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Thermodynamic properties of binary mixtures containing 1-alkanols

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Abstract Densities (ρ) of pure liquids and their mixtures have been measured at 303.15 and 313.15 K and atmospheric pressure over the entire composition range for the binary mixtures of benzylalcohol with 1-propanol, 1-butanol, 1-pentanol, and 1-hexanol by using Rudolph Research Analytical digital densitometer (DDM-2911 model). Further, the ultrasonic sound velocities for the above said mixtures were also measured at 303.15 and 313.15 K. The measured density data were used to compute excess molar volumes (V^E) and these were compared with the values obtained by Hwang equation. Isentropic compressibility (κ_S) and excess isentropic compressibilities (κ_S^E) were evaluated from experimental sound velocity and density data. Moreover, the experimental sound velocities were analyzed in terms of theoretic models namely, collision factor theory and free length theory. The experimental results were discussed in terms of intermolecular interactions between component molecules.

Keywords Density · Excess volume · Sound velocity · Theoretic analysis · Molecular interaction

Introduction

The important role played by solvents in chemistry has long been recognized. Mixing effects for a large number of compounds and their mixtures used in the petroleum processing industry are rather difficult be known, hence knowledge of thermodynamic properties such as densities, ultrasonic sound velocity, viscosity and refractive index of many organic mixtures at various temperatures is of great importance and consideration. Mixing of the compounds with different and complex molecular structure causes various intermolecular interactions, resulting in non-ideal behavior [1–5]. We report here the excess volume and excess isentropic compressibilities data of binary mixture of benzylalcohol with 1-propanol, 1-butanol, 1-pentanol, and 1-hexanol at 303.15 and 313.15 K. The organic liquids that were used in the present investigation are having many industrial applications. Benzylalcohol is an important as a solvent for gelatin, cellulose acetate, shellac [6, 7]. 1-Alkanols are interesting simple examples of biologic and industrial important amphiphilic materials [8]. Further, it has been reported [9–11] that the strength of association in alkanols decreases as the carbon chain length in the molecule increases for 1-propanol, 1-butanol, 1-pentanol, and 1-hexanol. A survey of the literature has shown that the thermodynamic properties for binary liquid mixtures of benzylalcohol with aromatic hydrocarbons [12], 1-propanol, and 1-octanol [13] were reported earlier. To our best of knowledge, no systematic studies on excess volumes (V^E) and excess isentropic compressibility (κ_S^E) were reported for a series of 1-alkanols with benzylalcohol, and hence, we report here new V^E and κ_S^E data for the binary mixtures of benzylalcohol with 1-alkanols (c_3–c_6). Further, the experimental sound velocity data were also compared with theoretic models proposed by Schaaff's collision factor theory (CFT) [14] and Jacobson's free length theory (FLT) [15, 16]. The present study was under taken to know the effect of temperature and chain length of 1-alkanols when mixed with benzylalcohol that may induce changes in sign and magnitude of excess thermodynamic functions.

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Table 1 Densities (ρ), sound velocity (u), thermal coefficient (α), and heat capacity (c_p) for pure component liquids at temperatures from 298.15 to 313.15 K along with the literature values

ρ/g cm^{-3}			u/m s^{-1}			
T/K	Exp.	Lit.	Exp.	Lit.	α/kK^{-1}	C_p/J mol^{-1} k^{-1}
Benzylalcohol						
303.15	1.03760	1.03700 [12]	1,514	1,511 [12]	0.7402	224.35 [24]
313.15	1.02691		1,519		0.7456	227.62
1-Propanol						
303.15	0.79559	0.79597 [18]	1,192	1,189 [18]	0.76827	147.80 [18]
313.15	0.78720	0.78770 [18]	1,158	1,155 [18]	0.53083	153.58 [18]
1-Butanol						
303.15	0.80206	0.80209 [18]	1,226	1,222 [18]	0.94756	177.10 [18]
313.15	0.79409	0.79430 [18]	1,191	1,189 [18]	0.89640	184.28 [18]
1-Pentanol						
303.15	0.80656	0.80650 [19]	1,262	1,258 [19]	0.89945	212.26 [19]
313.15	0.79905	0.79905 [19]	1,225	1,225 [19]	1.09881	220.94 [19]
1-Hexanol						
303.15	0.81215	0.81212 [19]	1,287	1,288 [19]	0.80783	245.30 [19]
313.15	0.80451	0.80451 [19]	1,254	1,252 [19]	0.98693	255.50 [19]

