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Behaviour of Sodium-2-Hydroxy Benzoic Acid in the Ternary Mixture with Water and Ethanol: An Ultrasonic Study

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***Abstract*-**Present paper, discusses about the measurement of ultrasonic speed, density and viscosity of sodium salicylate solution using ethanol and water in 1:1 ratio as a solvent at 298K, 303K, and 308K and ultrasonic speed at 4MHz frequency. By using these experimental values, various thermodynamic as well as acoustic parameters have been calculated. It is found that the value of these parameters have close relationship with the molecular interactions in the solutions .The data shows that the molecular interaction exists in ternary mixtures and also there is a strong solute–solvent interaction occurring in ternary solutions. This indicates that there may be hydrogen bonding between –COOH group of 2-hydroxy benzoic acid, ethanol and molecules of water.

Keywords- Ultrasonic Velocity, Relaxation Time, Acoustic Impedance, Wada′S Constant

I. INTRODUCTION

Ultrasonic technique is the most useful technique for studying the molecular behaviour of liquid mixtures[1].The study of liquid and liquid mixtures by using ultrasonic technique is much more important in studying the nature and relative strength of molecular interactions and finding the physicochemical characteristics of such system[2].In recent years measurement of ultrasonic velocity finds the number of applications in determination of different thermodynamic and physicochemical parameters[3-4]. Ultrasonic is the non-destructive technique which has been serving as a realistic technique applied to study the aqueous solutions, on-aqueous solutions and electrolytes. This modern approach gives new sight to enter into the process of ion-ion and ion-solvent interactions. Many of the researchers have measured the ultrasound speed for many binary and ternary liquid mixtures in polar and non-polar solvents [5].

These studies have conclusively established the existence of association, dissociation, complex formation and hydrogen bonding between the molecules. The parameters like acoustical and thermodynamic are widely used to study molecular association and different interactions in their solutions[6-7].The passing of ultrasonic waves through liquid is a tool used by researchers to probe into the properties of liquids. Like ultrasonic technique, other related technique like refractometry is used to study thermodynamic and physicochemical properties of binary and ternary liquid mixtures and presented in graphical manner to gain insight into the various interactions occurring in the mixtures [8]. The physical and chemical behaviour of solutions and molecular interactions can be studied by the measurement of the density, viscosity and ultrasonic velocity. These parameters are helpful for calculating the acoustic parameters such as adiabatic compressibility, free length, specific acoustic impedance, Rao′s constant, Wada′s constant. The present work deals with the investigation of behaviour of ternary solutions of sodium-2-hydroxy benzoic acid in 1:1 mixtures of water and ethanol with respect to adiabatic compressibility, intermolecular free length, acoustic impedance, Rao′s constant, Wada′s constant etc. by measurement of ultrasonic velocity at 298K,303K and 308K keeping frequency at 4 MHz. The obtained results are used to interpret the molecular interactions between the component molecules in the liquid mixtures.

II. EXPERIMENTAL SECTION

Materials

Sodium salicylate used was of A.R. grade. 1:1ratio of mixture of distilled water and 99.99% pure ethanol was used as a solvent for the preparation of solutions. Weights have been taken on digital electronic balance. (Model-CB/CA/AT-Series).

Methods

Ultrasonic Interferometer (Model-M-83). Mittal Enterprises, New Delhi) operating at4MHz frequency with an accuracy of ±2m/s. was used to measure the ultrasonic speed. The viscosities (η) of solution and solvent were determined using Ostwald's viscometer by calibrating with double distilled water. The densities (ρ) were measured accurately using digital densitometer (Model - DMA-35, Anton Paar). The ultrasonic speed was measured at4MHz frequency at 298K; 303K.and 308K.The temperature of cell was maintained with continuous circulation of water at constant temperature by using thermostat.

