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Excess isentropic compressibility and speed of sound of the ternary mixture 2-propanol + diethyl ether + n-hexane and the constituent binary mixtures at 298.15 K

GOKHAN SAVAROGLU* and ERTUNC ARAL
Department of Physics, Osmangazi University, 26480, Eskisehir, Turkey
*Corresponding author. E-mail: gsavarog@ogu.edu.tr

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Abstract. Speed of sound and densities of the ternary mixture 2-propanol + diethyl ether + n-hexane and also the binary mixtures 2-propanol + diethyl ether and 2-propanol + n-hexane have been measured at the entire composition range at 298.15 K. The excess isentropic compressibilities and the excess speed of the sound have been calculated from experimental densities and speed of sound. These excess properties of the binary mixtures were fitted to Redlich–Kister equation, while the Cibulka's equation was used to fit the values related to the values to the ternary system. These excess properties have been used to discuss the presence of significant interactions between the component molecules in the binary mixtures and also the ternary mixtures.

Speed of sound of the binary mixtures and the ternary mixture have been compared with calculated values from free length theory (FLT), collision factor theory (CFT), Nomoto's relation (NR), Van Deal's ideal mixing relation (IMR) and Junjie's relation (JR). The results are used to compare the relative merits of these theories and relations in terms of the root mean square deviation relative ($RMSD_r$).

Keywords. Densities; speed of sound; isentropic compressibilities; binary and ternary mixtures; 2-propanol; diethyl ether; n-hexane.

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1. Introduction

Because ethers are used as oxygenating agents in gasoline technology, the thermodynamic properties of hydrocarbons or alcohols + ether systems have been intensely studied [1–4]. The density, refractive index and speed of sound of the binary mixtures diethyl ether + n-hexane at $T = 298.15$ K can be found in literature [1].

Acoustic and thermodynamic parameters derived from these parameters have been used to understand different kinds of association, the molecular pacing, molecular motion, and various types of intermolecular interactions and their strength,

influenced by the size in pure components and in the mixtures. Therefore, this work was undertaken in order to understand possible association and intermolecular interaction in 2-propanol + diethyl ether + n-hexane and the constituent binary mixtures.

The present study reports measurements of densities, speed of sound for 2-propanol+diethyl ether, 2-propanol + n-hexane and 2-propanol + diethyl ether + n-hexane at 298.15 K and atmospheric pressure. The experimental data were used to calculate the isentropic compressibilities, κ_s, excess isentropic compressibilities, κ_s^E, and excess speed of sound, u^E. These properties have been used to understand different kinds of association in pure components as well as in the binary and the ternary mixtures.

The data of binary systems were fitted to the Redlich–Kister equation [5] and the same was done for the ternary mixture through Cibulka's equation [6]. Because one has to deal with the liquid state, theoretical models have been used to predict the speed of sound and the isentropic compressibility. Free length theory (FLT) [7,8], collision factor theory (CFT) [9], Nomoto's relation (NR) [10], Van Deal's ideal mixing relation (IMR) [11] and Junjie's relation (JR) [12] have been used in this research work to compare their predictions with the present experimental values at 298.15 K.

2. Experimental

The compounds used were 2-propanol (>99 mass%), diethyl ether (>99.5 mass%) and n-hexane (>99 mass%) obtained from Merck. The purity of these compounds were checked by comparing measured densities and the speed of sound with those reported in literature. These compounds were used as received without further purification. The pure compound densities and speed of sound and isentropic compressibilities at 298.15 K are shown in table 1 together with literature values [1,13–19].

The mixtures in this research were determined by measurement of mass. All the mass measurements were preformed using an electronic balance (Scaltec, SBC22)

Table 1. Physical properties of the pure components at 298.15 K.