Experimental

Materials and apparatus

All the chemicals used in the present study were of analytical reagent grade procured from S.D.Fine chemicals Ltd., India and Merck, and their purities were as follows: benzylalcohol 99.5 %, 1-propanol 99.5 %, 1-butanol 99.5 %, 1-pentanol 99.6 %, and 1-hexanol 99.5 %. Prior to experimental measurements, all the liquids were purified as described in the literature [17]. The purity samples were attained by fractional distillation, and the purity of chemicals were checked by comparing the measured densities and ultrasonic sound velocities, which were in good agreement with the literature values [12, 18, 19] and these are given in Table 1. The purity of the sample was further confirmed by GLC single sharp peak. Before use, the chemicals were stored over 0.4 nm molecular sieves for about 72 h to remove water and were later degassed.

All the binary liquid mixtures are prepared by weighting an appropriate amount of pure liquids and an electronic balance (Afoset, ER-120A, India) with a precision of ±0.1 mg by syringing each component into airtight stopper bottles to minimize evaporation losses. The uncertainty of the mole fraction was $\pm 1 \times 10^{-4}$. After mixing the sample, the bubble free homogenous sample was transferred into the U-tube of the densimeter through a syringe. The density measurements were reported earlier [20] with a Rudolph Research Analytical digital densimeter (DDM-2911 Model). A multi frequency ultrasonic interferometer (M-82 Model, Mittal Enterprise, New Delhi, India) operated at 2 MHz, was used to measure the ultrasonic velocities of the binary liquid mixtures at 303.15 and 313.15 K as described earlier in the literature [21, 22].

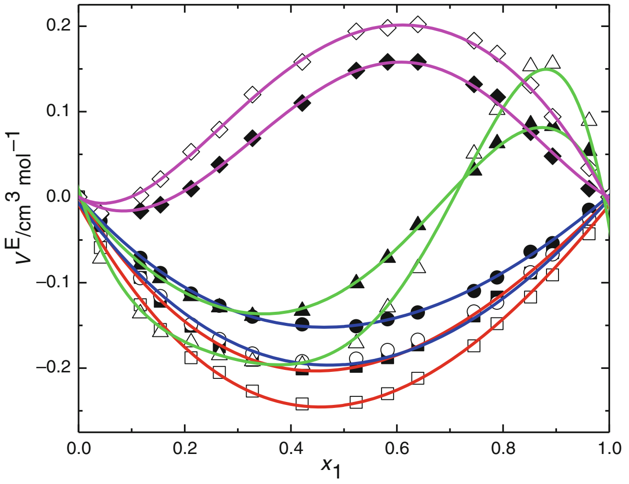

Fig. 1 Variation of excess volume (V^E) of the binary liquid mixture of benzylalcohol with 1-propanol (*filled square*), 1-butanol (*filled circle*), 1-pentanol (*filled triangle*), 1-hexanol (*filled diamond*) at 303.15 K and benzylalcohol with 1-propanol (*unfilled square*), 1-butanol (*unfilled circle*), 1-pentanol (*unfilled triangle*), 1-hexanol (*unfilled diamond*) at 313.15 K, respectively

Results and discussion

The experimental values of densities (ρ) and ultrasonic sound velocities (u) were used to calculate excess molar volume (V^E) and excess isentropic compressibilities (κ_S^E) for

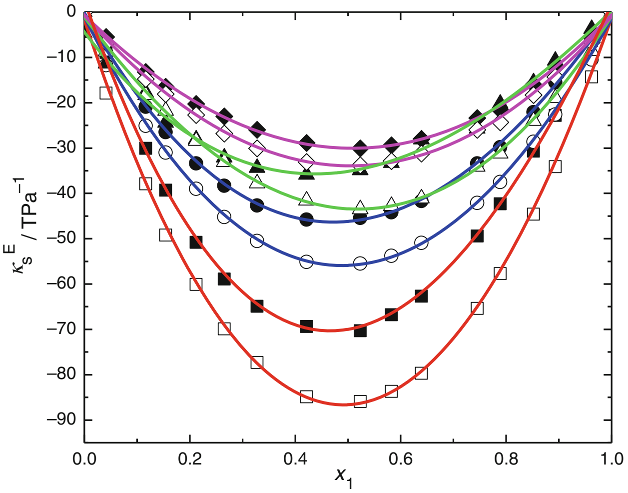

Fig. 2 Variation of excess isentropic compressibility (κ_S^E) of the binary liquid mixture of benzylalcohol with 1-propanol (*filled square*), 1-butanol (*filled circle*), 1-pentanol (*filled triangle*), 1-hexanol (*filled diamond*) at 303.15 K and benzylalcohol with 1-propanol (*unfilled square*), 1-butanol (*unfilled circle*), 1-pentanol (*unfilled triangle*), 1-hexanol (*unfilled diamond*) at 313.15 K, respectively

