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Acoustic properties of binary mixtures of 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin with 1-propanol and 2-propanol at 300, 303, and 307 K

Harsha V. Burghate* and Pravin B. Raghuwanshi

Abstract

Background: The present work deals with the study of acoustic parameters i.e. ultrasonic velocity, density, adiabatic compressibility (β_s), apparent molar compressibility (φ_k) and specific acoustic impedance (Z) which reflects structural interaction by interferometer by 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin at different percentage of 1-propanol-water and 2-propanol-water solvent system at 300K, 303K and 307K are studied.

Methods: In present investigation, ultrasonic velocity and density of 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin have been studied at 0.01 M concentration at different concentrations (70%, 75%, 80%, 85% and 90%) in 100% 1-propanol and 2-propanol at 300K, 303K and 307K. From these values, adiabatic compressibility (β_s), apparent molar compressibility (φ_k) and specific acoustic impedance (Z) are calculated.

Results: Measurement of ultrasonic velocity is the best tool to investigate solute-solvent, solute-solute and ion-solvent interactions. Therefore, in last four decades ultrasonic interferometric study created its own identity for determining solute-solvent interactions. By this study, adiabatic compressibility (β_s), apparent molar compressibility (φ_k) and specific acoustic impedance (Z) acoustic properties are determined which explain how these interactions occur and responsible for breaking and making of the structure in the solution. So in the present work these acoustic parameters were studied for synthesized ligands, which are used as solutes.

Conclusion: Trends of acoustic properties indicate the presence of molecular interaction in the present binary mixture under study. It may be qualitatively inferred that the interaction between unlike molecules is mainly due to hydrogen bonding. The present studies investigated that with increasing the concentration of 1-propanol and 2-propanol β_s and φ_k increases while Z decreases at 300 k, 303K and 307K by taking constant concentration of 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin (0.01M). Better values of parameters are found for the 2-propanol and water system in 90% ratio at 307K. From this study it is clear that properties, which are directly or indirectly responsible for this are protic nature of solvent, dielectric constant, polarity, density, tendency of forming hydrogen bonding, surface tension, viscosity of solvent etc.

Keywords: 3-Acetyl-4-methyl-6-chloro-8-nitrocoumarin, 1-Propanol, 2-Propanol, Solute-solvent interactions

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Background

Ultrasonic waves, in recent years, have acquired the status of an important probe for the study of structure and properties of matter in basic science. In the field of technology, the waves are being used for detection of flows, testing of materials, mechanical cleaning of surface, etc. The study of molecular interaction in liquids provides valuable information regarding internal structure, molecular association, complex formation, etc. Ultrasonic parameters are directly related to a large number of thermodynamic parameters, since various molecular theories of liquid state are based on thermodynamic considerations. Gopalrao and Jardar (1988) formulated the equation a state for a square-well fluid and obtained some thermodynamic parameters by extending the Florey's equation to mixture of unrelated type of molecules. Prigogine et al. (1956) have shown that the excess parameters such as excess volume V^E give interaction on the relative strengths of AA, AB, and BB interactions in the mixture of A and B liquids. In organic liquids, many records of measurements of ultrasonic velocity are found. Possibly the longest list being that due to Parthasarathy (1935) from the measurements of sound velocity in more than 60 organic liquids. He established empirical rules between velocity, molecular weight, density, and molecular size. One of the important rules of Parthasarathy states that aromatic compounds usually have high velocity than the corresponding aliphatic compounds. This is due to the larger densities of aromatic substances. Another rule of Parthasarathy points out that polar molecules favor higher velocities as in aniline, nitrobenzene, cyclohexanol, water, etc. Same liquids with small electric moment possess more velocity than in either benzene or cyclohexane.

The more important application of sound velocity measurements in liquids has been the evaluation of adiabatic compressibility and the ratio of specific heats, when isothermal compressibility had already been found out by static measurements. Most of the information procured from ultrasonic study of fluids is confined to the determination of hydration number and compressibility (Shilo 1958; Stuher et al. 1965; Braginskayu and Sodikharu 1975).

The successful application of acoustic methods to physico-chemical interactions of solution becomes possible after the development of adequate theoretical approaches and methods for precise ultrasound velocity measurements in small volumes of liquids (Sarvazyan and Khavakoz 1977; Bukin et al. 1979).

Kavitha et al. (2011) reported the measurements of the speed of sound in the binary liquid mixtures of acetone with toluene, chlorobenzene, and nitrobenzene measured at 303 K.

