Journal of Science and Technology
ISSN: 2456-5660 Volume 9, Issue 07 (July -2024)
www.jst.org.in *DOI:https://doi.org/10.46243/jst.2024.v9.i07.pp01- 12*

Ultrasonic Investigation of Molecular Interactions in Polymethylmethacrylate-TolueneBinary Liquid Mixture

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To Cite this Article

Indu Saxena,*Preeti Yadav, Divyanshi Mishra and Aditya Gupta**, "Ultrasonic Investigation of Molecular Interactions in Polymethylmethacrylate-TolueneBinary Liquid Mixture"** *Journal of Science and Technology, Vol. 09, Issue 07,- July 2024, pp01-12*

Article Info
Received: 26-02-2024 Revised: 09-04-2024 Accepted: 15-06-2024 Published: 30-07-2024

Abstract- Understanding molecular interactions in liquid mixtures is crucial for various industrial and scientific applications. In this study, ultrasonic technology was employed to investigate the nature and power of molecular interactions in a binary liquid mixture of polymethylmethacrylate (PMMA) and toluene at two different temperatures (308.15K and 298.15K) using a 2 MHz frequency. Acoustical characteristics including adiabatic compressibility (βad), intermolecular free length (L_f), acoustic impedance (ƶ), relaxation time (τ), free volume (V_f), and surface tension (S) were evaluated based on measured values of density (ρ), ultrasonic velocity (u), viscosity (η), and specific conductance. The results revealed significant variations in these parameters with temperature, indicating temperature-dependent molecular interactions. The calculated specific conductance provided additional insights into the conductivity of the liquid mixture. Overall, this study elucidates the complex molecular interactions in the PMMA-toluene binary system, providing valuable information for understanding its behavior and potential applications in various fields.

Keywords-- Ultrasonic technology, Molecular interactions, Binary liquid mixture, Polymethylmethacrylate (PMMA), Toluene, Temperature-dependent analysis

1. Introduction-

Polymers have become ubiquitous in modern life, permeating various aspects of our daily routines. From the adhesives that bond our materials together to the packaging that preserves our food, and from the fibers that clothe us to the high-tech composites in our electronic devices, polymers play an indispensable role in our world. Among these polymers, polymethylmethacrylate (PMMA) stands out as a versatile material with a myriad of applications spanning diverse industries [1]. The exploration of ultrasonic parameters has emerged as a burgeoning field of study in recent years, particularly in the context of liquid mixtures [2]. Understanding the intricate molecular interactions within these mixtures is essential for elucidating their physio-chemical behaviors, with implications for both industrial processes and product applications [3]. Utilizing ultrasonic technology for analyzing liquid mixtures offers a powerful means to delve into the nature of molecular interactions, providing valuable insights into their structural dynamics and functional properties [4,5]. In the realm of polymer science, investigating the molecular interactions between polymers and solvents, as well as among different polymer species, is of paramount importance [6,7]. Such research not only enhances our comprehension of polymer production processes but also informs the design and optimization of polymer-based products across a wide spectrum of applications. This paper focuses on the synthesis and properties of PMMA, a synthetic polymer derived from methyl methacrylate monomer units. PMMA, also known as acrylic or acrylic glass, exhibits unique characteristics owing to its molecular structure and polymer chain packing. Its rigid and transparent nature makes it an ideal substitute for glass in various applications, ranging from shatterproof windows to illuminated signs and aircraft canopies. The production process of PMMA involves several chemical transformations, starting from propylene, a component derived from crude oil. Through a series of reactions, propylene is converted into methyl methacrylate, which is then polymerized to form solid PMMA. This synthetic process underscores the

versatility and adaptability of PMMA as a material with diverse industrial applications. In this context, this paper aims to explore the molecular interactions within PMMA-toluene binary liquid mixtures using ultrasonic analysis. By investigating parameters such as adiabatic compressibility, intermolecular free length, and acoustic impedance, we seek to gain deeper insights into the structural modifications and molecular dynamics underlying these interactions. Such insights not only contribute to our fundamental understanding of liquid mixtures but also have practical implications for optimizing polymer processing and enhancing product performance. Through this study, we aim to contribute to the ongoing discourse surrounding the molecular interactions in polymer systems, shedding light on the intricate interplay between polymers and solvents and paving the way for advancements in polymer science and technology.