III. RESULTS AND DISCUSSION

The following formulae were used to calculate the acoustical parameters,

1) Adiabatic Compressibility$\beta= 1/v^2 \rho_S$…(1)

2) Free LengthL_f= K $\sqrt{\beta_S}$…(2)

3) Specific acoustic impedance Z = Vs . ρ_S…(3)

4) Rao's ConstantR = $(M_{eff}/\rho_S) \times V^{1/3}$…(4)

5) Wada's ConstantW = $(M_{eff}/\rho_S) \times \beta^{-1/7}$…(5)

6) Apparent Molar Compressibility -

$\Phi_K = [\ 1000\ (\ \beta_S\rho_0 - \beta_O\rho_S\ )\ /\ m\ \rho_S\rho_0\ ] + (\ \beta_S\ M\ /\ \rho_S)$ …(6)

7) Relative Association -R_A= $\rho_S / \rho_0[\ Vo\ /\ Vs\ ]^{1/3}$…(7)

8) Relaxation time – $\tau= 4/3\ \beta \times \eta$ …(8)

9) Free VolumeVf = $M_{eff} \times v_s / k \times \eta$ ….(9)

Where k = 4.28×10^9, Temperature Independent Constant for all liquids.

10) Equivalent Conductance -μ= Kc[1000/N] ….(10)

The symbols have their usual meaning.

The experimental data relating to viscosity, density and ultrasonic velocity at 298K,303K and 308K for frequency4MHz for the mixture are shown in Table no. 1.

The values of density (**ρ_S**), Viscosity(**η**), Ultrasonic velocity and different derived parameters at 298K,303K and 308K and at 4MHz Frequencyfor sodium salicylate solution in 50% alcohol are shown in the**table 1,2,3 and 4**.

Table 1: - Density, Velocity and Viscosity

Sr.No.	**Temperature (^{0}K.)**	**Concentration(M)**	**Density(ρ_S) (Kg/m^3)**	**Velocity(v_s) (m/s)**	**Viscosity(η) (Pa.S.) or Kg m^{-1} s^{-1}**
1	298	0.1	910.93	5309.4	2.29E-03
2		0.01	927.9	5187.04	2.28E-03
3		0.001	916.84	4394.78	2.15E-03
4	303	0.1	909.5	2946.08	1.99E-03
5		0.01	925.32	4027.11	2.01E-03
6		0.001	916.36	3112.7	1.98E-03
7	308	0.1	908	3663	1.75E-03
8		0.01	923.05	3612.1	1.73E-03
9		0.001	915.6	3349.9	1.73E-03

Table 2 :- Adiabatic Compressibility, Acoustic impedance and Free length at 298K, 303K, 308 K.

Sr.No.	Temperature (^{0}K.)	Concentration M	Adiabatic Compressibility (β_S) $N^{-1}m^2$	Acoustic Impedance Z ($Kgm^{-2}S^{-1}$)	Free length Lf (m)
1	298	0.1	3.8943E-11	4.84E+06	1.23E-11
2		0.01	4.0055E-11	4.81E+06	1.24E-11
3		0.001	5.64719E-11	4.03E+06	1.48E-11
4	303	0.1	1.2668E-10	2.68E+06	2.23E-11
5		0.01	6.6638E-11	3.73E+06	1.62E-11
6		0.001	1.1263E-10	2.85E+06	2.10E-11
7	308	0.1	8.2081E-11	3.33E+06	1.81E-11
8		0.01	8.3034E-11	3.33E+06	1.82E-11
9		0.001	9.7326E-11	3.07E+06	1.97E-11

Table 3 : Relative association, Apparent molar compressibility and relaxation time at 298K, 303K, 308 K. at 4MHz Frequency.

