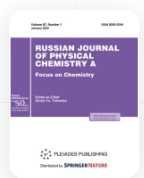


# Dielectric and Acoustic Characterization Study of Cyclohexane with *n*-Butanol at 298 K

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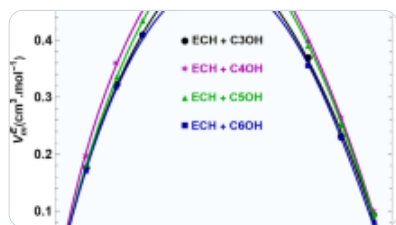
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## Abstract

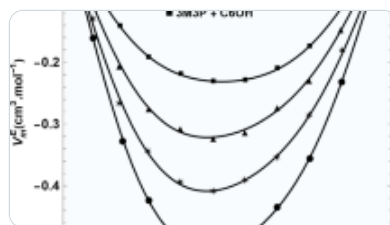
Present paper deals with study of dielectric constant, density and ultrasonic velocity and derived properties for the binary mixtures of cyclohexane and *n*-butanol over the entire concentration range measured at 298 K. The experimental data further used to calculate derived parameters like Bruggeman 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 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 intermolecular interactions between the molecules in the binary mixtures. Results confirm, strong bonded intermolecular interaction takes place between cyclohexane and *n*-butanol.

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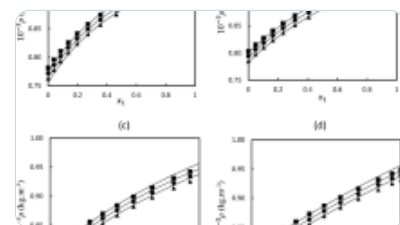
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## INTRODUCTION

Selection of solvent is a time-consuming process as 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 have been investigated till date that have been identified to be deciding the solubility properties, for instance, (i) strong bond or associativity in the solvent [1, 2], (ii) hydrogen bonding interaction in the solvent [1–6], (iii) polarity of the solvent [2, 6, 7], etc. A strong 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 figure of merits/demerits of a binary or higher complex mixture are required for designing of efficient chemical process. Alcohols like butanol, 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].

Both dielectric and 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 [36–60]. 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

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### Chemicals

In the present system of cyclohexane with *n*-butanol binary mixture, the liquid *n*-butanol is used of Analytical Reagent grade, with CAS no. 71-36-3, molecular mass 74.12 g/mol and is obtained from MERCK (99.99) and cyclohexane is of HPLC grade and CAS no. 110-82-7 and molar mass 84.162 g/mol. 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 (droplets of cyclohexane are mixed in *n*-butanol with increasing volume percentage). These concentrations were prepared for 5 mL solution samples at room temperature, assuming ideal mixing behavior, with an accuracy  $\pm 0.0006$  mL.

## DIELECTRIC MEASUREMENTS

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### 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 has been 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  $0.001 \text{ g/cm}^3$ . To reduce error three times measurements were done average of three readings is taken.

## Ultrasonic Velocity Measurements

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

## Viscosity Measurement

Viscosity of the sample in the present study were 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.

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

$$A^E = A_m - (x_A M_1 + x_B M_2),$$

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

**2.3.2. 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 ( $\epsilon_{sm}$ ), solute ( $\epsilon_{sA}$ ), and solvent ( $\epsilon_{sB}$ ) are related to the volume fraction of solvent ( $V$ ), which indicates the interaction between solvent and solute in the mixture is given as:

$$\left\{ \frac{\epsilon_{sm} - \epsilon_{sB}}{\epsilon_{sA} - \epsilon_{sB}} \right\} \left\{ \frac{\epsilon_{sA} - \epsilon_{sB}}{\epsilon_{sm} - \epsilon_{sB}} \right\}^{1/3} = 1 - V.$$

The specific acoustic impedance is given by,

$$Z = U\rho,$$

(1)

where  $U$  is the ultrasonic velocity (of the mixture), and  $\rho$  is the density of the mixture.

The adiabatic compressibility is given by,

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

(2)

where  $U$  and  $\rho$  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

$$A^E = x_1 x_2 \sum_{i=0}^n \lim_{i=0} A_i (1 - 2x_2)^i,$$

(4)

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

## RESULT AND DISCUSSION

Values in [Table 1](#) depicts that values of density, dielectric constant, viscosity decreases as concentration of cyclohexane increases whereas values of ultrasonic velocity increases as concentration of cyclohexane increases.

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**Table 1. Density, viscosity, ultrasonic velocity, dielectric constant, of cyclohexane + *n*-butanol at 298 K and atmospheric pressure**

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Comparison of experimental values with literature values are given. It shows that experimental values are in good agreement with literature values it also validates authenticity of the measured data ([Table 2](#)).

