

**GLOBAL JOURNAL OF ENGINEERING SCIENCE AND RESEARCHES**  
**INVESTIGATION OF ACOUSTICAL PARAMETERS OF N-(2'-HYDROXY-3'-**  
**NITROBENZYLIDENE) -3-SUBSTITUTED PYRIDINE-2-AMINE SCHIFF BASE**  
**DERIVATIVES IN DIOXANE-WATER MIXTURE AT 293, 297 AND 300 K**

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**ABSTRACT**

The density, viscosity and ultrasonic velocity of synthesized Schiff base N – (2'-hydroxy-3'-nitrobenzylidene) pyridine-2-amine and its substituted derivatives have been studied in 1,4-dioxane-water mixture at 293, 297 and 300 K, over a wide range of concentration. In present work, study of acoustical parameters like ultrasonic velocity (V), adiabatic compressibility ( $\beta_s$ ), apparent molar volume ( $\phi_v$ ) and intermolecular free length ( $L_f$ ) by ultrasonic interferometric measurements was done, which reflects structural interaction of water molecules and organic solvent molecules with substituted Schiff bases.

**Keywords:** Schiff bases, Density, ultrasonic velocity, acoustic, structural interaction.

**I. INTRODUCTION**

The structure making or breaking property of a solvent is associated with the study of number of acoustic parameters like adiabatic compressibility ( $\beta_s$ ), apparent molar volume ( $\phi_v$ ), intermolecular free length ( $L_f$ ), Specific acoustic impedance (Z), Relative association ( $R_A$ ), etc. To determine these parameters the ultrasonic velocity is a simple probe used by physicist along with density and viscosity [1]. Literature survey shows that ultrasonic study of liquid mixture is highly useful in understanding the nature of molecular interaction [2-4] and physicochemical behaviour of liquid mixture [5-6].

Ultrasound is used in many different fields like Ultrasound imaging is often used in quality control and medicine [7], in the non-destructive testing of products and structures, in industries for quality check, to speed up chemical processes, etc. Scientists are also studying application of the ultrasound in *ex-situ* form for cleaning the fouled membranes and pretreatment of wastewater prior to the membrane bioreactor system [8].

Ultrasonic technology has been adequately employed to investigate the properties of any substance to understand the nature of molecular interactions in pure liquid [9], liquid mixtures [10-12] and ionic interactions in electrolytic solutions [13]. One important ace for ultrasonic technique is that it can reveal very weak intermolecular interactions. Thus many researchers have made important advancements in understanding the nature of molecular interaction and physicochemical behavior of liquid mixtures by studying the acoustics at different concentrations and temperatures and results were interpreted in terms of solute-solvent and solvent-solvent interaction [14-24].

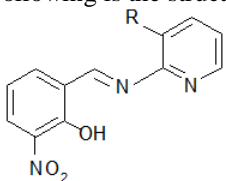
In the present investigation, acoustical parameters of Schiff base have been investigated. Schiff bases are having high synthesis flexibility, coordinating ability and medicinal utility and are an important class of ligands in the field of co-ordination chemistry [25]. A large number of Schiff bases have been found to possess important biological and catalytic activities [26-27] and their acoustical parameters were reported [28-29]. Recently a review was reported that covers a broad range of Schiff bases that are used in sensing applications for metallic cations and anions in

various kinds of environmental and biological media [30]. A correlation between anacoustic parameters and concentration of Schiff bases was reported which indicated existence of strong molecular interactions with temperature [31].

N – (2- hydroxyl-3-nitrobenzylidene) pyridine -2- amine and its substituted derivatives were synthesized in the laboratory preferring microwave radiations over conventional route [32]. The synthesized compounds were then characterized by their chemical properties, elemental and spectral analysis. In the present study, attempt has been made to study the molecular interactions of the following substituted Schiff base ligands in the suitable percentage of 1,4-dioxane – water mixture at different temperatures by acoustical investigations.