	2-Propanol		n-Hexane		Diethyl ether	
Property	Exp.	Lit.	Exp.	Lit.	Exp.	Lit.
ρ (g cm^{-3})	0.7813	0.7812[a] 0.78126[b] 0.78089[c]	0.6550	0.65493[f] 0.65484[b]	0.7083	0.7077[h]
u (m/s^{-1})	1140	1139[d] 1141[e] 1140.1[a]	1078	1077.7[f] 1076.42[g]	983	980[h]
κ_s (T Pa^{-1})	985		1314		1461	

[a]Ref. [13], [b]ref. [18], [c]ref. [14], [d]ref. [15], [e]ref. [16], [f]ref. [17], [g]ref. [19], [h]ref. [1].

accurate to 0.01 mg. The precision of the mole fraction is estimated to be better than $\pm 1 \times 10^{-4}$. The densities and speed of sound of both pure liquid and mixture were measured using a vibrating tube densimeter and sound analyser, Anton Paar DSA-5000, automatically thermostated at ±0.01 K. The calibration of the apparatus was done with air and deionised double-distilled water.

3. Results and discussion

The values of density, ρ, speed of sound, u, isentropic compressibilities, κ_s, excess isentropic compressibilities, κ_s^E and excess speed of sound, u^E, for 2-propanol + diethyl ether, 2-propanol + n-hexane shown in table 2 and for 2-propanol + diethyl ether + n-hexane are shown in table 3 at 298.15 K and atmospheric pressure. The values of density, ρ, speed of sound, u, isentropic compressibilities, κ_s, excess isentropic compressibilities, κ_s^E, of diethyl ether + n-hexane have been reported in [1]. The isentropic compressibilities, κ_s, has been calculated using the Newton–

Table 2. Experimental densities, ρ, experimental speed of sound, u, and calculated κ_s, κ_s^E, and u^E for 2-propanol (1) + diethyl ether (2) and 2-propanol (1) + n-hexane (3) binary mixtures at 298.15 K.

x_1	ρ (g cm^{-3})	u (m s^{-1})	κ_s (T Pa^{-1})	κ_s^E (T Pa^{-1})	u^E (m s^{-1})
2-Propanol (1) + diethyl ether (2)					
0.1139	0.7156	998	1403	−23	8
0.2221	0.7230	1011	1353	−38	12
0.2769	0.7268	1019	1325	−46	15
0.3866	0.7354	1035	1269	−59	20
0.5060	0.7441	1055	1207	−68	25
0.5915	0.7501	1068	1169	−66	26
0.6779	0.7559	1081	1132	−59	24
0.8041	0.7658	1104	1071	−46	21
0.9065	0.774	1123	1024	−27	13
0.9500	0.7772	1131	1006	−15	8
2-Propanol (1) + n-hexane (3)					
0.0977	0.6662	1075	1299	1	−3
0.1938	0.6717	1070	1300	20	−9
0.2979	0.6824	1070	1280	21	−11
0.4174	0.6957	1074	1246	17	−10
0.5132	0.7068	1079	1215	13	−9
0.6092	0.7186	1086	1180	9	−7
0.7145	0.7324	1095	1139	8	−6
0.8031	0.7449	1105	1100	7	−4
0.9035	0.7625	1122	1042	0	−1
0.9466	0.7704	1130	1017	−1	0

Table 3. Experimental, ρ, u, κ_s, κ_s^E, and u^E for the ternary mixture 2-propanol (1) + diethyl ether (2) + n-hexane (3) at 298.15 K.

