

*Original Article*

# Thermoacoustical parameters of dextran polymer in sodium hydroxide solutions

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**Abstract**

The physicochemical properties provide detailed information about the effect of solvent on structural changes in solutions. These properties are explained in terms of ion-solute or solute-solvent interactions. The density and viscosity of the polymer solution were measured at 303 K, 308 K, 313 K, 318 K and 323 K for 1% concentration of dextran. Ultrasound speed (U) in the solution was also measured at the four frequencies 1MHz, 5MHz, 9MHz, and 12MHz. Acoustic parameters such as adiabatic compressibility ( $\beta$ ), acoustic impedance (Z), relaxation time ( $\tau$ ), intermolecular free length ( $L_f$ ), and Gibbs energy ( $\Delta G$ ), were also computed for these 1% dextran polymer solutions. These data are intended for analyses of the solvent-solvent, solvent-ion, and ion-ion interactions.

**Keywords:** dextran, density, viscosity, ultrasound speed, acoustic parameters

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**1. Introduction**

In recent years the physicochemical properties of polymer solutions have been widely studied. These properties provide detailed information about the structural changes in polymer solutions. The solute-solvent and solvent-solvent interactions can also be studied using these properties (Fort & Moore 1966; Panda & Mahapatra, 2016;). This type of study helps us understand the structure formation and degradation properties of the solute.

Studies of molecular interactions and the variations in these interactions due to structural changes have been carried out by various experimental techniques such as infrared spectroscopy, nuclear magnetic resonance, Raman spectroscopy, and dielectric property measurements. A complete understanding of the nature of intermolecular and intermolecular interactions may not be revealed by any single method. Ultrasonic methods have the added advantage of being less costly, despite efficiency comparable to the other methods, and are nondestructive techniques. In this work, a new method for the determination of the thermodynamical properties of fluids, based on experimental speed-of-sound

measurements using an ultrasonic interferometer, is described.

The ultrasonic methods are important for analysis and design in many chemistry and industrial processes, such as in the pharmaceutical sector analysing blood for volume expander, decrease of viscosity etc.

Ultrasound is applied in various fields like biochemistry, electrolytic polymerization etc. Ultrasonic waves provide an important probe for the study of structure and properties of matter. The above mentioned interactions can be studied by ultrasonic, density and viscosity measurements (Rajagopal & Chenthilnath, 2010). The ultrasonic studies give an idea on the ion-association and complex formation processes. Thus ultrasonic parameters are extensively used to study the molecular interactions in pure liquids, liquid mixtures, and electrolyte solutions. The effects of temperature and frequency inform about adiabatic compressibility ( $\beta$ ), acoustic impedance (Z), relaxation time ( $\tau$ ), intermolecular free length ( $L_f$ ) and Gibbs energy ( $\Delta G$ ) in solute and solvent, giving important information about the intermolecular forces that determine the properties of solution. Such parameters elucidate the interactions like hydrogen bonding, ion-ion, ion-solute, and solvent-solvent interactions in solutions (Kawale, Tumberphale, Kalamse, & Kalamse, 2013).

The present paper involves the determination of experimental data on  $\rho$ ,  $\eta$  and U of dextran polymer at five

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temperatures from 303 K to 323K and at the four frequencies 1MHz, 5MHz, 9MHz, and 12MHz, in 1 M sodium hydroxide solvent. The acoustic parameters are intended for analysis of solvent-solvent, ion-solute, and ion-ion interactions. We have chosen dextran as the solute with 1 M sodium hydroxide as the solvent. This is the only polymer that is water soluble. It has been a frequent subject of analyses due to its flexible "pharmaceutical, biomedical and modern applications" (Armstrong, Wenby, Meiselman, & Fisher, 2004; Pribush, Zilberman-Kravits, & Meyerstein, 2007).

In this appealing research work, the main problem was to find out the molecular interactions of dextran with sodium hydroxide in an aqueous medium using a simple method. The outcomes give subjective data with respect to the nature and the quality of interactions between solute and solvent in the liquids solution.

In continuation of our prior work on various concentrations of dextran in water medium, we have made a further endeavor to contemplate efficiently the physical-chemical behavior of dextran in sodium hydroxide, i.e in an alkaline medium. This examination is motivated by the flexible pharmaceutical, biomedical and modern applications, as industry demands reliable data on the physical and chemical properties of a wide variety of solutions.