1. N-(2'-hydroxy-3'-nitrobenzylidene) pyridine-2-amine (B<sub>1</sub>)
2. N-(2'-hydroxy-3'-nitrobenzylidene)-3-hydroxy pyridine-2-amine (B<sub>2</sub>)
3. N-(2'-h
4. ydroxy-3'-nitrobenzylidene)-3-nitropyridine-2-amine (B<sub>3</sub>)
5. N-(2'-hydroxy-3'-nitrobenzylidene)-3-methylpyridine-2-amine (B<sub>4</sub>)

Following is the structure of the ligand –



Where,  
R = -H, -OH, -NO<sub>2</sub>, -CH<sub>3</sub>

N-(2-hydroxy-3-nitrobenzylidene)  
pyridine - 2- amine

## II. METHOD AND MATERIAL

### i.Method

N – (2- hydroxyl-3-nitrobenzylidene) pyridine -2- amine and its substituted derivatives were synthesized in the laboratory preferring microwave radiations. The density and the ultrasonic velocity measurements of the ligand solutions were done at 293, 297 and 300 K following the standard protocol.

### ii.Material

The chemicals used for synthesis were of L.R. grade. The ligands (B<sub>1</sub>-B<sub>4</sub>) were recrystallized before use. The solvent 1,4-dioxane was purified using standard procedure. All the working solutions were freshly prepared from the deionized water. To avoid any ionic contamination, deionized water was used for all the purpose in this study. The 0.01M solution of each ligand was prepared in different percentage (75%, 80%, 85%, 90%, 95% and 100%) of 1,4-dioxane-water mixture.

## III. INSTRUMENTATION

The densities of the solution were determined by standardize capillary pycnometer having a bulb of volume of about 10 cm<sup>3</sup> and capillary having an internal diameter of 1 mm. All the weighing was made on Citizen CY 104 one pan digital balance. In the present investigation, a variable path ultrasonic interferometer from Mittal enterprises, New Delhi, Model MX-3 was used to measure the ultrasonic velocity in liquid mixtures and solutions, having the working frequency of 1 MHz with accuracy of ±0.03%.

The principle used in the measurement of velocity (V) is based on the accurate determination of the wavelength (λ) in the medium.



Fig. 1: Ultrasonic Interferometer

#### IV. RESULT AND DISCUSSION

The acoustic parameters for ligands B<sub>1</sub>, B<sub>2</sub>, B<sub>3</sub> and B<sub>4</sub> in varying percentage of dioxane-water, studied at three different temperatures are calculated from the ultrasonic velocity obtained. The study of ultrasonic velocity (V), adiabatic compressibility ( $\beta_s$ ), apparent molar volume ( $\phi_v$ ) and intermolecular free length ( $L_f$ ) gives structural interaction of solvent with solute. It gives information regarding internal structure, molecular association, complex formation and stability of ligands under study. The results are given in table 1 to 12.

##### i. Adiabatic compressibility ( $\beta_s$ )

Compressibility of a liquid is one of the important physical quantities in fluid mechanics. It depends on the structure of the liquid which considers the geometrical fit such as shape, size, branching and presence of aromatic ring, etc of the solute into the ordered form of the aqueous solvent surrounding the solutes [33]. Adiabatic compressibility plays important role in the study of solute-solvent interactions by explaining the simple association or closed packing or clinging of molecules.

From table 1-12 and fig. 2 to 4, it is observed that the value of adiabatic compressibility of ligands mostly increases with increase in percentage of organic solvent. With higher concentration of solvent, the solvent molecules aggregate around the ions as the number of free ions decreases, showing the occurrence of ionic association due to strong ion-ion interaction [34]. It may also be due to departure of solvent molecules around the ions.