x_1	x_2	ρ (g cm^{-3})	u (m s^{-1})	κ_s (T Pa^{-1})	κ_s^E (T Pa^{-1})	u^E (m s^{-1})
0.0696	0.8725	0.7091	999	1413	−15	5
0.0705	0.8130	0.7053	1003	1409	−9	3
0.0480	0.0859	0.6626	1065	1331	15	−6
0.0500	0.0700	0.6604	1068	1328	14	−4
0.1044	0.7912	0.7087	1010	1383	−26	9
0.1310	0.5940	0.6965	1019	1383	8	−3
0.1084	0.5640	0.6939	1026	1369	−2	1
0.1100	0.4499	0.6862	1032	1368	14	−5
0.1135	0.3671	0.6833	1042	1348	6	−3
0.0970	0.2637	0.6734	1045	1360	27	−10
0.1216	0.1757	0.6700	1055	1341	25	−8
0.1187	0.0504	0.6688	1068	1311	12	−6
0.1146	0.0287	0.6674	1070	1309	11	−6
0.2133	0.6867	0.7166	1021	1339	−36	11
0.2455	0.5115	0.7086	1034	1320	−20	6
0.2009	0.4718	0.6965	1029	1356	12	−4
0.2081	0.3846	0.6914	1037	1345	15	−5
0.2292	0.2494	0.6881	1047	1326	19	−9
0.2294	0.1778	0.6821	1053	1322	26	−10
0.2150	0.0800	0.6812	1066	1292	7	−6
0.3291	0.5696	0.7208	1032	1303	−32	11
0.4000	0.4502	0.7167	1044	1280	−18	10
0.3305	0.3521	0.7051	1044	1301	2	−1
0.3348	0.2522	0.6984	1048	1304	20	−8
0.3222	0.1667	0.6915	1055	1299	24	−10
0.3090	0.1190	0.687	1058	1300	29	−12
0.4354	0.4632	0.7292	1045	1256	−36	12
0.4380	0.3471	0.718	1050	1263	−9	3
0.4238	0.2568	0.7100	1055	1265	3	−2
0.4416	0.1614	0.7075	1064	1249	4	−4
0.4274	0.0695	0.6971	1066	1262	26	−11
0.5383	0.3697	0.7365	1063	1202	−44	17
0.5500	0.2100	0.7292	1070	1198	−20	5
0.5303	0.1644	0.7194	1070	1214	−5	0
0.5304	0.0628	0.7131	1075	1213	10	−7
0.6146	0.3002	0.7421	1072	1173	−39	15
0.6335	0.1560	0.7322	1078	1175	−8	2
0.6285	0.0732	0.7248	1082	1178	6	−4
0.7398	0.1681	0.7518	1095	1109	−32	14
0.7340	0.0638	0.7398	1094	1129	−1	0
0.8263	0.0725	0.7559	1107	1080	−10	5
0.8746	0.0749	0.7635	1115	1054	−12	7
0.8553	0.0350	0.7570	1112	1068	−2	1
0.9082	0.0435	0.7681	1123	1032	−11	6

Laplace equation:

$$\kappa_{\mathrm{s}} = \frac{1}{\rho u^2}. \tag{1}$$

The excess isentropic compressibilities, $\kappa_{\mathrm{s}}^{\mathrm{E}}$, were estimated by means of the equation:

$$\kappa_{\mathrm{s}}^{\mathrm{E}} = \kappa_{\mathrm{s}} - \kappa_{\mathrm{s}}^{\mathrm{id}}, \tag{2}$$

where the isentropic compressibilities for the ideal mixture, $\kappa_{\mathrm{s}}^{\mathrm{id}}$, was calculated by the following relation [20,21]:

$$\kappa_{\mathrm{s}}^{\mathrm{id}} = \sum_{i=1}^{n} \phi_i \left\{ \kappa_{\mathrm{s},i} + \frac{TV_i(\alpha_i)^2}{C_{\mathrm{p},i}} \right\} - \frac{T\left(\sum_{i=1}^{n} x_i V_i\right)\left(\sum_{i=1}^{n} \phi_i \alpha_i\right)^2}{\sum_{i=1}^{n} x_i C_{\mathrm{p},i}}. \tag{3}$$

ϕ_i is defined by the relation:

$$\phi_i = \frac{x_i V_i}{\left(\sum_i x_i V_i\right)}. \tag{4}$$

V_i, α_i and $C_{\mathrm{p},i}$ are molar volume, cubic expansion coefficient, and the molar heat capacity of pure components, respectively.

The excess speed of sound, u^{E}, is estimated in binary and ternary mixtures using the following expression:

$$u^{\mathrm{E}} = u - u^{\mathrm{id}} = u - (\rho^{\mathrm{id}} \kappa_{\mathrm{s}}^{\mathrm{id}})^{-1/2}, \tag{5}$$

where ρ^{id} is the density of the corresponding ideal mixture [22].

$$\rho^{\mathrm{id}} = \sum_i \phi_i \rho_i. \tag{5a}$$

These calculated properties, isentropic compressibilities, κ_{s}, excess isentropic compressibilities, $\kappa_{\mathrm{s}}^{\mathrm{E}}$, and excess speed of sound, u^{E}, for the binary and the ternary mixtures are given in tables 2 and 3.