Table 1 Acoustic parameters at different percentages of 1-propanol-water mixture. System: 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin + 1-propanol + water

Concentration, 0.01 M			Ultrasonic frequencies, 1 MHz		
% 1-Propanol	V (m s^{-1})	$d_s \times 10^3$ (kg.m^{-3})	$\beta_s \times 10^{-7}$ (pa^{-1})	φ_k (m^3 mol^{-1} pa^{-1})	Z (kg m^{-2} s^{-1})
Temperature = 300 K					
70	1349.133	0.8651	6.3510	−0.0325	1193.5399
75	1334.000	0.8631	6.5106	−0.0305	1160.2758
80	1304.228	0.8498	6.9182	−0.0245	1121.7610
85	1278.971	0.8429	7.2535	−0.0199	1087.2964
90	1266.400	0.8321	7.4934	−0.0158	1056.7002
Temperature = 303 K					
70	1378.666	0.8713	6.0388	−0.0482	1201.1741
75	1320.666	0.8628	6.6458	−0.0405	1139.4136
80	1319.371	0.8483	6.7727	−0.0376	1119.1621
85	1278.400	0.8373	7.3077	−0.0302	1070.4043
90	1254.742	0.8263	7.6874	−0.0244	1036.7586
Temperature = 307 K					
70	1335.533	0.8721	6.4290	−0.0427	1164.6895
75	1280.571	0.8665	7.0383	−0.0352	1109.5532
80	1274.857	0.8531	7.2130	−0.0318	1087.5318
85	1270.000	0.8339	7.4349	−0.0272	1059.0530
90	1258.400	0.8293	7.6146	−0.0245	1043.5911

Table 2 Acoustic parameters at different percentages of 2-propanol-water mixture. System: 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin + 2-propanol + water

Concentration, 0.01 M			Ultrasonic frequencies, 1 MHz		
% 2-Propanol	V (m s^{-1})	$d_s \times 10^3$ (kg m^{-3})	$\beta_s \times 10^{-7}$ (pa^{-1})	φ_k (m^3 mol^{-1} pa^{-1})	Z (kg m^{-2} s^{-1})
Temperature = 300 K					
70	1379.333	0.8888	5.9139	−0.0493	1225.9218
75	1338.533	0.8736	6.3892	−0.0427	1169.3136
80	1331.133	0.8713	6.4775	−0.0415	1159.7874
85	1282.857	0.8501	7.1484	−0.0317	1090.5082
90	1246.742	0.8306	7.7461	−0.0225	1035.5090
Temperature = 303 K					
70	1331.533	0.8807	6.4045	−0.0567	1172.6520
75	1326.666	0.8640	6.5766	−0.0533	1146.1824
80	1290.666	0.8636	6.9519	−0.0489	1114.5621
85	1261.600	0.8383	7.4947	−0.0400	1057.5992
90	1221.142	0.8227	8.1518	−0.0303	1004.5989
Temperature = 307 K					
70	1377.333	0.8705	6.0558	−0.0625	1198.9396
75	1305.333	0.8573	6.8461	−0.0521	1119.0336
80	1279.942	0.8408	7.2603	−0.0456	1076.1399
85	1248.114	0.8279	7.7539	−0.0383	1033.3019
90	1211.600	0.8123	8.3861	−0.0287	984.1826

Active research work is still in progress to study the effects of ultrasonic waves in mechanical, biological, chemical, physical, and industrial fields. The most well-known application of ultrasound is its use in sonography to produce pictures of fetuses in the human womb. So, in the present day, applications of ultrasonics are emerging in the field of forensic sciences and space research and in wars also.

The present work deals with the measurement of density and ultrasonic velocity and the study of calculated acoustic parameters like adiabatic compressibility (β_s), apparent molar compressibility (ϕ_k), and specific acoustic impedance (Z) with the help of measured parameters, which reflects the structural interaction. So, we are interested to study 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin at different percentage of 1-propanol-water and 2-propanol-water solvent system at 300, 303, and 307 K to see any structural change occurring in the system.

Methods

In present investigation, ultrasonic velocity and density of 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin have been studied at 0.01 M concentration at different concentrations

