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Density, viscosity, ultrasonic speed and related acoustical parameters of symmetric double schiff bases of 1, 1'-bis (4-amino phenyl)cyclohexane in 1,4-dioxane solutions at 303.15, 308.15 and 313.15 K

B. J. Gangani and P. H. Parsania*

Department of Chemistry, Saurashtra University, Rajkot, Gujarat, India

ABSTRACT

The density (ρ), viscosity (η) and ultrasonic speed (U) (2MHz) of 1, 4-dioxane solutions of symmetric double Schiff bases (SB-1 and SB-2) were determined at 303.15, 308.15 and 313.15K. Various acoustical parameters such as specific acoustical impedance (Z), isentropic compressibility (K_s), Rao's molar sound function (R_m), Vander Waals constant (b), internal pressure (π), free volume (V_f), intermolecular free path length (L_f), viscous relaxation time (τ) and classical absorption coefficient (α/f^2)_{Cl}, were determined using ρ , η and U data. The results are interpreted in terms of molecular interactions occurring in the solutions at various temperatures and concentration. Linear increase of ρ , η U , Z , R_m , b , V_f (α/f^2)_{Cl} and τ , linear decrease of K_s , L_f and π with increasing concentration of SB-1 and SB-2 supported existence of strong molecular interactions in the solutions which suggested solvophilic nature of the Schiff bases. The structure, nature and size of solute and solvent, concentration and temperature affected molecular interactions.

Keywords: Ultrasonic speed, density, viscosity, symmetric double Schiff bases, molecular interactions and solvation number

INTRODUCTION

Symmetric double Schiff bases or bis Schiff bases are synthesized by acid catalyzed condensation of aromatic or aliphatic primary diamines with aromatic aldehydes by conventional and microwave irradiation methods [1-3]. Schiff bases are most widely used as pigments, dyes, catalyst, polymer stabilizers and intermediate in organic synthesis [4]. Schiff bases are well known due to their wide range of applications such as a corrosion inhibitors [5], analytical reagents, catalysis, for synthesis of metal complexes and additives in synthetic lubricant [6-7]. Schiff bases complexes show excellent catalytical activity for various reactions at high temperature and as homogeneous and heterogeneous catalysis [8]. Schiff bases and their metal complexes have attracted a lot of interest due to their potent biological activities such as antifungal, antibacterial [9], antimicrobial [10], antitumor [11], antiproliferative [12] anticancer, anti-inflammatory, antiphlogistic, anti-HIV activities [13] and antidepressant [14].

Ultrasonic technology can be employed to produce a range of ultrafine amorphous and nano structured powdered materials for use in activating catalyst, metal catalyzed hydrogenation and catalysis [15]. Ultrasound is also effective, in organo-metallic reactions to produce rapidly under mild conditions [16], to prepare finely divided precious metal product [17], in generation of free radicals for effluent treatment [18], in polymer synthesis as radical polymerization initiator [19] and number of Diels-Alder type of cycloaddition reactions promoted by the use of

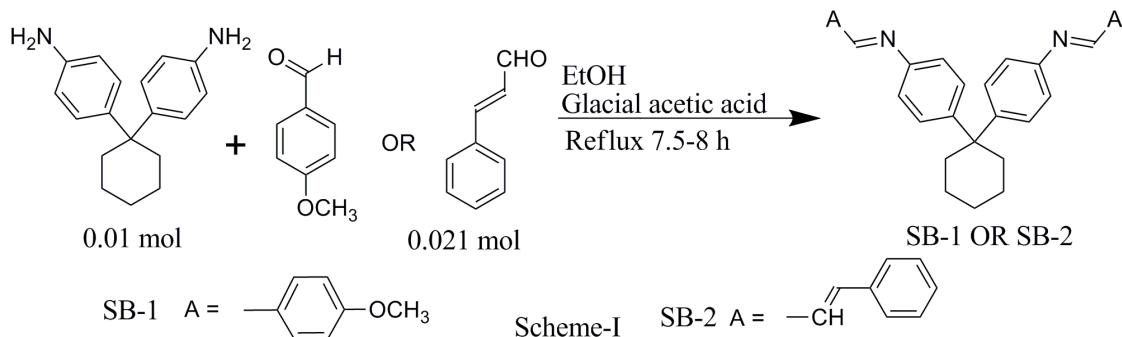
ultrasound [20]. At the present time ultrasound is most useful to study molecular interactions in solutions because close relation observed between ultrasonic velocity and chemical or structural characteristic of molecule of fluid [21-22]. Therefore by measuring density of liquid and its ultrasonic velocity many acoustical parameters can be determined [23]. Acoustical parameters provide information about molecular interactions, the nature and strength of interactions, solute-solute and solute-solvent interactions in pure solvents and its solutions [24]. Drug action is affected by various kinds of physico-chemical interactions e.g. ionic or covalent, charge-transfer, hydrogen bonding, ionic-dipole interaction, hydrophilic interaction [25], which can be understood by thermo physical and transport property [26] e.g. ultrasonic velocity, viscosity and thermal conductivity.