Excess isentropic compressibilities, $\kappa_{\mathrm{s}}^{\mathrm{E}}$, and excess speed of sound, u^{E}, of the binary mixtures were fitted to a Redlich–Kister equation [5]:

$$\Delta Q = x(1-x) \sum_{k=0}^{m} A_k (2x-1)^k, \tag{6}$$

where ΔQ is $\kappa_{\mathrm{s}}^{\mathrm{E}}$ or u^{E}, x_i is the mole faction of the component i, A_k is the polynomial coefficient, k is the number of polynomial coefficients, and m is the number of parameters.

The ternary excess and deviation magnitudes were fitted by means of the expression introduced by Cibulka [6].

Table 4. Values of the parameters A_i of the Redlich–Kister equation and C_i of Cibulka's equation, and the corresponding root mean square deviations σ, at 298.15 K.

2-Propanol (1) + diethyl ether (2)					
κ_s^E (T Pa^{-1})	$A_0 = -261.8$	$A_1 = -57.4$	$A_2 = 8.3$		$\sigma = 1.3$
u^E (m s^{-1})	$A_0 = 98.1$	$A_1 = 51.5$	$A_2 = -29.1$	$A_3 = -10.1$	$\sigma = 0.4$
	$A_4 = 91.4$				
2-Propanol (1) + n-hexane (3)					
κ_s^E (T Pa^{-1})	$A_0 = 49.3$	$A_1 = -107.8$	$A_2 = 233.1$	$A_3 = 143.3$	$\sigma = 1.4$
	$A_4 = -434.0$				
u^E (m s^{-1})	$A_0 = -35.2$	$A_1 = 35.1$	$A_2 = -56.2$	$A_3 = -30.1$	$\sigma = 0.2$
	$A_4 = 112.9$				
Diethyl ether (2) + n-hexane (3)					
κ_s^E (T Pa^{-1})[a]	$A_0 = 120.6$	$A_1 = 24.0$	$A_2 = 4.6$		$\sigma = 0.2$
u^E (m s^{-1})[b]	$A_0 = -44.2$	$A_1 = 0.1$	$A_2 = 6.3$	$A_3 = -41.3$	$\sigma = 0.3$
	$A_4 = -45.7$				
2-Propanol (1) + diethyl ether (2) + n-hexane (3)					
κ_s^E (T Pa^{-1})	$C_1 = 9.3$	$C_2 = 2.9$	$C_3 = -19.3$		$\sigma = 8.4$
u^E (m s^{-1})	$C_1 = -130.6$	$C_2 = -305.5$	$C_3 = 495.7$		$\sigma = 2.4$

[a]Obtained from literature [1]; [b]calculated from literature data [1].

$$\Delta_{123} = \Delta Q_{12} + \Delta Q_{13} + \Delta Q_{23} + x_1 x_2 (1 - x_1 - x_2)(C_1 + C_2 x_1 + C_2 x_2), \tag{7}$$

where ΔQ is κ_s^E or u^E, $\Delta Q = \Delta Q_{12} + \Delta Q_{13} + \Delta Q_{23}$ (with ΔQ having the same expression as of eq. (6)) and C_i are adjustable coefficients. C_i was obtained by the method of least squares along with the corresponding root mean square deviations, σ. The coefficients A_k of eq. (6) and adjustable parameters C_i of eq. (7), are presented in table 4.

The root mean square deviations were calculated using the expression

$$\sigma = \left[\frac{\sum_i^N (Z_{\text{exp}} - Z_{\text{pred}})^2}{N} \right]^{1/2}, \tag{8}$$

where Z is the value of the property and N is the number of experimental data.

The excess isentropic compressibilities, κ_s^E, and excess speed of sound, u^E, on the composition of 2-propanol + diethyl ether and 2-propanol + n-hexane at 298.15 K are plotted in figures 1a and 1b respectively. The curves of constant belonging to both κ_s^E and u^E for the ternary system are shown in figure 2. The ternary contributions of eq. (7) to κ_s^E and u^E are represented in figures 3a and 3b, respectively.

The excess isentropic compressibilities, κ_s^E, are positive over the entire composition for 2-propanol + n-hexane and diethyl ether + n-hexane; these excess properties are negative over the entire composition for 2-propanol + diethyl ether.