Sr.No.	Temperature(^{0}K.)	Concentration (M)	Relative Association (R_A)	Relaxation Time (τ) S	Apparent molar compressibility (ϕ_k)
1	298	0.1	9.05E-01	1.19E-13	-2.262E-10
2		0.01	9.29E-01	1.22E-13	-2.281E-09
3		0.001	9.70E-01	1.62E-13	-4.447E-09
4	303	0.1	1.09E+00	3.37E-13	6.88E-10
5		0.01	9.97E-01	1.79E-13	-5.52E-11
6		0.001	1.08E+00	2.97E-13	5.02E-08
7	308	0.1	9.94E-01	1.92E-13	1.14E-10
8		0.01	1.02E+00	1.91E-13	9.63E-10
9		0.001	1.04E+00	2.25E-13	2.58E-08

Table 4.Rao's Constant, Wada's Constant, Free volume and Equivalent conductance

Sr.No.	Temperature (^{0}K).	Concentration(M)	Rao's Constant (R)	Wada's Constant (W)	Free Volume (Vf) m^3mol^{-1}	λ=Kc*(1000/N)mhos eq.
1	298	0.1	1.79E-03	3.15E-03	3.6125E-07	34700
2		0.01	1.74E-03	3.07E-03	3.479E-07	71000
3		0.001	1.66E-03	2.95E-03	2.961E-07	350000
4	303	0.1	1.47E-03	2.65E-03	1.82231E-07	36100
5		0.01	1.59E-03	2.85E-03	2.852E-07	75000
6		0.001	1.48E-03	2.67E-03	1.988E-07	460000
7	308	0.1	1.57E-03	2.82E-03	3.043E-07	42200
8		0.01	1.53E-03	2.75E-03	3.02E-07	89000
9		0.001	1.51E-03	2.71E-03	2.693E-07	570000

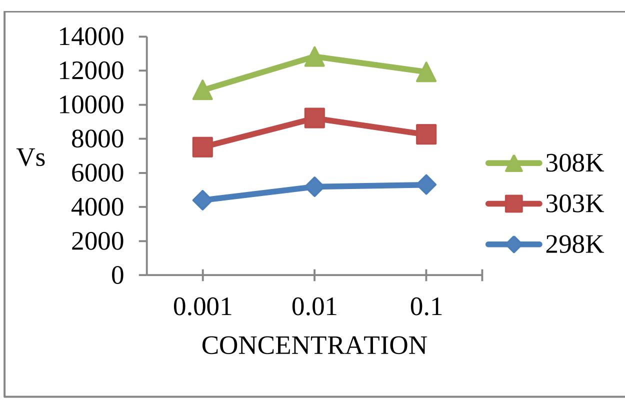
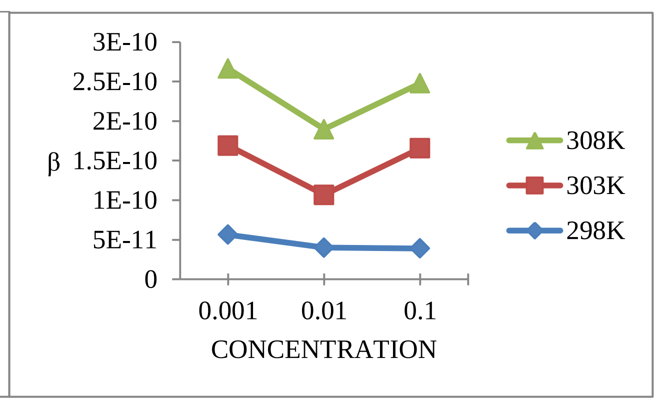


Fig.1



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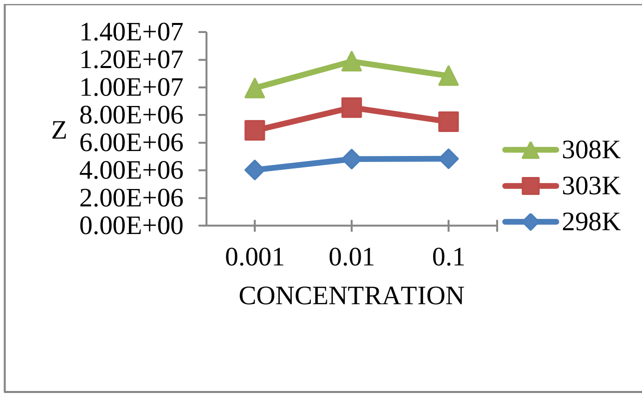


