers [7, 27] are given in Table 3, along with the standard deviations, σ.

Parameters obtained with the Redlich–Kister equation for the ternary mixtures (10) are given in Table 4.

Excess isentropic compressibilities of the ternary system cyclohexane+1,3-dioxolane+2-butanol are positive in almost the whole composition range and become slightly higher when the temperature increases from 298.15 to 313.15 K as we can see in Fig. 1. However, excess isentropic compressibilities of the ternary system *n*-hexane+1,3-dioxolane+2-butanol are negative at both temperatures, nearly in the entire composition range, as shown in Fig. 2.

Excess isentropic compressibility values are related to structural effects and packing phenomena as well as with energetic balance in the mixture (i.e. breaking of pure compounds molecular interactions and formation of cross-associates among different components). In our mixtures we can notice that differences in molecular size and shape are playing an important role, given that the energetic balance is quite similar in both mixtures. However, when cyclohexane is replaced by *n*-hexane the excess isentropic compressibility values change their sign.

Table 3 Coefficients of Redlich–Kister equation for the excess isentropic compressibilities of the constituent binary mixtures of the ternary systems cyclohexane(1)+1,3-dioxolane(2)+2-butanol(3) and *n*-hexane(1)+1,3-dioxolane(2)+2-butanol(3) at 298.15 and 313.15 K

System	T/K	A_0	A_1	A_2	A_3	A_4	σ/TPa^{-1}
Cyclohexane	298.15	102.9	52.4	–4.0	26.7	–	0.3
+1,3-dioxolane[a]	313.15	130.5	62.3	2.8	28.7		0.3
Cyclohexane	298.15	142.6	52.5	37.6	27.4	–	0.3
+2-butanol[a]	313.15	182.6	50.4	42.2	46.1		0.3
1,3-dioxolane(1)	298.15	–25.4	–52.7	–8.3	17.1	–	0.1
+2-butanol[b]	313.15	–35.6	–92.9	65.4	35.0		0.2
n-hexane	298.15	–230.1	239.0	–102.8	90.3	–	0.6
+1,3-dioxolane[a]	313.15	–287.5	303.6	–68.8	44.2		0.5
n-hexane	298.15	36.1	122.6	–51.8	47.0	104.4	0.4
+2-butanol[c]	313.15	18.3	147.2	–45.1	82.1	86.7	0.4

[a] To be published; [b] [7]; [c] [27]

Table 4 Coefficients of the Redlich–Kister equation for the excess isentropic compressibilities of the ternary systems cyclohexane(1)+1,3-dioxolane(2)+2-butanol(3) and *n*-hexane(1)+1,3-dioxolane(2)+2-butanol(3) at 298.15 and 313.15 K

System	T/K	C_1	C_2	C_3	σ/TPa^{-1}
Cyclohexane+1,3-dioxolane+2-butanol	298.15	208.4	–78.8	–155.4	0.3
	313.15	113.7	–20.7	113.1	1.0
n-Hexane+1,3-dioxolane+2-butanol	298.15	–285.2	255.1	5.1	0.7
	313.15	–77.0	–186.7	–98.8	1.0

Taking into account that *n*-hexane is a disordered flexible molecule with a large free volume [4] it is reasonable to think that when it is mixed with 1,3-dioxolane and 2-butanol the free volume decreases because of the interstitial accommodation. This effect makes the mixture less compressible, yielding to negative values of κ_S^E.

The FLT and CFT have been applied to predict the isentropic compressibilities of the ternary systems. Table 5 shows the root mean square deviation between the experimental and calculated isentropic compressibilities for the two models, defined by the expression:

$$RMSD(\%) = 100\left[\frac{1}{m}\sum\left(\frac{\kappa_S^{\mathrm{cal}} - \kappa_S^{\mathrm{exp}}}{\kappa_S^{\mathrm{exp}}}\right)^2\right]^{1/2} \tag{7}$$

