Excess thermodynamic and acoustic properties for the binary mixtures of Methyl Benzoate at \( T = (303, 308, 313, 318 \text{ and } 323) \) K

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Density and speed of sound data for (Methyl Benzoate + 1-Propanol), (Methyl Benzoate + 1-Butanol) and (Methyl Benzoate + 1-Pentanol) have been determined at temperatures = (303, 308, 313, 318 and 323) K. From this data, excess parameters like excess volume, \( V^E \), excess isentropic compressibility, \( K_s^E \), and excess intermolecular free length, \( L_f^E \), have been computed. The variations of these parameters with composition and temperature of the mixtures are discussed in terms of molecular interactions in these mixtures. The values of \( K_s^E \) and \( L_f^E \) for all the systems under study are negative.

Keywords: density; speed of sound; excess molar volume; excess isentropic compressibility

1. Introduction

In recent years, ultrasonic investigations find extensive applications in characterising the thermodynamic and physico-chemical aspects of binary and ternary liquid mixtures [1]. The thermodynamic and transport properties [2–4] of liquid mixtures provide important information with which to speculate the molecular liquid structure. These properties have been widely used to study the intermolecular interactions between various species present in the liquid mixtures. The excess thermodynamic functions [5] are sensitively dependent not only on the differences in intermolecular forces, but also on the differences in the size of the molecules. Several researchers [6–11] have measured the density, viscosity and velocity of sound for a wide range of binary mixtures containing alcohols as one of the components, and these properties were interpreted in terms of specific or nonspecific interactions. The study of excess values provides important information on molecular forces existing in the binary liquid mixtures. The variation of these excess values with temperatures and composition for mixtures containing polar molecules and hydrogen-bonded components may be complex due to a decrease or an increase in hydrogen-bonding interaction due to mixing; depending upon the nature of the liquids, whether they are polar or non-polar, the signs and magnitudes of these excess values can throw light on the strength of interactions. In continuation of our dielectric work [12–14], we report the values of density and speed of sound for (Methyl Benzoate + 1-Propanol), (Methyl Benzoate + 1-Butanol) and (Methyl Benzoate + 1-Pentanol) at temperatures \( T \) of 303, 308, 313, 318 and 323 K. Even though considerable work has been reported on

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the physicochemical properties of alcohols as one of the component in binary and ternary mixtures, the data on binary mixtures of alcohols with alkyl benzoates with temperature variation is scanty. The molecules with –OH group form associative liquids due to hydrogen bonding. The effect shown by the molecules with other functional groups on these molecules plays a vital role in understanding the behaviour of hydrogen bonding. Alcohols are strongly associated in solution because of dipole–dipole interaction and hydrogen bonding. They are of great importance for their vital role in chemistry, biology and studies on hydrogen bonding in liquid mixtures. Alcohols are widely used as solvents. On the other hand, alkyl benzoates are non-associated in solution, but are good hydrogen-bonding acceptors. They are widely used in perfumery, preservatives and pesticides. So in our present study, we have selected Methyl Benzoate as non-associative liquid and propanol, butanol and pentanol as associative liquid. Considering these aspects, the authors have made an attempt to report the influence of non-associative molecule on associative molecule in liquid state by calculating the excess properties of binary mixtures involving alcohols and Methyl Benzoate. The values of excess molar volume, $V^E$, excess isentropic compressibility, $K_s^E$, and excess intermolecular free length, $L_f^E$, were evaluated. The excess volumes, $V^E$, were correlated using the Redlich–Kister [15] equation to estimate the binary interaction parameters and standard errors.

2. Experimental procedure

2.1. Materials

Methyl Benzoate, 1-Propanol, 1-Butanol and 1-Pentanol of analytical reagent (AR) grade were obtained from Merck Co. Inc., Germany, with purities of >99% and were further purified by double distillation under reduced pressure and only middle fractions were collected. Before use, the chemicals were stored over 0.4 nm molecular sieves for 72 hours to remove water content and were then degassed. Further the purity of the chemicals was confirmed by gas–liquid chromatography (GLC) single sharp peaks. The density and viscosity were determined at a temperature of 303 K to evaluate the samples for the presence of large quantities of impurities.

