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Study of Dielectric and Acoustic Properties of Binary Liquid Mixtures of Cyclohexane with *n*-Butanol at 308 K

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Abstract

Present paper deals with study of dielectric constant, density and ultrasonic velocity and derived properties for the binary mixtures of cyclohexane and *n*-butanol over the entire concentration range measured at 308 K. The experimental data were further used to calculate derived parameters like Bruggeman factor, Gibbs free energy, enthalpy of activation, compressibility, and acoustic impedance, molecular free length, inverse relaxation time and excess parameters. The values of excess properties were further fitted with Redlich–Kister polynomial equation to estimate the binary coefficients and standard deviation. The resulting excess functions were interpreted in terms of the interactions between the molecules in the binary mixtures. Results confirm that weak bonded intermolecular interaction takes place between cyclohexane and *n*-butanol.

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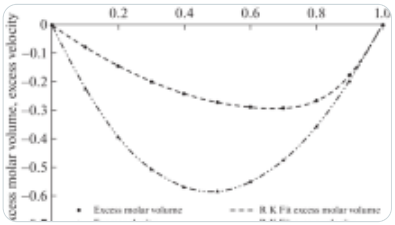

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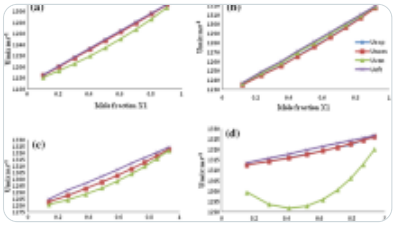

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INTRODUCTION

Choice of an ideal and suitable solvent in a large stratum of various industrial processes and phenomenon [1] is of paramount scientific interest to obtain desired and exact efficiency, selectivity and kinetics of a chemical reaction, extraction or separation [2]. Several factors of a solvent has been investigated till date that have been identified to be deciding the solubility properties, for instance: (i) weak bond or associativity in the solvent [1, 2], (ii) dipole–dipole interaction in the solvent [1–6], (iii) polarity of the solvent [2, 6, 7] etc. A weak bond, when present in a solvent, renders a considerable influential effect on the dielectric, physicochemical, and acoustic properties that shapes suitably enhanced solubility properties [1, 4–7]. Such phenomenon is more prominent in the solutions of polar–polar and polar–nonpolar group. Polar molecules constitute a large part of industrial reagents [8–11] and distinct knowledge of properties of a binary or higher complex mixture are required for designing of efficient chemical process. Butanol, as well as other alcohols, can be used as addition to fuel [8–11]. Here we have used *n*-butanol as a solvent. Alcohols are self-associated organic liquids, used for the synthesis of other organic compounds [10]. They are also widely used as solvent in various industries, coupling and dispersing agents in nanotechnology [12–14]. Also, they are used in the chemical and household industries and as carrier and extraction solvents for natural products and pharmaceutical [15–17].

Dielectric as well as ultrasonic study and its analysis for protic and aprotic binary liquid mixtures containing polar-polar and polar-non polar components is having significant importance in understanding intermolecular interaction and strength between the component molecules as they find application in various industrial and technological processes [1, 11–28]. A large number of studies talking about alcohols and their mixtures at various temperatures and pressures were carried out by molecular dynamics simulations and vibrational spectroscopy [18–35]. Dielectric properties viz. dielectric constant, excess dielectric constant, Bruggeman factor, enthalpy, ultrasonic velocity and its derived acoustical parameters like adiabatic compressibility, molecular free length, relaxation time, acoustic impedance, Gibbs free energy with their excess parameters, gives important information about the molecular interactions and their strengths [29–40]. In the present paper, variation of various dielectric as well as ultrasonic parameters of binary mixtures containing cyclohexane have been studied for entire range of concentration range (by adding solute percentage 10% in solvent 0–100%).

EXPERIMENTAL

Chemicals. *n*-Butanol (of analytical grade) and cyclohexane of HPLC grade were purchased from Merck. Both the liquids are used without further purification.

Solution preparation. The solutions were prepared at different volume percentages of cyclohexane in *n*-butanol in steps of 10% at room temperature. These concentrations were prepared for 5 mL solution samples at room temperature, assuming ideal mixing behavior, with an accuracy ±0.0006 mL.