In continuation of ultrasonic study of symmetric double Schiff bases [23,24,27,28] in this paper we have reported determination of density, viscosity and ultrasonic speed of the solution of symmetric double Schiff bases of 1,1'-bis (4-amino phenyl)cyclohexane (Scheme-I) in 1,4-dioxane at 303.15, 308.15 and 313.15 K and at different concentrations. Various acoustical parameters have been determined and discussed to understand molecular interactions in the solutions.

MATERIALS AND METHODS

1,4-Dioxane used in the present study was supplied by Allied Chemical Corporation Vadodara, and purified according to literature method [29]. Schiff bases SB-1 and SB-2 (Scheme-I) were synthesized and crystallized according to our previous work [1]. The stock Schiff bases solutions (0.10mol) were prepared and from them a series of solutions were prepared. The solutions were stored in air tight flasks.

The density (ρ), viscosity (η) and ultrasonic speed (U) measurements on 1, 4-dioxane, SB-1 and SB-2 solutions were measured at 303.15, 308.15 and 313.15 K by using specific gravity bottle, Ubbelohde suspended level viscometer and Mittal Enterprise Interferometer (New Delhi) Model No F-81, operating at 2 MHz, respectively. The ρ , η and U measurements were accurate to $\pm 0.1 \text{ kgm}^{-3}$, 0.01 mPa s and $\pm 0.15\%$, respectively.



RESULTS AND DISCUSSION

The ρ , η and U data of 1, 4-dioxane, SB-1 and SB-2 solutions at 303.15, 308.15 and 313.15 K are reported in Table 1. From Table 1, it is evident that ρ ($\gamma = 0.986$ to 0.999), η ($\gamma = 0.990$ - 0.999) and U ($\gamma = 0.985$ - 0.997) increased linearly with increasing concentration and decreased linearly with increasing temperature confirming existence of strong molecular interactions in the solutions [30]. Increase in ρ with C proved law of additivity due to higher density of SB-1 and SB-2 as compare to DO. Increase in temperature caused decreased in density due to volume expansion decrease in number of molecules in the given volume [31]. Strong molecular interactions caused increased in η with C, while increase in temperature resulted breaking of structure formed previously [32]. Molecular interactions depend upon concentration, temperature, pressure and nature of solvent and solute and ultimately ultrasonic speed is affected.

Using ρ , η and U data of Schiff bases solutions, various acoustical parameters such as specific acoustical impedance(Z), isentropic compressibility (κ_s), Rao's molar sound function (R_m), Van der Waals constant(b), internal pressure(π), intermolecular free path length(L_f), free volume(V_f), classical absorption coefficient(a/f^2_{Cl}) and viscous relaxation time(τ) were determine according to our previous work [22-23] and are reported in Table 2.These

parameters were correlated with concentration (C) of SB-1 and SB-2 at temperature (T). The least squares equation along with co-relation coefficient of SB-1 and SB-2 are presented in Tables 3 and 4 from which it is observed that good to excellent correlation ships are observed between a given parameters and C at different temperatures.

Table-1: The density (ρ), viscosity (η) and sound velocity (U) of SB-1 and SB-2 at 303.15, 308.15, 313.15K

