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Solvent-Induced Changes in the Acoustic Parameters and Intermolecular Forces of Polyethene Glycol (PEG) Solutions

Smita S. Kharkale-Bhuyar[1]*

[1] Department of Chemistry, Shri Lemdeo Patil Mahavidyalaya, Mandhal, Kuhi, Nagpur, Maharashtra, India,

Corresponding Author: smitakharkale2013@gmail.com

ABSTRACT: The present investigation focuses on the influence of solvent polarity on the physicochemical and acoustic behaviour of polyethene glycol (PEG-400) solutions in acetone and dimethylformamide (DMF) at 300 K. Ultrasonic velocity, density and viscosity of PEG-400 solutions having different concentrations were experimentally determined using a single-frequency ultrasonic interferometer operating at 2 MHz, a calibrated specific gravity bottle and an Ostwald viscometer, respectively. The experimentally measured values were utilised to evaluate various acoustical parameters, namely adiabatic compressibility, intermolecular free length, acoustic impedance, relative association, ultrasonic attenuation coefficient and relaxation time. The variation of these parameters with polymer concentration was analysed to understand the nature of solute-solvent interactions.

The results reveal that ultrasonic velocity, density, viscosity and acoustic impedance increase progressively with increasing PEG concentration in both solvents, whereas adiabatic compressibility and intermolecular free length decrease systematically. These observations indicate enhanced molecular packing and stronger intermolecular interactions resulting from hydrogen bonding, dipole-dipole interactions and polymer-solvent association. The magnitude of these changes is considerably greater in dimethylformamide than in acetone because of the higher polarity and superior solvating ability of DMF towards PEG molecules. Linear regression analysis further demonstrates excellent agreement between the experimental observations and concentration-dependent behaviour of all measured parameters. The findings establish ultrasonic investigation as a reliable and sensitive technique for evaluating molecular interactions and structural organisation in polymer solutions and provide useful information for designing polymer-based systems employed in pharmaceutical, chemical and industrial applications.

INTRODUCTION

Ultrasonic studies in polymeric solutions have drawn the attention of many researchers in recent years [1–4]. The extensive use of polymeric materials in technology has necessitated investigations of the molecular interactions of polymers and solvents [5–9]. The ultrasonic technique is a powerful and effective tool for investigating polymer solutions and the behaviour of polymer chains in an ultrasonic field. Polyethylene Glycol- 400 (PEG) is a semicrystalline polymer that has a simple chemical structure [- H-(O-CH2-CH2)n-OH]-]n. It is frequently used in research alongside ferroelectric materials. For instance, PEG is often used as a solvent or a polymer template in sol-gel chemistry to synthesise crack-free inorganic ferroelectric thin films (such as PZT or PMN-PT) [10-11]. The Polyethene Glycol- 400

(PFG). It is fully water-soluble and miscible with a vast range of organic solvents (like ethanol, water, or acetonitrile). When researchers perform ultrasonic or acoustic studies on PEG-400, they are usually investigating hydrogen bonding networks,

interstitial packing, and structural relaxation in liquid mixtures, rather than ferroelectric domains. In our experimental investigation, we used the ultrasonic technique to find the acoustic parameters such as adiabatic compressibility (βs), intermolecular free path length (Lf), acoustic impedance (Z), relative association (RA), ultrasonic attenuation, and relaxation time to correlate with the physical properties.

Polyethene glycol (PEG) is one of the most widely investigated water-soluble polymers because of its remarkable physicochemical properties, excellent chemical stability, non-toxicity and compatibility with a wide variety of organic solvents. Depending upon its molecular weight, PEG finds extensive applications in pharmaceutical formulations, biomedical engineering, cosmetics, polymer processing, drug delivery systems, lubricants, adhesives and separation science. Among various grades, polyethene glycol-400 (PEG-400) is particularly attractive because it exists as a viscous liquid at room temperature and exhibits excellent miscibility with both polar and moderately polar organic solvents.