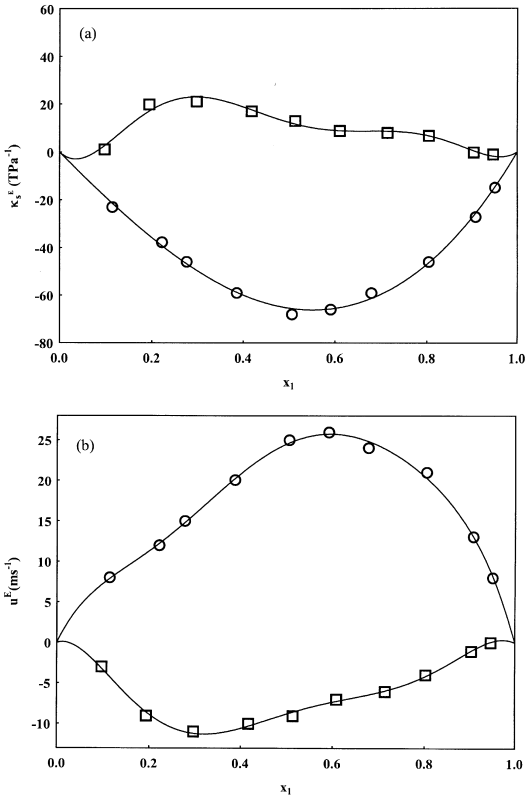

Figure 1. (**a**) Excess isentropic compressibilities, κ_s^E, and (**b**) excess speed of sound, u^E, for the binary mixtures at 298.15 K: 2-propanol (1) + diethyl ether (2) (○), 2-propanol (1) + n-hexane (3) (□), (—) Redlich–Kister's correlation.

For the same binary mixtures, excess speed of sound, u^E, is negative over the entire composition range for 2-propanol + n-hexane and diethyl ether + n-hexane and it is positive over the complete mole fraction range for 2-propanol + diethyl ether.

4. Theoretical

The empirical and semi-empirical theories of Jacobson, Schaaff, Nomoto, Van Deal and Junjie were used to obtain the speed of sound in the ternary and constituent binary liquid mixtures.

4.1 *The Jacobson's free length theory* (*FLT*) [7, 8]

$$u_m = \frac{K}{L\rho_m^{1/2}}. \tag{9}$$

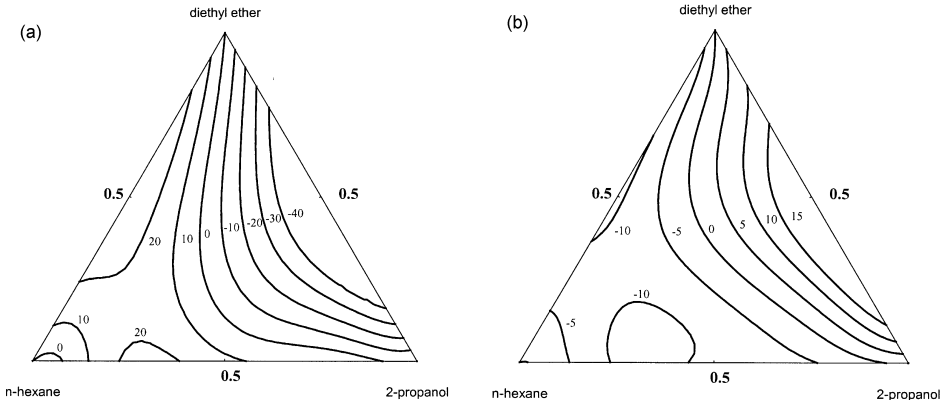

Figure 2. Curves of constant: (**a**) excess isentropic compressibilities, $\kappa_{\mathrm{s}}^{\mathrm{E}}$ and (**b**) excess speed of sound, u^{E} (m s^{-1}) for 2-propanol (1) + diethyl ether (2) + n-hexane (3) at 298.15 K.