Also increase in temperature shows very minor change in the  $\beta_s$  values of all ligands. It is seen that the trend in  $\beta_s$  values is affected due to the presence of electron withdrawing  $-\text{NO}_2$  group and electron releasing group ( $-\text{CH}_3$ ) on the another benzene nucleus in ligands.

##### ii. Apparent molar volume ( $\phi_v$ )

Apparent molar volume is related to the density, molarity of the solution and the molecular weight of the solute. It is the thermodynamic property of solutions, which express the solute-solvent interactions. Table 1 - 12 and Fig. 5-7 shows that, the values of apparent molar volume ( $\phi_v$ ) decreases with increase in the percentage of 1,4-dioxane at 293 K, 297 K and 300 K. The change in  $\phi_v$  with variation in concentration depends on the concentration of the salt [35], densities of solution and solvent, molecular weight of solute and molarity of solution.

##### iii. Intermolecular free length ( $L_f$ )

Intermolecular free length ( $L_f$ ) is the distance between the surfaces of the molecules. From Table 1-12 and Fig. 7-9, it can be noted that the  $L_f$  values are positive for all ligands in dioxane at different temperatures indicating existence of dispersive forces between molecules of mixture.

$L_f$  increases linearly with increase in concentration of organic solvent suggesting showing significant ion-solvent interaction. It is also observed that as the temperature increases  $L_f$  values are also increases, though the trend is not prominent. This leads to less ordered structure and hence increases in intermolecular free length, suggesting weak interactions at higher temperatures.

**Table 1: Acoustic Parameters at different percentages of dioxane-water mixture.**

System: Ligand – B<sub>1</sub>

Temp. = 293 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m <sup>-3</sup> )	$V \times 10^3$ (m sec <sup>-1</sup> )	$\beta_s \times 10^{-10}$ (pa <sup>-1</sup> )	$\phi_v \times 10^{-3}$ (m <sup>3</sup> mol <sup>-1</sup> )	$L_f \times 10^{-1}$ (m <sup>-1</sup> )
75	1.00093	1.7592	0.3228	610.73	351.13
80	1.00032	1.7642	0.3212	411.53	350.24
85	0.99927	1.8002	0.3088	422.12	343.42
90	0.99868	1.8018	0.3084	229.28	343.22
95	0.99743	1.8118	0.3054	189.32	341.54
100	0.99711	1.9582	0.2615	213.52	316.05

**Table 2: Acoustic Parameters at different percentages of dioxane-water mixture**

System: Ligand – B<sub>2</sub>

Temp. = 293 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m <sup>-3</sup> )	$V \times 10^3$ (m sec <sup>-1</sup> )	$\beta_s \times 10^{-10}$ (pa <sup>-1</sup> )	$\phi_v \times 10^{-3}$ (m <sup>3</sup> mol <sup>-1</sup> )	$L_f \times 10^{-1}$ (m <sup>-1</sup> )
75	1.00179	2.1388	0.2182	540.72	288.69
80	1.00157	2.1496	0.2161	302.44	287.27
85	1.00116	2.0262	0.2433	248.72	304.83
90	0.99884	2.026	0.2439	229.22	305.21
95	0.99698	2.0022	0.2502	250.73	309.13
100	0.99667	2.0064	0.2492	273.96	308.53

**Table 3: Acoustic Parameters at different percentages of dioxane-water mixture**

System: Ligand – B<sub>3</sub>

Temp. = 293 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m <sup>-3</sup> )	$V \times 10^3$ (m sec <sup>-1</sup> )	$\beta_s \times 10^{-10}$ (pa <sup>-1</sup> )	$\phi_v \times 10^{-3}$ (m <sup>3</sup> mol <sup>-1</sup> )	$L_f \times 10^{-1}$ (m <sup>-1</sup> )
75	1.00108	1.86	0.2887	640.67	332.08
80	1.00061	1.6162	0.3826	427.46	382.26
85	0.9999	1.6304	0.3762	403.92	379.07
90	0.99769	1.7726	0.3190	373.99	349.04
95	0.99762	1.774	0.3185	215.28	348.78
100	0.99653	1.8176	0.3037	317.19	340.60