2.2. Methods

All binary mixtures were prepared gravimetrically in air-tight bottles. The mass measurements were performed on a digital electronic balance (Mettler Toledo AB 135, Greifensee, Switzerland) with an uncertainty of ±0.00001 g. The binary mixtures were prepared just before use. The uncertainty in mole fraction was estimated to be less than ±0.0001. Caution was taken to prevent the evaporation of the solutions after preparation. The densities of the pure compounds and their mixtures were determined accurately using 10 ml specific gravity bottles. The overall accuracy in the density measurement was ±0.0001 kg m$^{-3}$. The speed of sound was measured with a single-crystal variable path interferometer (Mittal Enterprises, New Delhi, India) operating at a frequency of 2 MHz that had been calibrated with water and benzene. The uncertainty in the speed of sound was found to be ±0.5 m s$^{-1}$. In all property measurements, the temperature was controlled within ±0.1 K using a constant temperature bath (M/s Sakti Scientific Instruments Company, Chennai, India) by circulating water from the thermostat.

The measured values of density and speed of sound along with the literature data are given in Table 1. There is fairly good agreement between our data and the previously reported values.
Table 1. Experimental densities ($\rho$) and speed of sound (U) data for pure components with available literature values at 303 K, thermal expansion coefficients ($\alpha$) and specific heat capacity ($C_p$) values for the systems under study.

| Liquid          | $T$ (K) | Exp.      | Literature | Exp.  | Literature | $\alpha$ (k $K^{-1}$) | $C_p$ (J mol$^{-1}$ K$^{-1}$) | $C_p$ (J mol$^{-1}$ K$^{-1}$) |
|-----------------|---------|-----------|------------|-------|------------|------------------------|--------------------------------|--------------------------------|
| 1-Propanol      | 303     | 0.7981    | 0.7956$^a$| 1195.2| 1190.2$^b$| 1.237$^c$             | 149$^d$                        | 143.9$^e$  |
| 1-Butanol       | 303     | 0.8102    | 0.8018$^a$| 1217.4| 1225.8$^f$| 1.228$^c$             | 179$^d$                        | 177.2$^e$  |
| 1-Pentanol      | 303     | 0.8126    | 0.8127$^g$| 1248.4| 1254.9$^f$| 1.215$^c$             | 209$^d$                        | 208.1$^e$  |
| Methyl Benzoate | 303     | 1.0775    | 1.0787$^h$| 1399.4| 1392.0$^i$| 1.045$^e$             | 222$^d$                        | 221.3$^j$ 214 ± 45$^k$ |

Notes: $^a$Nikam et al. [16].
$^b$Palani et al. [17].
$^c$Derived from density data.
$^d$Estimated from group contribution method of Chueh–Swanson [18].
$^e$D.R. Lide et al. [19].
$^f$Palani et al. [20].
$^g$Riddick et al. [21].
$^h$Madhu Mohan et al. [22].
$^i$Aminabhavi et al. [23].
$^j$Y. Marcus et al. [24].
$^k$Carl Caleman et al. [25].
3. Results and discussion

The experimentally measured values of density, $\rho$, and speed of sound, $U$, at temperatures of (303, 308, 313, 318 and 323) K for (Methyl Benzoate + 1-Propanol), (Methyl Benzoate + 1-Butanol) and (Methyl Benzoate + 1-Pentanol) systems as a function of mole fraction of Methyl Benzoate are shown in Figures 1 and 2. For further information, please see the supplementary material.

The density values have been used to calculate the excess volumes, $V^E$, using the following equation:

$$V^E = \frac{X_1 M_1 + X_2 M_2}{\rho} - \left( \frac{X_1 M_1}{\rho_1} + \frac{X_2 M_2}{\rho_2} \right)$$

where $\rho$ is the density of the mixture and $X_1$, $M_1$ and $\rho_1$ and $X_2$, $M_2$ and $\rho_2$ are the mole fraction, molar mass and density of pure components 1 and 2, respectively. The density, $\rho$, and speed of sound, $U$, were used to calculate the isentropic compressibility, $K_s$, using the following relation:

$$K_s = \frac{1}{\rho U^2}$$

The excess values of isentropic compressibility were computed using

$$K_s^E = K_s - K_s^{id}$$

where $K_s^E$ is its excess value and $K_s$ represents the calculated value for the mixture; $K_s^{id}$ for an ideal mixture was calculated from the relation recommended by Benson and Kiyohara [26] and Douheret et al. [27]

$$K_s^{id} = \sum \phi_i \left( K_{s,i}^{o} + \frac{TV_i^{o} (a_i^{o})^2}{C_{p,i}^{o}} \right) - T \left( \sum x_i V_i^{o} \left( \frac{\sum \phi_i a_i^{o2}}{\sum x_i C_{p,i}^{o}} \right) \right)$$

in which $V_i^{o}$, $a_i^{o}$, $C_{p,i}^{o}$ are the molar volume, isobaric thermal expansion coefficient and molar isobaric heat capacity of pure component $i$, and $\phi_i$ is the volume fraction of $i$ in the mixture. The thermal expansion coefficient values were derived from the density data and the specific heat capacities were estimated from the group contribution method of Chueh–Swanson [18].