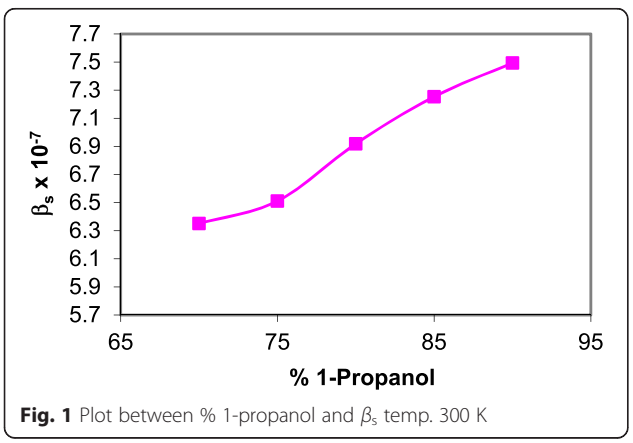

Fig. 1 Plot between % 1-propanol and β_s temp. 300 K

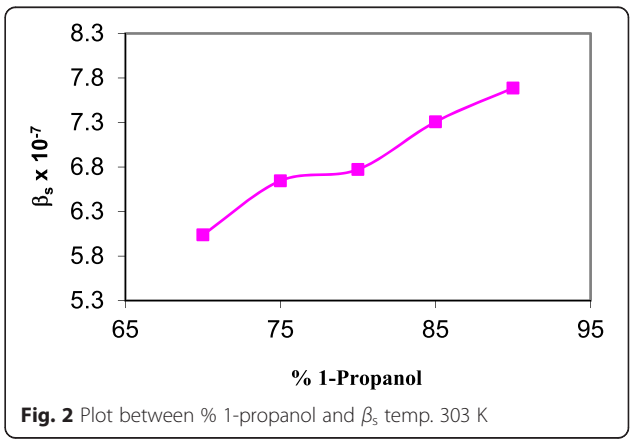

Fig. 2 Plot between % 1-propanol and β_s temp. 303 K

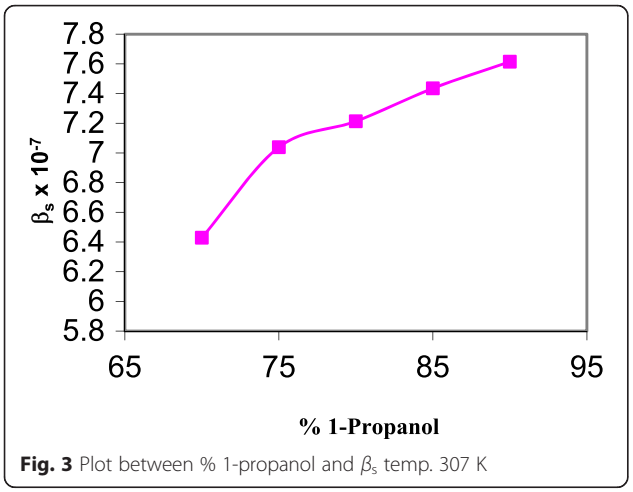

Fig. 3 Plot between % 1-propanol and β_s temp. 307 K

(70, 75, 80, 85, and 90 %) in 100 % 1-propanol and 2-propanol at 300, 303, and 307 K. From these values, adiabatic compressibility (β_s), apparent molar compressibility (ϕ_k), and specific acoustic impedance (Z) are calculated.

Using the measured data, the adiabatic compressibility (β) has been calculated.

$$\beta = 1/v^2 \times d$$

Apparent molar compressibility (ϕ_k) has been calculated from the relation,

$$\phi_k = \left[1000\ (\beta_s do - \beta_o ds)/m\ ds\ do\right] + (\beta_s M/ds)$$

Where,
do density of pure solvent
ds density of solution
m molarity of solution
M molecular weight of solute
β_o adiabatic compressibility of pure solvent
β_s adiabatic compressibility of solution

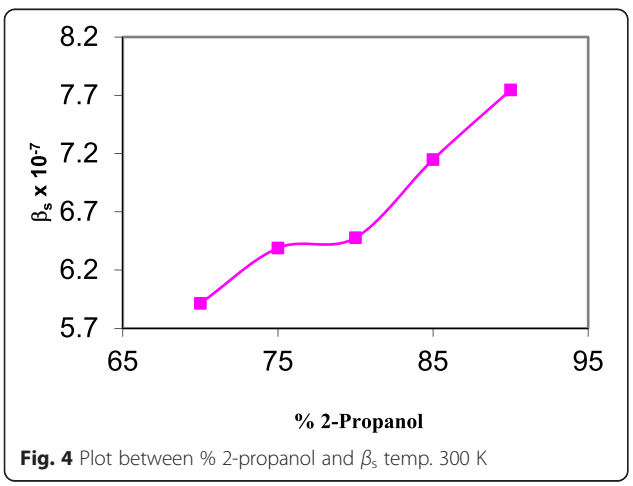

Fig. 4 Plot between % 2-propanol and β_s temp. 300 K

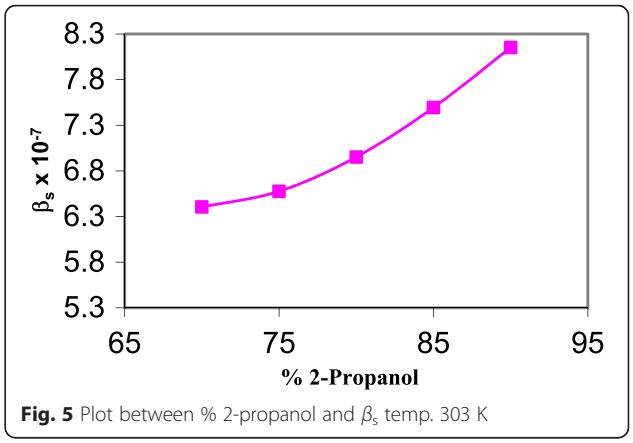

Fig. 5 Plot between % 2-propanol and β_s temp. 303 K

Specific acoustic impedance (Z) is determined from the measurement of ultrasonic velocity and density by the formula,

