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Interactions of poly (ethylene) glycols in aqueous solution at 288.0K: Ultrasonic studies

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Abstract

Alcohols are biologically and industrially important amphiphilic materials that exist in the liquid state which may due to hydrogen bonding of their O-H groups. They are polar and self associated liquids. Poly (ethylene) glycol (PEG) is polymeric in nature having different molecular weight. The PEGs exist in both liquid and solid state. The PEGs having lower molecular weight exist in liquid state but PEGs having higher molecular weight exist in solid state. The study of propagation of ultrasonic waves in liquids, liquid mixtures and in solutions is very useful for examining the nature of intermolecular interactions in the system. Ultrasonic velocity is an important parameter which depends upon the structure of the molecules and interactions among them. In the present investigation ultrasonic velocities (U) and densities (ρ) are measured for aqueous solutions of poly ethylene glycols 200, 400, 2000 and 4000 at 288.0K. Various acoustical parameters such as adiabatic compressibility (β) acoustic impedance (Z) and free length (L_f) are calculated from the measured value of 'U' and 'ρ'. The Hartmann and Balizer equation is used for the computation of nonlinear parameters using B/A ratio along with Ballou equation. The variation of B/A with concentration has almost linear relation which indicates that non complex formation between the constituents of the mixtures. Acoustical parameters have been used to obtain the presence of specific interactions solute – solvent type in the PEG - water system.

Keywords: Poly (ethylene) glycols, Acoustical parameters, Ultrasonic velocity

INTRODUCTORY

In recent years much importance has been given to the physicochemical properties of mixed -solvents rather than a single solvent because of their wide range of application in chemical industrial and biological processes. The physicochemical data of mixed solvents provide a significant knowledge of their solution structures and molecular influence on the intensity of the intermolecular interactions among component molecules **[1]**. Analysis of multi components liquid mixtures in terms of concentration and their thermodynamic, volumetric and viscometric properties are important for the designing of industrial and biological process. These studies are also important in the formulation of models to relate the molecular structure with macroscopic properties of liquids **[2]**. The knowledge of thermodynamic properties of non – electrolyte solutions are useful in chemical separation, heat transfer, fluid flow and mass transfer **[3]**. Spectroscopic methods have been used to study the molecular interactions in the mixture of ethylacetoacetate and tetrahydrofuran. The study is devoted to understand the molecular interactions using steady state ultraviolet-visible, fluorescence spectroscopy, fourier transform, infrared spectroscopy and proton – nuclear magnetic resonance measurements **[4]**. The deviation in the absorption as well as emission in binary mixture has been explained in terms of molecular interactions between solvent - molecules in ground as well as in excited state, respectively. The chemical shift in proton-nuclear magnetic resonance measurements have also been used to explain the molecular interactions in the binary mixture of the solvent. The fluorescent molecules such as coumarine dyes, 4-aminophthalimide, and rhodamine 6G have been used in UV-visible and fluorescence spectrophotometric methods and the solvatochromic behavior. These molecules have been utilized to determine in terms of stabilization which is directly proportional to the polar nature of the solvents **[5, 6]**. The structure-function relationship of bio-molecules have been determined with various techniques such as X-ray crystallography, chromatography, NMR, EPR, vibrational, and Raman spectroscopy, neutron and light scattering, circular dichroism (CD) and IR **[7,8].**

Though spectroscopic methods play major role in molecular interaction studies in liquids but ultrasonic velocity in liquid is also fundamentally related to binding force between atoms or molecules. In recent years, the measurements of ultrasonic velocity have been employed to investigate the nature of molecular interactions in pure liquids and mixtures. Acoustic and thermodynamic parameters have been employed to understand the different kinds of association, various types of intermolecular interactions with their strength, the molecular packing and molecular motion / the size of pure components and in the mixtures **[9-23]**. The literature survey has revealed that many attempts have been made to study the acoustic properties of the mixture of PEGs- 400 with water, polar and non – polar solvents **[24 -29]**. The ultrasonic investigation of water mixtures with poly (ethylene) glycols 200, 400 and

ethylene glycols have been reported, in this study. The structural interactions and the formation of compact pseudo-stable structure at very low concentration of ethylene glycols and poly ethylene glycols were observed **[26]**. Variations of the acoustic parameters in the mixture of lithium hydroxide and lithium bromide in PEG-400 solutions in different concentration have been studied. The study comprises the critical evaluation of acoustic non-linearity parameters B/A using Hartmann relations and Ballous empirical relation **[9]**.

The focus of present study is to confirm the nature of interactions present in PEGs – water system as we have obtained in viscometric study **[31]**

MATERIALS AND METHODS

(A) Materials

All the polyethylene glycols namely PEG 200 (CDH), PEG 400 (CDH), PEG 2000 (MERCK), PEG 4000 (CDH), were used as received without further purification. The aqueous solutions were obtained by dissolving the desired amount of the sample in conductivity water (conductivity < 1.0μS cm^{-1} at 15^0C).

(B) Measurements

The Ultrasonic interferometer model M – 81 S manufactured by Mittal Enterprises, New Delhi, having high frequency generator (1, 2, 3 and 4 MHz) with digital Vernier micrometer(LC 0.001 mm) controlled top assembly was used in the measurement of sound velocity. Sound velocity measurements are based on the measurement on wave length of wave in the medium. The ultrasonic waves of known frequency are produced by quartz plate at the bottom of the steal cell containing liquid. The wave reflected by a movable metallic plate kept parallel to the quartz plate. The acoustic resonance gives an electrical reaction on the generator and anode current of the generator becomes maximum. If the distance is now increased or decreased and the variation is exactly one half of the wave lengths (λ/2) or multiple of it, anode current again becomes maximum. The sound velocities of the solution were calculated from the relation; Velocity (U, ms-1) = 2 × Wave length (λ) × Frequency (f). The following **Equations** were used to calculate the parameters as reported in literature **[32]**.

