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Speeds of sound and isentropic compressibilities of binary mixtures of 1-heptanol + 1,2-dichloroethane, +1,1,1-trichloroethane, +1,1,2,2-tetrachloroethane, +trichloroethylene and +tetrachloroethylene at 303.15 K - comparison with theories

H. Iloukhani^a; B. Samiey^b

^a Faculty of Chemistry, University of Bu-Ali Sina, Hamadan, Iran ^b Faculty of Science, Department of Chemistry, University of Lorestan, Khoramabad, Iran

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Speeds of sound and isentropic compressibilities of binary mixtures of 1-heptanol + 1,2-dichloroethane, +1,1,1-trichloroethane, +1,1,2,2-tetrachloroethane, +trichloroethylene and +tetrachloroethylene at 303.15 K – comparison with theories

H. ILOUKHANI*[†] and B. SAMIEY[‡]

[†]Faculty of Chemistry, University of Bu-Ali Sina, Hamadan, Iran

[‡]Faculty of Science, Department of Chemistry, University of Lorestan, PO Box 465, Khoramabad, Iran

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Densities ρ , and speeds of sound u , of the binary mixtures of 1-heptanol with 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, trichloroethylene, and tetrachloroethylene have been measured over the complete composition range at 303.15 K. Isentropic compressibility κ_s , and Isentropic compressibility deviations $\Delta\kappa_s$ were calculated, and were correlated by a Redlich–Kister type function to determine the fitting parameters and the SDs. The results are consistent with the self-association of 1-heptanol and the polar and nonpolar characters of used chloroethanes or chloroethylenes, which produce the dissociation of 1-heptanol. Speeds of sound and isentropic compressibilities have been compared with calculated values from Jacobson free length theory, Schaaff collision factor theory, Nomoto relation, Junjie relation and Van Deal ideal mixing relation.

Keywords: Speed of sound; Isentropic compressibilities; Chloroethylenes; Chloroethanes; 1-Heptanol

1. Introduction

This study is a continuation of our earlier work related to the study of thermodynamic and acoustic properties of binary mixtures [1–4]. In recent years, measurements of thermodynamic, acoustic and transport properties have been employed in understanding the nature of molecular systems and physico-chemical behavior

*Corresponding author. Tel./Fax: +0098 0811 8271061. Email: iloukhani@basu.ac.ir

in liquid mixtures. The nonrectilinear behavior of the aforementioned properties of liquid mixtures with changing mole fractions is attributed to the difference in size of the molecules and strength of interactions. Here, we report densities ρ , speeds of sound u , isentropic compressibilities κ_s , and isentropic compressibility deviations $\Delta\kappa_s$, of 1-heptanol binary mixtures with 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, trichloroethylene and tetrachloroethylene at 303.15 K. This work will also provide a test of various empirical equations to correlate speeds of sound and isentropic compressibilities of the above mentioned binary mixtures.

2. Experimental

2.1. Materials

1-Heptanol ($\geq 99\%$), 1,2-dichloroethane ($> 99\%$), and 1,1,1-trichloroethane ($\geq 99\%$) were purchased from Merck. 1,1,2,2-Tetrachloroethane ($\geq 99\%$), trichloroethylene (99%), and tetrachloroethylene ($> 99\%$) were obtained from Fluka. All compounds used were purified by distillation using a 1 m fractionation column and stored in a glove prior to use. The purity of each compound was further ascertained by comparing the densities 298.15 K. Table 1 gives the density measurements, which agree with the values obtained from the literature [5–7].

2.2. Apparatus and procedure

Densities of the pure liquids and their mixtures were measured with an Anton Paar digital densimeter (Model DMA 4500) operated in the static mode and capable of a precision of better than $\pm 10^{-5} \text{ g cm}^{-3}$ and automatically thermostated within $\pm 0.01 \text{ K}$.

Speeds of sound at 303.15 K at a frequency of 1 MHz were determined using a quartz crystal interferometer (Mittal Enterprises, New Delhi, India). The uncertainty in the speed of sound measurement was $\pm 1 \text{ m s}^{-1}$. The temperature in the cell was regulated to $\pm 0.01 \text{ K}$.

Table 1. Source, purity grades, and density ρ of pure component 298.15 K.