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**Table 2. Comparison of measured net solvents properties with literature values at 298 K and atmospheric pressure**

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RK coefficients and values of standard error are given in [Table 3](#), the values of standard error or standard deviation is very less it indicates authentication of data.

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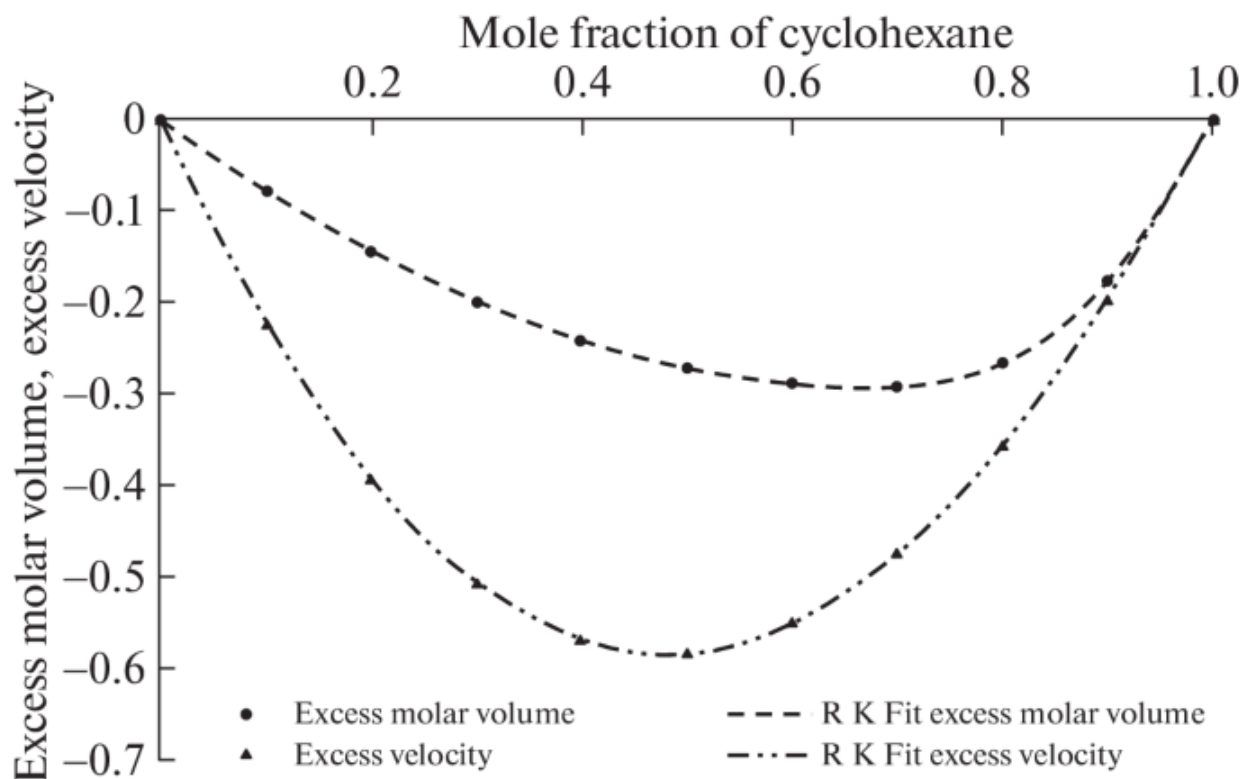
**Table 3. RK coefficients and values of standard deviation**

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Excess molar volume of cyclohexane + *n*-butanol is shown in [Fig. 1](#). As concentration of cyclohexane increases excess molar volume ( $(V_m^E)$ ) becomes negative. Negative values also attributed to strong interaction between unlike molecules through hydrogen bonding. Negative values indicate that volume contraction takes place upon mixing due to cross association between dissimilar molecules. This contraction gives more compactness i.e., molecules come closer to each other. This attributed to strong hydrogen bonded interaction between unlike molecules [[10](#)]. The negative variation of excess molar volume in cyclohexane-*n*-butanol mixtures indicates the structural contribution, of constituents arising from appropriate geometrical fitting in this case i.e., more interstitial accommodation of solute and solvent in the system. The molecules of cyclohexane cooperates with *n*-butanol is confirmed from our observation. This more cooperation causes the decrease in molar volume of the mixture. Leading to the formation more tightly packed strong-bonded aggregates (between unlike molecules). Negative values also indicates that the solute acts as a structure maker. The maximum deviation of excess molar volume shows the strength of strong bonded interaction. The negative  $V_m^E$  values of the system are attributed to interstitial accommodation of cyclohexane molecules in the hydrogen bonding bonded

network with *n*-butanol.