$$\boldsymbol{\beta = \frac{1}{U^2 \rho}} \ldots\ldots\ldots (\mathbf{1})$$

$$\boldsymbol{Z = \rho U} \ldots\ldots\ldots (\mathbf{2})$$

$$\boldsymbol{L_f = K\sqrt{\beta}} \ldots\ldots\ldots (\mathbf{3})$$

Where, 'β' is the adiabatic compressibility, 'U' speed of sound, 'ρ' density of medium and K is the temperature dependent constant (K = 93.875 + 0.375T) x 10^{-8} **[30].**

RESULTS AND DISCUSSION

Density and sound velocity measurement are performed with repetition for each binary liquid system namely PEG 200-water, 400- water, 2000- water, and 4000-water for different concentration of PEGs at 288K. Experimental values of densities and velocities of the solutions are listed as a function of molar concentration in Table 1

Table 1: Values of densities ultrasonic velocities, ρ, β, Z and L_f of water + PEGs mixture at 288K

PEG	CONC. mol.dm^{-3}	Density kg.m^{-3}	U m.sec^{-1}	β ×10^{-10} m^{2}N^{-1}	Z×10^6 kgm^{-2}s^{-1}	L_f×10^{-16} (m)
200	0.0496	992.2	1471	4.65	1.459	4.35
	0.2501	1000.6	1505	4.41	1.505	4.24
	0.5043	1008.6	1538	4.19	1.551	4.13
	0.7596	1012.8	1573	3.99	1.593	4.03
	1.0246	1024.6	1596	3.83	1.635	3.95
	1.2927	1034.2	1620	3.68	1.675	3.88
400	0.0254	1017.4	1447	4.69	1.472	4.37
	0.128	1024.2	1483	4.44	1.518	4.25
	0.258	1032.2	1504	4.28	1.552	4.17
	0.3902	1040.4	1538	4.07	1.600	4.07
	0.5249	1049.8	1570	3.86	1.648	3.97
	0.6612	1058.0	1572	3.82	1.663	3.94
	0.0050	1006.8	1479	4.54	1.479	4.30

2000	0.02524	1009.6	1494	4.37	1.508	4.22
	0.0510	1020.0	1514	4.28	1.544	4.17
	0.0770	1027.2	1565	3.96	1.607	4.01
	0.1032	1031.8	1583	3.86	1.633	3.96
	0.1303	1042.4	1603	3.13	1.671	3.57
4000	0.0025	1011.6	1434	4.80	1.450	4.42
	0.0127	1015.6	1494	4.41	1.517	4.23
	0.0256	1024.8	1527	4.18	1.514	4.12
	0.0385	1026.4	1611	4.03	1.593	4.05
	0.0517	1034.6	1645	3.53	1.666	3.79

It can be seen from the table that the density of the PEG solution increases in an almost linear manner with the concentration of the PEGs. The variation of density in a linear manner with solute concentration is the agreement with the previous report **[33]**. The density is a measure of solvent-solvent and solute-solvent interactions. The increase of density with the concentration indicates the presence of these interactions. A decrease in density may be interpreted the influence of solute on solvent as structure maker. The shrinkage in the volume results an increase in the density. The solvent – solvent interactions by bonding results and increase in size of the molecule, hence there will be a decrease in density **[34]**.

The measured sound velocity at different concentration of PEGs increases with the increase in concentration of PEGs. The plotbetween sound velocity and concentration of PEGs are shown in **Figure1A** for PEG 200, **Figure1B** for PEG 400, **Figure1C**for PEG 2000 and **Figure1D** for PEG 4000. In all the **Figures** from **Figure 1A to Figure1D,** the plots are straight line in nature. The linear plots suggest there is strong solute – solvent interactions in the solutions of a PEGs in water.