$$Z = V_s \times d_s$$

The value of these acoustic parameters has been used to discuss an important role in understanding the molecular interaction between the components of the mixtures and provides an insight into the physico-chemical properties of liquid mixtures.

Historical development

Interferometry is a method used to accurately measure the waves. This technique was invented by an American physicist called Albert Michelson. He conducted several experiments on light. He experimented by splitting the light into waves and combining them again. For these experiments, he invented the method of interferometry and created a device called interferometer. The size of the planets is calculated using the method of interferometry. Albert Michelson is the first American to receive the Noble Prize in Physics in 1907.

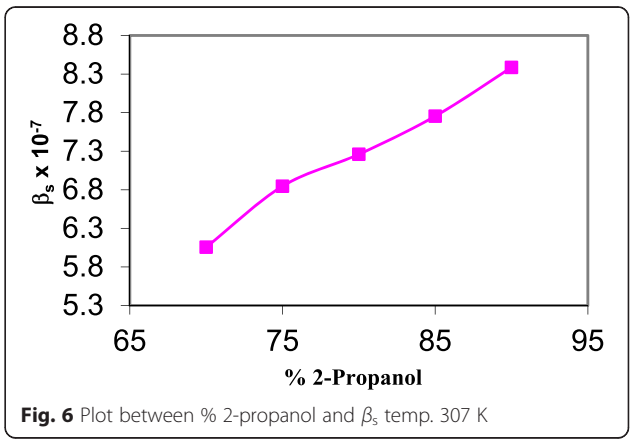

Fig. 6 Plot between % 2-propanol and β_s temp. 307 K

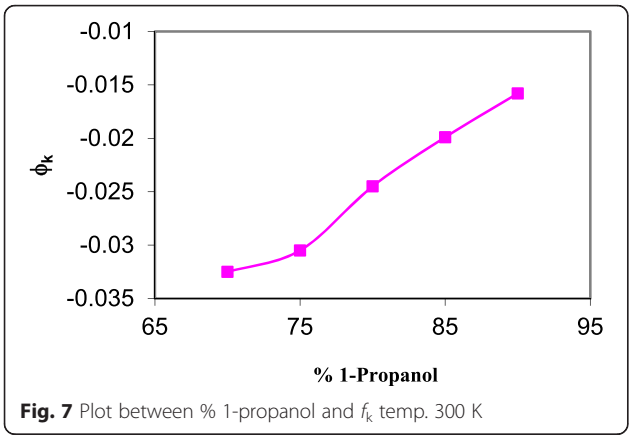

Fig. 7 Plot between % 1-propanol and f_k temp. 300 K

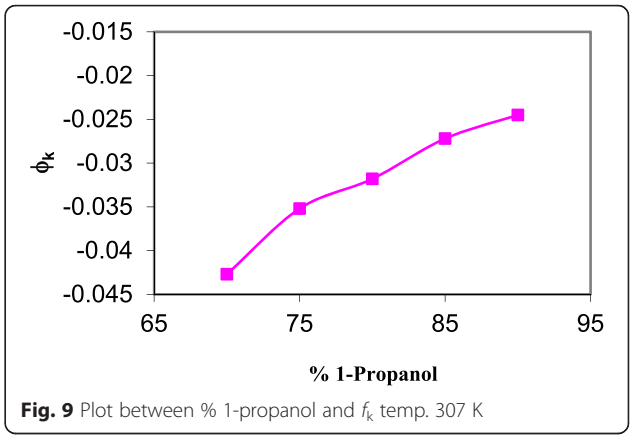

Fig. 9 Plot between % 1-propanol and f_k temp. 307 K

The volume of electrolyte solutions has been of scientific interest for a long time. The historical development of the volume of electrolyte solution can conveniently be divided into four major divisions with years 1770, 1887, 1923, and 1957 as points of change. Watson (1770) in 1970 made the first accurate measurements on the volume change of adding electrolytes to water. In 1887, Arrhenius (1887) presented his theory on the dissociation of electrolytes into ions. In 1923, Debye and Huckel (1923) presented their theory of interionic attraction, and in 1957, various workers like Ackerman (1957), Buckingham (1957), Eigen (1957), Frank and Wen (1957), Kaminsky (1957), Samcilov (1957), and Young et al. (1957) presented a number of papers on the structural hydration interactions.