*Table 4: Acoustic Parameters at different percentages of dioxane-water mixture*  
System: Ligand – B<sub>4</sub> Temp. = 293 K

% 1,4-Dioxane	$d_s \times 10^3$ ( $\text{kg m}^{-3}$ )	$V \times 10^3$ (m $\text{sec}^{-1}$ )	$\beta_s \times 10^{-10}$ ( $\text{pa}^{-1}$ )	$\phi_v \times 10^{-3}$ ( $\text{m}^3$ $\text{mol}^{-1}$ )	$L_f \times 10^{-1}$ ( $\text{m}^{-1}$ )
75	0.99913	2.131	0.2204	805.17	290.13
80	0.99899	2.0046	0.2491	558.96	308.45
85	0.99874	1.9882	0.2533	489.37	311.03
90	0.99688	1.9686	0.2588	424.57	314.42
95	0.99616	1.8574	0.2910	331.50	333.36
100	0.99554	1.9858	0.2547	386.13	311.91

*Table 5: Acoustic Parameters at different percentages of dioxane-water mixture*  
System: Ligand – B<sub>1</sub> Temp. = 297 K

% 1,4-Dioxane	$d_s \times 10^3$ ( $\text{kg m}^{-3}$ )	$V \times 10^3$ (m $\text{sec}^{-1}$ )	$\beta_s \times 10^{-10}$ ( $\text{pa}^{-1}$ )	$\phi_v \times 10^{-3}$ ( $\text{m}^3$ $\text{mol}^{-1}$ )	$L_f \times 10^{-1}$ ( $\text{m}^{-1}$ )
75	1.0021	1.7748	0.3168	493.80	351.22
80	1.0002	1.7564	0.3241	424.55	355.24
85	0.9987	1.6254	0.3790	483.39	384.16
90	0.9974	1.6030	0.3902	355.07	389.77
95	0.9960	1.8166	0.3042	331.59	344.18
100	0.9950	1.7762	0.3186	429.75	352.20

*Table 6: Acoustic Parameters at different percentages of dioxane-water mixture*  
System: Ligand – B<sub>2</sub> Temp. = 297 K

% 1,4-Dioxane	$d_s \times 10^3$ ( $\text{kg m}^{-3}$ )	$V \times 10^3$ (m $\text{sec}^{-1}$ )	$\beta_s \times 10^{-10}$ ( $\text{pa}^{-1}$ )	$\phi_v \times 10^{-3}$ ( $\text{m}^3$ $\text{mol}^{-1}$ )	$L_f \times 10^{-1}$ ( $\text{m}^{-1}$ )
75	1.0009	1.8206	0.3014	626.71	342.58
80	0.9993	1.9800	0.2553	529.83	315.26
85	0.9981	1.8012	0.3088	556.75	346.77
90	0.9973	1.8152	0.3043	380.19	344.22
95	0.9968	1.9786	0.2563	271.92	315.89
100	0.9956	1.8600	0.2903	385.10	336.23

*Table 7: Acoustic Parameters at different percentages of dioxane-water mixture*  
System: Ligand – B<sub>3</sub> Temp. = 297 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m <sup>-3</sup> )	$V \times 10^3$ (m sec <sup>-1</sup> )	$\beta_s \times 10^{-10}$ (pa <sup>-1</sup> )	$\phi_v \times 10^{-3}$ (m <sup>3</sup> mol <sup>-1</sup> )	$L_f \times 10^{-1}$ (m <sup>-1</sup> )
75	1.0015	1.8088	0.3052	602.66	344.73
80	0.9993	1.7984	0.3094	563.87	347.11
85	0.9984	1.8506	0.2925	550.58	337.45
90	0.9965	1.9514	0.2635	498.07	320.34
95	0.9957	1.8122	0.3058	414.19	345.08
100	0.9951	1.9558	0.2627	459.78	319.83