## 2. Materials and Methods

The aqueous solution of 70,000 Da dextran in 1 M sodium hydroxide were prepared as reported in (Panda, 2020). The  $\rho$  and  $\eta$  of the solution were measured by using a pycnometer and an Ostwald viscometer respectively, while the samples were kept in a temperature-controlled water bath. The ultrasound velocity was measured by an ultrasonic interferometer operated at various frequencies from 1 to 12 MHz. The dextran of molecular weight 70,000 Da was of analytical reagent (AR) grade, manufactured by HI Media Laboratories Private Limited, India. All the measurements have been carried out in a water bath with circulating medium having an uncertainty of  $\pm 0.1$  °C. The viscosity of the solution was determined using pre-calibrated Ostwald's viscometer having an uncertainty of  $\pm 0.06\%$ . The viscosity measurements are based on the measurement of flow time of the solution with an uncertainty up to  $\pm 0.01s$ . The density measurement has been done using a pycnometer with an uncertainty of  $\pm 0.04\%$ . Sound speed in the solution was measured using an ultrasonic interferometer having and uncertainty within  $\pm 0.05\%$ .

## 3. Theoretical Aspects

The ultrasonic velocity is determined by (Panda & Mahapatra, 2019).

$$U = \lambda \times \nu \quad (1)$$

Where,  $\nu$  is the frequency of the generator which is used to excite the crystal.

The density was estimated using the equation

$$\rho_2 = \frac{w_2}{w_1} \rho_1 \quad (2)$$

Where  $w_1$  denotes the weight of distilled water,  $w_2$  denotes the weight of experimental solution,  $\rho_1$  denotes the density of distilled water,  $\rho_2$  denotes the density of experimental solution

The viscosity was calculated using the equation,

$$\eta_2 = \eta_1 \left( \frac{t_2}{t_1} \right) \left( \frac{\rho_2}{\rho_1} \right) \quad (3)$$

Where,  $\eta_1$  is the viscosity of water,  $\eta_2$  is the viscosity of the aqueous solution of dextran,  $\rho_1$  is the density of water,  $\rho_2$  is the density of aqueous solution of dextran,  $t_1$  is the time of flow of water, and  $t_2$  is the time of flow of solution of dextran.

The thermoacoustic parameters were calculated using  $\rho$ ,  $\eta$  and  $U$  as input data by using standard formulas (Panda & Mahapatra, 2019).

The adiabatic compressibility plays a vital role in explaining the degree of compactness of a medium. Its variation with temperature and frequency gives a significant explanation for the propagation of ultrasonic waves, which is a major factor in determination of compactness of the medium in different technological processes and industries.

$$\text{Adiabatic compressibility } \beta = \frac{1}{\rho u^2} \quad (4)$$

Acoustic impedance ( $Z$ ) is the impedance offered to the sound wave by the components of the solution. Acoustic impedance is the complex ratio of effective sound pressure at a point to the effective particle velocity at that point.

$$\text{Acoustic impedance } Z = \rho \cdot U \quad (5)$$

Relaxation time and absorption coefficient are directly correlated. The absorption of sound waves is the result of the time lag between the passing of the ultrasonic wave and the return of the molecules to their equilibrium position.

$$\text{Relaxation time } \tau = \frac{4}{3} \cdot (\beta \cdot \eta) \quad (6)$$

Molecules in the liquid state are loosely packed leaving some free space between them, called available volume. This available volume leads to introducing the concept of intermolecular free length.

$$\text{Intermolecular free length } L_f = \frac{K_T}{u \rho^{1/2}} \quad (7)$$

The Gibbs energy reveals closer packing of the molecules due to the H-bonding of unlike molecules in the solution. A positive Gibbs energy indicates that less time is required for the cooperative process or the rearrangement of molecules in the solution.

$$\text{Gibbs energy } \Delta G = KT \ln \left( \frac{kT\tau}{h} \right) \quad (8)$$

Where  $KT = (93.875 + 0.375T) \times 10^{-8}$ , 'T' is the absolute temperature; 'k' is the Boltzmann's constant, and 'h' is the Planck's constant.

**4. Results and Discussion**

The  $\rho$  and  $\eta$  of dextran with sodium hydroxide at temperatures 303 K, 308 K, 313 K, 318 K and 323 K are presented in Table 1;  $U$  and  $\beta$  at different temperatures and frequencies in Table 2;  $Z$  and  $\tau$  at different temperatures and frequencies in Table 3; and  $L_f$  and  $\Delta G$  at different temperatures and frequencies in Table 4.