**Fig. 1.**

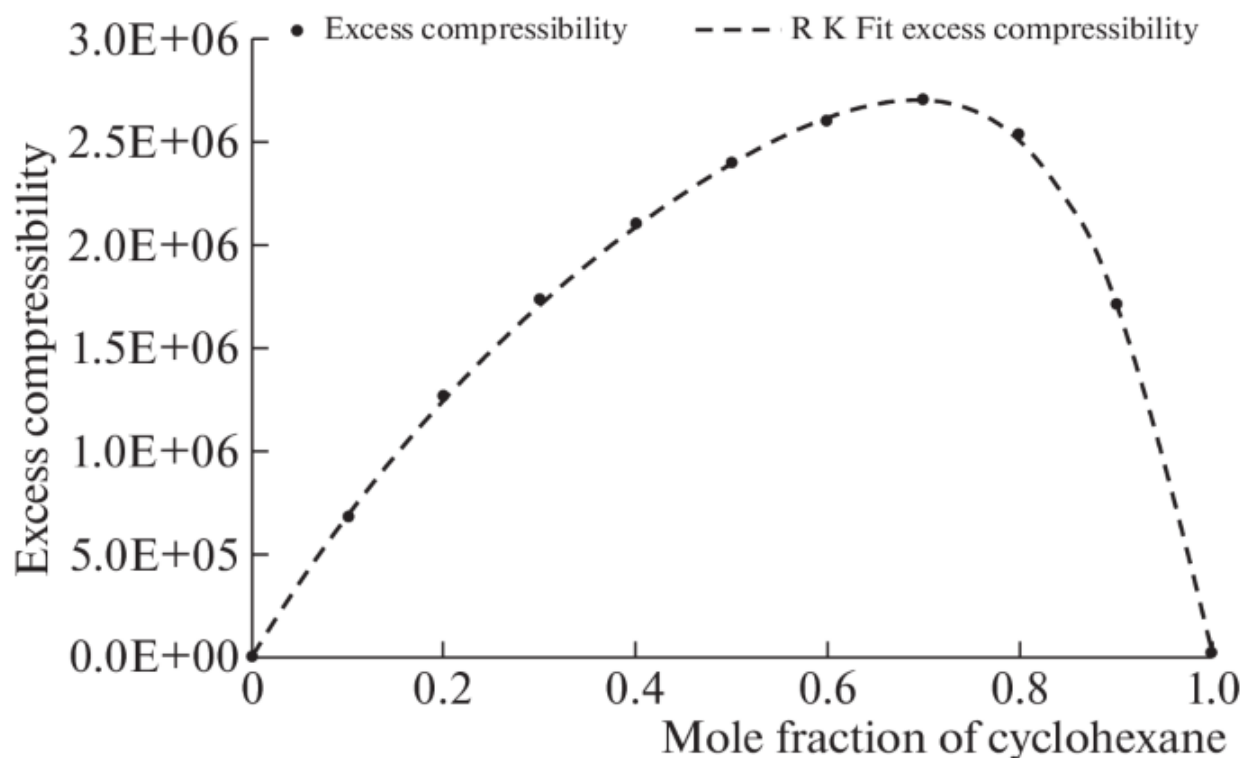


Excess molar volume, excess velocity of cyclohexane + *n*-butanol.

As shown in [Fig. 1](#) excess velocity becomes negative as concentration of cyclohexane increases. Negative deviation and non linear dependence suggests the presence of hydrogen bonding interaction between the components of the mixture negative excess velocity can be concluded as the formation of the structure [18–24]. strong interaction arises among the components of the mixture leading to the formation of molecular aggregates and more compact structure then sound will travel faster through the mixture by means of longitudinal waves and hence speed of sound with respect to linear behavior will be negative [10–15].

Excess compressibility of cyclohexane + *n*-butanol shown by [Fig. 2](#). Positive excess compressibility of values is due to more closed packed molecules, which accounts for the existence of hydrogen bonding 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 strong bonded interactions, leading to compact structure making positive excess compressibility [7–12].

Fig. 2.