The physical properties of polymer solutions are governed primarily by the nature and strength of intermolecular interactions existing between polymer chains and solvent molecules. These interactions influence molecular packing, free volume, compressibility, transport properties and solution structure. Consequently, investigations of polymer-solvent interactions are of considerable importance in understanding solution behaviour as well as in designing industrial formulations with desirable physicochemical characteristics.

Ultrasonic techniques have emerged as valuable, non-destructive and highly sensitive methods for probing molecular interactions in liquid mixtures. The propagation of ultrasonic waves through a liquid medium depends upon its elastic and structural properties, which are directly influenced by intermolecular forces. Measurements of ultrasonic velocity, density and viscosity enable the calculation of several acoustical parameters such as adiabatic compressibility, intermolecular free length, acoustic impedance, relaxation time, relative association and ultrasonic attenuation. These derived parameters provide valuable insight into molecular association, hydrogen bonding, dipole-dipole interactions and structural rearrangement occurring in complex liquid systems.

Several investigators have successfully employed ultrasonic techniques to study binary and ternary liquid mixtures, electrolyte solutions and polymer systems. Earlier studies have demonstrated that variations in ultrasonic velocity and related acoustic parameters are strongly dependent upon the polarity of the solvent, molecular size, hydrogen-bonding capability and concentration of dissolved species. Strong polymer-solvent interactions generally result in an increase in ultrasonic velocity, viscosity and acoustic impedance accompanied by a corresponding decrease in compressibility and intermolecular free length, indicating closer molecular packing and enhanced structural organisation.

Acetone and dimethylformamide (DMF) represent two solvents possessing distinctly different physicochemical characteristics. Acetone is a moderately polar aprotic solvent with relatively weak intermolecular association, whereas DMF is a highly polar aprotic solvent possessing a large dielectric constant and excellent solvating ability towards polar polymers. Owing to these differences, PEG-400 is expected to exhibit different interaction mechanisms in these two solvents. A comparative investigation therefore provides useful information regarding the influence of solvent polarity on polymer behaviour.

In the present work, ultrasonic velocity, density and viscosity of PEG-400 solutions in acetone and dimethylformamide were measured at 300 K over a range of concentrations. From these experimental data, several acoustical parameters were calculated to evaluate molecular interactions within the solution. The concentration dependence of these parameters has been analysed through regression analysis to establish correlations between acoustic behaviour and polymer-solvent interactions. The study contributes to a better understanding of the structural organisation of PEG solutions and demonstrates the usefulness of ultrasonic methods for investigating polymer systems employed in pharmaceutical, industrial and chemical processes.

MATERIALS AND METHODS

Analytical reagent (AR) grade Polyethene Glycol-400 (PEG-400) was utilised for this investigation. The polymer was dissolved in varying mass fractions (c = 0.00 to 0.50 %) of AR-grade acetone and dimethylformamide (DMF). To ensure uniform homogeneity and complete dissolution, the liquid mixtures were magnetically stirred prior to recording measurements.

EXPERIMENTAL

Materials

Polyethene glycol-400 (PEG-400) of analytical reagent (AR) grade was procured from a reputed chemical supplier and used

without further purification. Acetone and dimethylformamide (DMF) of analytical reagent grade having purity greater than 99.5% were obtained from Merck India Pvt. Ltd. Double-distilled water was used for cleaning all glassware employed during the experimental work. The physical properties of the solvents agreed well with the values reported in the literature, confirming their suitability for ultrasonic measurements.

Preparation of Solutions

PEG-400 solutions were prepared separately in acetone and dimethylformamide at concentrations of 0.00, 0.10, 0.20, 0.30, 0.40 and 0.50% (w/v). The required quantity of PEG-400 was accurately weighed using a digital analytical balance with an accuracy of ±0.1 mg and dissolved in the respective solvent with continuous magnetic stirring until a homogeneous solution was obtained. Fresh solutions were prepared immediately before each experimental measurement to avoid contamination or evaporation losses.

