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Frequency and Temperature Dependence of Ultrasonic Properties of Aqueous PVA solutions Using Pulse-Echo Technique

S. V. Khangar (Wagh)[1], O. P. Chimankar[2], R. Y. Bakale[1], Y.S Tamgadge[1], S. H. Nimkar[2]
[1]Department of Physics, S Mahatma Fule Arts, Commerce & Sitaramji Chaudhari Science Mahavidyalaya Warud, Dist. Amravati, SGBAU, Maharashtra, India
[2]PG department of Physics, RTM Nagpur University, Nagpur, Maharashtra, India
[3]Shivaji Arts, Commerce and Science College, Daryapur Road, Akot, Maharashtra, India

ABSTRACT
Molecular interaction study in polymers provides valuable information regarding internal structure, molecular association, etc. Ultrasonic investigations of polar- polar or polar- non-polar components are important in understanding the intermolecular interaction between component molecules. The ultrasonic pulse echo technique provides an effective and reliable tool to investigate properties of polymer solutions in the light of phase separation studies. The propagation of ultrasonic waves and the measurement of ultrasonic properties in solution form an important tool for the evaluation of various acoustical and thermo- dynamical parameters which give an insight into the nature of miscibility/compatibility and molecular interactions in polymer solution. The phenomenon polymer- solvent miscibility may arises due to any specific molecular interactions such as hydrogen bonding, dipole- dipole interactions and charge transfer complexes for homogeneous polymer- solvent mixture. Miscibility is an important phenomenon in polymer solution to achieve mechanical integrity, better adhesion, better processing and optimum property gain. Recently the use of ultrasonic waves has shown a great potential for characterization of polymer solutions.
In present work, aqueous PVA (Polyvinyl alcohol) are often used due to various useful properties in medical field. However, temperature changes also affect these properties. In this paper we present a method for measuring ultrasonic velocity, density, viscosity and ultrasonic absorption of polymer solutions. We also give experimental values for PVA solutions for temperatures between 288K to 308 and for 2 MHz ultrasound frequency.
Keywords : Ultrasonic, acoustic and optical parameters, PVA-polyvinyl alcohol, molecular interactions, pulse echo technique

I. INTRODUCTION
The propagation of ultrasonic waves and the measurement of their velocity in solution at different concentration (wt.%) and at different temperature form an important tool for the evaluation of various acoustical and thermo- dynamical parameters which give an insight into the nature of molecular interactions such as hydrogen bonding, dipole- dipole interactions and charge transfer complexes for homogeneous polymer- solvent mixture. Polyvinyl

Alcohol having molecular Formula $[C_2H_4O]_n$. For many applications Polyvinyl Alcohol is prepared in water solutions. On evaporation of water, transparent films are formed which have high tensile strength and tear resistance. The binder characteristics of Polyvinyl Alcohol offer excellent adhesion to porous, water-absorbent surfaces. Structural analysis of polymer is a subject of considerable interest in polymer science. It is an environmental friendly and water-soluble synthetic polymer with excellent film forming property, and emulsifying properties and outstanding resistance to oil, grease, and solvents. It has been extensively used in adhesive, in textile warp sizing and finishing, in paper size and coating, in the manufacturing of PVAc emulsion, in the suspension polymerization of PVC, and as binder for ceramics. It is sometimes supplied as beads or as solutions in water[1]. A review of literatur[2-5] reveals that ultrasonic parameters, such as adiabatic compressibility (β_a), relaxation time (τ), acoustic impedance (z), free length (L_f) and optical parameter refractive index have been used to study the molecular interactions in polymer solutions.

In present work, the authors have prepared the polyvinyl alcohol solutions at different concentrations (wt.%) with water solutions and have measured ultrasonic velocity, density and viscosity using ultrasonic pulse echo technique at temperature range 288K- 308K and at 2MHz and optical parameter refractive index (μ) in order to study the structural changes to the solutions, if any. Ultrasonic pulse-echo systems are widely used to estimate properties of liquids. A common principle is to use a buffer material (buffer-rod) fixed to the ultrasound transducer. Assuming the acoustic properties of the buffer-rod are known, it is then possible to calculate the acoustic impedance of the unknown material from reflections between the buffer-rod and the unknown material. From acoustic impedance and speed of sound it is the possible to calculate density and adiabatic compressibility of the material. The nature of polymer and polymer-solvent interactions and effect on molecular interactions have been studied.