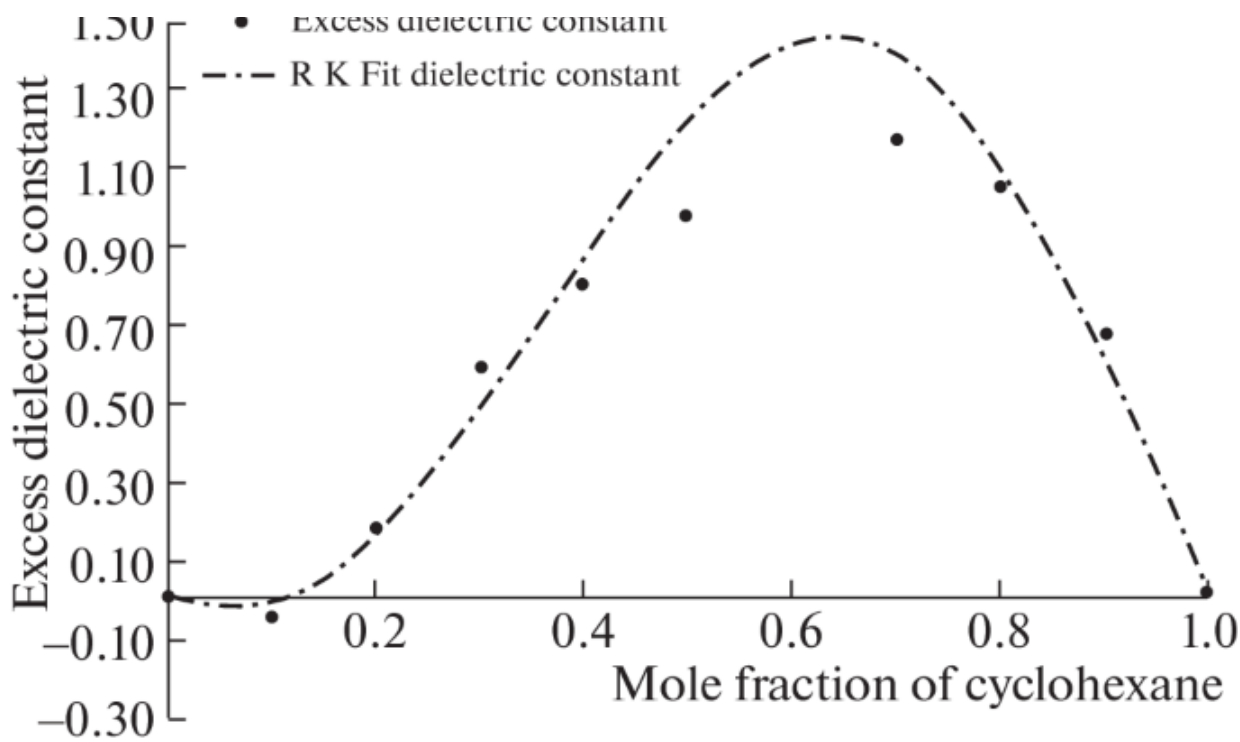
Excess compressibility of cyclohexane + *n*-butanol.

The positive value of excess static dielectric constant indicates as shown in Fig. 3, 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 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 hydrogen 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. 3.