the binary liquid mixtures of benzylalcohol with 1-alcohols (c_3–c_6) at 303.15 and 313.15 K. The V^E data are graphically represented in Fig. 1 and κ_S^E data in Fig. 2 for all the binary mixtures at two temperatures. The excess molar volume data of all the binary mixtures were calculated from the measured densities by using the following equation

$$V^E = [x_1M_1 + x_2M_2]/\rho - [x_1M_1/\rho_1 + x_2M_2/\rho_2] \quad (1)$$

where ρ is the density of the mixture, and x_1, M_1, ρ_1 and x_2, M_2, ρ_2 are the mole fractions, molecular masses and densities of pure components 1 and 2, respectively.

From the result of ultrasonic sound velocities (u) and densities (ρ), isentropic compressibilities (κ_S) were calculated as

$$\kappa_S = u^{-2}\rho^{-1} \quad (2)$$

The corresponding excess isentropic compressibilities (κ_S^E) were obtained from the relation [23]:

$$\kappa_s^E = \kappa_s - \kappa_s^{id} \quad (3)$$

where κ_S^{id} is the ideal value of the isentropic compressibility and was calculated from the following equation [23]:

$$\kappa_s^{id} = \sum_{i=1}^{2} \varphi_i\left[\kappa_{s.i} + TV_i\left(\alpha_i^2\right)/C_{p.i}\right] - \left\{T\left(\sum_{i=1}^{2} x_iV_i\right)\left(\sum_{i=1}^{2} \varphi_i\alpha_i\right)^2 \Big/ \sum_{i=1}^{2} x_iC_{p.i}\right\} \quad (4)$$

Here, C_{pi} and α_i are the molar heat capacity and the thermal expansion coefficient of the ith component, respectively. The value of C_{pi} and α_i obtained and evaluated from the literature [18, 19, 24] and these are mentioned in Table 1.

An examination of V^E data given in Fig. 1 show that it is negative over the entire composition range for the binary mixtures containing 1-propanol and 1-butanol whereas V^E data exhibit an inversion in sign in the mixtures containing 1-pentanol and 1-hexanol. The sign of excess volume (V^E) of system depends on the relative magnitude of expansion and contraction of two liquids. If the factors that cause expansion in volume dominate the factors creating contraction, then V^E becomes positive. On the other hand, if the contractive factors dominate the expansive factors than V^E become negative.

The factors those are responsible for expansion in volume, which are as follows:

(i) Loss of dipolar association i.e. rupturing of H-bonding of component by the other or breaking up of associates held by weaker forces, namely dipole–dipole or dipole-induced dipole interactions or by Vander Waals forces. (ii) The geometry of molecular structure which does not allow fitting of the one component into others. (iii) Steric hindrance which opposes proximity of the constituent molecules.

While the negative V^E values arise due to dominance of the following factors:

(i) Chemical interactions between constituent molecules, such as hetero molecule association through the formation of H-bond known as strong specific interactions. (ii) Accommodation of molecules of one component into the interstitials of the molecules of the other components.

(iii) Geometry of the molecular structure that favors fitting of the component molecules with each other [25]. From the Fig. 1, it is evident that the factors which are responsible for negative V^E values which are dominant in the mixtures of benzylalcohol with 1-propanol and 1-butanol. On the other hand, the factors those are responsible for both the positive and negative V^E data which are competing with each other in the binary mixtures containing 1-pentanol and 1-hexanol. Further, it is observed that the magnitude of negative V^E data decreases with increase in composition of benzylalcohol. According to Marcus [11], the molecules of 1-alkanols are associated through hydrogen bonding in pure state. When these alkanol molecules are mixed with polar molecule like benzylalcohol would induce mutual dissociation of the hydrogen-bonded structures present in pure alkanols with subsequent formation of strong intermolecular hydrogen bonding (O…H–O), relatively weak hydrogen bonding (π–H) between π-electrons of the benzene ring of benzylalcohol and protons of 1-alkanols [13]. Further, the curves in Fig. 1 also reveal that as the chain length of 1-alkanol molecule increases from 1-propanol to 1-hexanol, the negative excess volume tends to shift toward positive excess volume which implies that dipole–dipole interactions are becoming weak in higher 1-alkanols owing to the decrease in their polarizability [26]. Also the positive excess volumes suggest that the higher 1-alkanols pose less proton donating ability than the lower alkanols, and hence hetero-association effects decree in the binary mixtures with an increase of chain length of linear alkanols [27, 28]. The algebraic values of V^{E} for the mixtures of benzylalcohol with 1-alkanols fall in the order:

1-hexanol > 1-pentanol > 1-butanol > 1-propanol

A perusal of κ_S^E data in Fig. 2 suggests that the property is negative over the entire composition range for all the binary mixtures of benzylalcohol with 1-alkanols at both the temperatures. The κ_S^E values ascribed to the changes in intermolecular free space defined by Jacobson. These changes occur due to structure-breaking and structure-making effects of the components and the consequent change in geometrical factors. Structure-breaking effect contributes to increases in free space between the molecules and these results in the sound waves covering shorter distance in mixtures. This leads to the positive deviation in compressibilities. On the other hand, structure-making effects would contribute to decrease in free space and a negative deviation in isentropic compressibility. The actual deviation would depend upon the balance between the two opposing effects. The experimental results indicate that the structure-making effect is dominated in all binary mixtures of benzylalcohol with 1-alkanols. The algebraic κ_S^E values fall in the order:

1-hexanol > 1-pentanol > 1-butanol > 1-propanol

The above order indicates that as the chain length of alkanol increases, the hetero-association decreases due to decrease in polarity of alcohols molecules. Further, it is concluded that algebraic values of excess volume (V^{E}) and excess isentropic compressibility (κ_S^E) decrease not only the increasing the chain of 1-alkanols but also decrease in polarizability values of 1-propanol (3.09 D); 1-butanol (1.75 D); 1-pentanol (1.7 D); 1-hexanol (1.55 D) [17].

An examination of κ_S^E data in Fig. 2 suggests that mixing of benzylalcohol with 1-alkanols leads to interstitial accommodational molecules. This leads to decreasing intermolecular free space between component molecules and formation of closer molecular aggregates [29]. The effects of increasing temperature appear the increasing κ_S^E values suggesting the increasing the specific molecular interactions. κ_S^E data become more negative which may be due to high thermal dissociation of hetero-association in liquid mixtures and more interstitial accommodation of one component into another [30]. Over the entire investigated conditions, the absolute values of excess volumes and excess isentropic compressibilities are increased with increasing the temperature in all the binary systems of benzylalcohol with 1-alkanols.

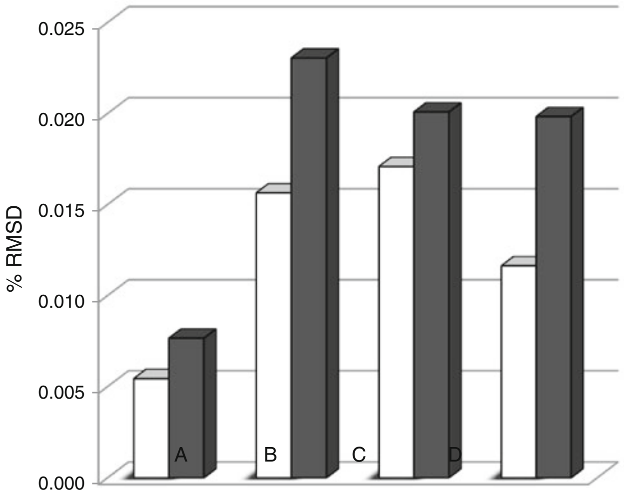

Fig. 3 Graphical representations of theoretical models FLT (*unfilled square*) and CFT (*filled square*) of benzylalcohol with 1-propanol (**a**); 1-butanol (**b**); 1-pentanol (**c**); 1-hexanol (**d**) at 303.15 K

Experimental speeds of sound were analyzed in terms of the CFT [14] and FLT [15, 16]. The pure component data namely, the molar volume (V_m), molar volume at absolute zero (V_0), molar available volume (V_a), free length (L_f), surface area (Y), collision factor (S), average molecular radius (r_m), actual volume of molecules per mole (B), and molecular sound velocity (R), which were used to calculate the above said theories were collected from the literature [31]. The methods and details of calculation of theories were discussed earlier [32]. The details of various theories and relevant equations are given as follows: A comparison between experimental speed of sound and theoretic speed of sound values suggests that the model proposed by Schaaff's CFT gives better estimation of sound velocity data. The methods of calculation of these theories were