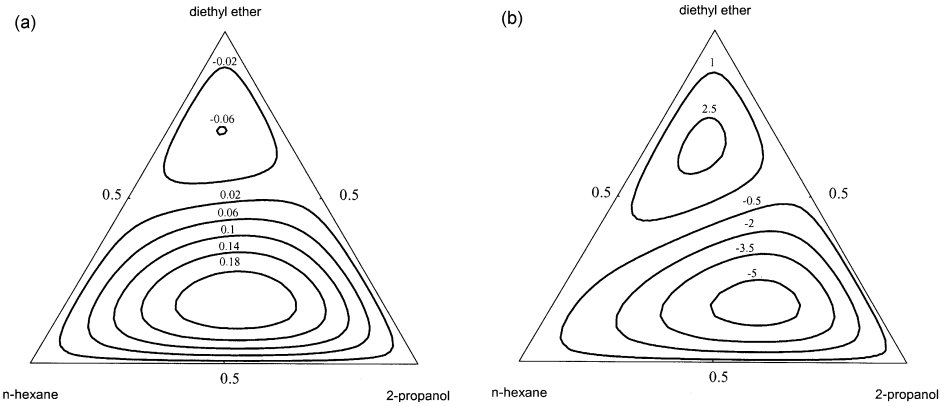

Figure 3. Curves of constant ternary contributions to: (**a**) excess isentropic compressibilities, $\kappa_{\mathrm{s}}^{\mathrm{E}}$, (**b**) excess speed of sound, u^{E}, at 298.15 K for 2-propanol (1) + diethyl ether (2) + n-hexane (3).

Here, K is a temperature dependent constant whose value at 298.15 K is 625, ρ_{m} is the density of the mixture and L is the free length of the mixture. Here

$$L = \frac{2\left((1/\rho) - \sum_{i=1}^{3}(w_i V_{0\mathrm{i}}/M_i)\right)}{\sum_{i=1}^{3}(w_i Y_i/M_i)}, \tag{9a}$$

where M_i is the molecular weight of component i, w_i is the weight fraction of component i, Y_i and V_{0i} are the surface area per mole and the molar volumes at absolute zero of temperature of component i, respectively. Y_i and V_{0i} are given by

$$Y = (36\pi N_{\mathrm{A}} V_{0i}^2)^{1/3}, \tag{9b}$$

$$V_{0i} = V_T(1 - T/T_{\mathrm{c}})^{0.3}, \tag{9c}$$

where V_T and T_c are respectively the molar volume of a pure component at temperature T K and the critical temperature.

4.2 *Schaaff's collision factor theory* [9]

$$u_m = u_\infty \frac{\left(\sum_{i=1}^{3} x_i s_i\right)\left(\sum_{i=1}^{3} x_i B_i\right)}{V_m}, \tag{10}$$

where $u_\infty = 1600$ m/s, s_i is the space filling factor of component i in the mixture. V_m, the molar volume of the mixture, can be evaluated as

$$V_m = \frac{\sum_{i=1}^{3} x_i M_i}{\rho_m}, \tag{10a}$$

where x_i is the mole fraction. B_i, the actual volume of the molecule per mole of component i, can be evaluated as

$$B = 4/3\pi r^3 N_A, \tag{10b}$$

where N_A is the Avogadro's number and r is the molecular radius of the pure component. Molecular radius is calculated as

$$r = \left[\frac{3b}{16\pi N}\right]^{1/3}, \tag{10c}$$

where b is the Van der Waals constant.

4.3 *Nomoto's relation* [10]

$$u_m = \left[\frac{\sum_{i=1}^{3} x_i R_i}{\sum_{i=1}^{3} x_i V_i}\right]^3, \tag{11}$$

where R_i and V_i are respectively the molar sound velocity and molar volume of component i in the mixture.

4.4 *Van Deal's ideal mixing relation* [11]

$$\left[\frac{1}{\sum_{i=1}^{3} x_i M_i}\right] \frac{1}{u_m^2} = \sum_{i=1}^{3} \frac{x_i}{M_i u_i^2}, \tag{12}$$

where M_i and u_i are respectively the molecular weight and speed of sound of the component i.