1.50 - Excess dielectric constant

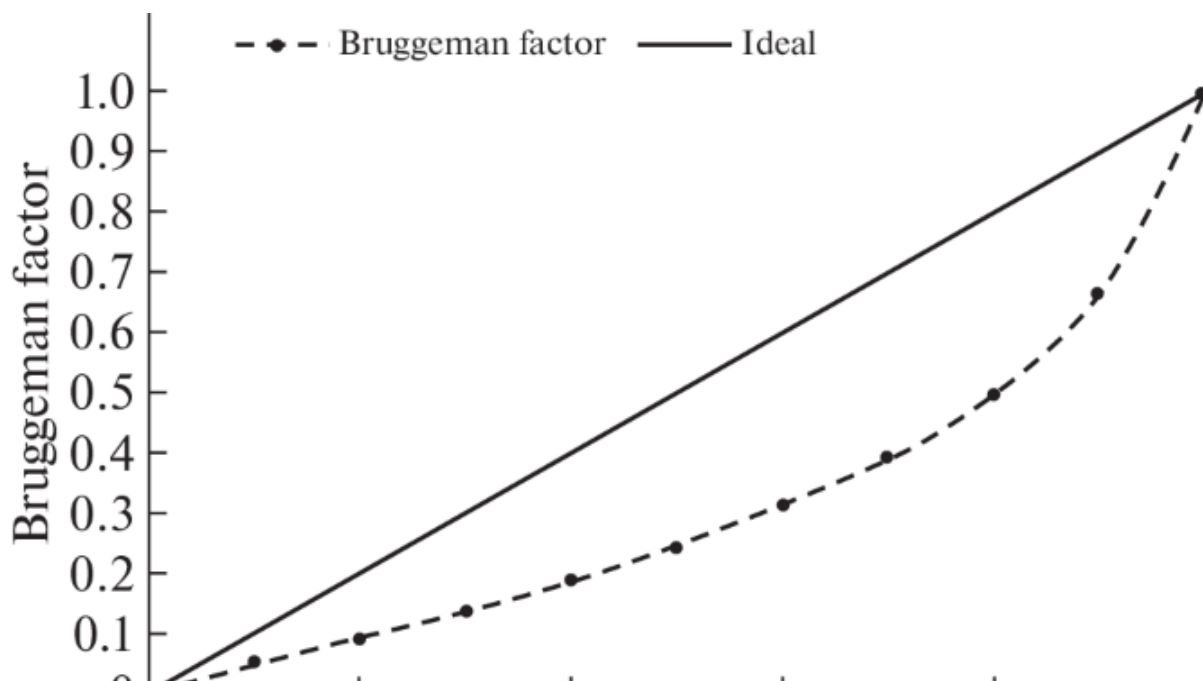


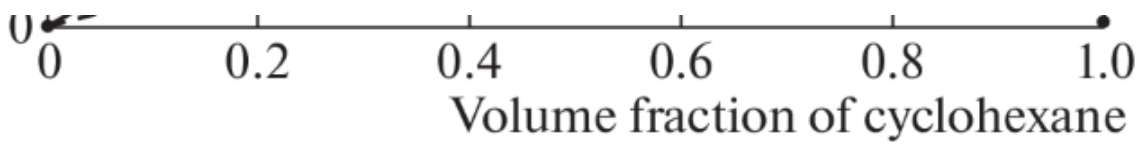


Excess dielectric constant of cyclohexane + *n*-butanol.

[Figure 4](#) 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. 4.**

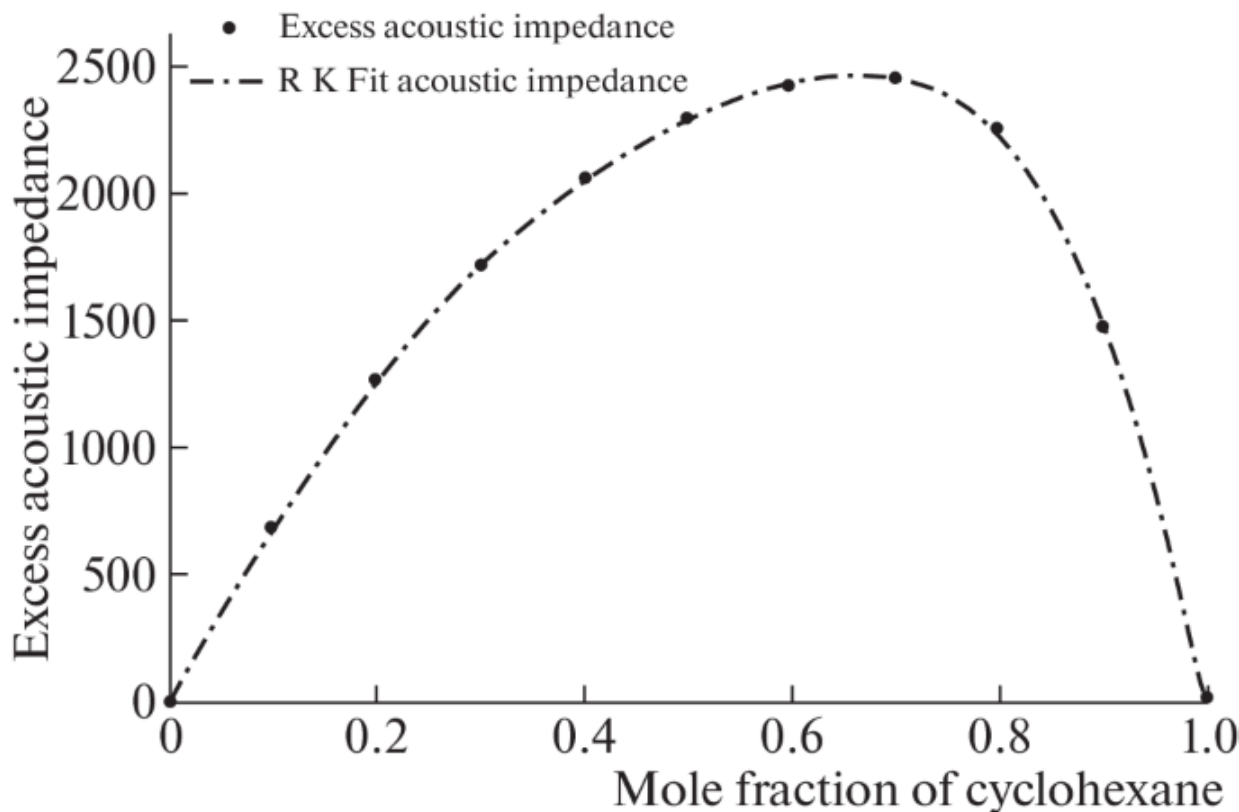




Bruggeman factor of cyclohexane + *n*-butanol.

Positive values of acoustic impedance hint to the possibility of presence of hydrogen bonded forces between the reacting components of the mixture [11–14]. Positive deviation also suggests that cyclohexane molecules cooperates with *n*-butanol molecules (Fig. 5) hence strong hydrogen bonding bonded intermolecular interactions occurs between them [10].