2.Materials and Methods-

Methyl methacrylate (MMA) was purified using an activated aluminum column. Polyvinyl alcohol and disodium hydrogen phosphate were sourced from Aldrich and CDH, respectively. Benzoyl peroxide served as the initiator, and toluene (99.9% purity) from Chennai, India, was used as the solvent. PMMA was weighed with high precision and dissolved in toluene at different concentrations (0.2M to 0.6M). The solutions were mixed using a REMI magnetic stirrer at 1000 rpm. Density measurements were performed using a Magnetic Float Densitometer, and relative viscosity was measured with Ostwald's viscometer at 295.15K and 308.15K. Ultrasonic velocities were measured at 298.15K and 308.15K using a Single Crystal Ultrasonic Interferometer at 2 MHz (Model-83S) from Mittal Enterprises. Conductivity was determined using a conductivity meter. The temperature was controlled at 298.15K and 308.15K with a thermostat (±0.1K stability). The ultrasonic interferometer used a double-walled metallic jacket for temperature control, with an embedded quartz crystal generating ultrasonic waves. Reflections were measured by a micrometer, forming standing waves, with distances between peaks and minima used to determine half the wavelength.

These preliminary parameters such as Density (ρ), Viscosity(η), Ultrasonic velocity (u)and specific conductance are calculated by using standard relations [8]

$$\text{Density } (\rho) = (W + w + f \times I)/ (V + w/ dpt) \quad (1)$$

$$\frac{y_s}{y_w} = \frac{\rho_s}{\rho_w} \times \frac{t_s}{t_w} \quad (2)$$

$$\text{Ultrasonic velocity } (u) = f\,\lambda \quad (3)$$

$$\text{Specific Conductance} = \text{Conductance} \times \text{Cell constant} \quad (4)$$

Where weight w used, current I passing in the circuit, ρ_{Pt} density of Pt and V, volume of float, η_w, ρ_w and t_w are the viscosity, density and time of flow of water respectively and η_s, ρ_s and t_s are the viscosity, density and time of flow of unknown experimental solution respectively and ƛ is wavelength and f is frequency.

The various acoustical parameters such as adiabatic compressibility (β_{ad}), Intermolecular free length (L_f), Acoustic impedance (ƶ), Relaxation time (τ), Free Volume (V_f) and Surface tension (S) have been calculated from the measured data using the following standard expressions:

$$\beta ad = 1/u^2\,\rho \quad (5)$$

$$L_f = K_T \beta_{ad}^{1/2} \quad (6)$$

$$Z = u \times \rho \quad (7)$$

$$\tau = 4\eta/3\beta_{ad} \quad (8)$$

$$V_f = [M_{eff}\, u/K\, \eta]^{3/2} \quad (9)$$

ρ, is density of the medium, u is the ultrasonic velocity in the medium, η is viscosity of the medium. K_T is a temperature dependent constant called Jacobson constant equal to (93.875 + 0.345T). M_{eff} is the effective molecular weight.

$M_{eff} = M_iX_i$ in which M_i and X_i are the molecular weight and the mole fraction of the individual constituents respectively. K is a temperature independent, constant which is equal to $4.28x10^9$ for all liquids.

3.Experimental Procedure-

The synthesis of Polymethylmethacrylate (PMMA) involved combining MMA, BPO, PVA, and disodium hydrogen phosphate in a flask, stirring at 80°C for an hour with nitrogen, followed by filtration, purification, and vacuum-drying at 100°C for 24 hours. PMMA was then dissolved in toluene at concentrations ranging from 0.2M to 0.6M. Experiments were conducted at 298.15K and 308.15K using a thermostat. Density was measured with a magnetic float densitometer, viscosity with Ostwald's viscometers, ultrasonic velocity with a Single Crystal Ultrasonic Interferometer, and conductance with an electronic conductivity meter. These measurements provided insights into PMMA's physicochemical properties under different conditions. In solution, PMMA's interaction with toluene involved significant polymer-solvent interactions, leading to swelling and expansion of the polymer matrix. This behavior was influenced by the temperature, as experiments at 298.15K and 308.15K revealed variations in density, viscosity, ultrasonic velocity, and conductance, contributing to a deeper understanding of PMMA's behavior in different environments.

