

The Study of Ultrasonic Velocity and Acoustical Parameters of the System Containing Aqueous Solution of Sodium Salt of 4-Amino Salicylic Acid at Different Temperatures

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Abstract

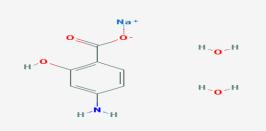
When acoustic waves are propogated through a liquid, dissipation of energy in sound waves takes place, the study of molecular interactions in various organic liquids and liquid mixtures is of considerable importance in recent years. 4-amino salicylic acid is most commonly used in the treatment of tuberculosis, known as Para- Amino Salicylic acid (PAS). It is antibiotic drug always used as Pro-drug. It is the most important member in the treatment of multidrug resistant treatment with few side effects. In the present work, ultrasonic velocity, density and viscosity of aqueous solutions of sodium salt of 4-amino salicylic acid have been measured experimentally using ultrasonic interferometer with 4 MHz frequency over entire range of concentration at temperatures 298.15K,303.15K and 308.15K. From this experimental data, other physical parameters such as acoustical impedance(Z), adiabatic compressibility (β), intermolecular free length (Lf), free volume(Vf), Rao's constant (R), Wada 's constant (W) have been determined for each composition. The computed acoustical parameters and their values point to the presence of specific molecular interaction in the mixtures and has been interpreted in light of solute-solvent interactions. Hence it is concluded that the association in these mixtures is the result of Hydrogen bonding in Binary liquid mixtures. The results may be helpful to understand the exact mode of action of the drug in vivo. **Keyword:** Ultrasonic velocity, Free volume(V_f), Rao's constant (R), Wada's constant (W).

Introduction

The study of intermolecular interactions plays an important role in the development of molecular sciences. A large number of studies have been made on the molecular interaction in liquid system by various physical methods like Infrared¹⁻², Raman effect³⁻⁴, Nuclear magnetic resonance, Dielectric constant⁵, Ultraviolet spectroscopy⁶ and Ultrasonic method⁷⁻⁸ In recent years, Ultrasonic technique has become a powerful tool in providing information regarding the molecular behavior of the medium. Ultrasonic's is an area of intense scientific and technological research. Science and technology of ultrasonic is widely sought in the recent years for industrial and medical application. Research and development in ultrasonic's have also maintained a steady pace for the past several decades to meet intense demands. The change in the wavelength of ultrasound waves in different media is due to the elastic properties and the induced particles vibrations in the medium. Further the wavelength of ultrasonic wave is small and hence it exhibits some unique phenomenon in addition to the properties of sound waves. The study of propagation of ultrasonic wave in liquid systems and solid is now rather well established. The ultrasonic velocity and density measurement of certain dielectric liquids can be used to compute acoustical parameters such as adiabatic compressibility, free length, Acoustic impedance, Rao's



Constant, and Wada's constant⁹⁻¹⁰. The present investigation deals with the study of molecular interaction in Sodium 4-Amino-Salicylate dihydrate in water at 298.15K, 303.15K and 308.15K. Further the data has been obtained at different concentration with a view to understand the effect of temperature on these properties



Sodium 4-Amino Salicylate dihydrate

Experimental Section:

Materials: Analytical Range (AR) sodium 4-amino salicylate dihydrate is used in the present work. The solutions were prepared by using double distilled water as solvent. The concentration range selected was 0.1M, 0.01M and 0.001M.

Methods: All the weighings was done on digital electronic balance Model-CB/CA/CT-Series Contech having accuracy \pm 0.0001g. The densities of the solutions were measured accurately using digital densitometer (Model-DMA Anton Paar). Viscosity of the solutions was measured by Ostwald's viscometer which was calibrated with benzene and double distilled water at all three temperatures. The values are accurate to \pm 0.001 cp.

The ultrasonic velocity was measured by using ultrasonic multi frequency interferometer (Model-M-83) supplied by Mittal Enterprises New Delhi, operating at 4MHz frequency with an accuracy of ± 2 m/s. The principle used in the measurement of ultrasonic velocity through medium is based on the accurate determination of wavelength of ultrasonic waves of known frequency produced by quartz crystal in measuring cell. The temperature of the solution was maintained by circulating water through the jacket of doubled walled cell. Measurements were made using constant temperature bath within ± 0.01 K.

Results and Discussions:

The experimentally measured values of density, viscosity and sound speed of solutions of 4amino salicylic acid sodium salt at 298.15K, 303.15K and 308.15K are given in **Table-1**.