Cone., mol/lit	Density $\rho, \text{kg/m}^3$	Viscosity $\eta, 10^3, \text{Pa s}$	U, ms^{-1}	Density $\rho, \text{kg/m}^3$	Viscosity $\eta, 10^3, \text{Pa s}$	U, ms^{-1}
DO-SB-1 at 303.15K				DO-SB-2 at 303.15K		
0	1043	1.103	1327.4	1043	1.103	1327.4
0.01	1048	1.161	1341.6	1047	1.112	1345.6
0.02	1052	1.249	1349.4	1050	1.132	1351.4
0.04	1056	1.261	1355.2	1053	1.168	1359.2
0.06	1059	1.314	1361.2	1057	1.207	1365.6
0.08	1064	1.377	1367.4	1060	1.245	1373.2
0.10	1068	1.409	1373.6	1063	1.287	1379.6
DO-SB-1 at 308.15K				DO-SB-2 at 308.15K		
0	1039	1.044	1319.6	1039	1.042	1319.6
0.01	1042	1.109	1332.8	1044	1.054	1339.2
0.02	1045	1.154	1341.0	1046	1.074	1345.6
0.04	1054	1.210	1347.6	1049	1.113	1353.4
0.06	1056	1.259	1353.4	1053	1.147	1361.4
0.08	1062	1.316	1359.2	1057	1.187	1367.2
0.10	1066	1.350	1365.6	1060	1.232	1373.2
DO-SB-1 at 313.15K				DO-SB-2 at 313.15K		
0	1035	1.015	1291.8	1035	1.015	1291.8
0.01	1040	1.085	1325.2	1042	1.034	1321.2
0.02	1046	1.139	1331.4	1045	1.050	1333.0
0.04	1051	1.023	1339.4	1048	1.069	1340.8
0.06	1055	1.250	1345.2	1053	1.128	1351.6
0.08	1062	1.303	1351.8	1056	1.179	1359.6
0.10	1065	1.341	1357.2	1059	1.210	1365.2

From Table 3 and 4 it is observed that Z ($\gamma = 0.991$ to 0.996), R_m ($\gamma = 0.993$ to 1.000), b ($\gamma = 0.999$ to 1.000), V_f ($\gamma = 0.843$ to 0.992), τ ($\gamma = 0.953$ to 0.998), $(\alpha/f^2)_{cl}$ ($\gamma = 0.974$ to 0.997) increased linearly, while K_s ($\gamma = -0.974$ to -0.997), L_f ($\gamma = -0.985$ to -0.997) π ($\gamma = -0.983$ to -0.997) decreased linearly with concentration of Schiff bases. For SB-1 and SB-2, V_f increased linearly with both C and T confirming presence of strong molecular interactions in the solutions. The decrease of K_s and L_f with increasing C, further confirmed aggregation of solvent molecules around solute molecules resulting in decrease of intermolecular distance. With increase in T, K_s and L_f both increased and hence intermolecular distance increased [33, 34]. The linearly decreased of π with increasing concentration and temperature of SB-1 and SB-2 solution suggested decrease of cohesive forces resulted in increase of free volume. The increase in free volume is due to loose packing of the molecules inside the shield, which may be brought about by decreasing strong molecular interactions with increase in C and T [35]. Thus, free volume is a inverse function of internal pressure and hence decrease in internal pressure led to increase in free volume [36].

Both $(\alpha/f^2)_{cl}$ and τ depend upon ρ , η and U at constant temperature and pressure. Increase of $(\alpha/f^2)_{cl}$ and τ with C can be explained in terms of motion of intermolecular forces. $(\alpha/f^2)_{cl}$ and τ increased linearly with concentration for SB-1 and SB-2 and decreased up to 303.15K and increased at 308.15K. A contribution of acoustical relaxation is accounted due to entropy fluctuation associated in solution of thermodynamically formed physical entity. The competing solute-solute interaction disrupts the structure formed as a result of solvent-solute interaction and consequently entropy fluctuation increases with increasing solute-solute interaction.

Table-2: Acoustical parameters of SB-1 and SB-2 in 1, 4-dioxane solutions at 303.15, 308.15 and 313.15 K

Conc., mol/lit	303.15K	308.15K	313.15K	303.15K	308.15K	313.15K
	Z $\times 10^{-6}, \text{kg.m}^{-2}\text{s}^{-1}$	Z $\times 10^{-6}, \text{kg.m}^{-2}\text{s}^{-18}$				
0.0	1.384	1.371	1.337	1.385	1.371	1.337
0.01	1.406	1.389	1.378	1.409	1.398	1.377
0.02	1.420	1.401	1.393	1.419	1.407	1.393
0.04	1.431	1.420	1.408	1.431	1.420	1.405
0.06	1.442	1.429	1.419	1.443	1.434	1.423
0.08	1.455	1.443	1.436	1.456	1.445	1.437