Most of the researchers showed that electrostriction of an ion should be inversely proportional to the compressibility of solvent. The researchers showed that the difference between the V_0 of ions in various solvents and water shows a good linear correlation with the compressibility of the solvent. Pavan Kumar et al. (2014) reported acoustical studies of binary liquid mixtures of p-chlorotoluene in benzene at different temperatures. Acoustical studies on binary liquid mixtures of some 1,3,4-oxadiazole derivatives with acetone at 303.15 K have been given by Alamelumangai and Santhi (2014).

Instruments

For the density measurement, bicapillary pyknometer is used, and for weighing purposes, single pan digital balance (Citizen CY104) is used.

Thermostat

A special thermostatic arrangement was done for density and ultrasonic velocity measurements. Elite thermostatic water bath was used, in which continuous stirring of water was carried out with the help of electric stirrer and temperature variation was maintained within ±0.1 °C.

Ultrasonic interferometer

Ultrasonic interferometer from Mittal Enterprises, Model M-81 with an accuracy of ±0.03 % and a frequency of 1 MHz, was used for the measurement of ultrasonic velocities of different solutions.

Ultrasonic velocity measurements

The sound velocities of ligands (L_1, L_2, L_3, L_4, and L_5) were measured at the same concentration (0.01 M) of

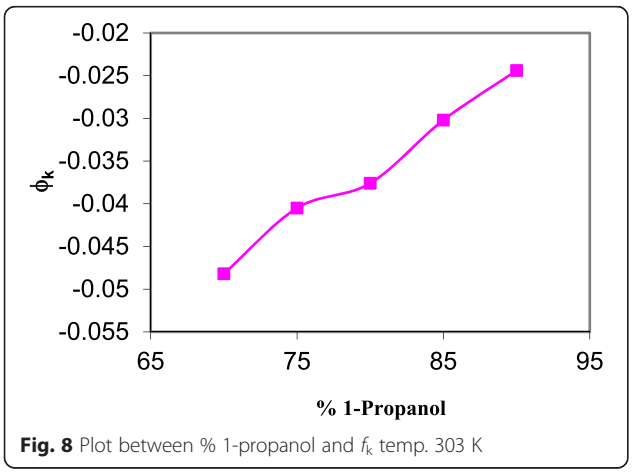

Fig. 8 Plot between % 1-propanol and f_k temp. 303 K

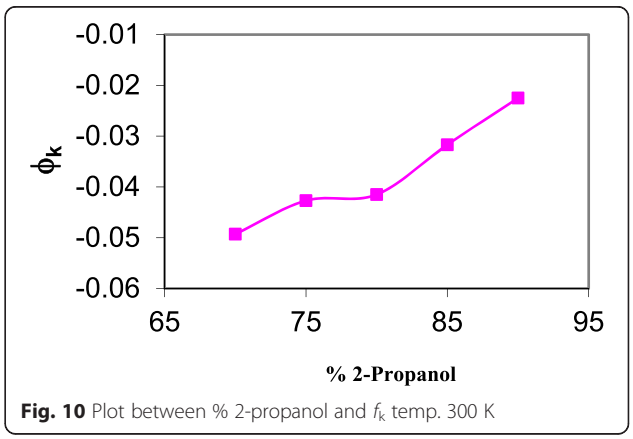

Fig. 10 Plot between % 2-propanol and f_k temp. 300 K

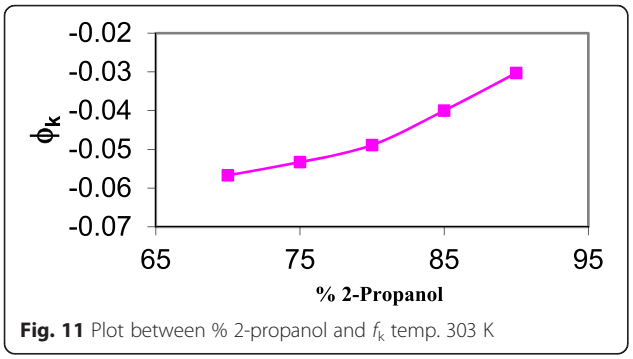

Fig. 11 Plot between % 2-propanol and f_k temp. 303 K

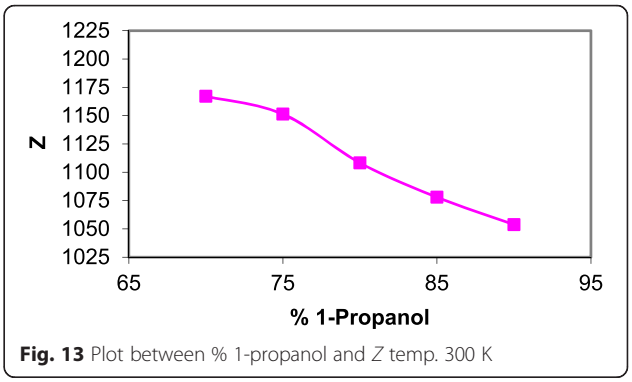

Fig. 13 Plot between % 1-propanol and *Z* temp. 300 K

ligand in the different percentages of 1-propanol-water and 2-propanol-water mixture in the concentration of 70, 75, 80, 85, and 90 % at 300, 303, and 307 K.