Table 1: At 308.15K, the following experimental measurements of density, viscosity, ultrasonic velocity, surface tension, and conductance were obtained.

Concentration (M)	Density (ρ) (Kgm^{-3})	Viscosity(η) (Nsm^{-2})	Ultrasonic velocity(u) (ms^{-1})	Surface Tension (S) $\times 10^{+4}$ ($Kgms^{-2}$)	Conductance(C) $\times 10^{-6}$ (Siemen)	Specific conductance(k) $\times 10^{-6}(Sm^{-1})$
0.2	855.3771	0.00088	1275.33	2.4543	2.565	2.3167
0.3	857.2714	0.00139	1307.33	2.5529	3.024	2.7312
0.4	860.5893	0.00152	1317.60	2.6050	3.663	3.3084
0.5	864.5659	0.00214	1344.00	2.6837	4.557	4.1158
0.6	866.5337	0.00309	1430.76	2.9544	4.608	4.1619

Table 2: At 298.15K, the following experimental measurements of density, viscosity, ultrasonic velocity, surface tension, and conductance were obtained.

Concentration (M)	Density (ρ) (Kgm^{-3})	Viscosity(η) (Nsm^{-2})	Ult/rasonic velocity(u) (ms^{-1})	Surface Tension (S) $\times 10^{+4}(Kgms^{-2})$	Conductance(C) $\times 10^{-6}$ (Siemen)	Specific conductance(k) $\times 10^{-6}(Sm^{-1})$
0.2	865.0227	0.00074	1269.00	2.4635	0.579	0.5229
0.3	867.4609	0.00126	1300.57	2.5632	1.182	1.0675
0.4	870.9506	0.00134	1313.71	2.6126	1.343	1.2129
0.5	873.7265	0.00199	1338.57	2.6957	1.487	1.3430
0.6	877.3306	0.00263	1426.00	2.9763	1.782	1.6095

Table 3: Theoretical values for various acoustical parameters, including Adiabatic compressibility, Acoustic impedance, Relaxation time, Intermolecular free pathlength and free volume at 308.15K.

Concentration (M)	Adiabatic Compressibility(β_{ad}) $\times 10^{-10}$	Acoustic Impedance(z) $\times 10^{6}$	Relaxation Time(τ) $\times 10^{-12}$	Intermolecular free length (L_f) $\times 10^{-11}$	Free Volume (V_f) x 10^{-2}
0.2	7.1878	1.0908	0.8440	5.3670	3.15123
0.3	6.8251	1.1207	1.2707	5.2298	2.08904
0.4	6.6932	1.1339	1.3639	5.1790	1.94730
0.5	6.4032	1.1619	1.8336	5.0656	1.43068
0.6	5.6374	1.2398	2.3276	4.7530	1.06994

Table 4: Theoretical values for various acoustical parameters, including Adiabatic compressibility, Acoustic impedance, Relaxation time, Intermolecular free pathlength and free volume at 298.15K.

Concentration (M)	Adiabatic Compressibility $\times 10^{-10}$	Acoustic Impedance $\times 10^{6}$	Relaxation Time (τ) $\times 10^{-12}$	Intermolecular Free length (L_f) $\times 10^{-11}$	Free Volume (V_f)x 10^{-2}
0.2	7.1787	1.0977	0.7159	5.2712	3.69158
0.3	6.8152	1.1281	1.1478	5.1360	2.29743
0.4	6.6528	1.1441	1.1907	5.0744	2.21055
0.5	6.3876	1.1695	1.2752	4.9722	1.53282

0.6	5.6052	1.2510	1.4762	4.6578	1.25387

The density, ultrasonic velocity, viscosity, and conductance of polymeric solutions at various temperatures were studied, specifically for PMMA (polymethylmethacrylate) in toluene at 298.15K and 308.15K and 2 MHz frequency (Tables 1-4).