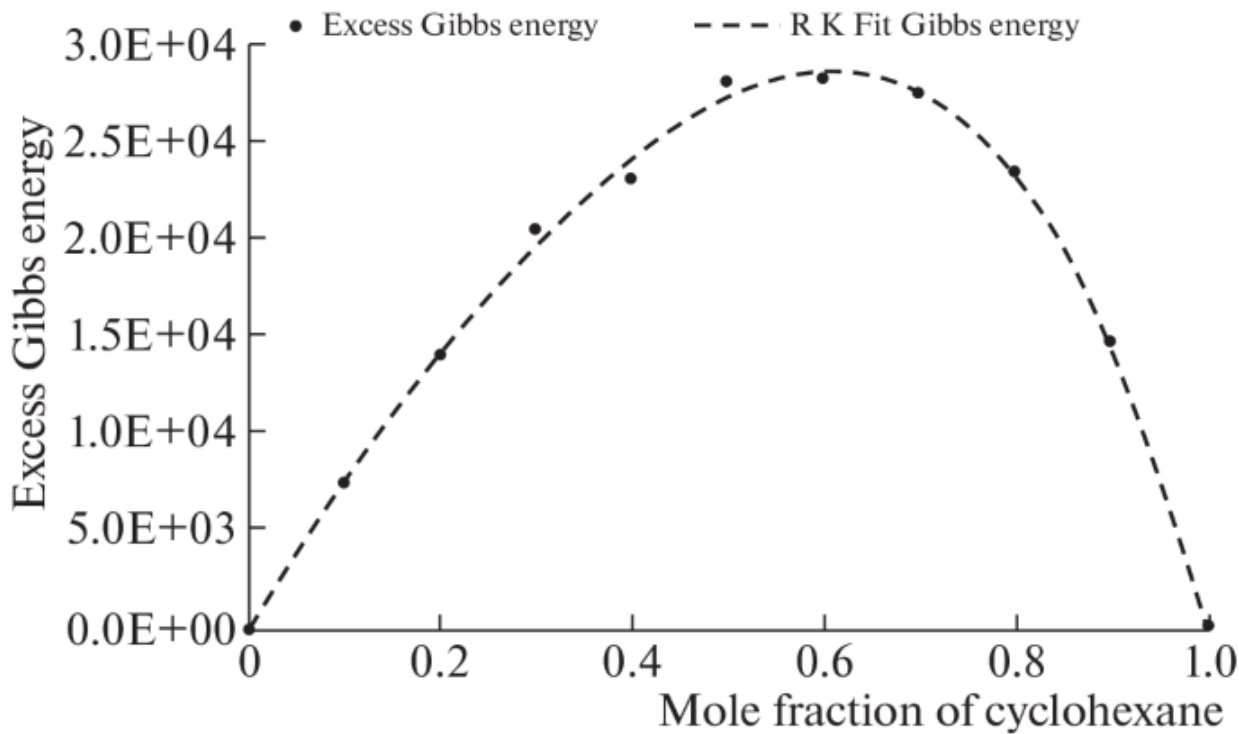
Fig. 5.



Excess acoustic impedance of cyclohexane + *n*-butanol.

Figure 6 depicts nonlinear deviation of Gibbs free energy of cyclohexane in *n*-butanol it clearly indicates presence of strong intermolecular interactions between solute and solvent.

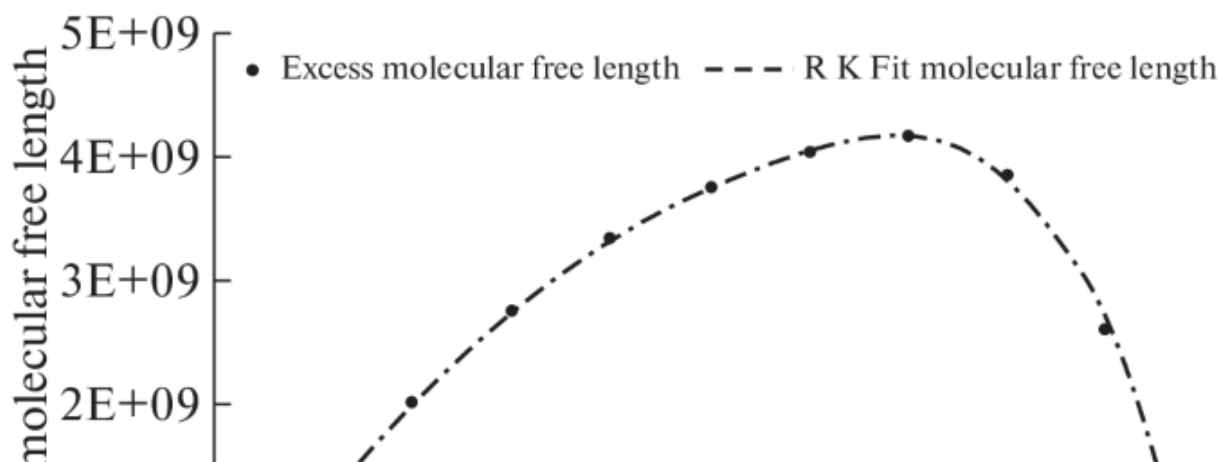
Fig. 6.