The excess values of free length, $L_f^E$, were calculated by using the expression

$$L_f^E = L_f - K_T (K_s^{id})^{1/2}$$

where $L_f$ represents the calculated value of free length for the mixture and $K_T$ represent a temperature dependent constant [28] whose value is $K_T = (91.368 + 0.3565T) \times 10^{-8}$.

The excess volumes, $V^E$, were fitted by the method of non-linear least-squares to a Redlich–Kister type polynomial [15].
Figure 1. Variation of density with respect to mole fraction at various temperatures for (a) Methyl Benzoate + propanol; (b) Methyl Benzoate + Butanol; and (c) Methyl Benzoate + Pentanol system.

\[ V^E = x_1(1-x_1)\sum_{i=1}^n A_i(2x_1 - 1)^{i-1} \]  \hspace{1cm} (6)
The values of coefficient $A_i$ were determined by a regression analysis based on the least-squares method and are reported along with the corresponding standard deviations between the experimental and the calculated values of the respective functions in Table 2.
It is observed from Figures 1 and 2 that the speed of sound and density values increase with increasing concentration of Methyl Benzoate in all the systems under study. This behaviour at such concentrations is different from the ideal mixtures behaviour and can be attributed to intermolecular interactions in the systems studied. Figures 3–11 show the dependence of $V^E$, $K_s^E$ and $L_f^E$ on composition. The values of $K_s^E$ and $L_f^E$ are negative for all the systems over the entire mole fraction range and their magnitude decreases with rise in temperature from 303 K to 323 K.

Figure 3. Variation of excess volumes with respect to mole fraction at various temperatures for Methyl Benzoate + Propanol system.
Alkanols are liquids which are associated through hydrogen bonding and in the pure state they exhibit equilibrium between multimer and monomer species. In order to understand the nature of interactions between the components of liquid mixtures, it is of interest to discuss the same in terms of excess parameter rather than actual values. Non-ideal liquid mixtures show considerable deviation from linearity in their physical behaviour with respect to concentration and these have been interpreted as arising from the presence of interaction forces between the components of the mixture.

Figure 4. Variation of excess volumes with respect to mole fraction at various temperatures for Methyl Benzoate + Butanol system.

Figure 5. Variation of excess volumes with respect to mole fraction at various temperatures for Methyl Benzoate + Pentanol system.
of strong or weak interactions. The effect of deviation depends upon the nature of constituents and composition of mixtures.

The deviations observed in the excess parameters indicate the strength of interactions present between the component molecules of the binary mixtures under study. In general, the values of excess thermodynamic functions are influenced by the following reasons:

Figure 6. Variation of excess isentropic compressibility with respect to mole fraction at various temperatures for Methyl Benzoate + Propanol system.

Figure 7. Variation of excess isentropic compressibility with respect to mole fraction at various temperatures for Methyl Benzoate + Butanol system.
The specific forces that exist between the molecules, like the charge transfer complexes and existence of hydrogen bonds, result in the negative excess values. Physical contributions comprised non-specific physical interactions like dispersion forces or weak dipole–dipole interactions. Structural characteristics, like the differences in the size and shape of the component molecules and their free volumes, cause the geometrical fitting of one component into the other.

Figure 8. Variation of excess iperatures for Methyl Benzoate + Pentanol system.