The cell of ultrasonic interferometer was filled fully with the solution, and the needle of the ammeter was adjusted in the range of 20 to 60 with the help of "Adj" knob. It was warmed for 10 min so that the range should remain steady. Micrometer reading was noted. A screw was moved anticlockwise to get the maximum deflection of the needle. Movement of the screw was continued to get five deflections. After returning back the needle to its original position, micrometer screw reading was noted. The difference between these two readings gave the distance traveled by the screw for getting five maxima. From this, the distance required through which a micrometer screw should move for one maxima was calculated just dividing it by 5 and multiplying by 2. The same procedure was repeated many times.

Experimental

The cell of ultrasonic interferometer was filled fully with the solution, and the needle of the ammeter was adjusted in the range of 20 to 60 with the help of "Adj" knob. It was warmed for 10 min so that the range should remain steady. Micrometer reading was noted. The screw was moved anticlockwise to get the maximum deflection of needle. Movement of screw was continued to get five deflections. After returning back the needle to its original position, micrometer screw reading was noted. The difference between these two readings gave the distance traveled by the screw for getting five maxima. From this, the distance required through which a micrometer screw should move for one maxima was calculated just by dividing it by 5 and multiplying by 2. The same procedure was repeated many times.

Results and discussion

The values of acoustic parameters (β_s, ϕ_k, and *Z*) in different percentage of 1-propanol-water and 2-propanol-water mixture for 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin at 300, 303, and 307 K are presented in Tables 1 and 2, and the graphs are shown in Figs. 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, and 18.

Adiabatic compressibility (β_s)

Adiabatic compressibility shows a strong correlation with hydrational behavior of the solute molecule and appears to be sensitive to the structural features of the solute, such as shape, size, branching, and the presence of an aromatic ring. It is argued that these aspects are important in terms of considering the geometrical fit of the solute into the ordered form of the aqueous solvent surrounding these solutes (Mohammad and Verrall 1989).

From Tables 1 and 2 and Figs. 1, 2, 3, 4, 5 and 6, it can be noted that the β_s values in 1-propanol medium are considerably and notably smaller than in 2-propanol medium.