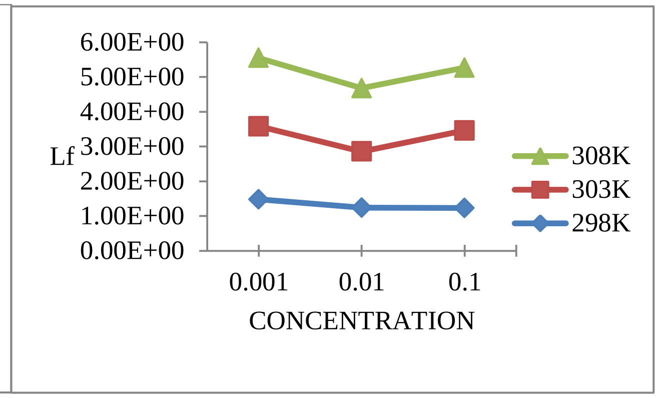
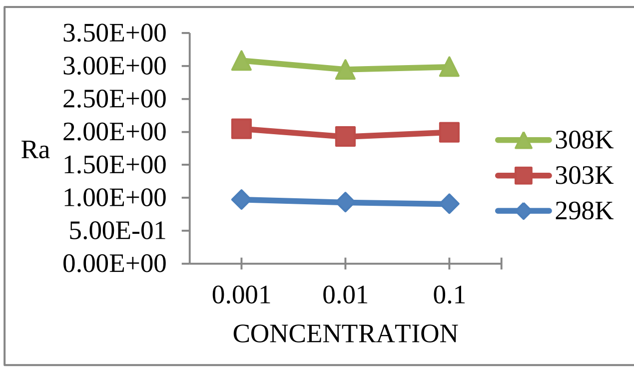


Fig.3

Fig.4



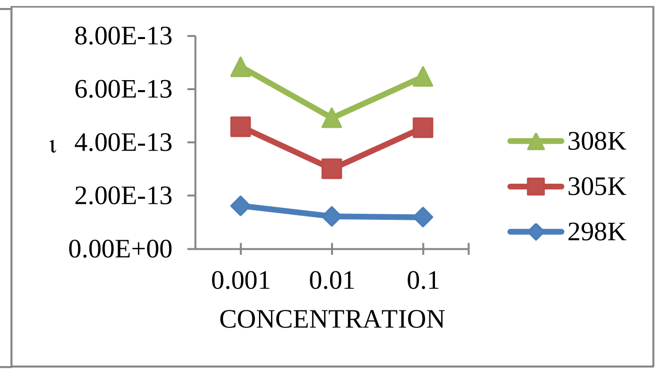
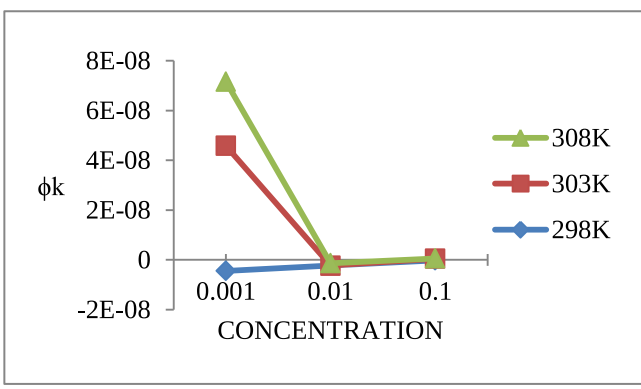