The acoustical parameters were calculated from v, η and ρ values using standard formulae, and

given in Table - 2, 3 and 4.		0 - 1/2	1		
1) Adiabatic Compressibility		$- \qquad \beta = 1/v^2 \rho$	I		
2) Intermolecular free length		- $L_f = K \sqrt{\beta_s}$	2		
3) Specific acoustic impedance	-	$\mathbf{Z} = \mathbf{v}_{s} \cdot \boldsymbol{\rho}$	3		
4) Rao's Constant		- $\mathbf{R} = (\mathbf{M}_{\rm eff} / \rho) \times \mathbf{v}^{1/3}$	4		
5) Wada's Constant		- $\mathbf{W} = (\mathbf{M}_{eff} / \rho) \times \beta^{-1/7}$	5		
6) Relative Association	-	$\mathbf{R}_{\mathbf{A}} = \rho_{\mathbf{S}} / \rho_{\mathbf{O}} \left[\mathbf{v}_{\mathbf{o}} / \mathbf{v}_{\mathbf{s}} \right]^{1/3}$	7		
7) Relaxation time		$- \qquad \tau = 4/3 \beta \times \eta$	8		
8) Free Volume	-	$\mathbf{V}\mathbf{f} = \mathbf{M}_{\mathbf{eff}} \times \mathbf{v}_{\mathbf{s}} / \mathbf{k} \times \mathbf{\eta}$	9		
Where $k = 4.29 \times 10^9$ Temperature Independent Constant for all liquids					

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Sr.N o.	Temperature (⁰ K.)	Concentration (M)	Density(ρ _S) (Kg/m3)	Velocity(v _s) (m/s)	Viscosity(η) (Pa.S.)
1		0.1	1005	8205.44	9.79E-04
2	298.15	0.01	998.6	11191.6	9.64E-04
3	-	0.001	997.4	11212.232	9.37E-04
4		0.1	1004.7	9444.4	9.16E-04
5	303.15	0.01	998.3	13312.26	8.60E-04
6		0.001	997	6523.7	8.84E-04
7		0.1	1003.5	9754.9	8.18E-04
8	308.15	0.01	997.7	11991.16	7.88E-04
9		0.001	996.6	7420.44	7.79E-04

Table - 1: Density, Velocity and Viscosity at 298, 303, 308 K (At Frequency-4MHz)

 Table 2 :- Adiabatic Compressibility, Acoustic impedance and Free length.

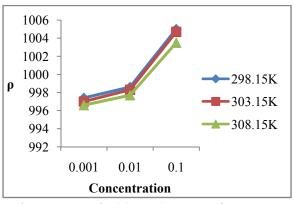
Sr.No.	Temperature	Concentration	Adiabatic	Acoustic	Free length
	(⁰ K.)	Μ	Compressibility	Impedance	L _f m
			(β _S)Pa-1	$Z x(Kgm^{-2}S^{-1})$	
1		0.1	1.478E-11	8.25E+06	7.56E-12
2	298.15	0.01	7.995E-12	1.12E+06	5.56E-12
3		0.001	7.9753E-12	1.12E+07	5.55E-12
4		0.1	1.116E-11	9.49E+06	6.61E-12
5	303.15	0.01	5.652E-12	1.33E+07	4.71E-12
6		0.001	2.357E-11	6.50E+06	9.61E-12
7		0.1	1.047E-11	9.79E+06	6.48E-12
8	308.15	0.01	6.971E-12	1.20E+07	5.28E-12
9		0.001	1.822E-11	7.40E+06	8.54E-12

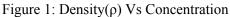
Table 3 : Relative association and relaxation time and apparent molar compressibility

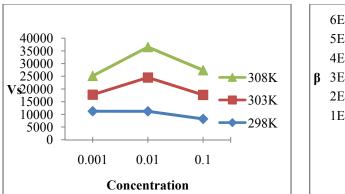
Sr.No ·	Temperatur e(⁰ K.)	Concentratio n(M)	Relative Association (R _A)	Relaxation Time τ	App. Molar Compressibility φ _k
1		0.1	6.20E-01	1.93E-14	-2.6E-09
2	298.15	0.01	6.16E-01	1.03E-14	-2.67E-08
3	1	0.001	5.55E-01	9.96E-15	-2.67E-07
4	303.15	0.1	5.75E-01	1.36E-14	-3.16E-09
5		0.01	5.13E-01	6.48E-15	-3.22E-08
6		0.001	6.45E-01	2.78E-14	-3.04E-07
7	308.15	0.1	5.43E-01	1.14E-14	-4.18E-09
8		0.01	5.04E-01	7.33E-15	-4.22E-08
9		0.001	5.91E-01	1.89E-14	-4.11E-07