*Table 8: Acoustic Parameters at different percentages of dioxane-water mixture.*  
System: Ligand – B<sub>4</sub> Temp. = 297 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m <sup>-3</sup> )	$V \times 10^3$ (m sec <sup>-1</sup> )	$\beta_s \times 10^{-10}$ (pa <sup>-1</sup> )	$\phi_v \times 10^{-3}$ (m <sup>3</sup> mol <sup>-1</sup> )	$L_f \times 10^{-1}$ (m <sup>-1</sup> )
75	1.0004	1.9652	0.2588	676.78	317.46
80	0.9993	1.8548	0.2909	529.83	336.55
85	0.9981	1.9842	0.2545	554.75	314.79
90	0.9974	1.7736	0.3187	370.12	352.28
95	0.9964	1.7880	0.3139	312.31	349.63
100	0.9951	1.8152	0.3050	427.62	344.60

*Table 9: Acoustic Parameters at different percentages of dioxane-water mixture*  
System: Ligand – B<sub>1</sub> Temp. = 300 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m <sup>-3</sup> )	$V \times 10^3$ (m sec <sup>-1</sup> )	$\beta_s \times 10^{-10}$ (pa <sup>-1</sup> )	$\phi_v \times 10^{-3}$ (m <sup>3</sup> mol <sup>-1</sup> )	$L_f \times 10^{-1}$ (m <sup>-1</sup> )
75	0.9991	1.5922	0.3948	799.19	394.61
80	0.9981	1.7858	0.3142	630.40	351.99
85	0.9974	1.7800	0.3165	615.24	353.28
90	0.9970	1.6412	0.3724	400.44	383.23
95	0.9965	1.7842	0.3152	279.07	352.59
100	0.9961	1.7972	0.3108	315.46	350.12

*Table 10: Acoustic Parameters at different percentages of dioxane-water mixture*  
System: Ligand – B<sub>2</sub> Temp. = 300 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m <sup>-3</sup> )	$V \times 10^3$ (m sec <sup>-1</sup> )	$\beta_s \times 10^{-10}$ (pa <sup>-1</sup> )	$\phi_v \times 10^{-3}$ (m <sup>3</sup> mol <sup>-1</sup> )	$L_f \times 10^{-1}$ (m <sup>-1</sup> )
75	0.9986	2.0264	0.2439	865.45	310.13
80	0.9984	1.9628	0.2600	625.30	320.22
85	0.9971	1.8110	0.3058	653.46	347.27
90	0.9969	1.9810	0.2556	424.56	317.50
95	0.9963	1.9996	0.2510	315.33	314.64
100	0.9959	1.8270	0.3008	354.77	344.44

*Table 11: Acoustic Parameters at different percentages of dioxane-water mixture*  
System: Ligand – B<sub>3</sub> Temp. = 300 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m <sup>-3</sup> )	$V \times 10^3$ (m sec <sup>-1</sup> )	$\beta_s \times 10^{-10}$ (pa <sup>-1</sup> )	$\phi_v \times 10^{-3}$ (m <sup>3</sup> mol <sup>-1</sup> )	$L_f \times 10^{-1}$ (m <sup>-1</sup> )
75	0.9993	2.0044	0.2491	824.14	313.43
80	0.9986	2.0122	0.2473	631.21	312.32
85	0.9983	2.0306	0.2429	561.65	309.53
90	0.9973	2.1490	0.2171	418.34	292.63
95	0.9968	2.0404	0.2410	296.97	308.27
100	0.9963	2.0048	0.2497	336.38	313.82