Fig.5

Fig.6



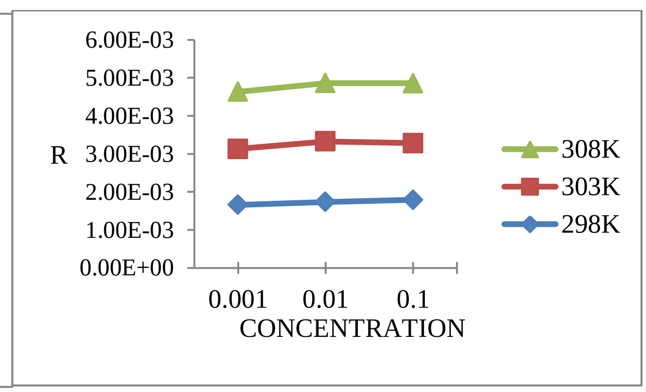
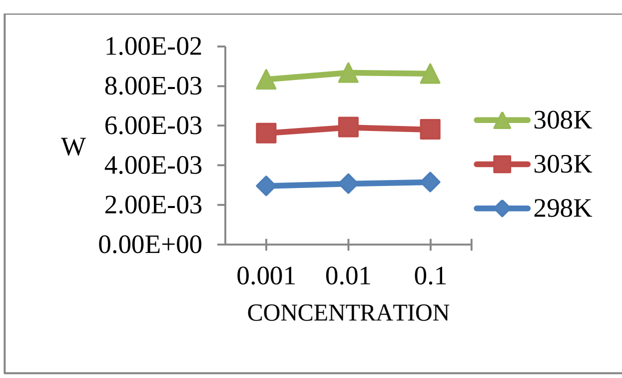


Fig.7

Fig.8



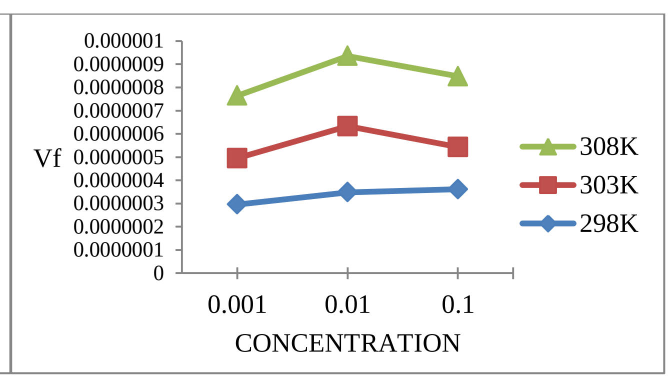


Fig.9

Fig.10

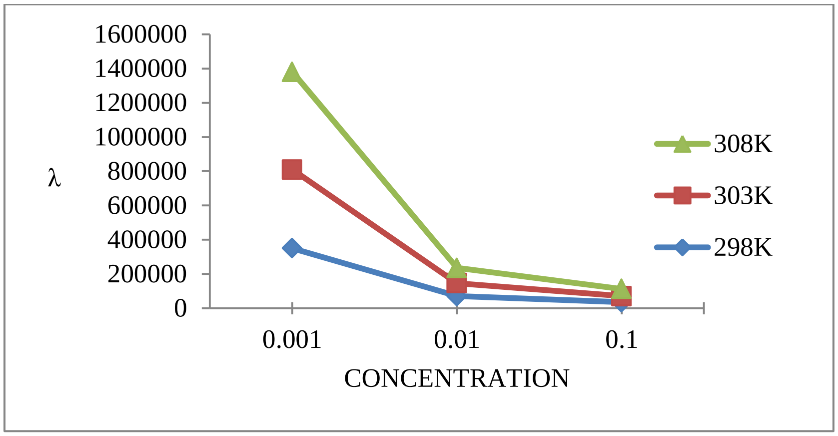


Fig.11

At 4MHz frequency, variation in ultrasonic speed of sodium salicylate solution is decreasing with temperature and increases with increase in concentrations(table 1 and fig.1.) ultrasonic velocity values changes with concentration in the binary mixture can be explained in terms of different types of interactions present in the system[9-12].The values of ultrasonic velocity depends on the structures of the solute. When solute is dissolved in the solvent, due to the changes in molecular free length, the values of ultrasonic velocity also changes which can be explained on the basis of Eryingand Kincarl [13].model. The ultrasonic velocity increases as concentration increases which shows that that there may be formation of hydrogen bond between solute and solvent molecules leads to the closed packed structures. Table 2 and fig.2 shows variation in adiabatic compressibility with concentration. Adiabatic compressibility decreases with increasing concentration, it is due to the effect of the electrostatic field of the ions on the surrounding solvent molecules of ethanol and water. The lower values of adiabatic compressibility reveal that the medium is tightly packed [14].