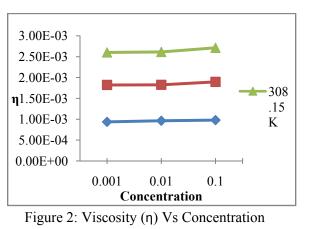
Sr.No.	Temperature (⁰ K).	Concentration(M)	Rao's Constant (R)	Wada's Constant (W)	Free Volume(V _f)
1	298.15	0.1	2.01E-03	3.50E-03	2.738E-06
2		0.01	2.23E-03	3.84E-03	4.45E-06
3	-	0.001	2.24E-03	3.85E-03	4.654E-06
4		0.1	2.10E-03	3.65E-03	3.73E-06
5	303.15	0.01	2.37E-03	4.04E-03	6.843E-06
6		0.001	1.87E-03	3.30E-03	2.252E-06
7		0.1	2.13E-03	3.68E-03	4.643E-06
8	308.15	0.01	2.29E-03	3.92E-03	6.664E-06
9		0.001	1.95E-03	3.42E-03	3.299E-06

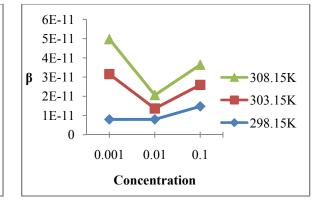
 Table - 4 : Rao's Constant, Wada's Constant, Free volume.











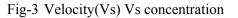
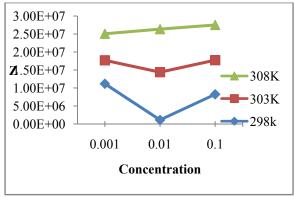


Fig-4 Adiabatic compressibility Vs concentration

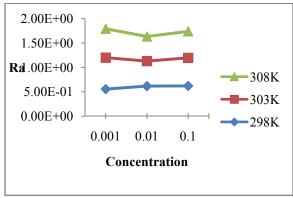
This paper deals with the study of molecular interaction in binary liquid mixtures of Sodium 4amino salicylate dihydrate at 298.15K, 303.15K and 308.15K. We have reported ultrasound velocity (Vs), density (ρ_s) and viscosity (η) of binary liquid mixture. With the help of experimental data, the thermodynamic and acoustic properties like adiabatic compressibility (β_s), intermolecular free length (L_f),

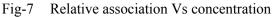


free volume (V_f), and their excess values have been computed at three different temperatures 298.15K, 303.15K, 308.15K. The above functions and their excess values are tabulated and in figures. The 4-amino salicylate in water acts as polar molecule.









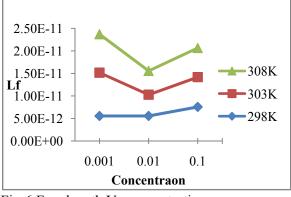


Fig-6 Free length Vs concentration

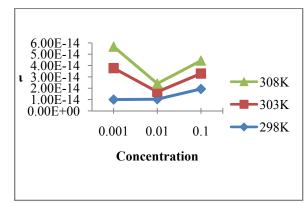
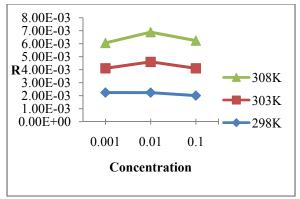
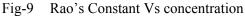


Fig-8 Relaxation time Vs concentration





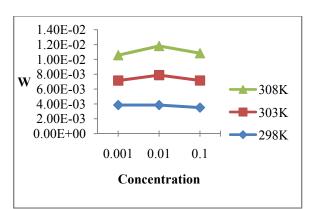
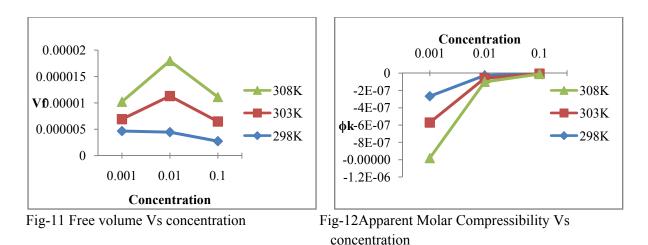