Component	$\rho \text{ (g cm}^{-3}\text{)}$		Source	Purity (mass %)
	Experimental	Literature		
1-Heptanol	0.81909	0.8186 ^a	Merck	99.0
1,2-Dichloroethane	1.24564	1.24561 ^b	Merck	99.0
1,1,1-Trichloroethane	1.31455	1.32963 ^b	Merck	99.0
1,1,2,2-Tetrachloroethane	1.58585	1.58683 ^b	Fluka	99.0
Trichloroethylene	1.45465	1.45572 ^c	Fluka	99.5
Tetrachloroethylene	1.61420	1.61309 ^c	Fluka	99.0

^a[5].

^b[6].

^c[7].

3. Results and discussion

3.1. Speeds of sound and isentropic compressibility deviations

Isentropic compressibility was calculated using Laplace–Newton equation

$$\kappa_s = \frac{1}{\rho u^2} \quad (1)$$

The data of experimental ρ , u , and κ_s are listed in table 2. Speeds of sound and isentropic compressibilities of the binary mixtures were fitted to the following polynomial equation:

$$Q = \sum_{i=1}^2 \sum_{j=1}^m B_{ij} x_i^j \quad (2)$$

where Q stands for u or κ_s and x_i is the mole fraction of component i in the binary mixture. The adjustable coefficients B_{ij} and SDs are presented in table 3. The speeds of sound were also calculated using Jacobson free length theory (FLT) [8,9], Schaaff collision factor theory (CFT) [10], Nomoto relation (NR) [11], Junjie relation (JR) [12], and Van Deal ideal mixing relation (IMR) [13,14].

The speeds of sound in the n -component mixture are given by the equations following:

(i) Jacobson free length theory (FLT)

$$u = \frac{K}{L_f \rho^{1/2}} \quad (3)$$

The value of K for the individual n -component mixture has been taken as [15]

$$K = \sum_{i=1}^n K_i x_i \quad (4)$$

where the value of K_i for each pure component in the mixture are obtained from equation (3).

The free length of the mixture L_f is obtained by

$$L_f = \frac{2(V - \sum_{i=1}^n x_i V_{0,i})}{\sum_{i=1}^n x_i Y_i} \quad (5)$$

where $V_{0,i}$ is the molar volume of pure component i at absolute zero temperature, Y_i the surface area per mole for pure component i and V is the molar volume of mixture. For spherical molecules the surface area per mole of the pure liquid is given by

$$Y_i = \left(36\pi N_A V_{0,i}^2\right)^{1/3} \quad (6)$$

Table 2. Speeds of sound, isentropic compressibilities and isentropic compressibility deviations for binary mixtures at 303.15 K.

x_1	u (m s ⁻¹)	κ_s (T Pa ⁻¹)	$\Delta\kappa_s$ (T Pa ⁻¹)	x_1	u (m s ⁻¹)	κ_s (T Pa ⁻¹)	$\Delta\kappa_s$ (T Pa ⁻¹)
1-Heptanol + 1,2-dichloroethane							
0.0000	1168	591.9	0.0	0.5451	1220	710.8	33.9
0.1156	1169	634.1	18.6	0.6027	1232	712.1	29.1
0.2024	1173	660.4	19.6	0.6949	1250	714.6	22.6
0.3011	1183	681.9	35.8	0.7970	1272	713.7	12.6
0.3272	1187	685.6	35.7	0.8849	1290	713.0	4.8
0.3944	1196	695.9	36.9	1.0000	1308	716.7	0.0
0.5056	1215	705.8	33.3				
1-Heptanol + 1,1,1-trichloroethane							
0.0000	959	832.4	0.0	0.5537	1131	786.5	27.5
0.1036	982	838.3	22.0	0.6039	1151	777.0	23.4
0.2046	1012	831.0	29.2	0.6786	1180	764.7	18.8
0.3039	1044	820.1	31.6	0.7402	1205	753.8	13.9
0.3013	1062	811.0	28.4	0.7920	1225	746.4	11.4
0.3987	1077	806.6	29.9	0.9032	1269	730.2	5.3
0.5031	1115	790.5	25.9	1.0000	1308	716.6	0.0
1-Heptanol + 1,1,2,2-tetrachloroethane							
0.0000	1141	486.7	0.0	0.5489	1195	634.1	4.9
0.0938	1141	517.8	3.1	0.6002	1205	644.9	4.6
0.2016	1145	551.6	6.8	0.6963	1228	661.5	1.3
0.3005	1154	578.9	8.2	0.7994	1254	679.5	-0.9
0.4146	1169	607.4	8.7	0.8970	1280	696.8	-1.7
0.5099	1187	626.6	6.0	1.0000	1308	716.7	0.0
1-Heptanol + trichloroethylene							
0.0000	1012	674.2	0.0	0.5549	1160	722.3	19.7
0.0937	1026	699.9	19.0	0.6022	1176	721.3	17.0
0.1993	1052	712.7	25.9	0.7082	1213	717.7	9.7
0.2671	1070	718.5	28.2	0.8021	1244	716.8	5.8
S0.3050	1082	718.9	26.8	0.8686	1266	716.1	3.1
0.4021	1112	720.9	24.4	0.8928	1274	716.4	2.7
0.5059	1145	721.4	20.7	1.0000	1308	716.7	0.0
1-Heptanol + tetrachloroethylene							
0.0000	1025	593.3	0.0	0.5529	1150	683.4	12.3
0.1026	1036	622.5	12.4	0.5981	1163	688.7	12.4
0.2029	1055	642.4	17.1	0.6923	1194	696.2	9.6
0.2878	1075	655.1	17.6	0.7982	1233	701.6	4.0
0.4069	1106	670.1	16.8	0.8963	1270	708.0	0.8
0.4960	1131	680.1	15.8	1.0000	1308	716.7	0.0