Measurement of Ultrasonic Velocity

The ultrasonic velocity (U) of the prepared solutions was measured using a single-frequency crystal-controlled ultrasonic interferometer (Mittal Enterprises, Model F-81) operating at a fixed frequency of 2 MHz. The instrument was calibrated with distilled water before measurements. The temperature of the sample was maintained at 300 ± 0.1 K using a thermostatically controlled circulating water bath throughout the experiment. The uncertainty in ultrasonic velocity measurements was estimated to be within ±0.5 m s^{-1}.

Density Measurement

The densities (ρ) of PEG-400 solutions were determined using a calibrated specific gravity bottle (pyknometer) having a capacity of 25 ml. The pyknometer was thoroughly cleaned, dried

and weighed before and after filling with the sample solution. All measurements were carried out at 300 ± 0.1 K. Density values were calculated using the standard gravimetric method, and the estimated uncertainty was ±0.5 kg m^{-3}.

Viscosity Measurement

The viscosity (η) of each solution was measured using a calibrated Ostwald capillary viscometer immersed in a constant-temperature water bath maintained at 300 ± 0.1 K. The flow time was measured using a digital stopwatch with an accuracy of ±0.01 s. Each measurement was repeated at least three times, and the average value was considered for calculation. The uncertainty in viscosity measurement was estimated to be less than ±1%.

Evaluation of Acoustic Parameters

The experimentally determined values of ultrasonic velocity, density and viscosity were employed to calculate various acoustical parameters using standard thermodynamic relations.

Adiabatic Compressibility (β)

$$[\beta = \frac{1}{\rho \upsilon 2}]$$

where

β = Adiabatic compressibility (m^2 N^{-1})

U = Ultrasonic velocity (m s^{-1})

ρ = Density (kg m^{-3})

Intermolecular Free Length (Lf)

$$[Lf=K\beta 1/2]$$

Where K_t is Jacobson's temperature-dependent constant.

Acoustic Impedance (Z)

$$[Z=\rho \upsilon\ ]$$

Where, Z represents the resistance offered by the medium to the propagation of ultrasonic waves.

Relative Association (RA)

$$[RA=\left(\frac{\rho}{\rho 0}\right)\left(\frac{\upsilon 0}{\upsilon}\right)]$$

Where, ρ_0 and U_0 denote the density and ultrasonic velocity of the pure solvent, respectively.

Relaxation Time (τ)

$$[\tau = \frac{4\eta}{3\rho\upsilon 2}]$$

Relaxation time provides valuable information regarding the structural rearrangement occurring within the liquid medium during ultrasonic wave propagation.

Ultrasonic Attenuation Coefficient ($\alpha/_{f2}$)

The ultrasonic attenuation per unit frequency square was calculated using

$$[\alpha/_{f2} = \frac{8\pi 2\eta}{\rho\upsilon 3}]$$

Where

α = Ultrasonic attenuation coefficient

f = Frequency of ultrasonic wave

η = Viscosity

ρ = Density

U = Ultrasonic velocity

Statistical Analysis

The concentration dependence of the experimentally determined and derived acoustic parameters was analysed by least-squares linear regression. The coefficient of determination (R^2) was calculated to assess the degree of linearity between PEG concentration and each acoustical parameter. Regression equations were obtained separately for PEG-400 solutions in acetone and dimethylformamide to compare the influence of solvent polarity on molecular interactions.

All experimental measurements were carried out in triplicate, and the average values were used for subsequent calculations. The maximum experimental uncertainty in the calculated acoustic parameters was estimated to be within ±1.5%, indicating satisfactory precision and reproducibility of the measurements.

RESULTS AND DISCUSSION

The experimentally measured values of density (ρ), viscosity (η), and ultrasonic velocity (U) for PEG-400 solutions in acetone and dimethylformamide (DMF) at 300 K are presented in Tables 1 and 2. These primary experimental parameters provide useful information about the molecular interactions and structural changes occurring in the polymer–solvent systems at different concentrations of PEG-400.