Tables 1 and 2 show that as PMMA concentration in toluene increases, the density of the solution rises due to polymer-solvent interactions. Toluene molecules penetrate the polymer matrix, causing the polymer chains to swell and increase the solution's volume and density. Higher PMMA concentrations lead to more polymer chains absorbing toluene, expanding and increasing density. As temperature rises, PMMA chains spread out and move faster, reducing the solution's density since they occupy more space. These molecular interactions, including relaxation, can be better understood through density measurements at various frequencies.[9]

Figure1. *The graph of density of polymeric solution (PMMA + Toluene) of different concentrations at two different temperatures are 298.15K and 308.15K*

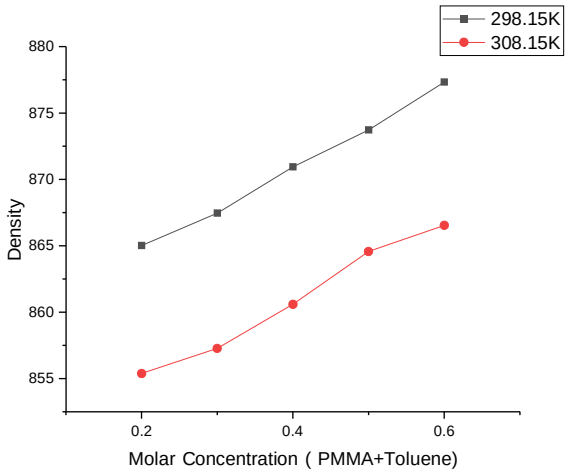
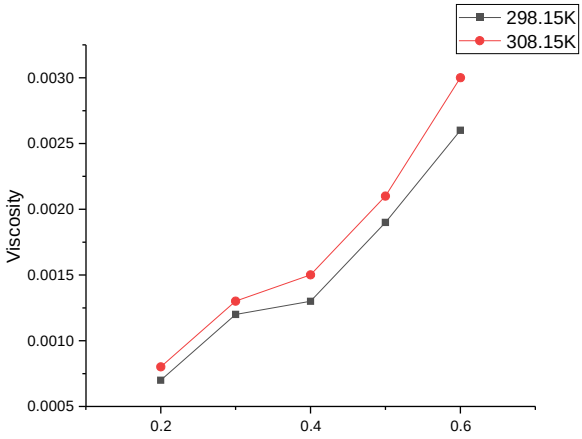


Figure2. *The graph of Viscosity of polymeric solution (PMMA + Toluene) of different concentrations at two different temperatures are 298.15K and 308.15K*



Molar Concentration (Pmma+Toluene)

Figure 3. *The graph of Ultrasonic velocity of polymeric solution (PMMA + Toluene) different concentrations at different temperatures are 298.15K and 308.15K*

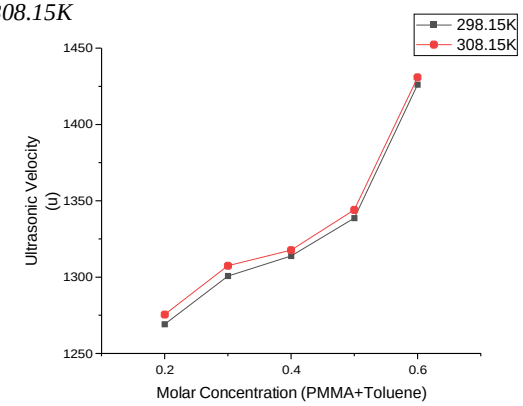


Figure 4*. The graph of Surface Tension of polymeric solution (PMMA + Toluene) different concentrations at different temperatures are 298.15K and 308.15K*