Table 3. Parameter values of equation (2), B_{ij} , and the corresponding SDs for binary mixtures at 303.15 K.

System	B_{11}	B_{12}	B_{13}	B_{21}	B_{22}	B_{23}	σ
$\sigma(u)$ (m s ⁻¹)							
1-Heptanol + 1,2-dichloroethane	1578	-134	-135	738	415	15	1
1-Heptanol + 1,1,1-trichloroethane	1515	-131	-76	629	336	-7	1
1-Heptanol + 1,1,2,2-tetrachloroethane	1541	-148	-85	712	419	11	0
1-Heptanol + trichloroethylene	1545	-118	-119	654	341	16	1
1-Heptanol + tetrachloroethylene	1517	-135	-74	649	362	13	1
$\sigma\kappa_s$ (T Pa ⁻¹)							
1-Heptanol + 1,2-dichloroethane	1093.9	-445.2	66.9	388.3	341.4	-138.5	0.9
1-Heptanol + 1,1,1-trichloroethane	1119.8	-461.2	58.9	484.0	442.1	-91.1	1.8
1-Heptanol + 1,1,2,2-tetrachloroethane	1049.8	-400.0	66.3	261.8	259.0	-35.0	0.7
1-Heptanol + trichloroethylene	1081.2	-460.8	97.7	407.8	381.6	-112.1	1.4
1-Heptanol + tetrachloroethylene	1073.7	-427.1	70.2	339.6	318.8	-64.1	0.9

N_A is Avogadro number. The molar volume of pure component i at absolute zero temperature $V_{0,i}$ is obtained by the Sugden formula

$$V_{0,i} = V_i \left(1 - \frac{T}{T_C}\right)^{0.3} \tag{7}$$

where T_C and V_i are the critical temperature and molar volume of pure component i .

(ii) Schaaff collision factor theory (CFT)

$$u = u_\infty \frac{(\sum_{i=1}^n x_i S_i)(\sum_{i=1}^n x_i B_i)}{V} = u_\infty S r_f \tag{8}$$

where $u_\infty = 1600 \text{ m s}^{-1}$, S = collision factor and $r_f = B/V$, space filling factor. B is the actual volume of the molecules per mole. For n -component mixtures S and B are related to pure component values through

$$S = \sum_{i=1}^n x_i S_i \tag{9}$$

$$B = \sum_{i=1}^n x_i B_i \tag{10}$$

B_i can be evaluated as

$$B_i = \frac{4}{3} \pi r^3 N_A \tag{11}$$

where r is the molecular radius of the pure component. Molecular radius is calculated as

$$r = \left[\frac{3b}{16\pi N_A} \right]^{1/3} \tag{12}$$

where b is the van der Waals constant.