Density (ρ) reflects the mass and molecular packing of the solution, while viscosity (η) represents the resistance offered by the solution to molecular flow. Changes in these properties with increasing PEG-400 concentration are associated with changes in molecular arrangement, polymer chain mobility, and polymer–solvent interactions. Ultrasonic velocity (U) is highly sensitive to the intermolecular forces and compressibility of the medium; therefore, its concentration dependence provides valuable information about the nature of interactions between PEG-400 and the solvent molecules.

Using the experimentally determined values of density and ultrasonic velocity, various acoustical parameters were calculated by applying standard thermodynamic relationships. These parameters include adiabatic compressibility (β), intermolecular free length (Lf), acoustic impedance (Z), relative association (RA), relaxation time (τ), and ultrasonic attenuation coefficient (α/f^2). Adiabatic compressibility gives information about the ease of compression of the solution, whereas intermolecular

free length provides an indication of the average distance between interacting molecules. Acoustic impedance represents the resistance offered by the medium to ultrasonic-wave propagation and depends on both density and ultrasonic velocity.

The relative association (RA) provides information regarding the degree of molecular association between PEG-400 and solvent molecules. Relaxation time (τ) is related to the molecular rearrangement and energy dissipation processes occurring during ultrasonic propagation, while the ultrasonic attenuation coefficient (α/f^2) gives information about the loss of ultrasonic energy within the solution.

The variation of these acoustical parameters with PEG-400 concentration helps to understand the strength and nature of polymer–solvent interactions. A comparative study of PEG-400 solutions in acetone and DMF is particularly useful because the two solvents provide different molecular environments for the polymer. The combined analysis of density, viscosity, ultrasonic velocity, and derived acoustical parameters therefore provides insight into molecular association, packing, compressibility, and relaxation behaviour of PEG-400 in the investigated solvent systems at 300 K.

Table 1: Variation of density, viscosity, and ultrasonic velocity of PEG-400 with different concentrations of acetone at 300K

Conc. (%)	Density kg/m	Viscosity Nsm^{-2}	Ultrasonic velocity m/s
.00	873.00	0.3835	1265.00
0.10	873.56	0.3865	1265.90
0.20	874.10	0.3972	1266.50
0.30	874.68	0.4047	1268.00
0.40	877.76	0.4036	1269.00
0.50	890.36	0.4193	1270.80

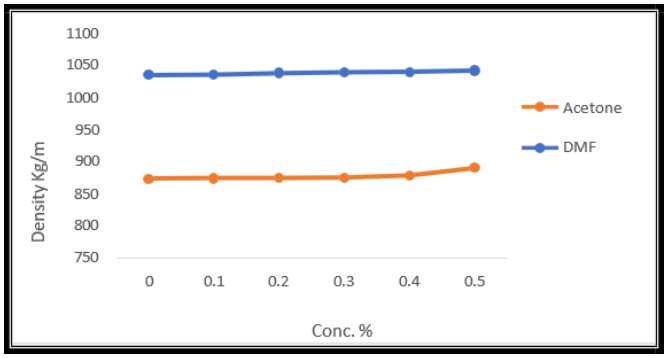
Table 2: Variation of density, viscosity, and ultrasonic velocity of PEG-400 with different concentrations of dimethylformamide [DMF]at 300 K

