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Acoustical Investigation of N–(2-hydroxybenzylidene)-3-Substituted Pyridine -2- Amine Schiff bases by Ultrasonic Velocity Measurements in Dioxane at Different Temperatures

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ABSTRACT

The present work deals with the study of acoustic parameters like ultrasonic velocity (V), adiabatic compressibility (β_s), apparent molar volume (ϕ_v) and intermolecular free length (L_f) by ultrasonic interferometric measurements which reflects structural interaction of water molecules and organic solvent molecules with substituted Schiff bases. The study of N–(2-hydroxybenzylidene)-3-substituted pyridine -2- amine Schiff bases were carried in different percentage of 1,4-dioxane-water medium, at 293, 297 and 300 K. The densities and velocities thus obtained were used to evaluate acoustic parameters for all the ligands.

Keywords: Acoustic, structural interaction, substituted Schiff bases, densities, velocities.

1. Introduction

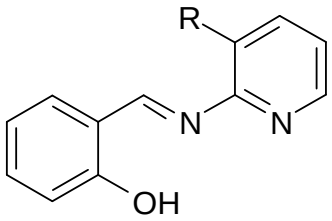
Ultrasounds are sound waves with frequencies higher than the upper audible limit of human hearing. Ultrasound is used in many different fields like ultrasonic devices are used to detect objects and measure distances. Ultrasound imaging is often used in quality control and medicine [1-2]. In the nondestructive testing of products and structures, ultrasound is used to detect invisible flaws. Industrially, ultrasound is used for cleaning, mixing, and to accelerate chemical processes. Scientists are also studying ultrasound using graphene diaphragms as a method of communication [3].

The unique feature of sound wave property is that it gives direct and precise information of adiabatic properties of solution. The anomalous behavior of non-electrolytic solutions and the properties of liquid systems, which are otherwise not easily deduced directly, can be determined from speed of sound and density data in conjunction with equation of state. Ultrasonic technology has been adequately employed to investigate the properties of any substance to understand the nature of molecular interactions in pure liquid [4], liquid mixtures [5-7] and ionic interactions in electrolytic solutions [8]. As ultrasonic technique can reveal very weak intermolecular interactions due to its useful wavelength range, 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 [9-18].

Schiff base have high synthesis flexibility, varied coordinating ability and medicinal utility and are an important class of ligands in the field of co-ordination chemistry [19]. A large number of Schiff bases have been found to possess important biological and catalytic activities [20-21] and their acoustical parameters were reported [22-24]. Hence 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’-hydroxybenzylidene) pyridine-2-amine (A_1)
2. N-(2’-hydroxybenzylidene)-3-hydroxy pyridine-2-amine (A_2)
3. N-(2’-hydroxybenzylidene)-3-nitropyridine-2-amine (A_3)
4. N-(2’-hydroxybenzylidene)-3-methylpyridine-2-amine (A_4)

Following is the structure of the ligand –



Where,
R = -H, -OH, $-NO_2$, $-CH_3$

N - (2 - hydroxybenzylidene)
pyridine - 2 - amine

2. Experimental and Instrumentation

2.1 Experimental

The chemicals used for synthesis were of L.R. grade. The ligands (A_1-A_4) 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. The density and the ultrasonic velocity measurements of the ligand solutions were done at 293, 297 and 300 K following the standard protocol.

2.2 Instrumentation

The densities of the solution were determined by standardize capillary pyknometer having a bulb of volume of about 10 cm^3 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%.

3. Result and Discussion

The study of ultrasonic velocity (V), adiabatic compressibility (β_s), apparent molar volume (ϕ_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 acoustic parameters for ligands A_1, A_2, A_3 and A_4 in varying percentage of dioxane-water, studied at three different temperatures are calculated from the ultrasonic velocity obtained. The results are given in table 1 to12.

3.1 Adiabatic compressibility (β_s)

Adiabatic compressibility plays important role in the study of solute-solvent interactions by explaining the simple association or closed packing or clinging of molecules. It depends on the structure of the liquid which considers the geometrical fit of the solute into the ordered form of the aqueous solvent surrounding the solutes [25].