*Table 12: Acoustic Parameters at different percentages of dioxane-water mixture*  
System: Ligand – B<sub>4</sub> Temp. = 300 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m <sup>-3</sup> )	$V \times 10^3$ (m sec <sup>-1</sup> )	$\beta_s \times 10^{-10}$ (pa <sup>-1</sup> )	$\phi_v \times 10^{-3}$ (m <sup>3</sup> mol <sup>-1</sup> )	$L_f \times 10^{-1}$ (m <sup>-1</sup> )
75	0.9989	1.5794	0.4013	830.28	397.84
80	0.9983	1.7576	0.3243	628.32	357.61
85	0.9971	1.7298	0.3352	655.49	363.58
90	0.9966	1.6326	0.3765	450.80	385.31
95	0.9963	1.7982	0.3104	321.40	349.89
100	0.9958	1.7740	0.3191	360.85	354.75

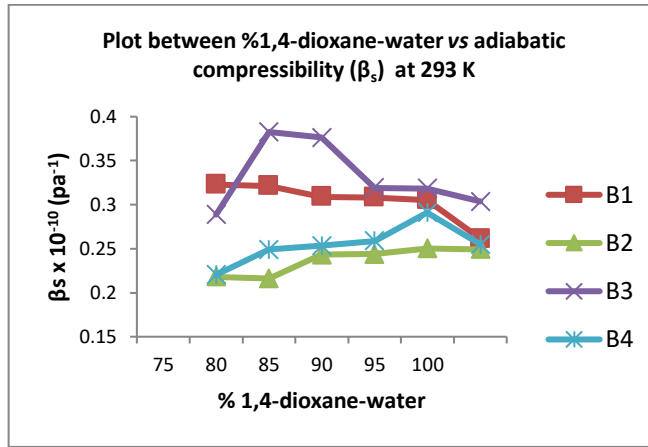