The ultrasound speed increases with temperature at a particular frequency (Figure 1). This is due to structural changes occurring in the polymer solution on addition of sodium hydroxide, resulting in associations of the constituents (Panda & Mahapatra, 2014). There are molecular associations between the components of the solution due to dipole-dipole, dipole-induced dipole, charge transfer, and hydrogen bonding, which vary with the frequency of ultrasound (Panda & Mahapatra, 2018; Pawar, Chimankar, Bhandakkar, & Padole 2012). At constant temperature, ultrasound speed drops with frequency (Table 2). The decline in speed is an indication of weak molecular associations between the solute and solvent, as an agitation of the molecules reduces the speed.

The  $\beta$  decreases with temperature at a given frequency, and increases with frequency at a given temperature (Figures 2 and 3), which are reversed from velocity variation trends. This type of behaviour may be due to the break-up of associated clusters of dextran releasing

Table 1. Density ( $\rho$ ) and viscosity ( $\eta$ ) at various temperatures

T in kelvin	( $\rho$ ) kg.m <sup>-3</sup>	( $\eta$ ) 10 <sup>-3</sup> N.s.m <sup>-2</sup>
303	1039.00	1.256
308	1038.21	1.126
313	1036.78	1.070
318	1034.76	0.989
323	1031.74	0.893

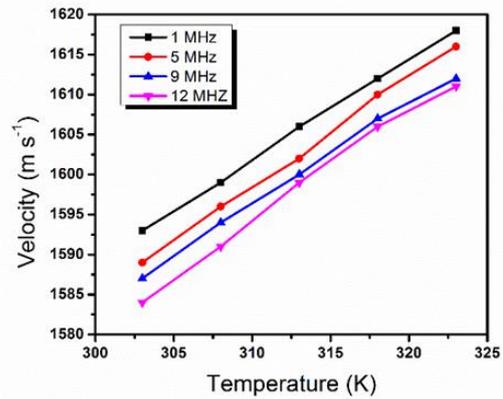


Figure 1. Ultrasound velocity versus temperature

Table 2.  $U$  and  $\beta$  at various temperatures and frequencies

T (K)	U (m.s <sup>-1</sup> )				$\beta$ (10 <sup>10</sup> m <sup>2</sup> N <sup>-1</sup> )			
	1MHz	5MHz	9MHz	12 MHz	1MHz	5MHz	9MHz	12 MHz
303	1593	1589	1587	1584	3.793	3.812	3.821	3.836
308	1599	1596	1594	1591	3.767	3.781	3.791	3.805
313	1606	1602	1600	1599	3.740	3.758	3.768	3.772
318	1612	1610	1607	1606	3.719	3.728	3.742	3.747
323	1618	1616	1612	1611	3.702	3.711	3.730	3.735

Table 3.  $Z$  and  $\tau$  at various temperatures and frequencies

T (K)	$Z$ (×10 <sup>4</sup> )kg.m <sup>-2</sup> .s <sup>-1</sup>				$\tau$ (× 10 <sup>10</sup> )s			
	1MHz	5MHz	9MHz	12 MHz	1MHz	5MHz	9MHz	12 MHz
303	1.655	1.651	1.649	1.646	6.350	6.382	6.398	6.423
308	1.660	1.657	1.655	1.652	5.654	5.675	5.689	5.711
313	1.665	1.661	1.659	1.658	5.334	5.361	5.374	5.381
318	1.668	1.666	1.663	1.662	4.905	4.917	4.935	4.942
323	1.669	1.667	1.663	1.662	4.407	4.418	4.440	4.446

Table 3.  $L_f$  and  $\Delta G$  at various temperatures and frequencies

T (K)	$L_f$ (10 <sup>-10</sup> m)				$\Delta G$ (10 <sup>-23</sup> (kJ/mol)			
	1MHz	5MHz	9MHz	12 MHz	1MHz	5MHz	9MHz	12 MHz
303	3.864	3.874	3.879	3.886	252.28	253.20	253.66	254.34
308	3.884	3.892	3.897	3.904	238.02	238.72	239.18	239.87
313	3.904	3.913	3.918	3.921	233.99	234.93	235.39	235.63
318	3.926	3.931	3.938	3.941	224.74	225.21	225.92	226.16
323	3.950	3.955	3.965	3.968	210.59	211.06	212.02	212.26