4.5 *Junjie's relation* [12]

$$u_{\mathrm{m}} = \frac{\sum_{i=1}^{3}(x_i V_i)}{\sum_{i=1}^{3}(x_i M_i)^{1/2}\left[\sum_{i=1}^{3}(x_i V_i/\rho_i u_i^2)\right]^{1/2}}. \tag{13}$$

The root mean square deviation relative ($\mathrm{RMSD_r}$) between the experimental and calculated speed of sound and the isentropic compressibilities for FLT, CFT, NR, IMR is defined by the equation:

$$\mathrm{RMSD_r} = \left[\frac{1}{m}\sum\left(\frac{Z^{\mathrm{teor}} - Z^{\mathrm{exp}}}{Z^{\mathrm{exp}}}\right)^2\right]^{1/2}, \tag{14}$$

where m is the number of experimental points and Z is the value of the property.

Table 5 comprises the root mean square deviation relative to the speed of sound obtained by Jacobson's free length theory (FLT), Schaaff's collision factor theory (CFT), Nomoto's relation (NR), Van Deal's ideal mixing relation (IMR) and Junjie's relation (JR). A critical examination of table 5 shows that for the 2-propanol + diethyl ether mixture, minimum $\mathrm{RMSD_r}$ is observed in the case of IMR, followed by CFT, NR, FLT and JR. For 2-propanol + n-hexane mixture, IMR predicts the data with minimum $\mathrm{RMSD_r}$ followed by JR, NR, CFT and FLT. For the diethyl ether +n-hexane mixture, again IMR predicts the estimation of speed of sound with minimum $\mathrm{RMSD_r}$, followed by FLT, CFT, JR and NR. For 2-propanol + diethyl ether + n-hexane, speed of sound predicted by IMR and JR show minimum $\mathrm{RMSD_r}$ followed by CFT and NR, while maximum deviations are observed from FLT. Thus, the estimation of the speed of sound in all the presently investigated binary mixtures and the ternary mixture by IMR is reasonably well. The assumption for the formation of the ideal mixing relation is that, the ratio of specific heats of ideal mixtures and the volumes are also equal [23]. IMR makes accurate predictions of the speed of sound in all the studied binary mixtures and the ternary mixture.

5. Conclusion

For the 2-propanol + diethyl ether mixture, negative values of excess isentropic compressibilities, $\kappa_{\mathrm{s}}^{\mathrm{E}}$, and positive values of the excess speed of sound, u^{E}, are

Table 5. Root mean square deviation relative to the predicted speed of sound by means of FLT, CFT, NR, IMR and JR theories.

	$\mathrm{RMSD_r}$ (%)				
298.15 K	u_{FLT}	u_{CFT}	u_{NR}	u_{IMR}	u_{JR}
2-Propanol (1) + diethyl ether (2)	0.8	0.3	0.4	0.2	1.2
2-Propanol (1) + n-hexane (2)	2.9	1.7	1.6	1.1	1.1
Diethyl ether (2) + n-hexane (3)[a]	0.3	0.4	0.9	0.2	0.8
2-Propanol (1) + diethyl ether (2) + n-hexane (3)	1.7	1.2	1.2	0.8	0.8

[a]Calculated from literature data [1].

explained by the interactions between 2-propanol and diethyl ether. The magnitude and sign of excess isentropic compressibilities are reflections of the type of interaction taking place in the mixture. These interactions are proposed to occur via complex formation between the two species or n–π interaction. Diethyl ether has been known to participate in cross-association effect with the hydrogen present in alcohol. Positive values of excess isentropic compressibilities, $\kappa_{\mathrm{s}}^{\mathrm{E}}$, and negative values of the excess speed of sound, u^{E}, for diethyl ether + n-hexane and 2-propanol + n-hexane mixtures give a more packing effect in 2-propanol + diethyl ether mixture. Some information about diethyl ether + n-hexane mixture has been published previously [1]. The effective packing is less when the mixture is 2-propanol + diethyl ether, than when the mixture is diethyl ether + n-hexane because the effective packing is less when the length of chain of 2-propanol is higher. For 2-propanol + n-hexane mixture, the accommodation effect of n-hexane molecules is stronger than the dissociation of hydrogen bonding, or self-association in 2-propanol except for the mixture with low 2-propanol mole fraction.

For the ternary mixture the observed values of the excess isentropic compressibilities, $\kappa_{\mathrm{s}}^{\mathrm{E}}$, and the excess speed of sound, u^{E}, could be explained in terms of the same effects as a function of the mole fraction of the components.

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