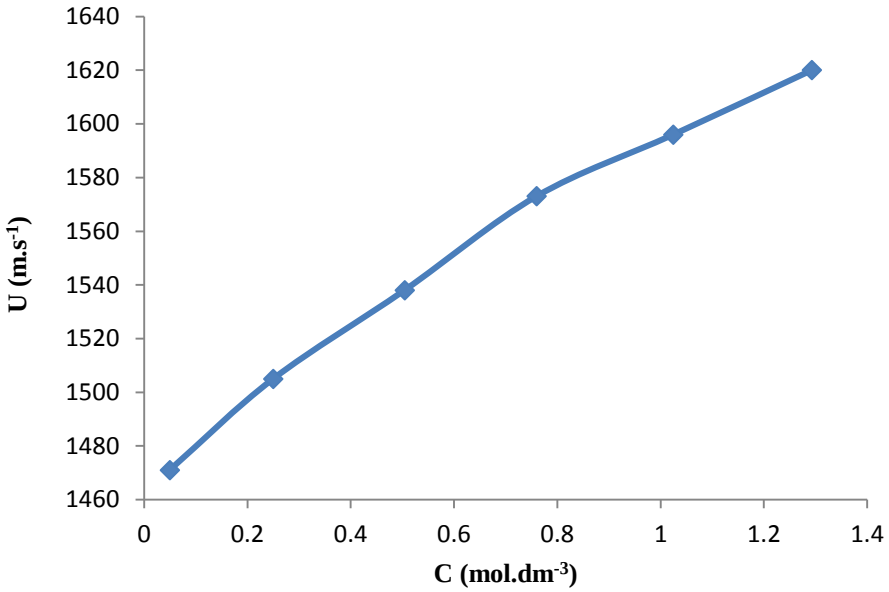

Figure 1A, Plot of Sound velocity with concentration for PEG – 200

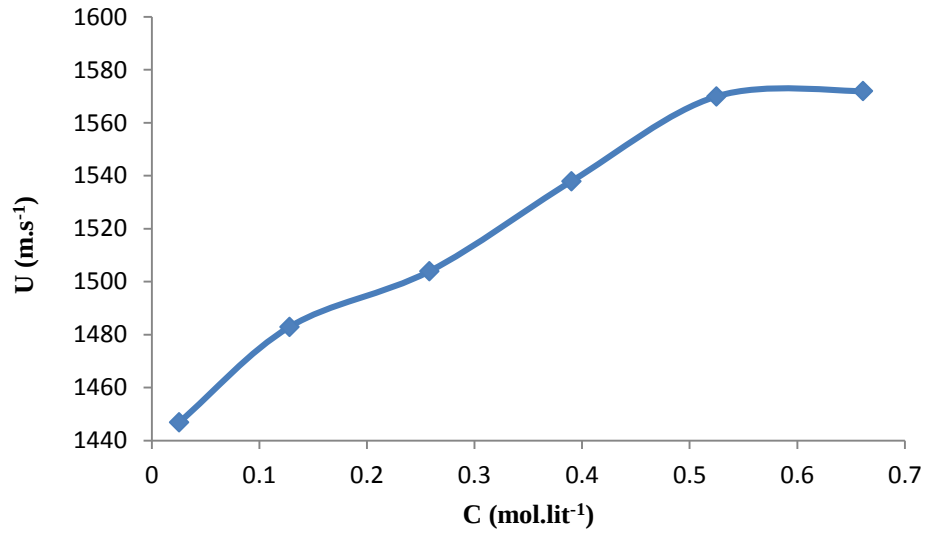

Figure 1B, Plot of Sound velocity with concentration for PEG – 400

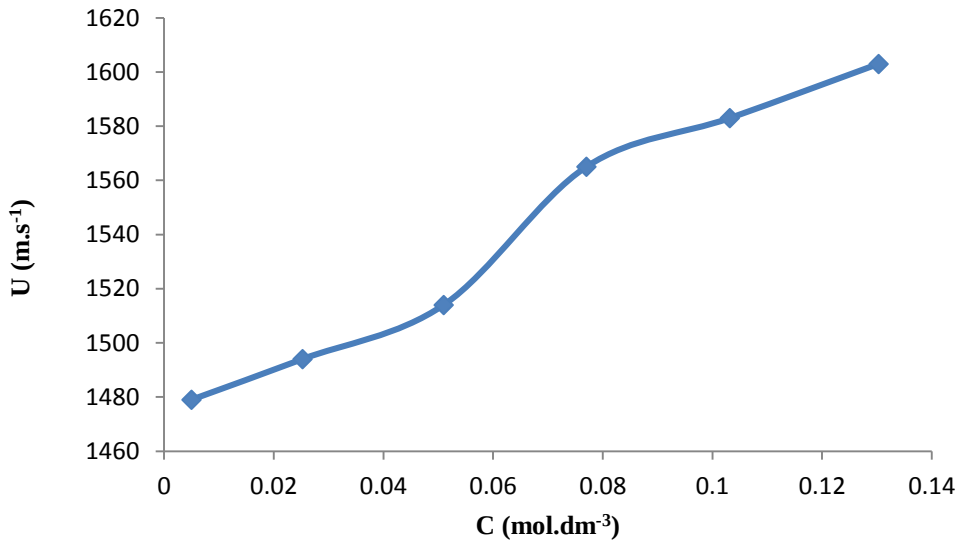

Figure 1C, Plot of Sound velocity with concentration, for PEG – 2000

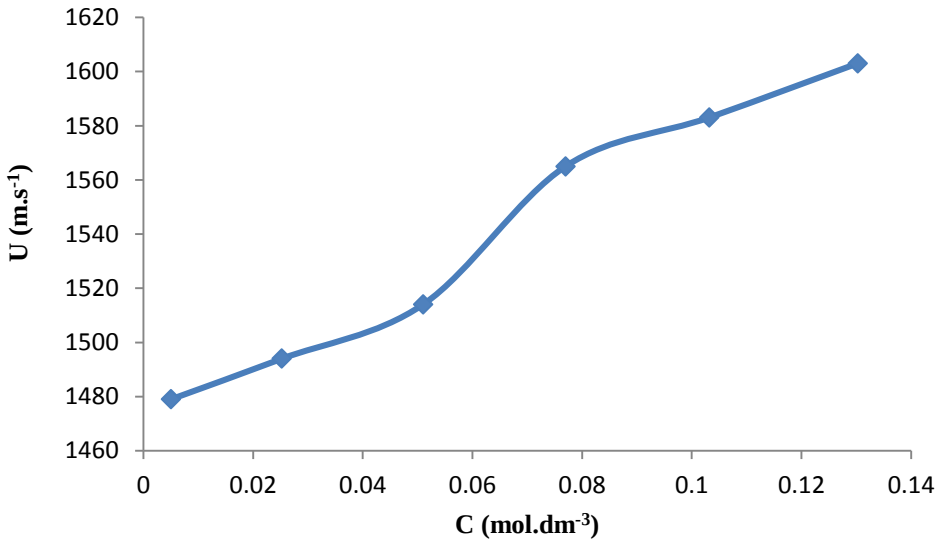

Figure 1C, Plot of Sound velocity with concentration, for PEG – 2000

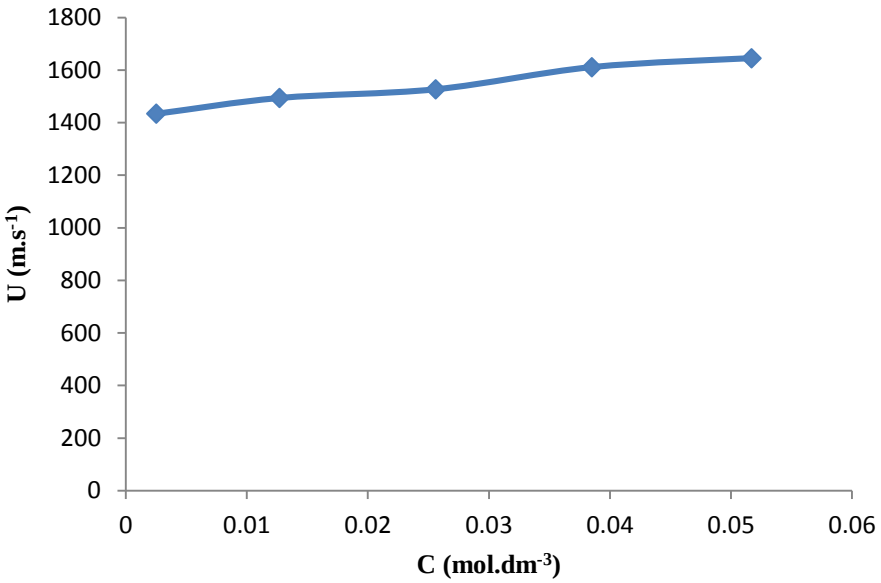

Figure 1D, Plot of Sound velocity with concentration, for PEG – 4000

The sound velocity and density data has seen used to calculate the different parameters like adiabatic compressibility, intermolecular free lengthand acoustic impedance to explore and confirm the nature and type of existing interactions in aqueous solutionsof PEGs.