Dielectric measurements: TDR setup and data acquisition. The complex permittivity spectra were studied using the time domain reflectometry (TDR) method as described in [1, 2]. The Hewlett Packard HP 54750 sampling oscilloscope with HP54754A TDR plug-in module was used.

Density measurement. The density measurements were carried out by portable digital density meter (DMA-35, Anton Paar) for pure liquids and binary mixture. Accuracy of the instrument used is 0.001 g/cm^3. To reduce error three times measurements were done average of three readings is taken.

Ultrasonic velocity measurement. The ultrasonic velocity measurements were performed using ultrasonic interferometer (Model F-05, Mittal Enterprises, New Delhi). It is single crystal interferometer operating at 2 MHz fixed frequency.

Viscosity measurement. Viscosity was measured by using Brookfield Viscometer (Brookfield Viscometer, Model: LV DV-II+ Pro, Cone-plate Model with CPE-40 spindle). The accuracy of the

instrument is 0.01 cP.

Excess parameters. The general formula for calculating the excess parameters [18–42] is given below:

$${{A}^{E}} = {{A}_{m}} - ({{x}_{{\text{A}}}}{{M}_{1}} + {{x}_{{\text{B}}}}{{M}_{2}}),$$

where A^E is the excess parameter such as excess density, x_A is mole fraction of solute, and x_B is mole fraction of solvent.

Bruggeman factor. Bruggeman's mixture formula [4] can be used as the first evidence of molecular interactions in binary mixtures. The effective volume of the solute gets modified by solute–solvent interactions and is best illustrated by the non-linearity of the Bruggeman factor.

This formula states that static permittivity of the binary mixtures (ε_{sm}), solute (ε_{sA}), and solvent (ε_{sB}) are related to the volume fraction of solvent (V), which indicates the interaction between solvent and solute in the mixture is given as:

$${{f}_{{\text{B}}}} = \left({\frac{{{\varepsilon }_{{\text{sm}}}} - {{\varepsilon }_{{\text{sB}}}}}{{{\varepsilon }_{{\text{sA}}}} - {{\varepsilon }_{{\text{sB}}}}} \right){{\left({\frac{{{\varepsilon }_{{\text{sA}}}}}{{{\varepsilon }_{{\text{sm}}}}} \right)}^{{1/3}}} = 1 - V.$$

The specific acoustic impedance is given by,

$$Z = U\rho ,~$$

(1)

where U is the ultrasonic velocity (of the mixture) and ρ is the density of the mixture.

The adiabatic compressibility is given by,

$$\beta = 1{\text{/}}({{U}^{2}}\rho),$$

(2)

where U and ρ are the velocity and density of liquid mixture.

The general formula for calculating the excess parameters is given below;

$${{A}^{E}} = {{A}_{m}} - ({{x}_{1}}{{M}_{1}} + (1 - {{x}_{1}}){{M}_{2}}),$$

(3)

where A^E is the excess parameter such as excess density x_1 mole fraction.

And the excess parameters are fitted to the Redlich–Kister polynomial equation [8] of third order and this equation is given by

$${{A}^{E}} = {{x}_{1}}{{x}_{2}}\mathop \sum \limits_{i = 0}^n \,{{A}_{i}}{{(1 - 2{{x}_{2}})}^{i}},$$

(4)

where x_i is the mole fraction of pure component 1 and 2.

RESULTS AND DISCUSSION

Table 1 depicts that values of dielectric constant, viscosity decreases as concentration of cyclohexane increases whereas values of density, ultrasonic velocity increases as concentration of cyclohexane increases.

Table 1. Density (ρ), viscosity (η), ultrasonic velocity (*U*), dielectric constant (ε) of cyclohexane + *n*-butanol at 308 K and atmospheric pressure (*V* is volume fraction of cyclohexane)

Table 2 depicts RK coefficients and values of standard error prove correctness of data.