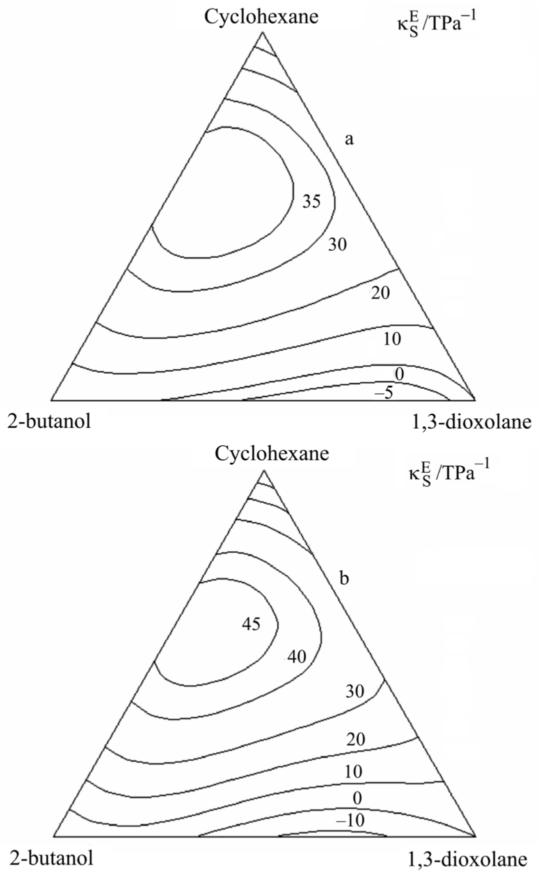

Fig. 1 Excess isentropic compressibilities of the ternary mixture cyclohexane+1,3-dioxolane+2-butanol at: a – 298.15 K, b – 313.15 K

where *m* is the number of experimental points, κ_S^{cal} is the calculated value for the isentropic compressibility of the mixture and κ_S^{exp} is the experimental value of the property.

From Table 5 it can be concluded that isentropic compressibility predictions of both models are reasonably close to experimental results.

Table 5 Root mean square deviation of isentropic compressibilities predicted by FLT and CFT theories

System	*RMSD*/%			
	T=298.15 K		*T*=313.15 K	
	FLT	CFT	FLT	CFT
Cyclohexane+1,3-dioxolane+2-butanol	7.6	5.7	8.0	6.0
n-Hexane+1,3-dioxolane+2-butanol	8.1	6.5	9.1	7.2

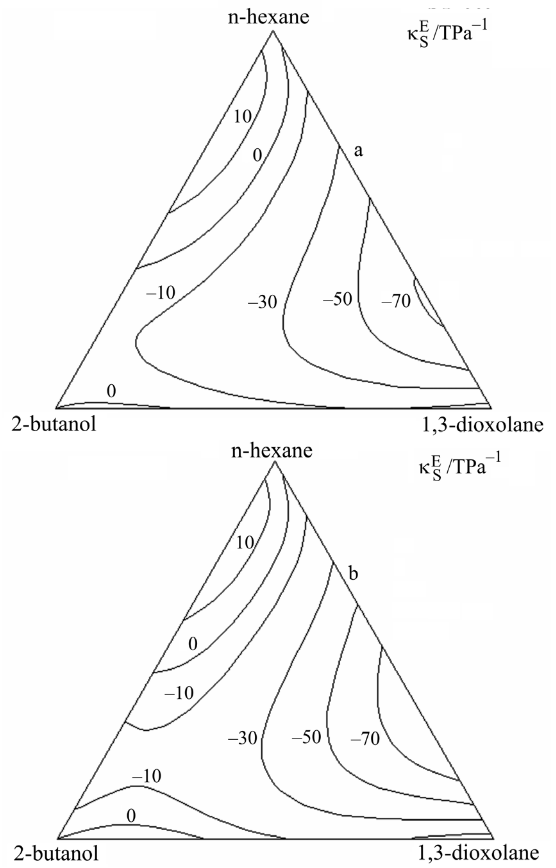

Fig. 2 Excess isentropic compressibilities of the ternary mixture *n*-hexane+1,3-dioxolane+2-butanol at: a – 298.15 K, b – 313.15 K

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