0.10	1.467	1.456	1.445	1.467	1.456	1.446
	$\kappa_s \times 10^{+10} \text{ Pa}^{-1}$					
0.0	5.441	5.527	5.790	5.441	5.527	5.79
0.01	5.301	5.403	5.475	5.275	5.321	5.498
0.02	5.220	5.321	5.393	5.215	5.280	5.385
0.04	5.156	5.224	5.304	5.141	5.204	5.308
0.06	5.096	5.170	5.238	5.073	5.124	5.198
0.08	5.027	5.097	5.153	5.003	5.061	5.123
0.10	4.963	5.030	5.098	4.943	5.003	5.067
	$R_m \times 10^4, \text{ m}^{10/3} \text{ s}^{1/3} \text{ mol}^{-1}$					
0.0	9.282	9.299	9.269	9.282	9.299	9.269
0.01	9.479	9.514	9.514	9.490	9.503	9.479
0.02	9.668	9.715	9.682	9.677	9.702	9.681
0.04	10.056	10.058	10.068	10.068	9.95	10.074
0.06	10.450	10.463	10.453	10.440	10.474	10.449
0.08	10.818	10.820	10.801	10.823	10.843	10.834
0.10	11.193	11.196	11.185	11.200	11.220	11.211
	$b \times 10^6, \text{ m}^3$					
0.0	8.323	8.355	8.385	8.322	8.353	8.384
0.01	8.472	8.521	8.537	8.472	8.497	8.512
0.02	8.626	8.685	8.676	8.628	8.622	8.670
0.04	8.961	8.979	9.006	8.962	8.998	9.007
0.06	9.302	9.331	9.340	9.281	9.320	9.320
0.08	9.617	9.638	9.637	9.607	9.638	9.647
0.10	9.938	9.959	9.970	9.928	9.961	9.971
	$L_f \times 10^{11}, \text{ m}$					
0.0	4.88	4.92	5.04	4.88	4.92	5.04
0.01	4.82	4.87	4.90	4.81	4.84	4.91
0.02	4.78	4.83	4.86	4.78	4.81	4.86
0.04	4.75	479	4.82	4.75	4.78	4.82
0.06	4.73	4.76	4.79	4.72	4.74	4.77
0.08	4.69	4.73	4.75	4.68	4.71	4.74
0.10	4.66	4.70	4.73	4.65	4.68	4.71
	$\pi \times 10^8, \text{ Pa}$					
0.0	5.256	5.111	5.083	5.429	5.279	5.251
0.01	5.242	5.121	5.073	5.294	5.155	5.135
0.02	5.300	5.087	5.075	5.211	5.071	5.036
0.04	5.074	4.978	4.581	5.043	4.920	4.840
0.06	4.943	4.841	4.836	4.901	4.771	4.748
0.08	4.846	4.744	4.733	4.765	4.651	4.644
0.10	4.699	4.606	4.601	4.646	4.545	4.514
	$V_f \times 10^7, \text{ m}^3$					
0.01	1.200	1.272	1.304	1.283	1.382	1.393
0.02	1.120	1.250	1.261	1.298	1.397	1.424
0.04	1.183	1.248	1.592	1.329	1.418	1.487
0.06	1.189	1.257	1.259	1.350	1.451	1.472
0.08	1.181	1.254	1.262	1.372	1.465	1.469
0.10	1.213	1.283	1.285	1.386	1.471	1.498
	$\tau \times 10^{13}, \text{ s}$					
0.0	8.005	7.682	7.832	8.005	7.682	7.832
0.01	8.205	7.991	7.922	7.824	7.504	7.582
0.02	8.694	8.189	8.191	7.873	7.588	7.542
0.04	8.668	8.430	7.233	8.003	7.725	7.565
0.06	8.926	8.677	8.734	8.161	7.833	7.815
0.08	8.226	8.941	8.950	8.306	8.011	8.051
0.10	9.322	9.052	9.111	8.484	8.220	8.176
	$(\alpha/f^2)_{cl} \times 10^{14}, \text{ s}^2 \text{m}^{-1}$					
0.01	1.206	1.182	1.179	1.147	1.105	1.132
0.02	1.270	1.204	1.213	1.149	1.108	1.116
0.04	1.261	1.234	1.065	1.161	1.126	1.113
0.06	1293	1.264	1.280	1.178	1.135	1.140
0.08	1.330	1.297	1.306	1.193	1.155	1.168
0.10	1.338	1.307	1.324	1.213	1.180	1.181