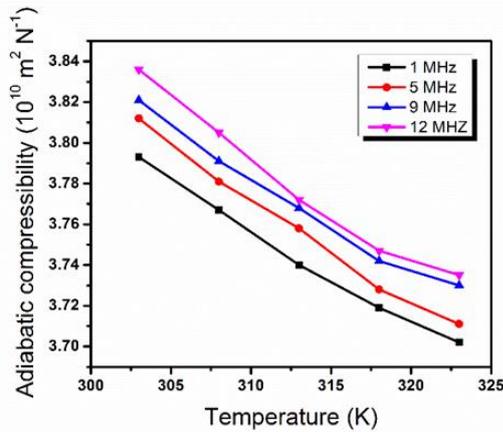


Figure 2. Adiabatic compressibility versus temperature

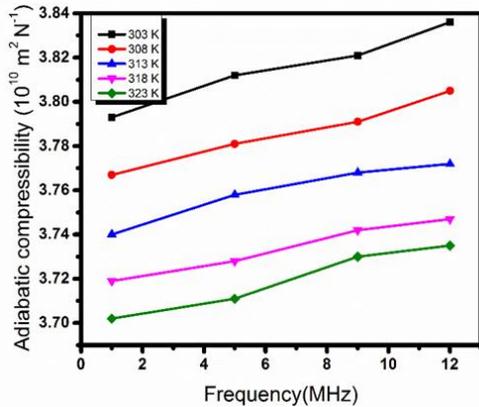


Figure 3. Adiabatic compressibility versus frequency

several dipoles and ions, which in turn induce dipole moments in sodium hydroxide, resulting in dipole-induced dipole and solute-ion interactions (Panda & Mahapatra, 2017).

Z increases with temperature (Figure 4). The solute-solvent molecules collide with each other more frequently, hence resisting the propagation of ultrasonic waves through the medium, so the acoustic impedance tends to increase with temperature. This indicates strong molecular interactions between the components of the solution at a given frequency (Figure 5). Z decreases with frequency indicating weak interactions between unlike molecules at a given temperature (Bhandakkar, Chimankar, & Pawar, 2010).

The  $\tau$  of dextran with sodium hydroxide at different temperatures and frequencies is shown in Figures 6 and 7. Figure 6 indicates that the relaxation time decreases with temperature. This is due to the increased number of collisions, and their higher frequency allows less relaxation time. The variation of  $\tau$  with frequency is almost parallel to the frequency axis (Figure 7) indicating very small changes in  $\tau$  with frequency (Tabhane, Chimankar, & Tabhane, 2012).

$L_f$  increases with temperature (Figure 8) due to thermal energy increasing the volume of the system, hence increasing  $L_f$ . Further, in the higher frequency range (Figure 9) intermolecular gap increases, which leads to a less ordered structure and more spacing between the molecules, decreasing the velocity that is observed (Panda & Mahapatra, 2019).