From table 1-12 and fig. 1 to 3, 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 [26]. Also it can be noted that with increase in temperature, there is slight increase in the □$_s$ values of all ligands. Also presence of the electron withdrawing $–NO_2$ group in ligand A_3 decreases its □$_s$ values.

3.2 Apparent molar volume ($\square_v$)

Apparent molar volume is the thermodynamic property of solutions, which express the solute-solvent interactions, and is related to the density, molarity of the solution and the molecular weight of the solute. Table 1 - 12 and Fig. 4-6 shows that, the values of apparent molar volume (ϕ_v) decreases with increase in the percentage of 1,4-dioxane at 293 K, 297 K and 300 K. The change in ϕ_v with variation in concentration depends on the concentration of the salt, the nature and size of electrolyte ions and the dielectric constant [27].

3.3 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. The positive value of excess free length shows the existence of dispersive forces between molecules of mixture.

L_f increases linearly with increase in concentration of organic solvent suggesting structure promoting behavior of the added solute, as there is 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. Increase in temperature leads to less ordered structure and volume expansion and hence in intermolecular free length [28]. It reveals that interaction become weaker at higher temperatures.

4. Conclusion

From the present study it can be concluded that $\square_s$ and L_f values mostly increases with increase in percentage of organic solvent while the values of ϕ_v decreases at 293, 297 and 300 K. Thus we can say that these acoustic properties helps in explaining how solute-solvent, solute-solute and ion-solvent interactions occur and are responsible for breaking and making of the structure in the solution.

Acknowledgement

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(Tables & Figures)

Table 1: Acoustic Parameters at different percentages of dioxane-water mixture.
System: Ligand - A_1 Temp. = 293 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	0.99882	2.014	0.2468	777.24	307.03
80	0.99859	1.763	0.3222	540.08	350.79
85	0.99855	1.674	0.3574	449.39	369.44
90	0.99703	1.7982	0.3102	350.26	344.19
95	0.99562	1.739	0.3321	326.83	356.16
100	0.99547	1.6566	0.3660	333.94	373.90

Table 2: Acoustic Parameters at different percentages of dioxane-water mixture.
System: Ligand – A_2 Temp. = 293 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	1.00168	2.0528	0.2369	506.79	300.80
80	1.0007	1.9326	0.2676	344.50	319.66
85	1.00052	1.8012	0.3081	267.80	343.02
90	0.99907	1.788	0.3131	161.07	345.80
95	0.99714	1.8176	0.3036	189.46	340.50
100	0.99599	1.7808	0.3166	297.46	347.73

Table 3: Acoustic Parameters at different percentages of dioxane-water mixture.
System: Ligand – A_3 Temp. = 293 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	0.99903	1.9814	0.2550	801.20	312.05
80	0.99878	1.794	0.3111	566.04	344.69
85	0.99863	1.7886	0.3130	486.41	345.76
90	0.99744	1.824	0.3013	354.07	339.25
95	0.99577	1.785	0.3152	356.86	346.95
100	0.99535	1.9552	0.2628	391.29	316.82

Table 4: Acoustic Parameters at different percentages of dioxane-water mixture.

System: Ligand – A_4 Temp. = 293 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	1.00086	1.8872	0.2805	586.76	327.33
80	0.9993	2.1456	0.2174	482.79	288.13
85	0.99795	1.9966	0.2514	523.74	309.84
90	0.99667	1.9938	0.2524	400.61	310.48
95	0.99655	1.8464	0.2943	246.96	335.28
100	0.99552	1.9472	0.2649	342.95	318.09

Table 5: Acoustic Parameters at different percentages of dioxane-water mixture.

System: Ligand – A_1 Temp. = 297 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	1.0013	2.2452	0.1981	527.78	277.74
80	0.9989	2.2288	0.2015	507.93	280.12
85	0.9986	2.1714	0.2124	443.36	287.57
90	0.9973	2.1710	0.2127	326.08	287.82
95	0.9972	2.1760	0.2118	165.36	287.16
100	0.9962	2.1804	0.2111	260.18	286.73

Table 6: Acoustic Parameters at different percentages of dioxane-water mixture.