The values of calculated parameters adiabatic compressibility, acoustic impedance and intermolecular free length are also given in **Table 1**.

The plots between adiabatic compressibility and concentration for all the PEGs are shown in **Figure 2A, Figure 2B, Figure 2C** and **Figure 2D** for PEG 200, PEG 400, PEG 2000 and PEG 4000 respectively.

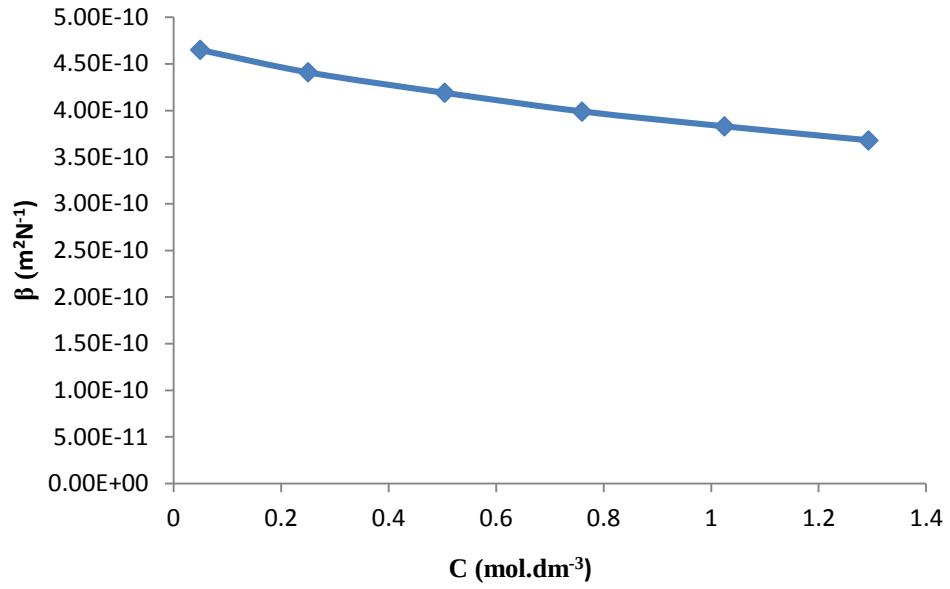

Figure 2A, Plot between adiabatic compressibility, β and concentration, C of PEG 200

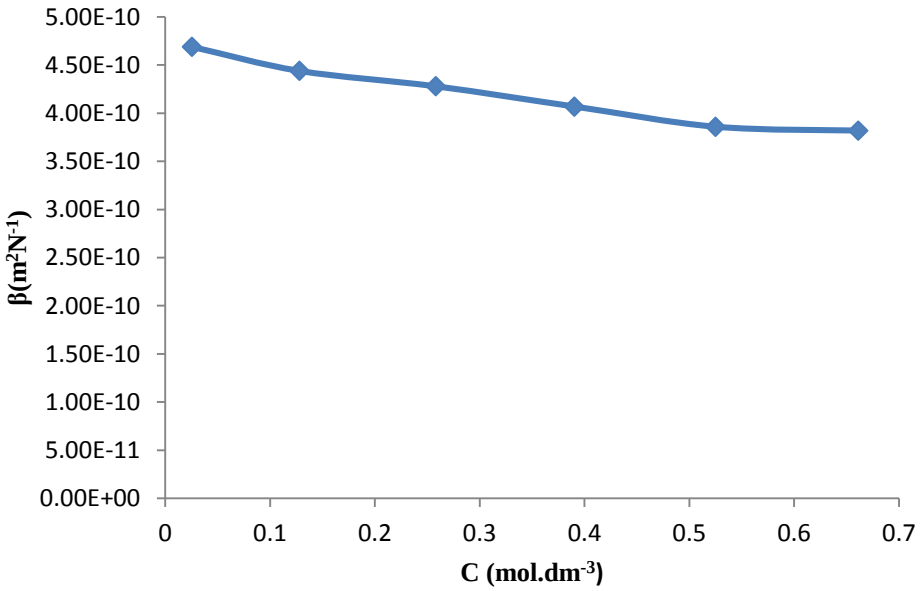

Figure 2B, Plot between adiabatic compressibility, β and concentration, C of PEG 400

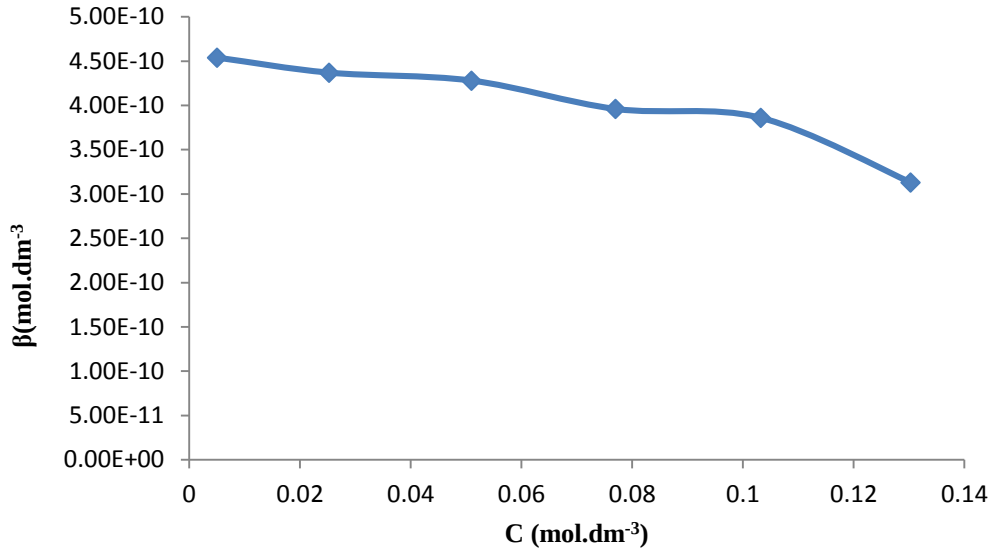

Figure 2C, Plot between adiabatic compressibility, β and concentration, C of PEG 2000