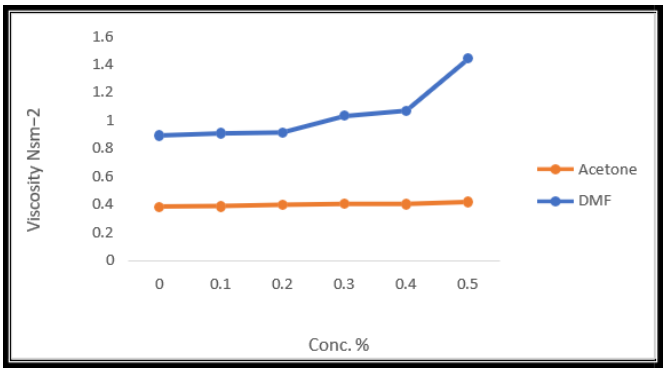
Conc. (%)	Density kg/m	Viscosity Nsm^{-2}	Ultrasonic velocity m/s
0.00	1034.20	0.8921	1554.00
0.10	1034.92	0.9084	1574.00
0.20	1037.04	0.9146	1578.00
0.30	1038.16	1.0320	1582.50
0.40	1039.04	1.0701	1585.00
0.50	1040.88	1.4442	1586.60

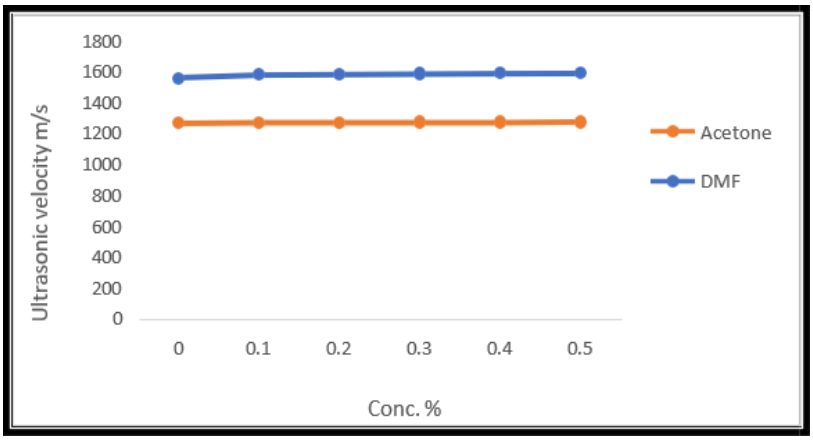
Table 3: The least-squares equations and regression coefficients for PEG-400 solution in acetone and DMF.

Parameters	Regression coefficients (R^2)	
	Acetone	DMF
ρ kg/m^3	14.28c + 782.00 (0.929)	13.394c + 934.024 (0.992)

$\eta\ 10^{-3}$ Nsm^{-2}	$5.257 \times 10^{-5}c + 3.611 \times 10^{-4}$ (0.961)	$9.685 \times 10^{-4}c + 7.7219 \times 10^{-4}$ (0.949)
V m/s	11.37c + 1164.69 (0.987)	57.285c + 1462.36 (0.890)
$\beta\ 10^{-10}$ m^2/N	−0.35097c + 9.42638 (−0.968)	−0.45354c + 5.0065 (−0.916)
$Z\ 10^{6}$ kgm^{2}s^{-1}	0.02309c + 0.91106 (0.971)	0.07346c + 1.36582 (0.940)
$Lf\ 10^{-11}$ m	−0.11817c + 6.31044 (−0.968)	−0.21003c + 4.59886 (−0.917)
RA	$2.2614 \times 10^{-5}c + 2.3064 \times 10^{-14}$ (0.957)	$2.207 \times 10^{-14}c + 2.095 \times 10^{-14}$ (0.931)
$\tau\ 10^{-13}$ s	$4.913 \times 10^{-14}c + 4.534 \times 10^{-13}$ (0.962)	$5.1685 \times 10^{-13} + 5.7522 \times 10^{-13}$ (0.938)
$\alpha/f^2\ 10^{-14}$ s^{2}m^{-1}	0.96994c + 8.95398 (0.962)	$2.207 \times 10^{-14}c + 2.0959 \times 10^{-14}$ (0.931)
ρ kg/m^3	14.28c + 782.00 (0.929)	13.394c + 934.024 (0.992)
$\eta\ 10^{-3}$ Nsm^{-2}	$5.257 \times 10^{-5}c + 3.611 \times 10^{-4}$ (0.961)	$9.685 \times 10^{-4}c + 7.7219 \times 10^{-4}$ (0.949)







Effect of PEG Concentration on Density

The density of PEG-400 solutions increased continuously with increasing polymer concentration in both acetone and DMF. In acetone, the density increased from 873.00 to

890.36 kg m^{-3}, whereas in DMF it increased from 1034.20 to 1040.88 kg m^{-3}. The increase in density indicates progressive incorporation of PEG molecules into the solvent matrix, resulting in closer molecular packing and reduced free volume.