System: Ligand – A_2 Temp. = 297 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	0.9990	1.7930	0.3114	773.17	348.19
80	0.9984	1.6592	0.3638	580.22	376.40
85	0.9974	1.7716	0.3194	577.09	352.68
90	0.9966	1.6688	0.3603	405.64	374.55
95	0.9953	1.7914	0.3131	380.32	349.16
100	0.9947	1.6326	0.3772	427.94	383.23

Table 7: Acoustic Parameters at different percentages of dioxane-water mixture.

System: Ligand – A_3 Temp. = 297 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	1.0061	2.2282	0.2002	93.12	279.19
80	1.0012	2.3592	0.1795	324.45	264.34
85	0.9990	2.2534	0.1971	445.21	277.05
90	0.9964	2.2160	0.2044	455.94	282.09
95	0.9956	1.9656	0.2600	379.11	318.17
100	0.9948	1.9972	0.2520	447.98	313.25

Table 8: Acoustic Parameters at different percentages of dioxane-water mixture.

System: Ligand – A_4 Temp. = 297 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	1.0020	1.7830	0.3139	469.85	349.62
80	1.0004	1.6828	0.3530	375.53	370.74
85	0.9982	1.7870	0.3137	495.58	349.50
90	0.9973	1.6594	0.3642	339.11	376.55
95	0.9953	1.7652	0.3224	370.22	354.33
100	0.9946	1.5474	0.4199	441.13	404.36

Table 9: Acoustic Parameters at different percentages of dioxane-water mixture.

System: Ligand – A_1 Temp. = 300 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	0.9975	1.5988	0.3922	913.01	393.29
80	0.9969	1.3500	0.5504	707.15	465.90
85	0.9959	1.5150	0.4375	718.41	415.38
90	0.9952	1.6942	0.3501	538.09	371.57
95	0.9946	1.7888	0.3142	434.09	352.03
100	0.9944	1.6196	0.3834	446.30	388.85

Table 10: Acoustic Parameters at different percentages of dioxane-water mixture.

System: Ligand – A_2 Temp. = 300 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	0.9973	1.6088	0.3874	950.21	390.89
80	0.9970	1.6032	0.3902	714.13	392.30
85	0.9965	1.5508	0.4173	674.90	405.67
90	0.9960	1.5938	0.3953	472.27	394.82
95	0.9953	1.6444	0.3716	372.22	382.80
100	0.9948	1.6406	0.3735	421.87	383.79

Table 11: Acoustic Parameters at different percentages of dioxane-water mixture.

System: Ligand – A_3 Temp. = 300 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	0.9992	1.8520	0.2918	781.11	339.22
80	0.9984	1.9984	0.2508	605.25	314.50
85	0.9976	1.8050	0.3077	586.02	348.33
90	0.9975	1.9626	0.2603	351.04	320.39
95	0.9972	1.9994	0.2508	208.47	314.53
100	0.9964	1.8240	0.3017	290.21	344.93

Table 12: Acoustic Parameters at different percentages of dioxane-water mixture.

System: Ligand – A_4 Temp. = 300 K

% 1,4-Dioxane	$d_s \times 10^3$ (kg m^{-3})	$V \times 10^3$ (m sec^{-1})	$\beta_s \times 10^{-10}$ (pa^{-1})	$\phi_v \times 10^{-3}$ (m^3 mol^{-1})	$L_f \times 10^{-1}$ (m^{-1})
75	0.9988	2.0264	0.2438	798.29	310.10
80	0.9983	1.7850	0.3144	579.23	352.11
85	0.9970	1.7768	0.3177	619.43	353.98
90	0.9962	1.6696	0.3601	444.01	376.85
95	0.9961	1.7514	0.3273	296.43	359.28
100	0.9957	1.7784	0.3176	325.76	353.89