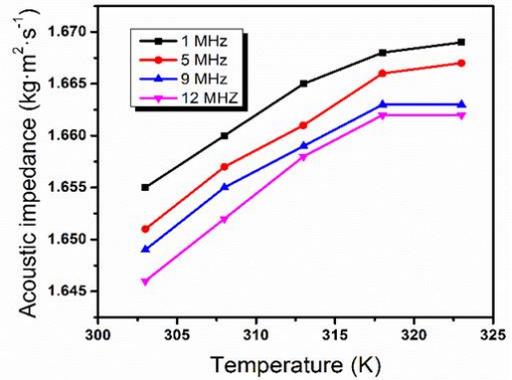


Figure 4. Acoustic impedance versus temperature

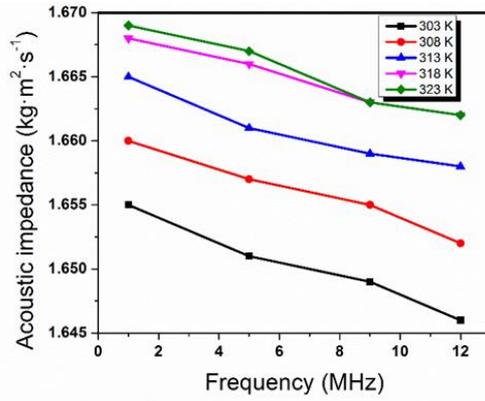


Figure 5. Acoustic impedance versus frequency

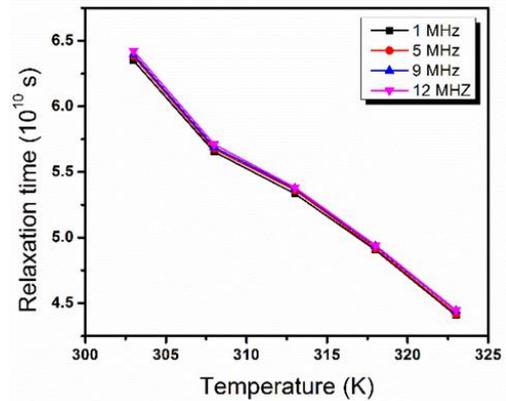


Figure 6. Relaxation time versus temperature

Gibbs energy of dextran with sodium hydroxide (Figure 10) falls with increasing temperature. The trend is the same at all frequencies (Figure 11) as if the frequency does not influence the Gibbs energy considerably.

### 5. Conclusions

When a polymer fragment is added to a solvent, forces of attraction or dispersion start acting between them, depending on their chemical nature, polarity and their solubility parameters. When the solvent-solute interactions

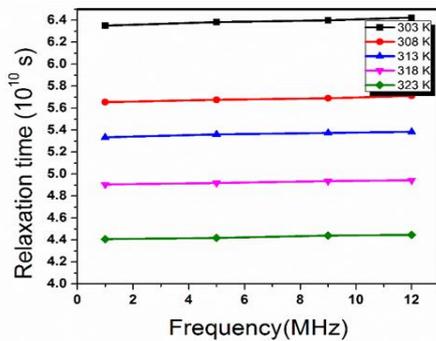


Figure 7. Relaxation time versus frequency

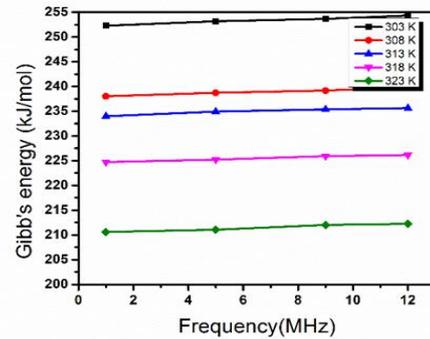


Figure 11. Gibbs energy versus frequency

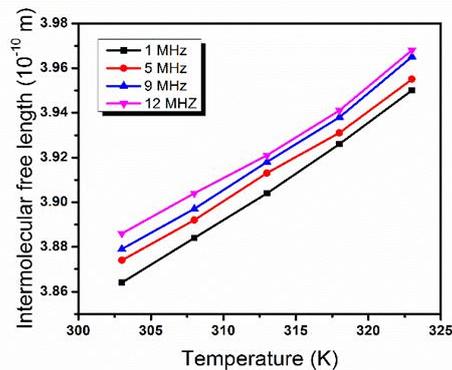


Figure 8. Intermolecular free length versus temperature

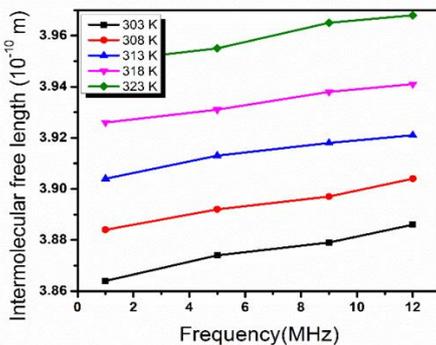


Figure 9. Intermolecular free length versus frequency

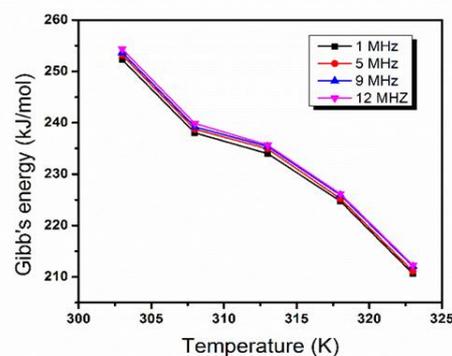


Figure 10. Gibbs energy versus temperature

become more powerful than the solute-solute attractions, the forces holding the polymer segments together become weak. So, the solvent molecules enter between the polymer segments and break the segment-segment contacts. They surround the individual segments and establish contact with them.

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