The observed increase is attributed to enhanced intermolecular attraction between PEG chains and solvent molecules through dipole-dipole interactions and hydrogen bonding involving the ether oxygen atoms of PEG. Since DMF possesses a higher dielectric constant and stronger polarity than acetone, the interaction between PEG and DMF molecules is comparatively stronger, leading to a denser molecular arrangement. The linear regression equations obtained for density exhibit excellent correlation coefficients, indicating that density varies almost linearly with PEG concentration.

Variation of Viscosity

Viscosity is highly sensitive to molecular association and structural organisation in liquid mixtures. Experimental observations reveal a gradual increase in viscosity with increasing PEG concentration for both solvent systems.

The increase is considerably more pronounced in DMF than in acetone. The higher viscosity values obtained in DMF indicate stronger polymer-solvent interaction and greater resistance to molecular flow. The long polymer chains of PEG become increasingly entangled at higher concentrations, thereby restricting molecular mobility and increasing internal friction. The increase in viscosity also supports the existence of extensive intermolecular association between PEG molecules and solvent molecules, confirming the formation of a more structured liquid system.

Ultrasonic Velocity

Ultrasonic velocity increases progressively with increasing PEG concentration in both solvents. The velocity increases from 1265.0 to 1270.8 m s^{-1} in acetone and from 1554.0 to 1586.6 m s^{-1} in DMF.

An increase in ultrasonic velocity generally indicates stronger intermolecular attraction and improved transmission of ultrasonic waves through the liquid medium. As PEG concentration increases, solvent molecules become more closely associated with polymer chains, reducing intermolecular voids and enhancing the elasticity of the solution. The comparatively higher ultrasonic velocity observed in DMF confirms stronger molecular association because of its superior solvating ability and greater polarity.

ADIABATIC COMPRESSIBILITY

Adiabatic compressibility decreases steadily with increasing PEG concentration for both solvent systems

A decrease in compressibility signifies that the solution becomes less compressible owing to increased cohesive forces between molecules. Strong polymer-solvent interaction reduces the available free space within the liquid, making compression under ultrasonic waves more difficult. The lower compressibility values obtained for DMF solutions further demonstrate stronger

molecular interaction compared to acetone.

Intermolecular Free Length

The intermolecular free length follows the same trend as adiabatic compressibility and decreases continuously with increasing PEG concentration.

According to Jacobson's theory, free length represents the average distance separating neighbouring molecules. The decrease in free length indicates that PEG molecules occupy intermolecular spaces effectively, resulting in closer molecular packing. This behaviour confirms increasing attractive forces between polymer chains and solvent molecules.

Acoustic Impedance

Acoustic impedance increases almost linearly with PEG concentration in both solvent systems. Since acoustic impedance is directly proportional to density and ultrasonic velocity, its increase reflects improved resistance of the medium towards ultrasonic wave propagation.

Higher impedance values indicate stronger cohesion and greater compactness of the liquid structure. The larger impedance observed for PEG-DMF solutions clearly demonstrates stronger intermolecular interaction than that observed in PEG-acetone solutions.

Relative Association

Relative association provides valuable information regarding the extent of molecular association and solvent structure around polymer molecules.

The observed increase in relative association with concentration suggests progressive association between PEG molecules and surrounding solvent molecules. The ether oxygen atoms present in PEG interact strongly with the carbonyl oxygen of DMF through dipolar interactions, producing greater structural organisation compared with acetone.