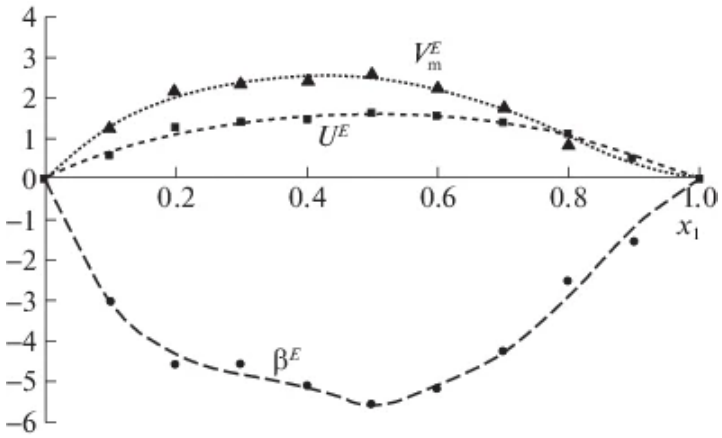
Table 2. RK coefficients and values of standard error

Table 3 gives comparison of experimental values with literature values. Values depends upon instrument used and its accuracy, various parameters like temperature, pressure, humidity/presence of moisture, purity of liquids, use of fresh/sealed or old liquids. Variation in values can occur due to the change in any factor/parameter mentioned.

Table 3. Comparison of measured net solvents properties with literature values at 308 K and atmospheric pressure

Figure 1 gives excess molar volume of cyclohexane + *n*-butanol. As concentration of cyclohexane increases excess molar volume (\(V_{{\text{m}}}^{E}\)) becomes positive. Positive values indicate that volume expansion takes place upon mixing due to cross association between dissimilar molecules. Positive values also attributed to weak interaction between unlike molecules through dipole–dipole interactions.

Fig. 1.



Excess molar volume (\(V_{{\text{m}}}^{E}\)), excess velocity (U^E), excess compressibilty (β^E) of cyclohexane + *n*-butanol.

Positive values indicate that volume expansion takes place upon mixing due to cross association between dissimilar molecules. This expansion gives less compactness i.e., molecules randomly moves far from each other. This attributed to weak dispersive or dipole-dipole bonded interaction between unlike molecules [10]. The positive variation of excess molar volume in cyclohexane–*n*-butanol

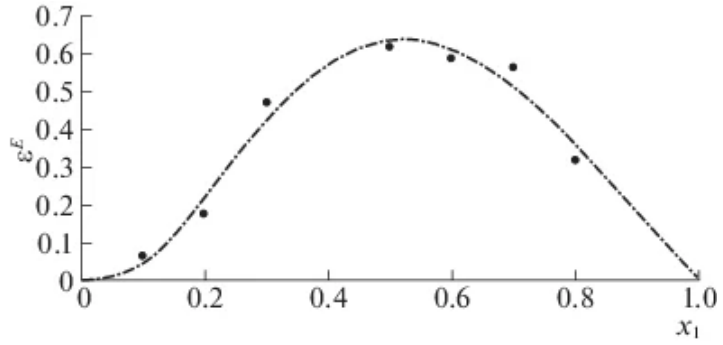
mixtures indicates the structural contribution, of constituents arising from inappropriate geometrical fitting in this case i.e., less interstitial accommodation of solute and solvent in the system. The molecules of cyclohexane do not cooperate with *n*-butanol is confirmed from our observation. This less cooperation causes the increase in molar volume of the mixture. Leading to the formation less of tightly packed weak-bonded aggregates (between unlike molecules). Positive value also indicates that the solute acts as a structure breaker. The maximum deviation of excess molar volume shows the strength of weak bonded interaction. The positive V^E values of the system are attributed to interstitial accommodation of cyclohexane molecules in the dipole–dipole bonded network with *n*-butanol.

As shown in Fig. 1 excess velocity becomes positive as concentration of cyclohexane increases. Positive deviation and non linear dependence suggests the presence of dipole–dipole interaction between the components of the mixture positive excess velocity can be concluded as the formation of the structure [18–24]. Weak interaction arises among the components of the mixture leading to the formation of molecular aggregates and less compact structure then sound will travel slower through the mixture by means of longitudinal waves and hence speed of sound with respect to linear behavior will be positive [10–15].

Figure 1 indicates excess compressibility of cyclohexane + *n*-butanol. Negative excess compressibility of values are due to less closed packed molecules, which accounts for the existence of dipole-dipole specific molecular interaction between unlike molecules [6–10]. Sign of compressibility plays vital role in assessing the compactness due to molecular interaction in liquid mixture through weak bonded interactions, leading to compact structure making negative excess compressibility [7–12].