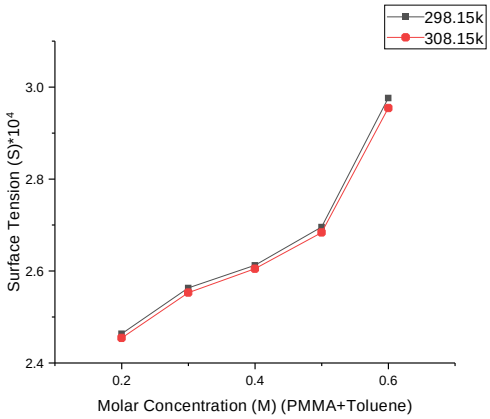
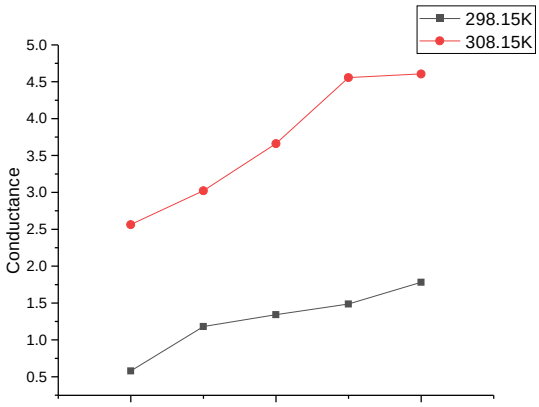


Figure 5. *The graph of Conductance of polymeric solution (PMMA + Toluene) different concentrations at different temperatures are 298.15K and 308.15K*



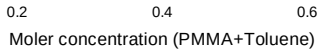


Figure 6. *The graph of Specific Conductance of polymeric solution (PMMA+ Toluene) different concentrations at different temperatures are 298.15K and 308.15K*

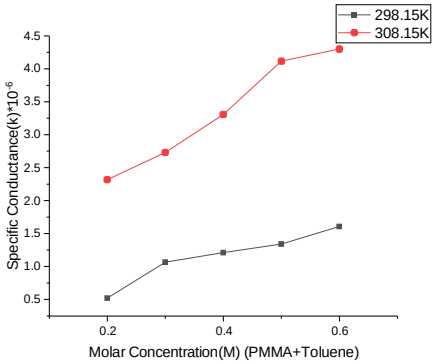


Figure 7. *The graph of Adiabatic Compressibility of polymeric solution (PMMA + Toluene) different concentrations at different temperatures are 298.15K and 308.15K*

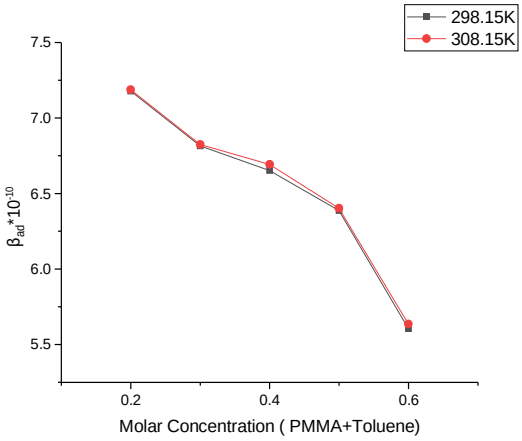


Figure 8. *The graph of Acoustic Impedance of polymeric solution (PMMA + Toluene) different concentrations at different temperatures are 298.15K and 308.15K*

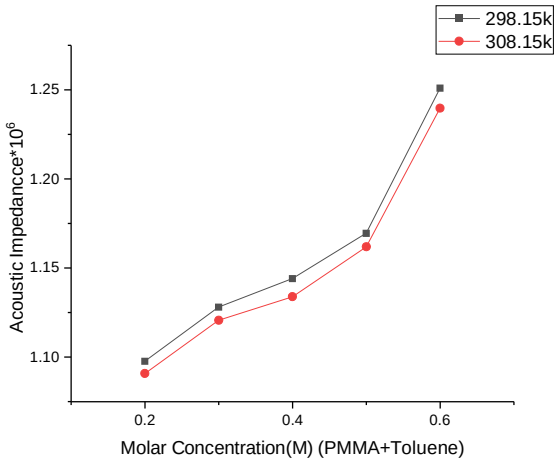


Figure 9- *The graph of Free Volume (Vf)of polymeric solution (PMMA + Toluene) different concentrations at different temperatures are 298.15K and 308.15K*