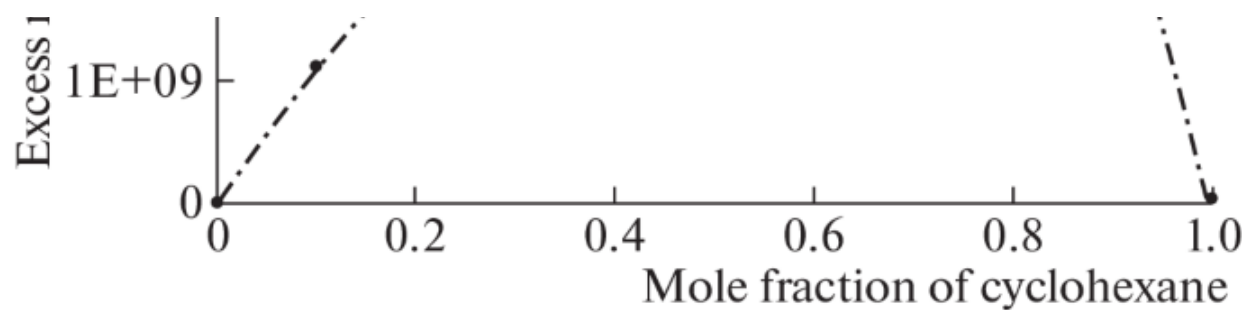


Excess Gibbs free energy of activation of cyclohexane + *n*-butanol.

Positive values of excess molecular free length exhibit strong interaction. Increase in values of free length with concentration can be concluded as there is significant interaction between two liquids [26–36]. Positive values also suggests that as cyclohexane molecules are mixed with *n*-butanol molecules (Fig. 7) their intermolecular distance decreases and gives rise to hydrogen bonding bonded interaction between them [36–46, 60].

Fig. 7.