Fig. 2

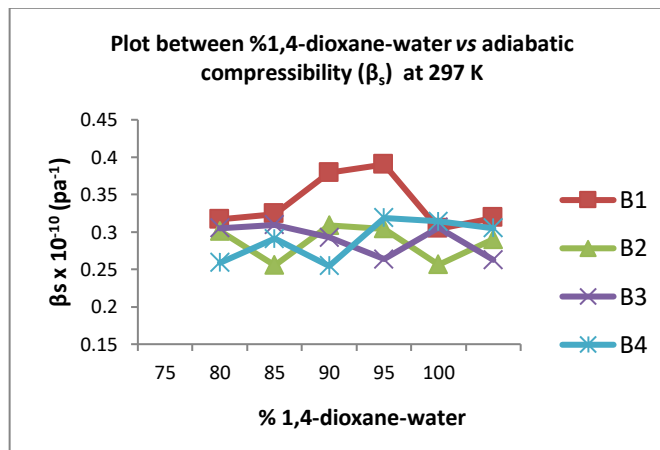


Fig. 3

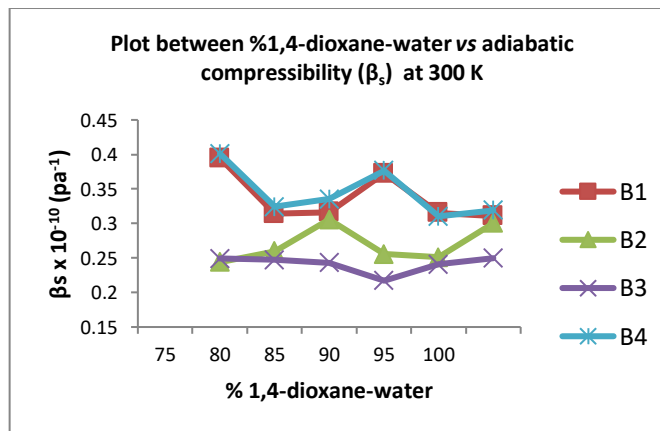


Fig. 4



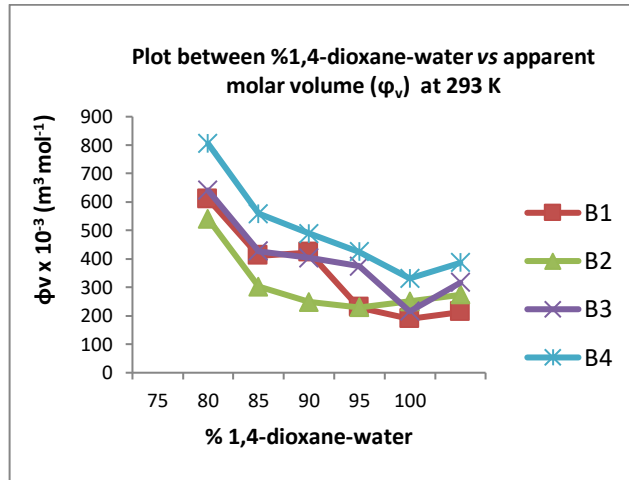


Fig. 5

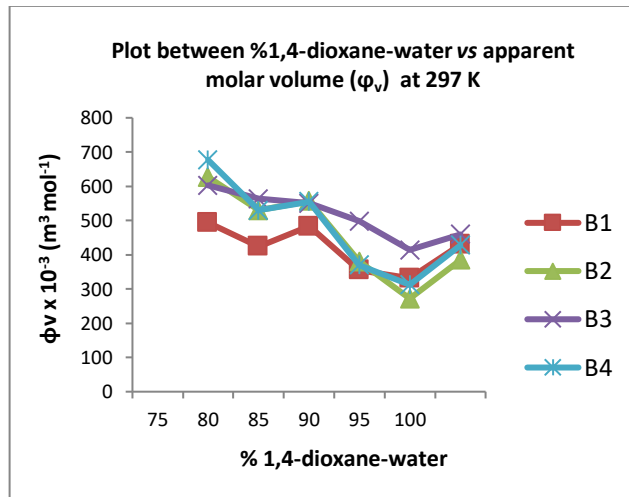


Fig. 6

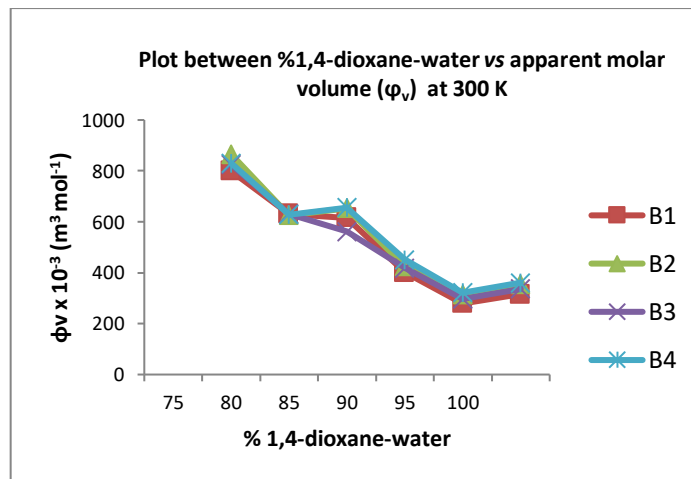


Fig. 7

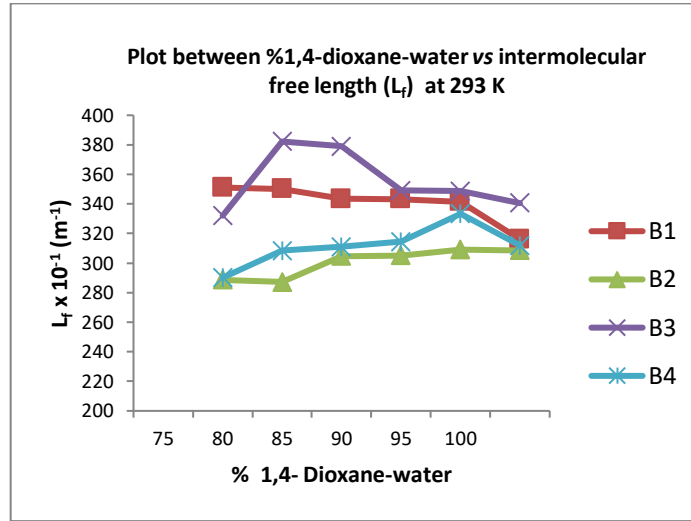


Fig. 8

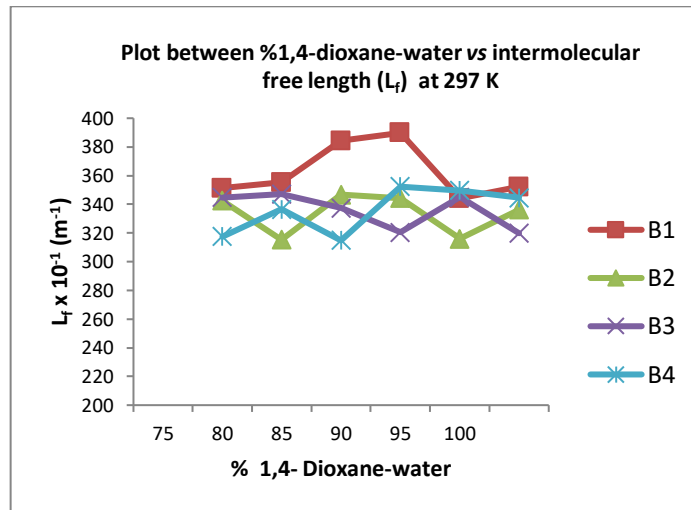


Fig. 9

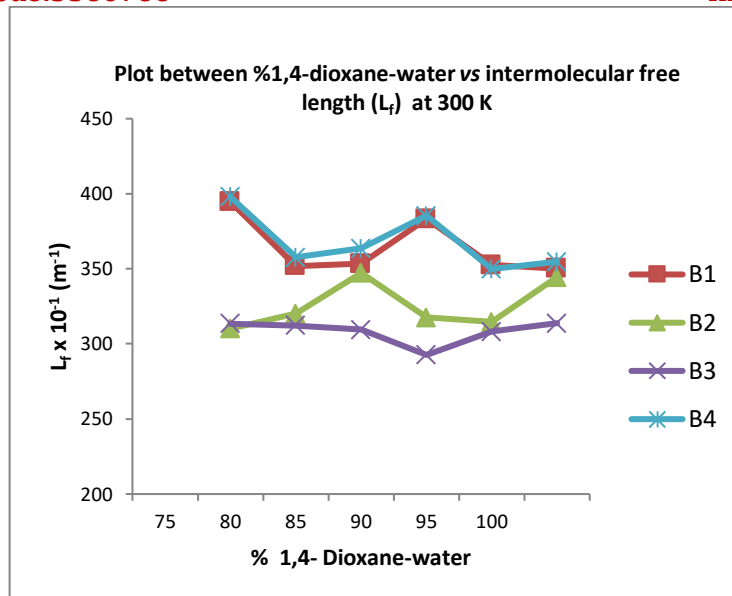


Fig. 10

## V. CONCLUSION

From the present study it can be concluded that lower  $\beta_s$  values shows the increased association of molecules, expressing the solute-solvent interactions. Positive  $\phi_v$  values in dioxane shows existence of strong ion-solute interaction. When polar solute is added in dioxane, there is compact packing of molecules and free space ( $L_f$ ) decreases. Thus we can say that measurement of ultrasonic velocity can be used to investigate solute-solvent, solute-solute and ion-solvent interactions and acoustic properties explains how these interactions occur and responsible for breaking and making of the structure in the solution. With this knowledge, one can easily find out the stability of the ligands in the solution.

## VI. ACKNOWLEDGEMENT

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