The positive value of excess static dielectric constant indicates (Fig. 2) that the solute and solvent interact in such a manner so as to increase the effective dipole moment and leads to the formation of dimers.The excess static dielectric constant values of the binary mixtures of polar solvents is commonly used to obtain insight into the strength of hetero-molecular bonded structures and dipolar ordering and also the stoichiometric composition corresponding to the formation of a stable complex product [14]. The concentration, X_{CYH} corresponding to the maximum magnitude of excess static dielectric constant, is (at) 0.50 (Fig. 2) suggesting the formation of a stable complex with 1 : 1 mole ratio, which governs their molecular dielectric polarization [15], cyclohexane molecule, when mixed with *n*-butanol, it does not cooperate with *n*-butanol molecule and thus there is possibility to form dipole–dipole bonding with each other. The positive excess static dielectric constant also indicates that one of the mixture constitute acts as a structure maker in lesser extend for another molecule during the cooperation with dipoles of similar orientation. Hence there is increase in the total number of parallel aligned effective dipoles that contribute to the mixture dielectric polarization.

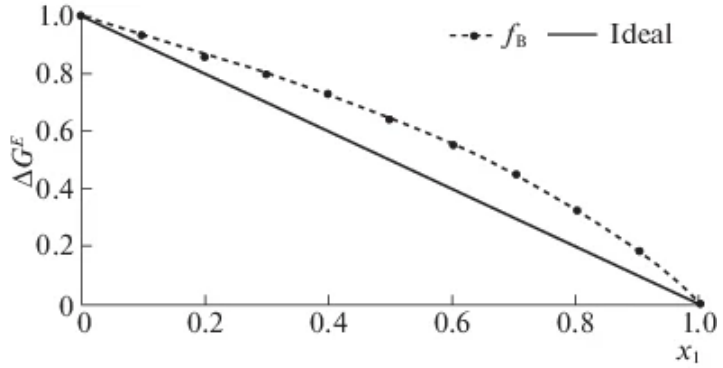
Fig. 2.



Excess dielectric constant (ε^E) of cyclohexane + *n*-butanol, x_1 is mole fraction of cyclohexane.

Figure 3 shows the variation of Bruggeman factor [5] with volume fraction of cyclohexane. For the system, it is observed that the value of Bruggeman factor (f_B) deviates from ideal line. The nonlinearity of the curve indicates existence of intermolecular interactions in the system.

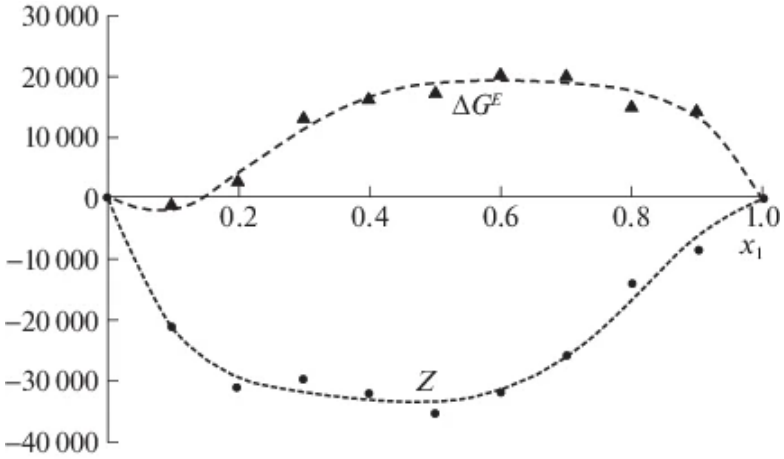
Fig. 3.



Bruggeman factor of cyclohexane + *n*-butanol.

Negative values of acoustic impedance as shown in Fig. 4 hint to the possibility of presence of dipole forces between the reacting components of the mixture [11–14]. negative deviation also suggests that cyclohexane molecules does not cooperate with *n*-butanol molecules hence weak dipole-dipole bonded intermolecular interactions occurs between them [10]. Figure 4 depicts nonlinear deviation of Gibbs free energy of cyclohexane in *n*-butanol it clearly indicates presence of weak intermolecular interactions between solute and solvent.