Fig-10, Wada's Constant Vs concentration





In the present investigation, the measured values of ultrasonic velocity for the liquid mixture of sodium-4amino-2-hydroxy benzoic acid dihydrate in water are reported in Table -1. It is observed from Table-1 and Figure3 for liquid mixture of sodium-4-amino-2-hydroxy benzoic acid dihydrate in water that ultrasonic velocity values shows similar trend for all concentration at different temperature. The adiabatic compressibility is the fractional decrease of volume per unit increase of pressure, when no heat flows in or out. These changes are related to the compressibility of the medium by thermodynamic relation

 $\beta = 1/v^2 \rho_S$

From the **Table.1**, it is noted that the density and viscosity decreases with decrease in concentration for all the cases. The variation in velocity may be due to the variation in free length and adiabatic compressibility of the liquid mixtures (**Figure 4 and 6**) which shows the change in the structural arrangement of the molecules in the mixture as stated in some other aqueous binary mixtures¹¹⁻. The adiabatic compressibility depends on the values of ultrasonic velocity. The value of adiabatic compressibility for system sodium-4-amino-2-hydroxy benzoic acid dihydrate in water decreases at 0.01 M Concentration and increases again with increase in concentration at 0.1 M shown in Figure4. The decrease in Adiabatic Compressibility values suggests presence of solvent-solvent interactions and increase in Adiabatic Compressibility values suggests solute-solvent molecular interactions. This may lead to the presence of specific molecular interaction between the molecules of the liquid mixture. For the system sodium-4-amino-2-hydroxy benzoic acid dihydrate +Water, the values of intermolecular free length show somewhat irregular trend with increase in concentration as shown in Figure6. As concentration increases at 0.01M, L_f decreases. The reduction in L_f indicates that the components are much closer. The adiabatic compressibility and free length are the deciding factors of the ultrasonic velocity in liquid systems.

According to Eyrings liquid state theory, the acoustic wave, which was excited in the liquid, is transmitted momentarily to the intermolecular free length and the rate process in liquids is determined by the free volume. Free volume plays an important role in ultrasound wave propogation. The values are reported in Table 4. for the system sodium-4-amino-2-hydroxy benzoic acid dihydrate + water shows that the free volume increases with increase in concentration of solute at temperature 298.15K. It indicates that the addition of salt in solution enhances the structural arrangement of the system. Free volume is one of the significant factors in explaining the free space and its dependent properties have close connection with molecular structure and it may show interesting features about interactions between components in liquid mixtures.

The values of acoustic impedance increases with increase in concentration, but the trend is not regular at temperature 298.15K. (Figure 5) Increase in the values of acoustic impedance shows the presence of solute-solvent interactions in the system. It may be due to the interactions between the functional group $-NH_2$ or -COOH of sodium-4-amino-2-hydroxy benzoic acid dihydrate and -H or -OH of water. The relaxation time (t) varies with increasing concentration for all the three systems (**Figure8**). The dispersion of the ultrasonic velocity in the system should contain information about the characteristic time of the relaxation process that causes dispersion. The relaxation time which is in the order of 10^{-14} sec is due to structural relaxation process¹³ and in such a situation it is suggested that the molecules get rearranged due to co-operative process¹⁴.

For the system sodium-4-amino-2-hydroxy benzoic acid dihydrate + water, the value of Rao's constant shows non linearity with increasing concentration at all temperatures. The variation of molar sound velocity or Rao's Constant and molar compressibility or Wada's Constant are found to be non linear (**Table.4 and Figure 9 and 10**). It shows the presence of solute-solvent interaction¹⁵.

The relative association depends on either the breaking up of the solvent molecules on addition of solute to it or the salvation of ions that are present. In the present case for the system sodium-4-amino-2-hydroxy benzoic acid dihydrate + water, the values of relative association increases with increase in concentration which is due to the salvation of solute on addition of it in solvent at temperature 298.15K. But at temperature 303.15K and 308.15K, the variation in R_A is irregular (Figure 7). Systematic variations are observed in apparent molar compressibility with concentration of solute for system sodium-4-amino-2-hydroxy benzoic acid dihydrate + water over the entire range of composition and at the temperatures studied (Figure 12). The values of apparent molar compressibility increases with increase in concentration showing all the values are negative. It indicates occurrence of molecular interactions in the system.

Coclusion:

The computed acoustical parameters and their values point to the presence of specific molecular interaction in the mixtures. Hence it is concluded that the association in these mixtures is the result of Hydrogen bonding in Binary liquid mixtures.

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