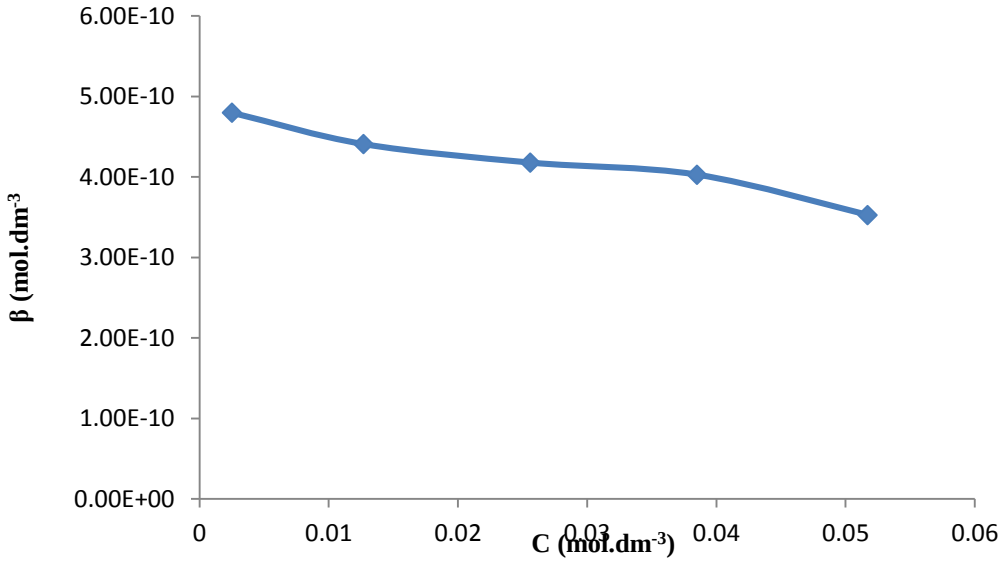

Figure 2D, Plot between adiabatic compressibility, β and concentration, of PEG 4000

Adiabatic compressibility is the reciprocal of bulk modulas; it is the property of the substance capable of bringing reduction in volume by application of pressure. The plot shown in **Figure 2A, Figure 2B, Figure 2C and Figure 2D** indicates that adiabatic compressibility decreases with increase in concentration for all the PEGAs. Decrease in adiabatic compressibility indicates that the medium become more compressible and it also suggest that the interactions of water molecules around PEGs molecules. The observed trained in adiabatic compressibility indicate the formation of a relatively incompressible moiety.

The intermolecular free length is again a predominant factor in determining the existing interactions among the component of the solutions. Analysis of the data of **Table 2** reflects a similar trend for all the PEGs as that of adiabatic compressibility. The linear variation of intermolecular free length with the concentration of PEGs are shown in **Figure 3A, Figure 3B, Figure 3C and Figure 3D** for PEG 200, PEG 400, PEG 2000 ,and PEG 4000 respectively.

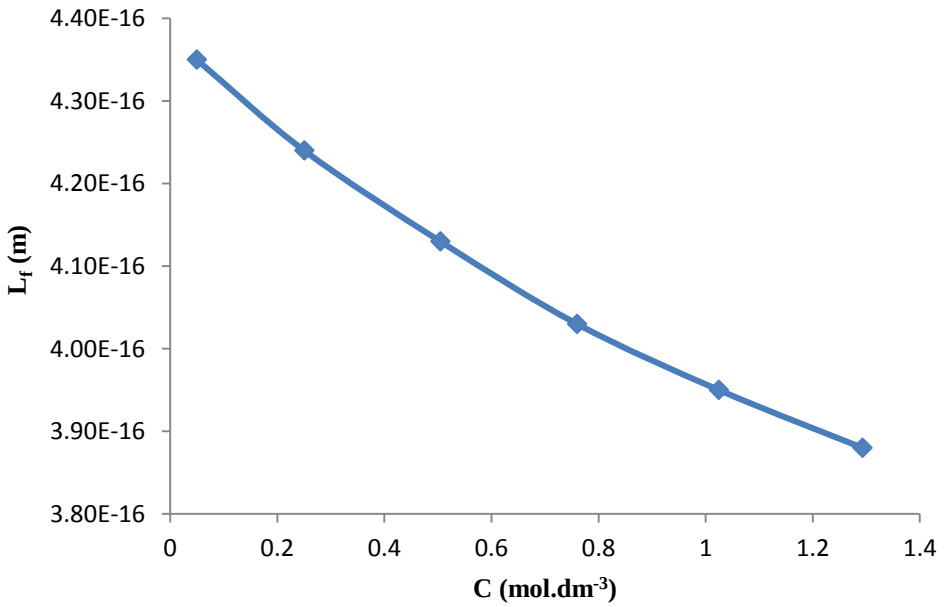

Figure 4.3A,Plot between inter molecular free length, L_f and concentration, C for PEG 200