Intermolecular free length is defined as it is the distance between surfaces of the adjacent molecules. It has direct proportion to the adiabatic compressibility and inverse proportion to ultrasonic velocity as shown in table2and fig.4. It shows same approach to that of adiabatic compressibility and opposite approach to that of ultrasonic velocity. According to Erying and Kincaid model there is inverse relation between the intermolecular free length and ultrasonic velocity in the liquid mixtures. Therefore intermolecular free length is the predominant factors for deciding ultrasonic parameters in the liquid mixtures.

The variation in specific acoustic impedance with concentration is shown inFig.3 and table 2. The acoustic impedance is influenced by inertial and elastic properties of the medium [15]. With increase in concentration, the acoustic impedance increases which supports the presence of molecular interaction between solute and solvent. The increase in acoustic impedance values with concentration is responsible for the effective solute-solvent interactions [16].This suggests the molecular packing in the medium which shows the hydrogen bonding between sodium salicylate and solvent molecules.

Relative association is the measure of extent of association of components in the medium. It is the property of understanding the interactions between molecules in liquid mixtures and solutions. Fig. 5 and table 3 shows variation of relative association with concentration. Relative association decreases with concentration at 4 MHz frequency shows strong solute-solvent interactions and the molecules are closer to each other and there may exist ion dipole type of interactions between component molecules in the binary liquid mixture [17].

The values of Rao′s constant(R) and Wada′s constant (W) increase linearly with the concentration (table.4, Fig.8 and Fig.9.) The linear increase in Rao′s constant and Wada′s constant shows the presence of specific interactions in the liquid mixture without complex formation.

Acoustical relaxation time (ι) decreases as there is increase in concentration but this decrease is not regular. By comparing the values of adiabatic compressibility and acoustical relaxation time shows that the acoustical relaxation time varies mainly due to the variations in the viscosity of the solutions due to both concentration and temperature. It is seen that the free volume rises with rise in salt concentration. This is due to the loosely packed solvent molecules of ethanol and water around the ions at higher concentrations and indicates significant interaction between solute and solvent molecules.

Like adiabatic compressibility (β), apparent molar compressibility (ϕ_k) is another important acoustic parameter which explains the solute-solvent and solute-solute interactions in solutions. The variation in the values of apparent molar compressibility (Fig.7 and table 3) may explain the interactions between molecules, like structure making and breaking nature of solute. Apparent molar compressibility property is related to structural changes especially in highly structured solvent like water/ethanol. The values of apparent molar compressibility decreases with rise in concentration shows loosely packing of solute-solvent molecules also shows the interactions between the molecules of solute-solvent.

The equivalent conductance values are decreasing for increase in concentration while increasing for increase in the temperature as expected (table 4 and fig. 11.) There is inverse relation in between specific conductance and equivalent conductance. The decrease in equivalent conductance with rise in concentration indicates presence of fewer ions concentration in particular zone of the solution and correlates with the loosely packed structure of solute-solvent molecules.

IV. CONCLUSIONS

Experimental data of density, velocity, related acoustical parameters and some of their excess values for binary liquid mixture shows that there exist interactions between molecular components in the liquid mixture of sodium salicylate, water and ethanol. The ultrasonic velocity values increase as concentration of sodium -2-hydroxy benzoic acid increases. The decreased values of intermolecular free length and adiabatic compressibility indicate the strong molecular interaction in the liquid mixture.