II. METHODS AND MATERIAL

polyvinyl alcohol (AR grade) from Otto Chemi was used. Ultrasonic velocity, density, viscosity and refractive index (μ) values are estimated in the aqueous polyvinyl alcohol solutions. The solution was prepared by adding a known weight of polymer (i.e. polyvinyl alcohol) to a fixed volume of distilled water and then stirring until clear solution were obtain. The concentration range chosen in the solution are 0.05, 0.1, 0.15, 0.2, 0.25, & 0.3 wt. %. Velocity measurements are carried out using ultrasonic pulse echo technique by using MHF-400 High frequency pulser- receiver supplied by Roop Telsonic Ultrasonic Limited; Mumbai- (India) at different frequencies of 1 MHz, 2MHz & 5MHz and at temperature range 288K-308K with an accuracy of ±0.1m/sec. Density has been measured by pycnometer method. The viscosity of liquid was measured by Oswald's viscometer. And refractive index is measured by Abbes refractometer. Temperature is maintained at a constant range by Plasto Crafts (LBT-10) Thermostat and other related parameters calculated by standard formulae10.

III. RESULTS AND DISCUSSION

The variations of ultrasonic velocity (u), density (ρ), viscosity (η), refractive index (μ) and the related parameters such as adiabatic compressibility (β_a), acoustic impedance (z), relaxation time (τ), free length (L_f), volume expansivity(α) and Molwen Moelwyn-Hughes(C_1) of the various concentrations (wt.%) at temperature range 288K-308K and at 2MH frequency for aqueous PVA are given below:

Figure1. represents the variations of ultrasonic velocity with concentration (wt%). From the graph It is observed that ultrasonic velocity increases with increase in concentration (wt. %) of polyvinyl alcohol

in water indicating association in the component molecules. The increment in velocity with concentration implies a decrement in the compressibility. This behavior suggests the formation of a more rigid structure, possible due to polymer-water hydrogen bonding at sites on the polymer's hydroxyl (OH) group[3]. Ultrasonic velocity increases with increase in temperature. At higher temperature ultrasonic velocity is high and at lower temperature ultrasonic velocity is low indicates the strong association between solute and solvent. Oxygen atom (O) in polyvinyl alcohol can form hydrogen bond with hydrogen of hydroxyl molecules through hydrogen bonding.

Figure 2. shows variation of density with concentration (wt.%). From graph It is observed that density increases with increase in concentration of polyvinyl alcohol in water, increase in density decreases the volume indicating association in component molecules.