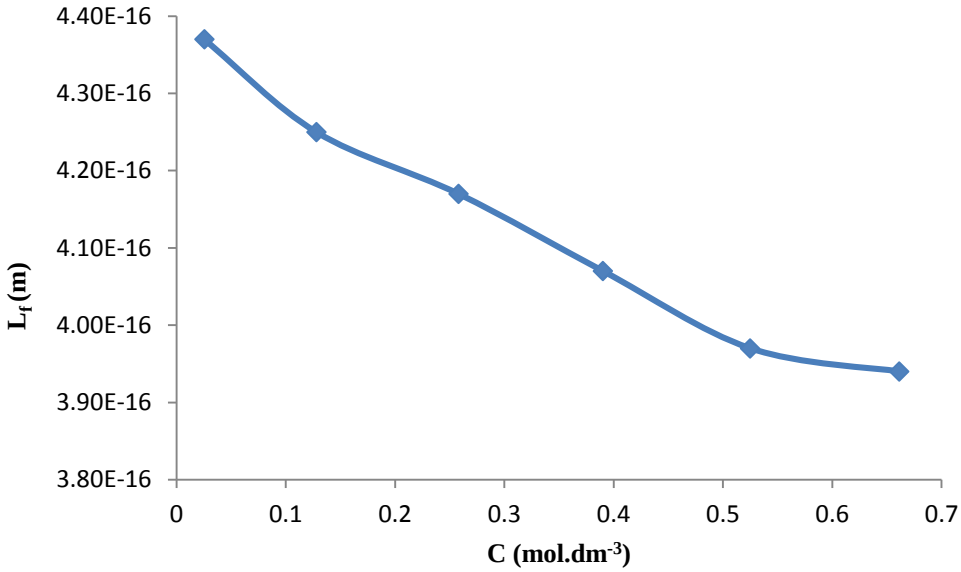

Figure3B,Plot between inter molecular free length, L_f and concentration, C for PEG 400 concentration, C for PEG 400

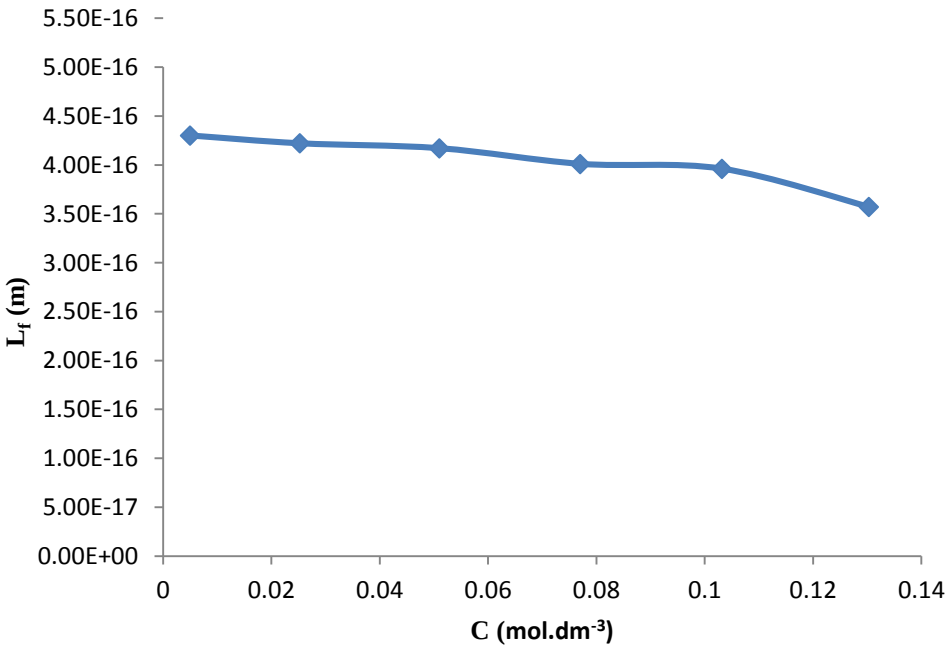

Figure 3C,Plot between inter molecular free length, L_f and concentration, C for PEG2000

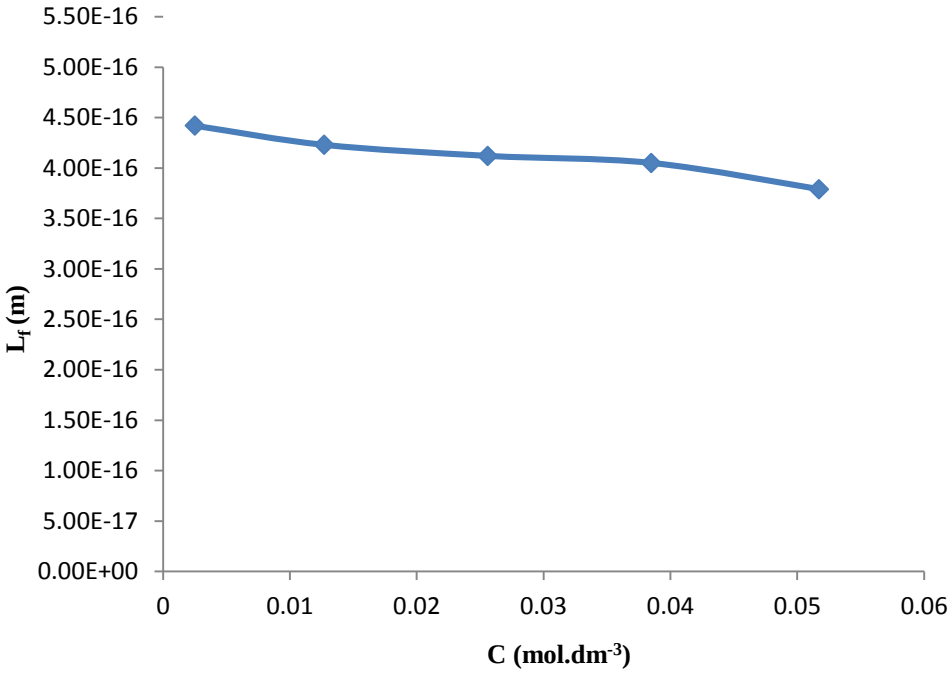

Figure 3D,Plot between inter molecular free length, L_f and concentration, C for PEG 4000

These plots indicate that intermolecular free length decreases with increase in concentration of PEGs. The decreases in the intermolecular free length confirm the presence of significant interactions between PEG and solvent molecules. Hence the PEGs may be considered as structural promoter in the systems investigated. These results also supports to our results obtained in the viscometric studies in the previous study **[31]**. Analysis of the data of **Table 2** reflects a similar trend in acoustic impedance calculated for PEGs solution in different concentration in water. The values of Z increase with the concentration of PEGs can be explained on the basis of hydrophobic interactions between PEG and water molecules which increases the intermolecular distance, resulting relatively wider gap between the molecules and take part for the propagation of sound wave. The linear variation of acoustic impedance with the concentration of PEGs are shown in **Figure 4A, Figure4B, Figure 4C and Figure 4D** for PEG 200, PEG 400, PEG 2000 ,and PEG 4000 respectively.