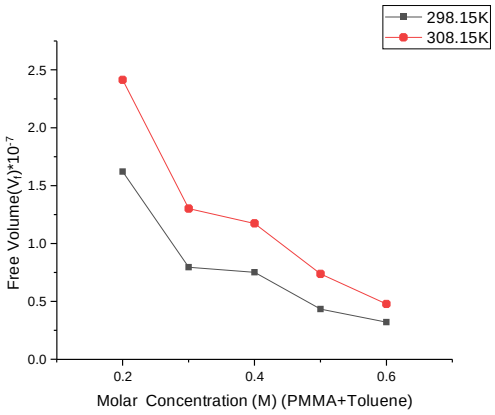


Figure 10. *The graph of Relaxation Time of polymeric solution (PMMA + Toluene) different concentrations at different temperatures are 298.15K and 308.15K*

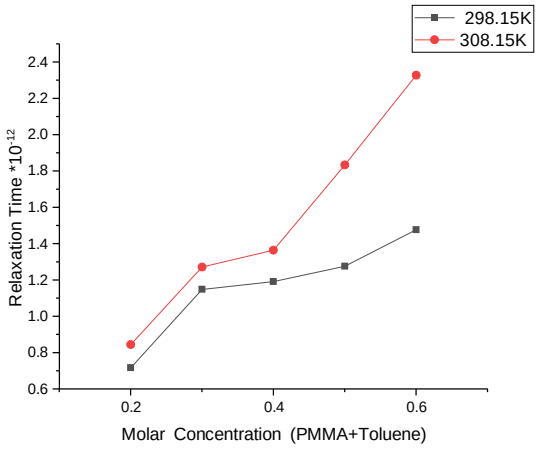
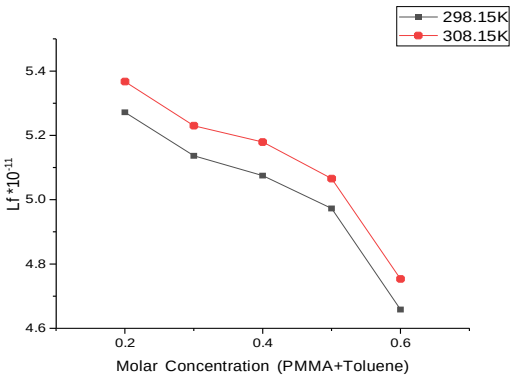


Figure 11- The graph of Intermolecular Free Path length (Lf)of polymeric solution (PMMA+ Toluene) different concentrations at different temperatures are 298.15K and 308.15K



4.Result and Discussion

Ultrasonic velocity measures the speed of sound through a solution, indicating changes in elastic properties and molecular interactions. As polymer concentration increases, a more rigid molecular structure develops due to bonds between larger polymer molecules. Tables 1 and 2 show that ultrasonic velocity increases with both temperature and concentration, suggesting enhanced associations between solute and solvent molecules【9-11】.

Viscosity refers to the resistance of a fluid to flow and is influenced by polymer chain length, concentration, and solvent interactions in polymeric solutions. As the concentration of PMMA in toluene increases, the hydrodynamic interactions between PMMA chains and solvent molecules also increase, leading to higher viscosity. This is due to the attraction between PMMA segments and toluene molecules, causing greater resistance to flow. Additionally, higher concentrations increase the likelihood of chain overlap and collisions, further increasing viscosity by hindering polymer movement [12]. Measuring viscosity in dilute macromolecular solutions is crucial for determining molecular weights and evaluating key parameters to understand polymer conformations.

According to Figure 4, surface tension of the PMMA solution increases with concentration as more polymer molecules occupy the surface, disrupting the interaction between toluene and air. PMMA chains adopt extended conformations near the surface, increasing the surface area and surface tension. Conversely, increasing the temperature decreases surface tension. Higher thermal energy enhances molecular motion, weakening cohesive forces and reducing surface tension. Additionally, increased solubility of PMMA in toluene at higher temperatures creates a more homogeneous mixture, further lowering surface tension.

Conductance measures a solution's ability to conduct electricity, influenced by the presence of ions or charged species. As shown in Tables 1 & 2 and Figures 5 & 6, specific conductance increases with polymer concentration. This is because higher concentrations mean more polymer chains, enhancing interactions with charged groups and increasing ion mobility within the polymer network. Additionally, specific conductance rises with temperature. Elevated temperatures improve interactions between toluene molecules and PMMA chains, facilitating better solvation and increasing the mobility of charged species.