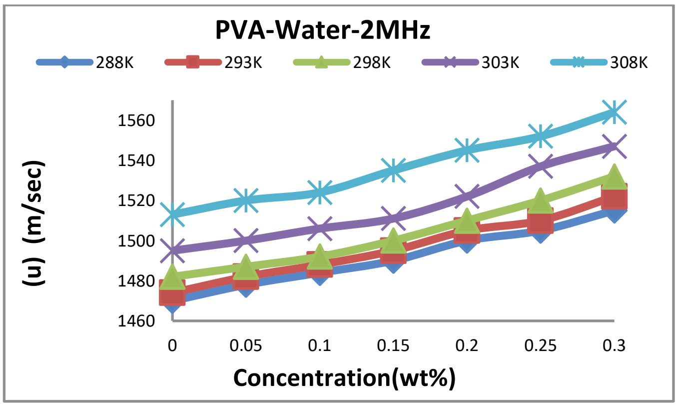

Fig.1

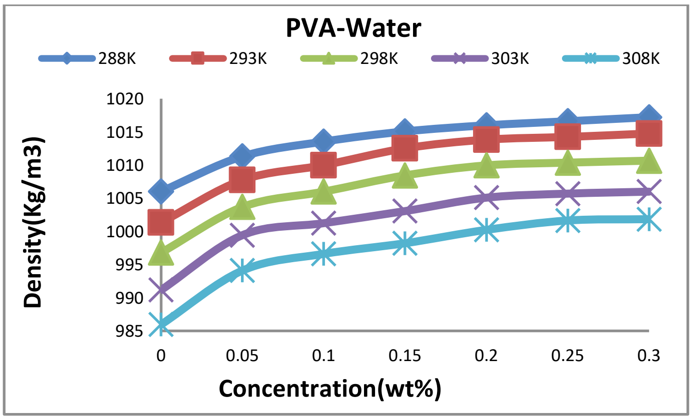

Fig.2

Increasing the temperature of the substance decreases its density (ρ) and viscosity (η). Decrease in density (ρ) and viscosity (η) with increase in temperature indicated decrease in cohesive forces. The increasing temperature has two opposite effects namely increase of molecular interactions (structure formation) and destruction of structure formed previously as a result of thermal fluctuation. Thus, increase of temperature favors increase of kinetic energy and volume expansion and hence decrease of density (ρ) and viscosity (η), while it increases intermolecular distance (free length).

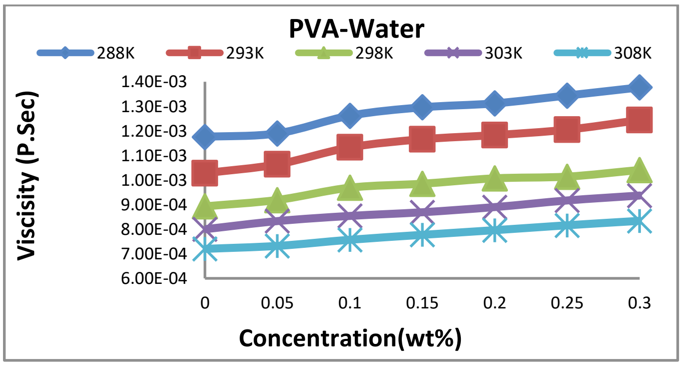

Fig.3

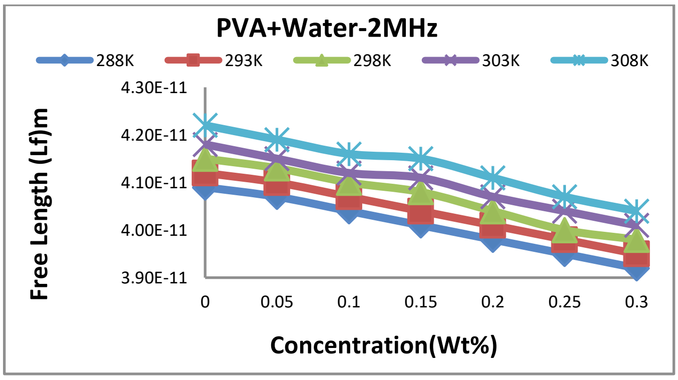

Fig. 4

Figure 3 and 4 shows the plot of viscosity (η) and free length versus concentration (wt. %) in aqueous PVA solutions. It is observed that viscosity slightly increases with increase in concentration (wt. %) of polyvinyl alcohol in water while free length decreases. Measurement of viscosity in binary mixture yields some reliable information in the study of molecular interaction. The viscosity gives the strength of molecular interaction between interacting molecules. The dipole-dipole interactions of permanent dipoles in constituent molecules increase the viscosity in the polyvinyl alcohol + water system7.