Relaxation Time

Relaxation time increases gradually with increasing polymer concentration. An increase in relaxation time indicates that molecular rearrangement following ultrasonic perturbation becomes progressively slower owing to increased structural rigidity of the liquid.

The higher relaxation times observed for DMF indicate stronger molecular association and slower configurational changes compared with acetone.

Ultrasonic Attenuation

Ultrasonic attenuation also increases with PEG concentration. The increase in attenuation arises from enhanced viscous losses and energy dissipation occurring during ultrasonic wave propagation through increasingly structured polymer solutions.

The larger attenuation values observed in DMF indicate more effective absorption of ultrasonic energy owing to stronger molecular interaction.

Regression Analysis

Least-squares regression analysis was employed to establish mathematical relationships between PEG concentration and various physicochemical parameters.

The regression coefficients (R^2) obtained for density, viscosity, ultrasonic velocity and derived acoustic parameters are close to unity, indicating excellent agreement between experimental observations and the fitted linear equations. These results confirm that concentration is the dominant factor controlling the physicochemical behaviour of PEG solutions under the present experimental conditions.

Molecular Interaction Mechanism

The collective behaviour of all measured acoustic parameters clearly demonstrates significant polymer-solvent interaction in both solvent systems. Increasing density, viscosity, ultrasonic velocity, acoustic impedance, relative association and relaxation time, together with decreasing compressibility and intermolecular free length, indicate strengthening cohesive forces within the

solution.

The interaction mechanism primarily involves dipole-dipole attraction between PEG ether oxygen atoms and solvent molecules, accompanied by hydrogen-bond-assisted molecular association. Owing to its higher dielectric constant and stronger electron-donating ability, DMF exhibits greater affinity towards PEG molecules than acetone.

Consequently, PEG-DMF solutions possess a more compact molecular structure, lower free volume, higher resistance to compression and greater ultrasonic wave propagation efficiency than PEG-acetone solutions. The results clearly establish that solvent polarity plays a decisive role in determining the extent of molecular association and structural organisation in PEG solutions.

CONCLUSION

The present ultrasonic investigation demonstrates that the physicochemical properties of PEG-

400 solutions are significantly influenced by the nature of the solvent and polymer concentration. The experimentally determined density, viscosity, and ultrasonic velocity increased progressively with increasing PEG-400 concentration in both acetone and dimethylformamide (DMF), whereas adiabatic compressibility and intermolecular free length exhibited a corresponding decrease. These observations clearly indicate enhanced molecular association, closer packing of molecules, and a reduction in the free volume of the solution.

The calculated acoustic impedance, relative association, relaxation time, and ultrasonic attenuation coefficient increased with polymer concentration, confirming the existence of strong polymer-solvent interactions. Comparative analysis revealed that PEG-400 exhibits stronger intermolecular interactions in DMF than in acetone because of the higher polarity, dielectric constant, and superior solvating ability of DMF. The regression analysis further demonstrated excellent linear relationships between concentration and the measured acoustic parameters, indicating the reliability and reproducibility of the experimental results.

The present study confirms that ultrasonic techniques provide an effective, rapid, and non-destructive approach for investigating molecular interactions in polymer solutions. The findings contribute to a better understanding of polymer-solvent interactions and may be useful in the design and optimisation of polymeric systems employed in pharmaceutical formulations, chemical processing, coatings, lubricants, and advanced materials.

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CONFLICT OF INTEREST

The author declares that there is no conflict of interest regarding the publication of this manuscript.

AUTHOR CONTRIBUTIONS

Smita S. Kharkale-Bhuyar conceived the research work, designed the experimental methodology, prepared the polymer solutions, performed all ultrasonic, density, and viscosity measurements, analysed the experimental data, calculated the acoustic parameters, interpreted the results, prepared the figures and tables, wrote the manuscript, and approved the final version for publication

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