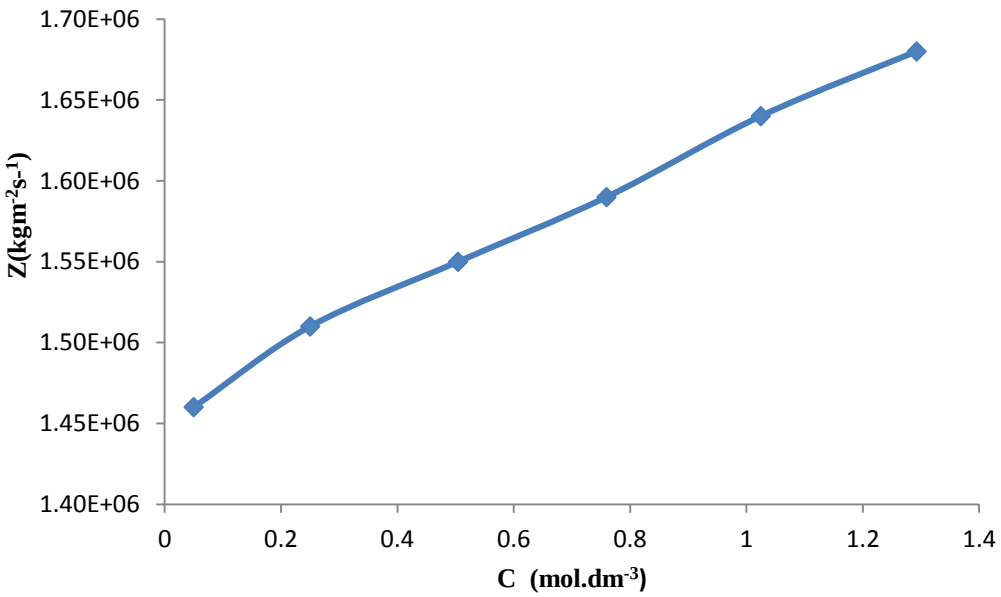

Figure 4A, Plot between acoustic impedance, Z and concentration C of PEG 200

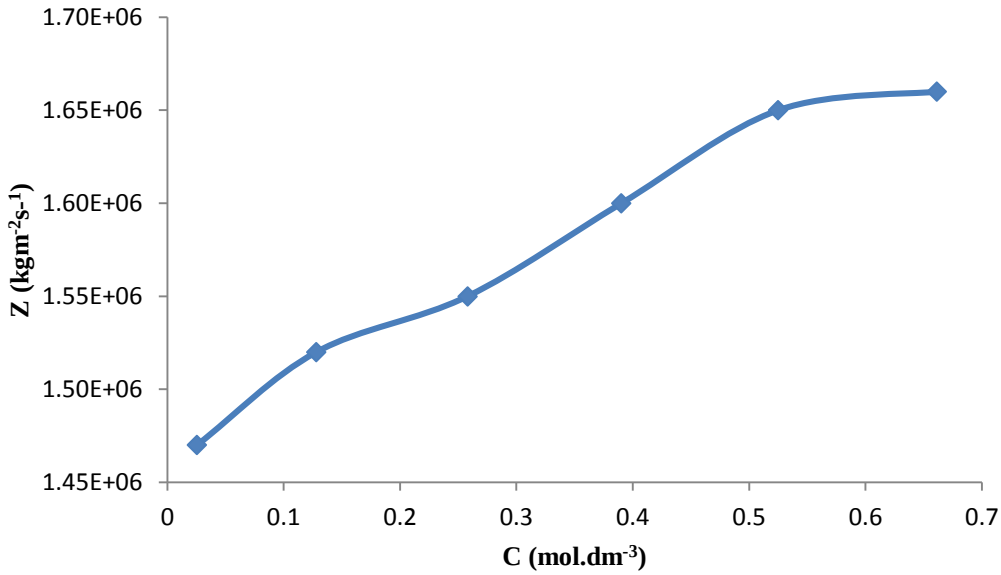

Figure 4B,Plot between acoustic impedance, Z and concentration, C of PEG 400

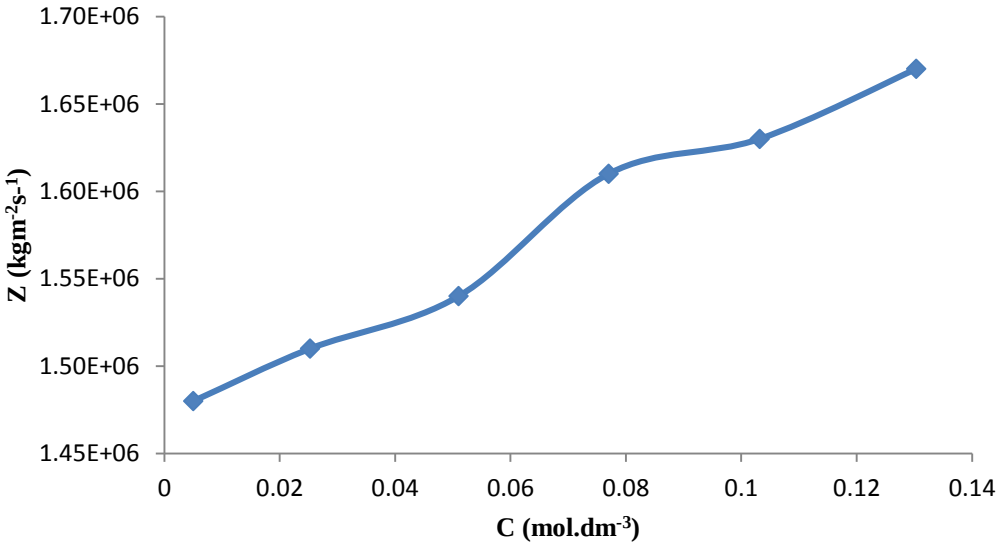

Figure 4C, Plot between acoustic impedance, Z and concentration, C of PEG 2000

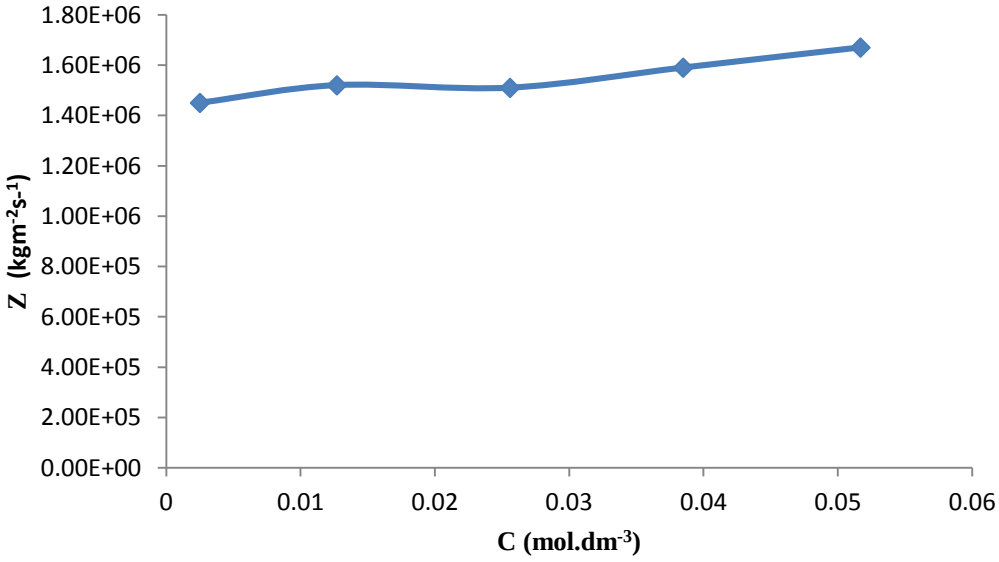

Figure 4D, Plot between acoustic impedance, Z and concentration, C of PEG 4000

The results obtained from all the parameter acoustic impedance, adiabatic compressibility and intermolecular free length are in good agreement with each other all these results confirms the presence of specific interactions solute – solvent type in all the PEG –water systems.

At the last the computation of non linear parameters using B/A ratio has been used to results obtained in the study. The velocity of the solution has been used to calculate the B/A value.

COMPUTATION OF NONLINEAR PARAMETER USING B/A RATIO

A number of theoretical methods have been purposed for calculating nonlinearity parameters (B/A) for pure liquids and liquid mixtures **[35]**. The B/A values for the liquids have been interpreted as the quantity representation the magnitude of the hardness of the liquid. These values are concerned with interaction between compounds of the binary systems. In the present study the following relation have been used for calculating the B/A values **[36, 37]**

Hartmann and Balizer equation

$$\frac{\boldsymbol{B}}{\boldsymbol{A}} = \mathbf{2} + \frac{\mathbf{0.98 \times 10^4}}{\boldsymbol{U}} \ldots\ldots\ldots (\mathbf{4}) Uinms^{-1}$$

Ballou equation

$$\frac{\boldsymbol{B}}{\boldsymbol{A}} = -\mathbf{0.5} + \frac{\mathbf{1.2 \times 10^4}}{\boldsymbol{U}} \ldots\ldots\ldots (\mathbf{5}) Uinms^{-1}$$