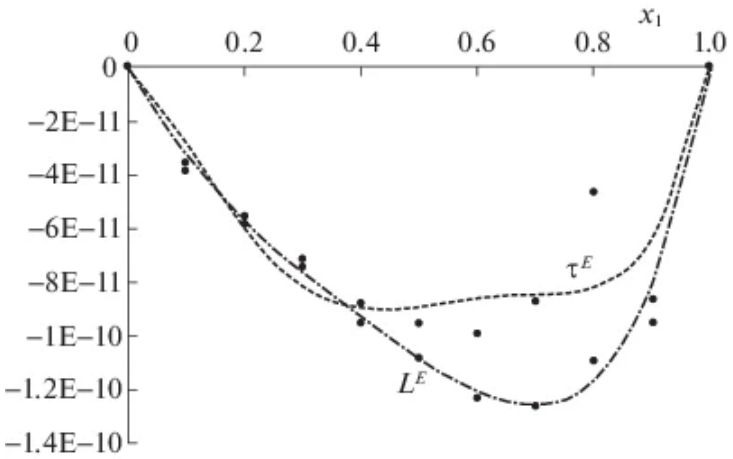
Fig. 4.



Excess acoustic impedance (Z), excess Gibbs free energy (ΔG^E) of activation of cyclohexane + n-butanol.

Negative values exhibit weak interaction. Increase in values of free length with concentration can be concluded as there is significant interaction between two liquids [26–36]. negative values also suggests that as cyclohexane molecules are mixed with n-butanol molecules their intermolecular distance increases and gives rise to dipole-dipole bonded interaction between them [36–46, 54]. Figure 5 indicates negative deviation in excess inverse relaxation time which indicates presence of intermolecular interactions within the system.

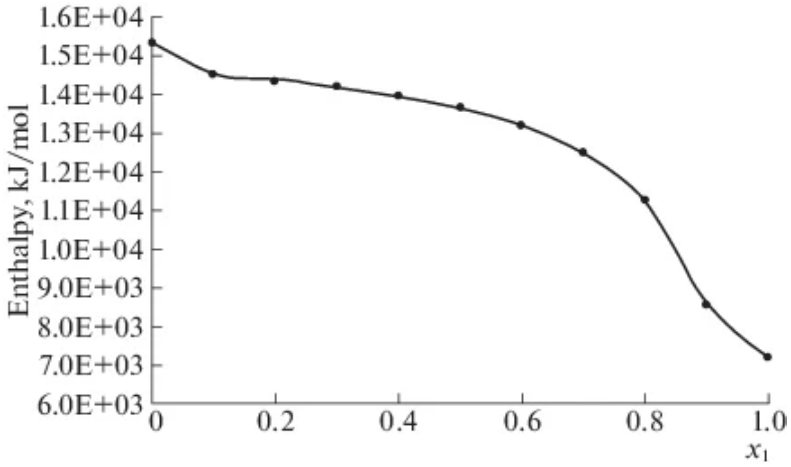
Fig. 5.



Excess molecular free length (L^E), excess inverse relaxation time (τ^E) of cyclohexane + n-butanol.

Figure 6 depicts variation of enthalpy of activation. Enthalpy of activation is obtained from Eyring's rate equation [38, 39]. The value of enthalpy of activation gradually decreases with increase in volume fraction of cyclohexane. This indicates that less energy is needed for dipole reorientation.

Fig. 6.



Enthalpy of activation (ΔH^E) of cyclohexane + *n*-butanol.

CONCLUSION

In this work, the measurement of dielectric constant, density, ultrasonic velocity and other dielectric, acoustical parameters of cyclohexane in *n*-butanol solution was studied in different concentrations at 308 K. Positive excess molar volume (\(V_{{\text{m}}}^{E}\)) values indicate the presence of weak dipole–dipole bonded interactions. It has been observed that the excess velocity values become more positive with rise in concentration of cyclohexane. The experimental dielectric parameters and ultrasonic velocity data and other acoustical parameters contain valuable information regarding the solute-solvent interactions in the measurements, it can be concluded that the concentration of cyclohexane affects and gives rise to dipole–dipole bonded interaction. Our dielectric study and ultrasonic study both suggest that cyclohexane acts as structure breaker. Increase in concentration of cyclohexane plays an important role in forming dipole–dipole bonded interactions in the solutions.

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