(iii) Nomoto relation (NR)

$$u = \left[\frac{\sum_{i=1}^n x_i R_i}{\sum_{i=1}^n x_i V_i} \right]^3 \tag{13}$$

$$R_i = V_i u^{1/3} \tag{14}$$

where R_i and u_i stand for Rao molar sound velocity and speed of sound of the pure component i , respectively.

(iv) Junjie relation (JR)

$$u = \frac{\sum_{i=1}^n x_i V}{(\sum_{i=1}^n x_i M_i)^{1/2} (\sum_{i=1}^n x_i V_i / \rho_i u_i^2)^{1/2}} \tag{15}$$

(v) Van Deal ideal mixing relation (IMR)

$$\left(\frac{1}{\sum_{i=1}^n x_i M_i} \right) \frac{1}{u^2} = \sum_{i=1}^n \frac{x_i}{M_i u_i^2} \quad (16)$$

Table 4 lists various basic parameters of the pure components namely molar volume, V^0 , molar volume at absolute zero, V_0 , available volume, V_a , free length L_f , molar surface factor, Y , molecular radius, r , molar actual volume of molecule, B , and collision factor, S at 293.15 K.

The calculated speeds of sound of the binary mixtures are presented in table 5. The SDs for speeds of sound $\sigma(u)$ and isentropic compressibilities $\sigma(\kappa_s)$ predicted by FLT, CFT, NR, JR, and IMR are given in table 6.

Isentropic compressibility deviations $\Delta\kappa_s$, were calculated using the relation

$$\Delta\kappa_s = \kappa_s - \sum_{i=1}^2 x_i \kappa_{s,i} \quad (17)$$

where κ_s is the isentropic compressibility of mixture and $\kappa_{s,i}$ is the isentropic compressibility of the component i . The corresponding $\Delta\kappa_s$, values of binary mixtures of [x_1 1-heptanol + $(1 - x_1)$ chloroethanes or chloroethylenes] measured at 303.15 K are also presented in table 2 and are plotted against mole fraction of 1-heptanol in figure 1. Each set of results were fitted using a Redlich–Kister polynomial [16] which for binary mixtures is

$$\Delta\kappa_s = x_1(1 - x_1) \sum_{k=0}^N A_k (1 - 2x_1)^k \quad (18)$$

where $\Delta\kappa_s$ are deviation values of isentropic compressibility deviations and A_k is the polynomial coefficient. A_k values were obtained by a nonlinear least-squares fitting procedure.

In each case, the optimum number of coefficients was ascertained from an examination of the variation of SD σ with

$$\sigma = \left[\frac{\sum (\Delta\kappa_{s,\text{exp}} - \Delta\kappa_{s,\text{cal}})}{(n - p)} \right]^{1/2} \quad (19)$$

Table 4. Molar volume V^0 , molar volume at absolute zero (V_0), available volume (V_a), free length (L_f), molar surface factor (Y), molecular radius (r), molar actual volume of molecule (B), and collision factor (S) of the pure components at 293.15 K.

Component	$(10^{-6} \text{ m}^3 \text{ mol}^{-1})$			$(\text{m}^2 \text{ mol}^{-1})$	(10^{-10} m)			
	V^0	V_0	V_a	B	Y	r	L_f	S
1-Heptanol	141.2646	117.1999	24.0647	53.7689	977,912	2.7727	0.4963	2.2036
1,2-Dichloroethane	78.9921	63.2794	15.7127	26.9934	648,420	2.2039	0.4840	2.2094
1,1,1-Trichloroethane	100.8520	80.0034	20.8487	32.9347	758,148	2.3550	0.5501	1.8966
1,1,2,2-Tetrachloroethane	105.3243	88.3472	16.9771	29.4178	809,985	2.2680	0.4204	2.6136
Trichloroethylene	89.8086	71.2036	18.6023	28.1523	701,499	2.2354	0.5305	2.0835
Tetrachloroethylene	102.2058	84.3530	17.8529	33.8536	785,384	2.3767	0.4534	1.9926

Table 5. Experimental and predicted speeds of sound by means of FLT, CFT, NR, JR, and IMR theories for binary mixtures at 303.15 K.