The velocity of sound has been used to calculate the B/A values by using the **Equation 4** and **5**. The obtained values are collected in **Table 3**. It is evident from **Table 3** the interaction between PEG and water in binary mixture is stronger at lower concentration of PEGs while it is weaker at higher concentration of PEGs. The variation of B/A with concentration has almost linear relation. It indicates that the no complex formation between the constituents of the mixtures. In case of PEG 400 in presence of lithium hydroxide and lithium bromide the formation of complex have been concluded from the B/A values **[9]**. The magnitudes of B/A values for all the PEGs solutions calculated by both the equations are nearly same lies between 8.83 to 6.79 these results are in good agreement with reported literature **[9]**. These values are inverse proportional to the concentration of PEG.

Table 3: The values of B/A with concentration.

PEG	CONC. mol.dm^{-3}	(B/A)^a m^{-1}.sec	(B/A)^b m^{-1}.sec
200	0.0496	8.66	7.66
	0.2501	8.51	7.47
	0.5043	8.37	7.30
	0.7596	8.23	7.13
	1.0246	8.14	7.02
	1.2927	8.05	6.91
400	0.0254	8.77	7.79
	0.128	8.61	7.59
	0.258	8.51	7.48
	0.3902	8.37	7.30
	0.5249	8.24	7.14
	0.6612	8.23	7.13
2000	0.0050	8.63	7.61
	0.02524	8.56	7.53
	0.0510	8.47	7.43
	0.0770	8.26	7.17
	0.1032	8.19	7.08
	0.1303	8.11	6.98
4000	0.0025	8.83	7.87
	0.0127	8.56	7.53
	0.0256	8.42	7.36
	0.0385	8.08	6.95
	0.0517	8.07	6.79

'a' values obtained from Hartmann 'b' from Ballou equation.

CONCLUSION

In the present study the nature of the interactions in PEG – water system with different molecular mass (200, 400, 2000 and 4000) have been examined using density and acoustical parameters. The acoustical parameters β and L_f indicates the presence of solute – solvent interactions in all the systems. In case of all the PEGs the value of B/A ratio obtained from Hartmann, Balizer and Ballous equation also support

the equation results with conclusion of acoustical parameters.The results obtained from different parameters are in good agreement with each other. The nature of all the PEGs in water as structure maker has been confirmed obtained in viscometric study **[31]** in the same temperature and concentrations range.

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