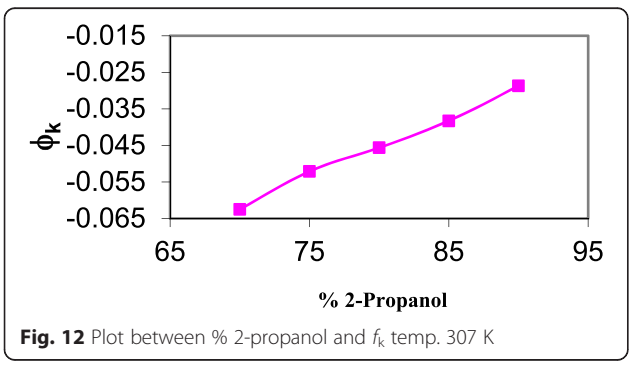

Fig. 12 Plot between % 2-propanol and f_k temp. 307 K

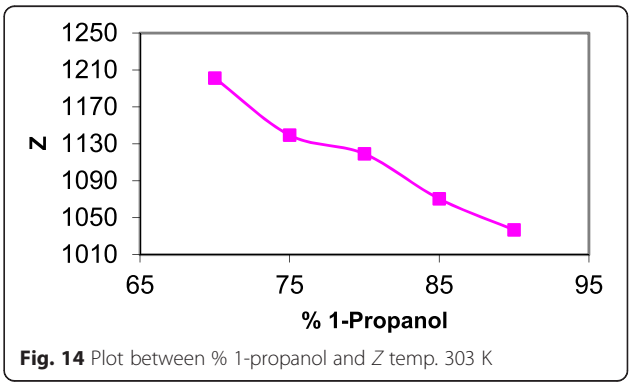

Fig. 14 Plot between % 1-propanol and *Z* temp. 303 K

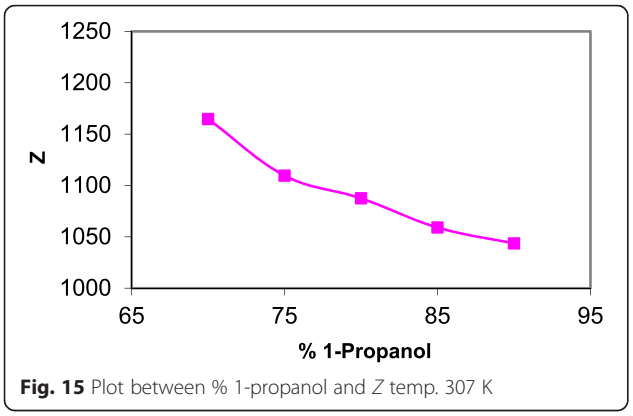

Fig. 15 Plot between % 1-propanol and *Z* temp. 307 K

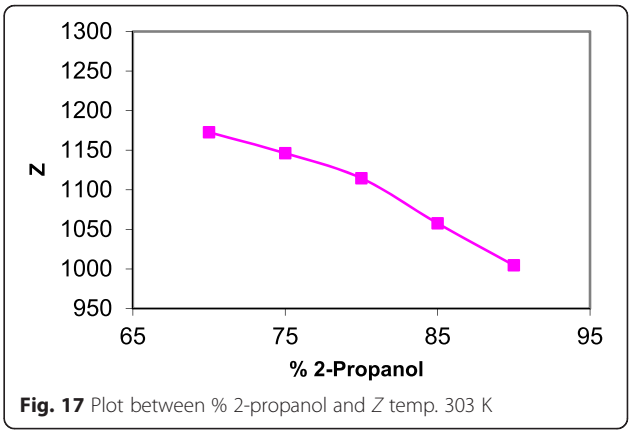

Fig. 17 Plot between % 2-propanol and *Z* temp. 303 K

This may due to the nature of solvent of 2-propanol at different temperatures.

$$\text{1-Propanol} < \text{2-Propanol}$$

It is also observed that the values of adiabatic compressibility of 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin are increased with increase in percentage of organic solvent. As the percentage of organic solvent increases, it decreases the number of free ions due to aggregation of solvent molecules around the ions (Pandey et al. 1989) showing the occurrence of ionic association due to strong ion-ion interaction. It may also be due to departure of solvent molecules around the ions. It can be noted that in both the medium, i.e., 1-propanol and 2-propanol, the β_s values of all ligands increase with increase in temperature.

It appears from Figs. 1, 2, 3, 4, 5, and 6 that when atom like chlorine is present in the molecules attached to phenyl ring, the compressibilities are increased meaning thereby decrease compactness, in spite of their polar nature and high electronegativity. The reason for this may be looked into the negative inductive and positive mesomeric effects of these groups, which are operating simultaneously but in opposite directions, neutralizing the effect of negative charge on the halogen atom. Therefore, the close packing at these ends may not be possible leaving voids between molecular species, i.e., making the solution more compressible. Similarly, decrease in compressibility because of close packing due to polar nitro group may be explained.

Apparent molar compressibility (ϕ_k)

Apparent molar compressibility (ϕ_k) is another important acoustic parameter, which explains the solute-solvent and solute-solute interactions in solutions. Thus, the structure of solute and the number of atoms present in it will have a direct effect on ϕ_k values. From Tables 1 and 2 and Figs. 7, 8, 9, 10, 11 and 12, it is observed that ϕ_k values increase with increase in the percentage of 1-propanol solvent. Similar results are observed in 2-propanol medium at 300, 303, and 307 K.

It could be observed that the ϕ_k values are negative. This interprets in terms of loss of compressibility of solute due to strong electrostatic solvation of ions. The appreciable negative values ϕ_k for all of the system reinforce our earlier view about the existence of ion-solvent interactions (Dhanlakshmi and Jasmine Vasantharani 1999). Negative values of ϕ_k show that the interactions are insensitive to the solvent. It could be also explained by postulating that