x_1	u (m s ⁻¹)	u_{FLT} (m s ⁻¹)	u_{CFT} (m s ⁻¹)	u_{NR} (m s ⁻¹)	u_{JR} (m s ⁻¹)	u_{IMR} (m s ⁻¹)
1-Heptanol + 1,2-dichloroethane						
0.0000	1168	1170	1168	1165	1168	1168
0.1156	1169	1175	1190	1191	1189	1178
0.2024	1173	1183	1205	1208	1204	1187
0.3011	1183	1195	1220	1226	1219	1198
0.3272	1187	1198	1225	1230	1223	1201
0.3944	1196	1209	1235	1240	133	1209
0.5056	1215	1227	1251	1256	1248	1223
0.5451	1220	1234	1256	1261	1254	1229
0.6027	1232	1244	1264	1268	1261	1237
0.6949	1250	1259	1275	1279	1273	1251
0.7970	1272	1276	1287	1289	1285	1269
0.8849	1290	1290	1296	1298	1296	1285
1.0000	1308	1310	1308	1308	1308	1308
1-Heptanol + 1,1,1-trichloroethane						
0.0000	959	958	959	959	959	959
0.1036	982	990	998	1003	996	985
0.2046	1012	1023	1053	1044	1031	1012
0.3039	1044	1057	1070	1083	1066	1041
0.3013	1062	1073	1087	1100	1082	1055
0.3987	1077	1090	1104	1118	1098	1070
0.5031	1115	1127	1141	1154	1135	1104
0.5537	1131	1145	1158	1172	1152	1121
0.6039	1151	1164	1176	1188	1169	1139
0.6786	1180	1191	1201	1213	1195	1167
0.7402	1205	1214	1222	1232	1217	1191
0.7920	1225	1233	1239	1248	1235	1212
0.9032	1269	1274	1276	1281	1274	1261
1.0000	1308	1311	1308	1308	1308	1308
1-Heptanol + 1,1,2,2-tetrachloroethane						
0.0000	1141	1143	1141	1140	1141	1141
0.0938	1141	1144	1172	1159	1145	1151
0.2016	1145	1152	1204	1181	1153	1164
0.3005	1154	1164	1228	1199	1163	1176
0.4146	1169	1182	1252	1219	1178	1192
0.5099	1187	1200	1269	1236	1193	1207
0.5489	1195	1208	1275	1242	1200	1214
0.6002	1205	1218	1282	1250	1210	1222
0.6963	1228	1239	1292	1265	1229	1240
0.7994	1254	1263	1301	1280	1253	1261
0.897	1280	1286	1306	1294	1278	1283
1.0000	1308	1310	1308	1308	1308	1308
0.0000	1012	1013	1012	1008	1012	1012
0.0937	1026	1042	1048	1047	1041	1033
0.1993	1052	1076	1086	1087	1072	1059
0.2671	1070	1098	1109	1111	1092	1075
0.3050	1082	1110	1122	1124	1103	1085
0.4021	1112	1141	1152	1155	1132	1111
0.5059	1145	1174	1183	1187	1162	1140
0.5549	1160	1189	1197	1200	1176	1155
0.6022	1176	1203	1210	1213	1190	1169
0.7082	1213	1234	1238	1241	1221	1203
0.8021	1244	1260	1262	1264	1249	1234
0.8686	1266	1278	1278	1279	1269	1258
0.8928	1274	1284	1284	1285	1276	1267
1.0000	1308	1310	1308	1308	1308	1308

(Continued)

Table 5. Continued.

x_1	u (m s ⁻¹)	u_{FLT} (m s ⁻¹)	u_{CFT} (m s ⁻¹)	u_{NR} (m s ⁻¹)	u_{JR} (m s ⁻¹)	u_{IMR} (m s ⁻¹)
1-Heptanol + tetrachloroethylene						
0.0000	1025	1024	1205	1024	1025	1025
0.1026	1036	1043	1057	1060	1047	1048
0.2029	1055	1065	1087	1094	1070	1071
0.2878	1075	1085	1113	1121	1091	1092
0.4069	1106	1117	1147	1156	1121	1122
0.4960	1131	1143	1173	1182	1145	1146
0.5529	1150	1160	1189	1198	1161	1162
0.5981	1163	1174	1201	1210	1174	1176
0.6923	1194	1203	1227	1234	1203	1204
0.7982	1233	1238	1255	1261	1237	1238
0.8963	1270	1271	1281	1284	1271	1271
1.0000	1308	1310	1308	1308	1308	1308

Table 6. The SDs of predicted speeds of sound and isentropic compressibilities by means of FLT, CFT, NR, JR, and IMR theories of binary mixtures at 303.15 K.