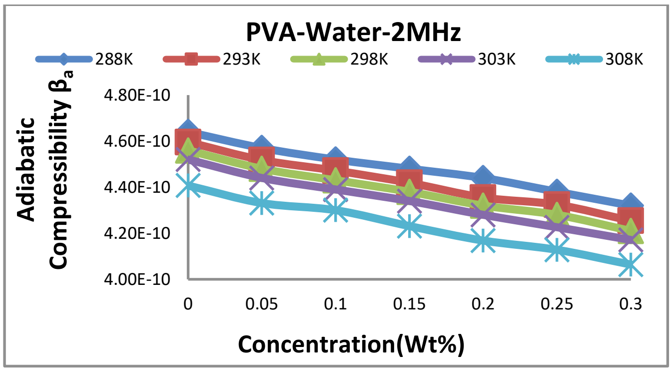

Fig.5

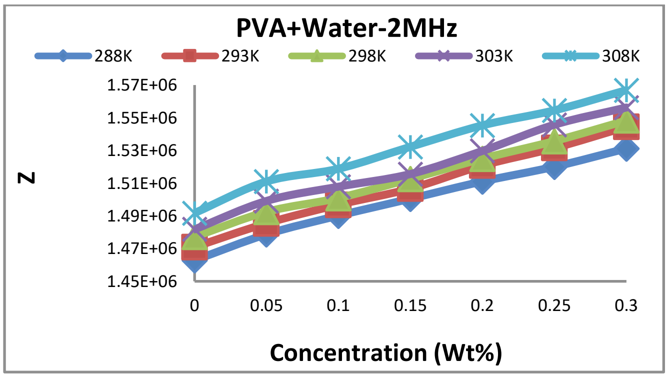

Fig.6

From figure 5, It is also observed that adiabatic compressibility decreases with increase in concentration (wt. %) of polyvinyl alcohol in water indicating strong intermolecular interaction in the component molecules in this system shows associating tendency of the component molecules. The observed decrease of adiabatic compressibility with concentration indicates the enhancement of degree of association in the component molecules. Hence the intermolecular distance decreases with increase in concentration. It is primarily the compressibility that changes with structure which leads to change in ultrasonic velocity[8].

From figure 6, It is observed that, the values of acoustic impedance increase with increase in concentration. It is in good agreement with the theoretical requirements because ultrasonic velocity increases with increase in concentration. The increase in acoustic impedance (Z) with concentration can be explained on the basis of intermolecular interaction between component molecules, which decreases the intermolecular distance, making relative fewer gaps between the component molecules[9].

From fig. 7, it is observed that relaxation time increases slightly with increase in concentration of polyvinyl alcohol in water indicating high stability of polyvinyl alcohol molecules. Polyvinyl alcohol is a polymer having compact structure and hence molecules are not stable which increases the relaxation time. The relaxation can be caused by the energy transfer between translational and vibrational degrees of freedom and all these degrees take part in the observed process[9]. Its behavior depends on viscosity and adiabatic compressibility of the liquid solution. In this system viscosity plays very important role for increasing relaxation time with increase in concentration (wt.%).

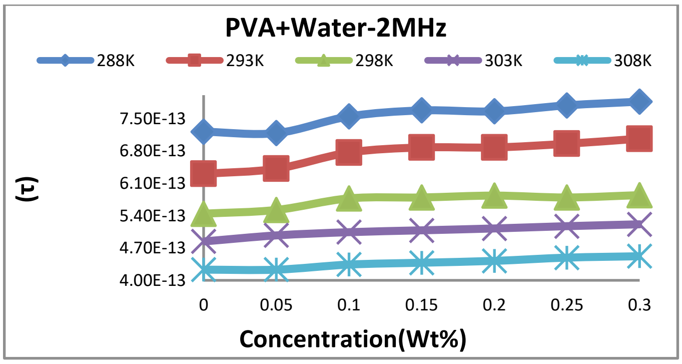

Fig.7

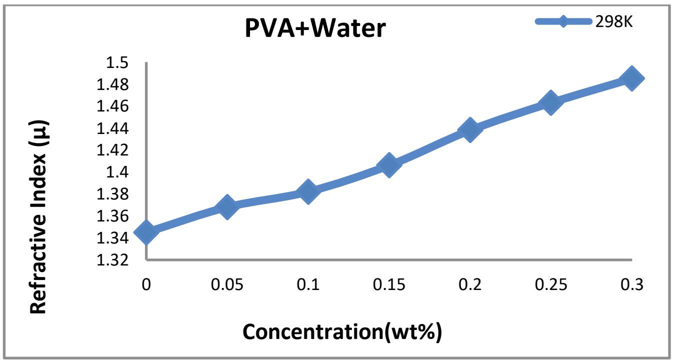

Fig.8

Figure 8 shows the variation of refractive index with concentration in aqueous PVA respectively. Refractive index (μ) increases with increase in concentration of polyacrylamide in water. It may be due to possible specific interactions like hydrogen bonding to some extent between the hydroxyl group of water. Hence the present study indicates the existence of miscibility windows only.

IV. CONCLUSION

1. The linear nature of ultrasonic velocity suggests miscibility of polymers indicated by single phase formation and thus suggesting compatibility of the polymers.
2. Ultrasonic velocity, density, viscosity, adiabatic compressibility, acoustic impedance, relaxation time, free length, etc. indicates the strength of molecular interactions in the binary liquid mixtures of polymer.
3. Decrease in adiabatic compressibility and free length with increase in concentration (Wt. %) is due to strong association.

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