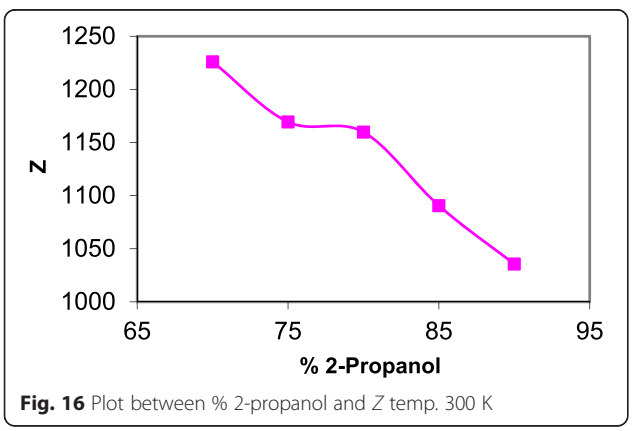

Fig. 16 Plot between % 2-propanol and *Z* temp. 300 K

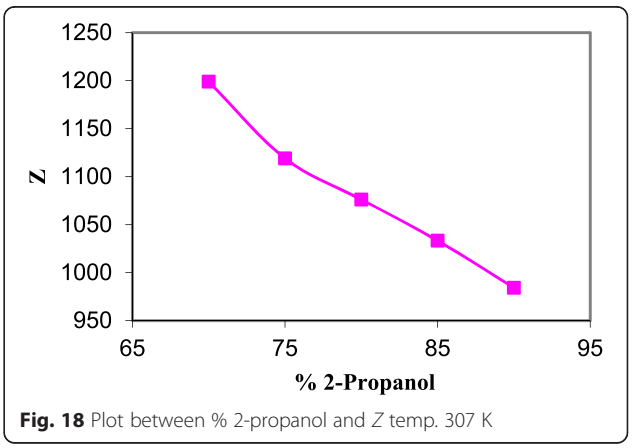

Fig. 18 Plot between % 2-propanol and *Z* temp. 307 K

the polar –OH group interacts with the surrounding organic solvent through dipole-dipole interaction in such a way that the surrounding solvent molecule loses its own compressibility to a certain extent.

From the Tables 1 and 2 and Figs. 7, 8, 9, 10, 11 and 12, the general trend for ϕ_k is seen as follows

$$\text{1-Propanol} > \text{2-Propanol}$$

The ϕ_k values of 2-propanol are lower than 1-propanol solvent. This appears to be a reverse trend than which is observed for β_s. Here, the difference between the two properties can be clearly seen. The adiabatic compressibility may just explain the simple association or close packing or clinging of molecules. But on the contrary, apparent molar compressibility is a property with difference, which may explain the molecular interactions like the structure making and structure breaking nature of a solute. Apparent molar compressibility property fairly sensitive to structural changes especially like water/ethanol and is hence expected to throw interesting light (Kaulgud et al. 1996).

From the difference in the trends in two compressibilities, adiabatic and apparent molar, it may be predicted that adiabatic compressibility can detect gross changes in interaction but minute changes due to change in structure may only be noticed by apparent molar compressibility.

Specific acoustic impedance (*Z*)

Specific acoustic impedance also makes the contribution in explaining molecular interactions. Literature survey shows that the impedance approach to explain the molecular interactions in liquid mixtures has been rather less commonly employed. This is one of the reasons why the impedance approach has been adopted here, to examine the behavior of the solutions regarding molecular interactions. The mathematical relation for specific acoustic impedance $Z = V \times d$ and adiabatic compressibility $\beta = 1/V^2 \times \mathrm{d}$ shows that their behavior is opposite.

The conventional approach based on compressibility is both useful and fundamental; however, the impedance approach serves equally well. In fact, it constitutes an additional probe for studying molecular interactions. Specific acoustic impedance is the complex ratio of the effective sound pressure at a point to the effective particle velocity at that point (Parkar 1982).

It is also observed that the Z values of 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin at 300, 303, and 307 K in 2-propanol solvents are higher than in 1-propanol solvent. This can happen only when the effective particle velocity increases, this turn means that dispersion forces should be active in mixtures, a result anticipated in the absence of any specific interactions such as hydrogen bonding. On the contrary, the Z values in 1-propanol are lower indicating hydrogen bonding. The values of specific acoustic impedance (Z) decreased with increase in percentage of 1-propanol and 2-propanol at different temperatures (Figs. 13, 14, 15, 16, 17 and 18). Therefore, the specific acoustic impedance depends upon the various structures of liquids and molecular packing of the medium.

Conclusions

Trends of acoustic properties indicate the presence of molecular interaction in the present binary mixture under study. It may be qualitatively inferred that the interaction between unlike molecules is mainly due to hydrogen bonding. The present studies investigated that with increasing the concentration of 1-propanol and 2-propanol, β_s and ϕ_k increases while Z decreases at 300, 303, and 307 K by taking constant concentration of 3-acetyl-4-methyl-6-chloro-8-nitrocoumarin (0.01 M). Better values of parameters are found for the 2-propanol and water system in 90 % ratio at 307 K. From this study, it is clear that properties, which are directly or indirectly responsible for this are protic nature of solvent, dielectric constant, polarity, density, tendency of forming hydrogen bonding, surface tension, viscosity of solvent, etc.

Authors' contributions

HV. Burghate carried out the synthesis and measured acaustic parametes of given compound under theguidance of P.B.Raghuwanshi also participated in the sequence alignment and drafted the manuscript.Soall authors read and approved the final manuscript.

Competing interests

The authors declare that they have no competing interests.

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