System	FLT	CFT	NR	JR	IMR
$\sigma(u)$ (m s ⁻¹)					
1-Heptanol + 1,2-dichloroethane	9	27	32	26	9
1-Heptanol + 1,1,1-trichloroethane	10	20	31	16	9
1-Heptanol + 1,1,2,2-tetrachloroethane	10	55	35	5	16
1-Heptanol + trichloroethylene	22	27	31	14	6
1-Heptanol + tetrachloroethylene	8	28	37	10	12
$\sigma(\kappa_s)$ (T pa ⁻¹)					
1-Heptanol + 1,2-dichloroethane	10.7	30.9	35.0	29.3	9.9
1-Heptanol + 1,1,1-trichloroethane	14.5	28.8	42.3	22.8	11.8
1-Heptanol + 1,1,2,2-tetrachloroethane	9.9	52.8	34.9	5.5	15.9
1-Heptanol + trichloroethylene	26.6	35.2	37.5	17.4	7.6
1-Heptanol + tetrachloroethylene	9.9	34.6	39.6	12.3	13.7

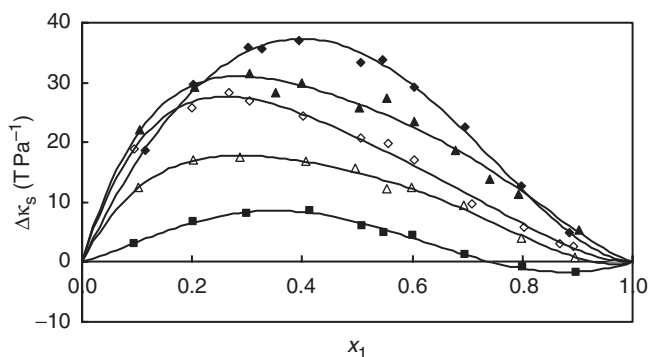


Figure 1. Isentropic compressibility deviations vs. mole fraction of 1-heptanol for binary mixtures of 1-heptanol with (◆) 1,2-dichloroethane, (▲) 1,1,1-trichloroethane, (■) 1,1,2,2-tetrachloroethane, (◇) trichloroethylene, and (△) tetrachloroethylene at 303.15 K. The solid curves were calculated from equation (18) using the coefficients from table 7.

Table 7. Parameters of adjustable coefficient A_i in equation (18) and the SDs σ , in equation (19) for binary mixtures at 303.15 K.

System	A_0	A_1	A_2	A_3	σ
1-Heptanol + 1,2-dichloroethane	141.0	79.2	-39.6	16.3	0.9
1-Heptanol + 1,1,1-trichloroethane	105.9	68.0	63.9	68.9	1.5
1-Heptanol + 1,1,2,2-tetrachloroethane	26.9	46.1	-27.4	-18.0	0.5
1-Heptanol + trichloroethylene	83.1	86.1	57.3	55.7	0.9
1-Heptanol + tetrachloroethylene	59.7	46.4	18.7	52.9	0.7

where $\Delta\kappa_s$ is the calculated values of isentropic compressibility deviations and n and p are the number of experimental points and number of parameters retained in the respective equations, respectively. The adjustable coefficients and SDs are given in table 7. As seen from figure 1, the $\Delta\kappa_s$ values are large and positive over the entire range of composition for mixtures of 1-heptanol with 1,2-dichloroethane, 1,1,1-trichloroethane, trichloroethylene and tetrachloroethylene and for mixtures of 1-heptanol with 1,1,2,2-trichloroethane change sign from positive in the 1,1,2,2-trichloroethane rich region to negative in 1-heptanol rich regions. The positive $\Delta\kappa_s$ values indicate that the mixed species are overall more compressible and hence have large volumes due to the structure disruption effect of chloroethanes or chloroethylenes on 1-heptanol.

The negative $\Delta\kappa_s$ values, in 1-heptanol rich region of the binary mixture containing 1,1,2,2-trichloroethane indicate the dominance of structure making polar interactions between -OH group of 1-heptanol and hydrogen atoms of 1,1,2,2-trichloroethane molecules.

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