Figure 9. Variation of excess free length with respect to mole fraction at various temperatures for Methyl Benzoate + Propanol system.
The variation of excess molar volume, $V^E$, with respect to mole fraction, $x_1$, is shown in Figures 3–5 over the entire composition range and at different $T = (303, 308, 313, 318$ and $323)$ K. The $V^E$ values are negative mostly in the case of 1-propanol but the trend changed to positive as the alcohol chain increases, because when the alcohol chain increases, it has less proton donating ability, the self aggregates break easily resulting in positive values of molar volume. Similar behaviour of $V^E$ was observed by Garcio et al. [29] while experimenting with alkanols and alkyl benzoate systems at 298.15 K, considering the effect of temperature, the calculated values of $V^E$ are in good agreement. This
kind of behavior of $V^E$ can be attributed to the formation of H-bond, disruption of alcohol self-associations and the structural characteristics like geometrical fitting of one component into the other. It is clear from Figures 3–5 that the positive values of $V^E$ are in the following order: (Methyl Benzoate + Pentanol) > (Methyl Benzoate + Butanol) > (Methyl Benzoate + Propanol). Similar results were observed by earlier workers [29,30]. The expansion in molar volume can be attributed to the presence of weak intermolecular forces of attraction [30,31]. The values of $V^E$ decrease with the rise in temperature for the binary liquid mixtures under study as the interactions between unlike molecules become weaker due to rise in thermal motions.

Benson and Kiyohara [26] stated that the thermodynamic properties of an ideal mixture must be mutually related in the same way as those of pure substances and real mixtures. Douheret et al. [27] suggested that the interpretation of the nature of molecular interactions in mixtures requires a correct calculation of a thermodynamic property of the ideal liquid mixtures by the application of correct ideal mixing rules. So the excess values of isentropic compressibility were calculated by using the relation proposed by them.

Figures 6–8 display the variation of excess isentropic compressibility, $K^E_s$, for the binary liquid mixtures of Methyl Benzoate with Propanol or Butanol or Pentanol over the entire mole fraction range and at different $T = (303, 308, 313, 318, 323)$ K. It is clear from Figures 6–8 that the $K^E_s$ values are negative over the entire mole fraction range and the changes in $K^E_s$ values with respect to temperature are small in all the mixtures. The negative values of $K^E_s$ are of the order (Methyl Benzoate + Propanol) > (Methyl Benzoate + Butanol) > (Methyl Benzoate + Pentanol). The negative values of $K^E_s$ suggest that the mixtures are less compressible than the corresponding ideal mixtures, which signifies the H-bond formation, and other chemical effects like charge transfer forces and complex forming interactions. As the alkanol chain increases, the methylene group increases, which produces greater steric hindrance to the formation of hydrogen bonds between the Methyl Benzoate and alkanol molecule. So, a relatively weaker hydrogen bonding is expected in Methyl Benzoate + Butanol and Methyl Benzoate + Pentanol systems which are reflected from the less negative values of $K^E_s$ in these mixtures. It is observed that there is a decrease in magnitude of negative $K^E_s$ and $V^E$ with the rise in temperature. This suggests that the interactions between unlike components tend to reduce due to increase in thermal motions.

Fort and Moore [32] suggested that the liquids having different molecular sizes and shapes mix well thereby reducing the volume which causes the values of $K^E_s$ to be negative. It also suggests that the liquids are less compressible when compared with their ideal mixtures signifying the chemical effects including charge transfer forces, formation of H-bond and other complex forming interactions. It can also be said that the molecular interactions are strong in these binary liquid mixtures and that the medium is highly packed. Similar results were obtained by earlier workers [33,34].

The sign of excess isentropic compressibility plays a vital role in assessing the compactness due to molecular interaction in liquid mixtures through charge transfer, dipole–dipole interactions and dipole-induced dipole interactions, interstitial accommodation and orientational ordering [35] leading to a more compact structure-making, which enhances the excess isentropic compressibility to have negative values. The negative excess value of isentropic compressibility in the present investigation is an indication of strong interaction in the liquid mixtures [36].

It can be observed from Figures 9–11 that the $L^E_f$ values have a negative trend similar to what we have observed in case of the $K^E_s$ at all the temperatures under study. The negative values of $L^E_f$ suggest that specific interactions are present between unlike
molecules in these binary systems [37,38]. Considering the excess values, it is clear that the strength of specific interactions between unlike molecules varies in the order (Methyl Benzoate + Propanol) > (Methyl Benzoate + Butanol) > (Methyl Benzoate + Pentanol).

4. Conclusions

The excess molar volume is negative mostly in case of 1-propanol but the trend changed to positive as the alcohol chain increases and excess values of isentropic compressibility and excess free length are found to be negative for all the binary systems over the entire range of composition and at all the temperatures considered in the present study. This is a clear indication of the presence of specific interactions between component molecules through hydrogen bonding between these unlike molecules. The strength of interaction is decreasing with increase in chain length of alcohol in the binary mixtures.

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