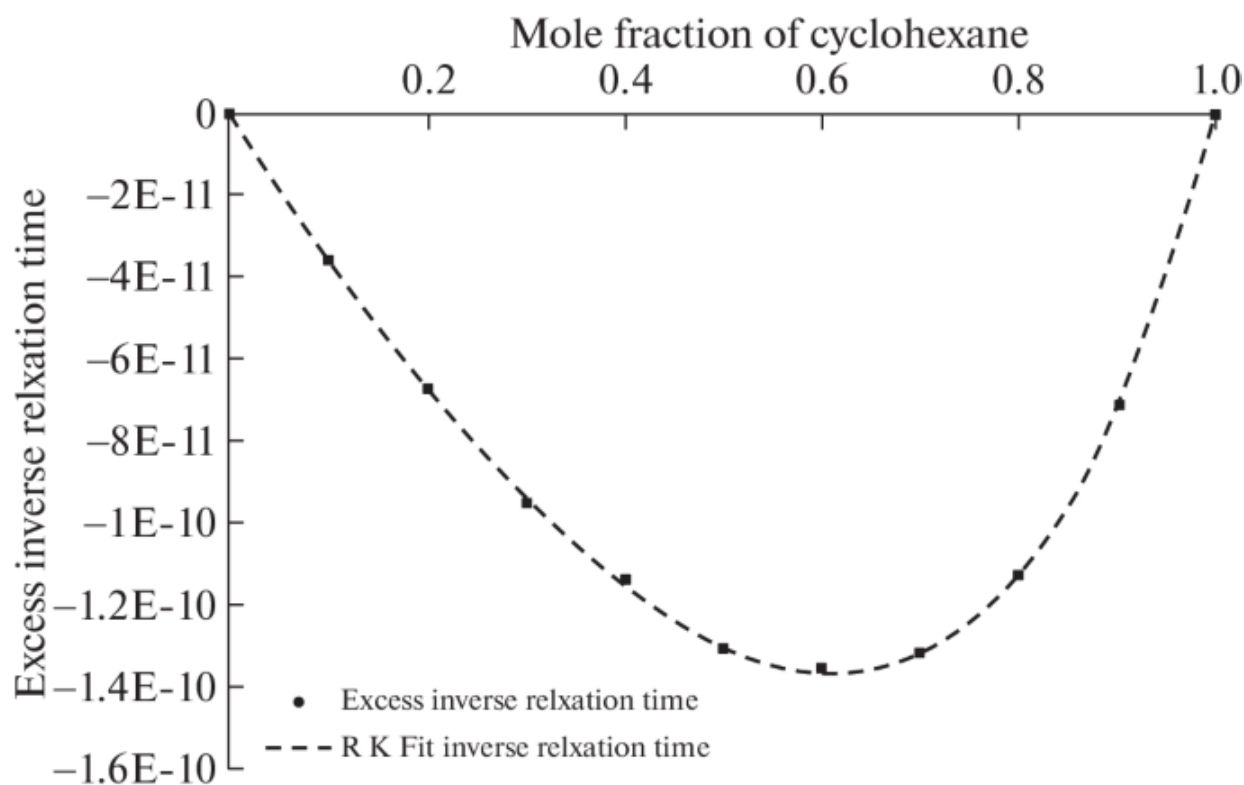




Excess molecular free length of cyclohexane + *n*-butanol.

[Figure 8](#) indicates negative deviation in excess inverse relaxation time which indicates presence of the strong intermolecular interactions within the system.

**Fig. 8.**

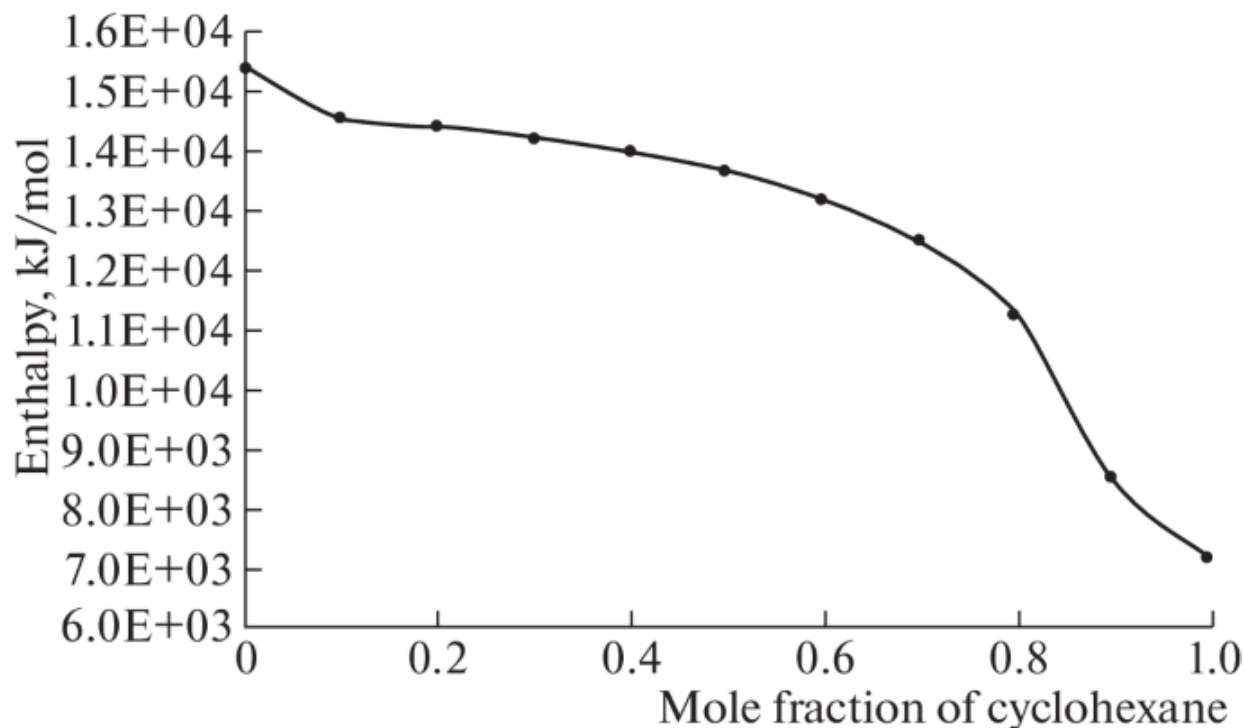


Inverse relaxation time of cyclohexane + *n*-butanol.

[Figure 9](#) 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. 9.



Enthalpy of activation of cyclohexane + *n*-butanol.

## CONCLUSIONS

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 298 K. Negative excess molar volume ( $(V_m^E)$ ) values indicate the presence of strong hydrogen bonding interactions. It has been observed that the excess velocity values become more negative 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 the cyclohexane affects and gives rise to hydrogen bonded interaction. Dielectric study and ultrasonic study both suggest that cyclohexane acts as structure maker. Increase in concentration of cyclohexane plays an important role in forming hydrogen bonding bonded interactions in the solutions.

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## Ethics declarations

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The authors declare that they have no conflicts of interest.

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