Fig. 1

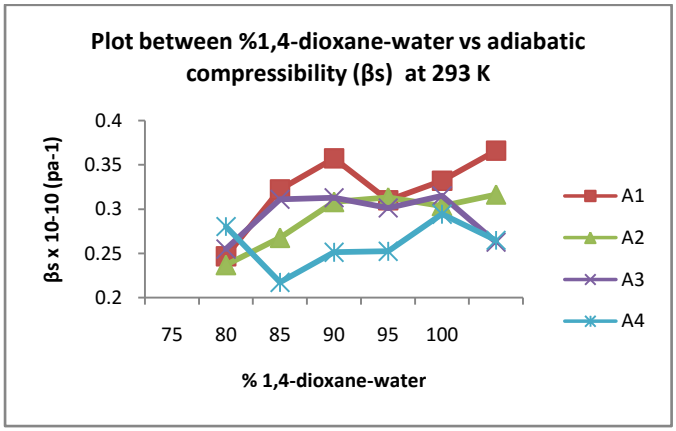


Fig. 2

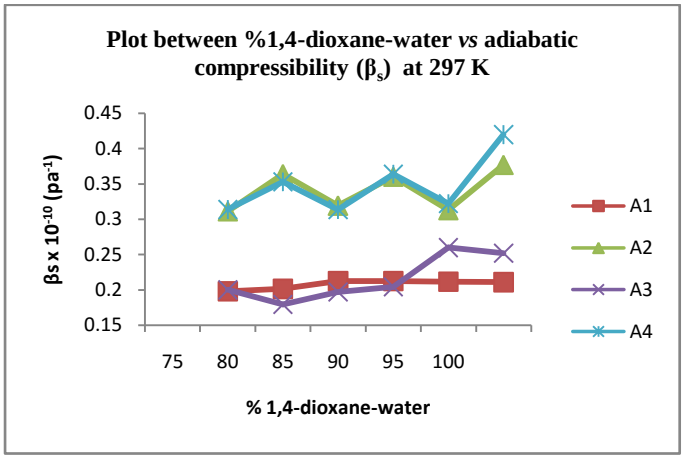


Fig. 3

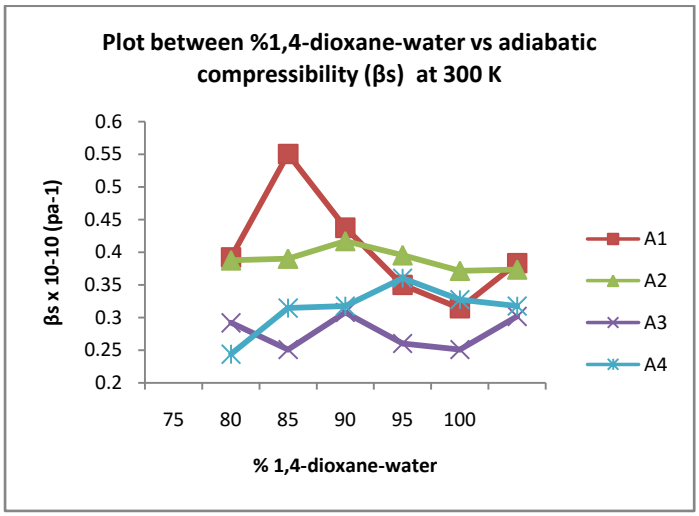


Fig. 4

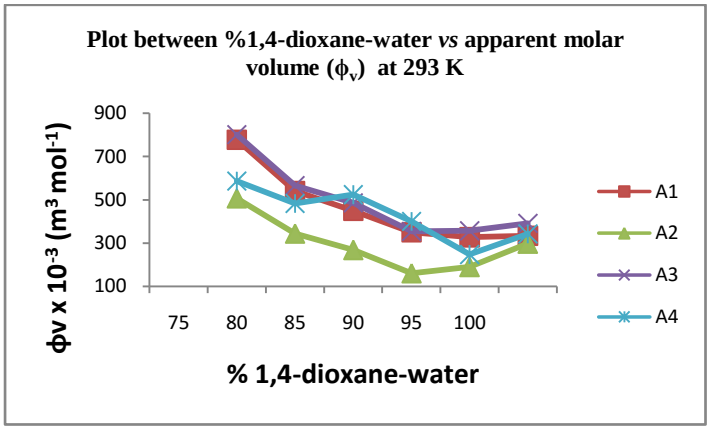
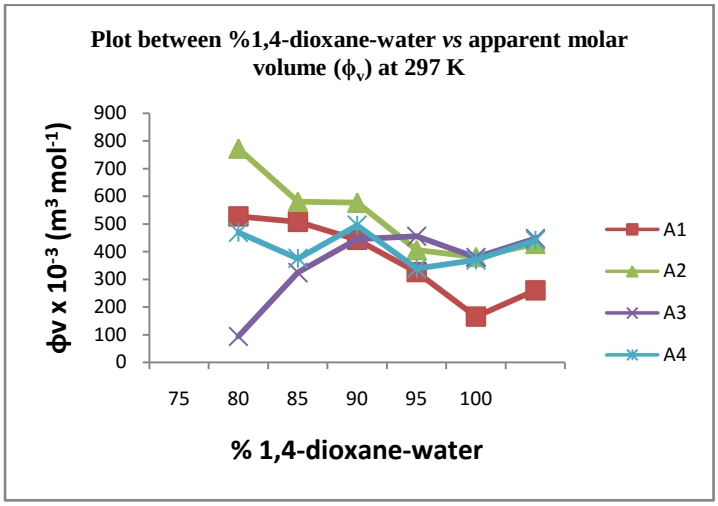