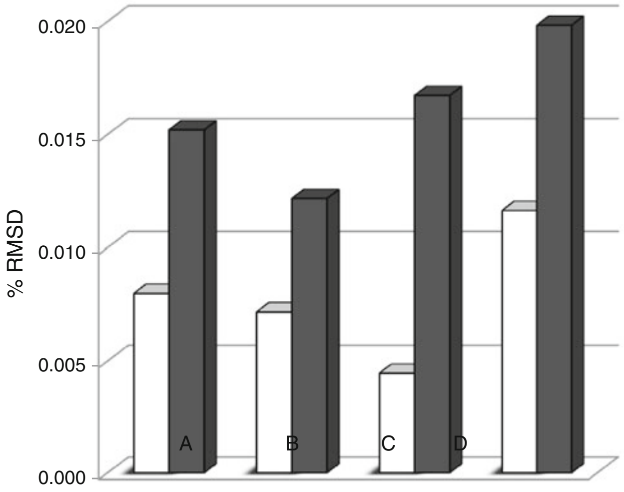

Fig. 4 Graphical representations of theoretic models FLT (*unfilled square*) and CFT (*filled square*) of benzylalcohol with 1-propanol (**a**); 1-butanol (**b**); 1-pentanol (**c**); 1-hexanol (**d**) at 313.15 K

Table 2 Coefficients a_i of Redlich–Kister Eq. (6) and the corresponding standard deviations (σ) of all the binary systems

Temperature/K	Function	a_0	a_1	a_2	σ (y^E)
Benzyl alcohol (1)+1-propanol (2)					
	V^E				
303.15		−0.797	0.157	−0.043	0.001
313.15		0.917	0.176	−0.388	0.005
	κ_S^E				
303.15		−294.27	34.62	78.747	2.264
313.15		−335.61	18.62	−77.932	1.727
Benzyl alcohol (1)+1-butanol (2)					
	V^E				
303.15		−0.621	0.085	0.073	0.003
313.15		−0.781	0.102	−0.081	0.002
	κ_S^E				
303.15		−183.86	23.73	−11.89	0.497
313.15		−213.65	6.34	−67.95	1.615
Benzyl alcohol (1)+1-pentanol (2)					
	V^E				
303.15		−0.443	1.069	0.854	0.003
313.15		−0.668	1.832	1.271	0.001
	κ_S^E				
303.15		−137.68	−10.93	−43.21	0.982
313.15		−165.26	−12.51	−54.13	1.403
Benzyl alcohol (1)+1-hexanol (2)					
	V^E				
303.15		0.599	0.471	−0.771	0.007
313.15		0.791	0.662	−0.549	0.009
	κ_S^E				
303.15		−115.71	−4.671	−31.280	0.685
313.15		−128.96	−4.604	−44.952	1.039

described in earlier. The merits of these theories were compared in terms of relative root mean deviation by using the following formula [19].

$$\mathrm{RMSD} = \left[\frac{1}{n}\sum_{i=1}^{n}\left[\frac{y_{\mathrm{exp}} - y_{\mathrm{pred}}}{y_{\mathrm{exp}}}\right]^2\right]^{1/2} \quad (5)$$

It is evident from the Figs. 3 and 4 that the RMSD data for all the binary systems, CFT model gives better estimation in speed of sound for the liquid mixtures under the investigation.

The experimental V^E values and κ_S^E data have been fitted to Redlich–Kister type polynomial equation [33]

$$y^E = x_1 x_2 \sum_{i=0}^{n} a_i (x_1 - x_2)^i \quad (6)$$

where y^E are V^E and κ_S^E, and while x_1 and x_2 refer to the mole fractions of the pure components. The equation takes value from 0 to 2; a_i is the adjustable parameter of the function and is determined using the least-squares method. The corresponding standard deviations σ (y^E) have been computed using the relation.

$$\sigma\left(y^E\right) = \left[\Sigma\left(y^E_{\mathrm{exp}} - y^E_{\mathrm{cal}}\right)^2 \Big/ (m-n)\right]^{1/2} \quad (7)$$

where m is the total number of experimental points, and n is the number of coefficients in Eq. (6), and the standard deviations of all the binary mixtures have been presented in Table 2.

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