Adiabatic compressibility (β) measures volume change under pressure in an adiabatic process. In polymer solutions, it reflects the elasticity and flexibility of polymer chains and their interactions with solvent molecules. Tables 3 & 4 and Figure 7 show that as PMMA concentration in toluene increases, stronger interactions between polymer and solvent molecules reduce free volume, decreasing compressibility. At higher concentrations, PMMA and toluene may form complexes, further reducing compressibility. Conversely, rising temperature increases adiabatic compressibility due to higher kinetic energy, which allows molecules to move apart more easily under pressure.

In polymer solutions, acoustic impedance (Z) indicates density, compressibility, and polymer-solvent interactions. Increasing PMMA concentration in toluene raises the mixture's density and acoustic impedance due to stronger polymer-solvent interactions. PMMA exhibits viscoelastic behavior, affecting acoustic impedance. Additionally, from table 3&4 acoustic impedance decreases with rising temperature as both density and sound speed in PMMA drop, assuming effective solute-solvent interactions.[14]

Relaxation time (τ) measures how long a system takes to return to equilibrium after a disturbance. In polymer solutions, it indicates polymer chain relaxation and reorganization. Longer relaxation times mean slower dynamics and more entanglement. Increasing PMMA concentration in toluene raises viscosity, slowing polymer and solvent movement, and lengthening relaxation time. Higher viscosity and PMMA aggregation create obstacles, delaying equilibrium. As shown in Tables 3 & 4 and Fig. 10, relaxation time increases with temperature, influenced by changes in density, viscosity, and ultrasonic velocity, highlighting significant system interactions. [15,16]

Free volume is the unoccupied space within a polymer matrix, influenced by polymer-solvent interactions, chain flexibility, and solvent penetration. It affects physical properties like density, viscosity, and diffusion coefficients. Higher free volume indicates greater molecular mobility and solvent accessibility. In PMMA-toluene solutions, increasing PMMA concentration causes polymer chains to swell, reducing free volume due to closer chain packing and increased entanglement. As shown in Tables 3 & 4 and Fig. 10, higher concentrations result in decreased free volume. Temperature increases lead to greater thermal energy, causing polymer chains to vibrate and expand, increasing free volume. Higher temperatures also enhance chain mobility, creating temporary voids and further increasing free volume.

Free length measures the average distance between entanglement points in a polymer chain, reflecting chain mobility and flexibility. Longer free length indicates greater mobility, while shorter free length suggests more entanglement. From tables 3 & 4 and fig. 11: Higher PMMA concentrations enhance interactions between polymer chains (e.g., van der Waals forces, hydrogen bonding), leading to a compact arrangement and shorter free lengths. This reduced segmental mobility confines polymer chains, limiting free length. Fig. 11 shows that in PMMA-toluene blends, free path length increases with temperature. Higher temperatures increase thermal energy, free volume, and solvent interactions, creating more space in the polymer matrix and allowing segments to travel further, thus increasing free path length.

5. Conclusions-

The density, viscosity, and other pertinent parameters, together with the ultrasonic velocity, were computed. The binary solution of Polymethylmethacrylate in toluene exhibits non-linear fluctuation in ultrasonic velocity (u), density (ρ), and viscosity (V), as well as other relevant thermodynamic and acoustical parameters such as a, L_f, Z, etc. The systems examined at provide confirmation of weak dispersive type intermolecular interactions and Specific conductance is increases with rise the concentration as well as temperature due to rise in temperature and concentration of conductance. Acoustic parameter experimental determinations are all highly associated with one. Heteromolecular contact in the liquid mixture is caused by hydrogen bond forms, which account for the molecular interaction that was observed. This gives important details regarding the inter and intramolecular interactions of the mixture as they exist in the liquid system. They are then helpful for the production and use of polymers in pharmaceuticals and industry.

Note:

The authors thankful to UPCST for providing financial support for the award of project of fellowship and making the research easier.

Acknowledgment:

The authors thank The Head of the Department, Lucknow University for providing the research facility in the Department.

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