Fig. 5



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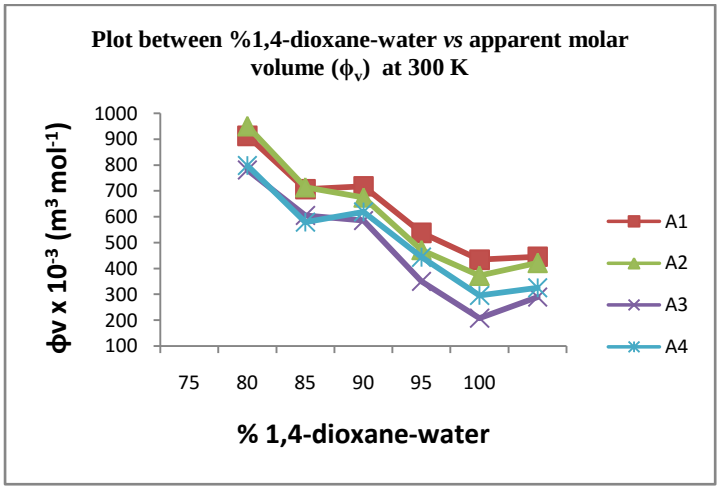

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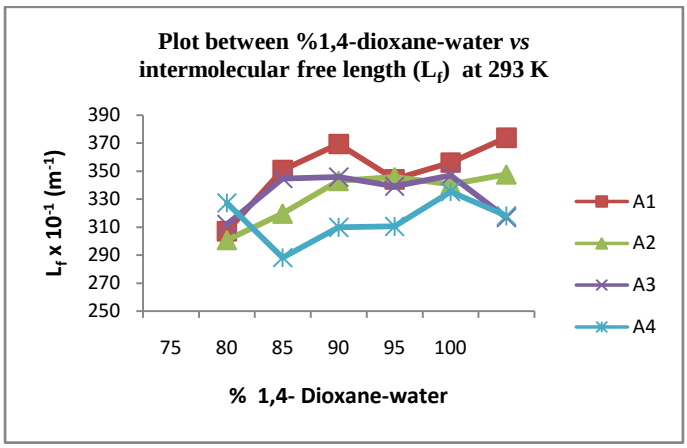

Fig. 8

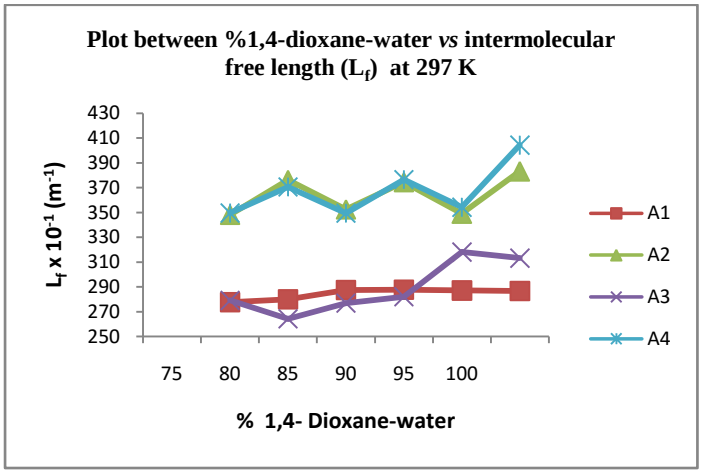

Fig. 9