The negative excess values of apparent molar compressibility and increased values of acoustic impedance predict the existence of dispersive forces between component molecules in the solutions. The nonlinear behaviour in the parameters confirms the presence of solute-solute, solute-solvent and dipole-dipole interactions. All the values of acoustic parameters show that solute and solvent molecules may be loosely packed due to salvation between solute molecules and mixture of ethanol and water solvent molecules. Observed molecular interactions are due to heteromolecular interactions in the solutions by the formation of hydrogen bonds. This gives valuable information about the existence of intermolecular and intra-molecular interactions in the liquid mixtures.

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Dr. ShashikantRajeshwarAswale did M. Phil. From Department of Chemistry, Nagpur University; Nagpur in the Session 1987-1988 under UGC FIP, Fifth Five years plan. The topic of his Research was "Partial molal and excess molar volumes in ternary liquid mixtures (Methanol-Dioxane-Water)" under the supervision of Dr. A. A. Bhalekar.

He worked as **Associate Professor** for more than 25 years in LokmanyaTilakMahavidyalaya, Wani. He has more than 50 publications in International and National journals to his credit and presented more than 50 papers in National and International conferences. He has organized a National Conference in Chemistry at Wani and published a "Proceedings of National Conference" with ISBN NO. 978-81-905776-12-1.He is also an Editor of"A Text Book of Chemistry", B. Sc. Sem.II, SantGadgeBaba Amravati University, having ISBN NO. 13-978-81-926163-3-9. He has completed one Minor Research Project funded by U.G.C. (WRO) Pune. Five students are awarded Ph.D. degree under his supervision and three students are doing research pertaining to Ph. D.

He also worked as **Principal** of LokmanyaTilakMahavidyalaya, Wani and Mahatma Gandhi College of Science, Gadchandur for more than 8 years and during this tenure he has organized a number of students centric programs like -Campus Interview (RojgarMelawa),"Wiwidh Kala Mahotsav" sponsored by Maharashtra Government Cultural Ministry,"Lok Kala Mahotsav" Sponsored by Dakshin Madhya Kala SanskrutikManch, GraminSahityaSammelan,ShetkariMelawa,Golden Jubilee of college, workshops on Information Technology, Radiology, Self-employment, Sculpture, Video shooting, Photography, Bag making,MIHAN etc.

As a Social Consultant he motivated "HENKEL" Mumbai Company to donate a] Rs. 5 lacks to "BAL-SADAN" for children of Prostitutes at Wani, b] Rs. 4 lacks to "NRHUSINGH BAL-SADAN" for orphan children at Wani.

He was an Associate **NCC officer** for 21 years and leadtheMaharashtra Directorate, NCC boys, All India ThalSena Camp, New Delhi 1999, as Contingent Commander and for the same he was awarded a "Certificate of Merit" by Amravati NCC Group for training BLC team, 1999.

He is member of **Advisory Board**of RNB Global University, Bikaner, Rajasthan, **Review committee**of a) Wyno Journal of Agricultural Sciences, Libiya, b)Journal of Pure and Applied Ultrasonics, India,c) International journal of Analytical Chemistry, **Editorial board** of a) Wyno Journal of Agricultural Sciences, Libiya, b) Natural Science Society Ajmer, India, c) Advances in Natural Science, Canada and **Life member**of a) Indian Council of Chemist, Agra, Journal, b) Indian Science Congress, c) Natural Science Society Ajmer d) Ultrasonic Society of India, e) Association of Chemistry Teachers, Mumbai, f) American Chemical Society.

He received different awards like "**Best Service Award**", category-**Principal**, by S.G.B. Amravati University, on 1st May, 2014, "**Innovative Award**" by SGB Amravati University, on 1st May 2013,"**ShikshakRatna**" A best teacher National level award by SamtaSahitya Academy, 2011, "**Collection of Blood**" by SGB Amravati University and Chandrapur Blood Bank etc.

Presently he is working as **Finance and Accounts Officer** in SantGadge Baba Amravati University, Amravati since last one and half years and is a member of various authorities and committees of SantGadge Baba Amravati University, Amravati.