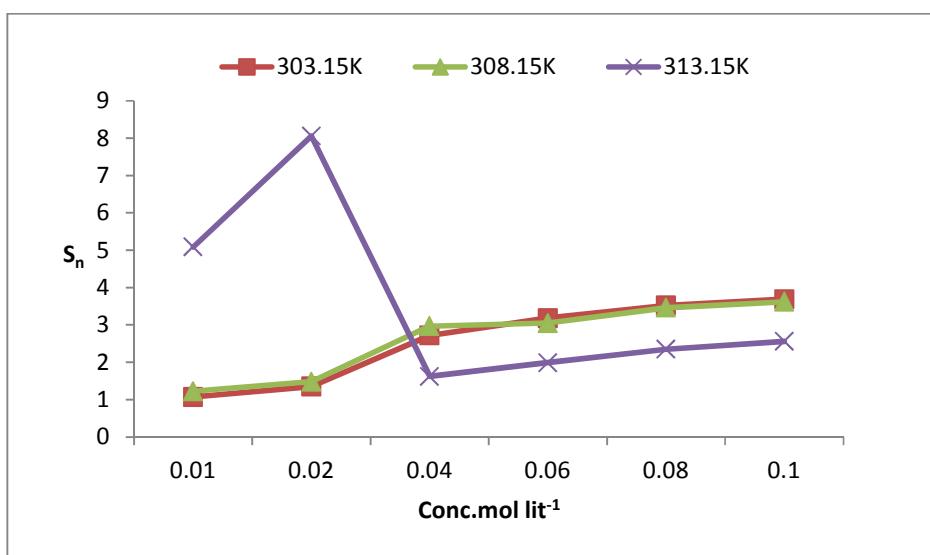
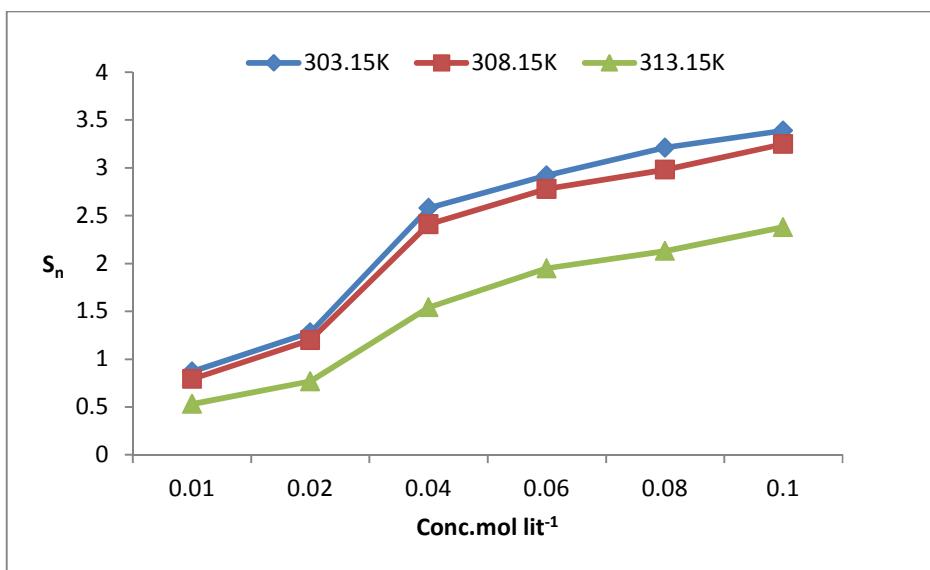
Table-3: The least squares equations and correlation coefficients for SB-1 + DO

Parameter,	Least squares equation (Correlation coefficients γ)		
	303.15K	308.15K	313.15K
ρ , kg.m ⁻³	212.33C+1046.9 (0.996)	253.97C+1041.4 (0.986)	269.32 C+1039.3 (0.989)
η , mPa s	2.7652C+1.0982 (0.995)	2.577C+ 1.0962 (0.991)	2.758 C+1.078 (0.991)
U, ms ⁻¹	334.58 C+1340.8 (0.992)	340.71C+1332.3 (0.989)	346.03C+1323.8 (0.993)
Z x 10 ⁻⁶ , kg.m ⁻² s ⁻¹	0.6414C+1.4037 (0.994)	0.7167C+1.386 (0.992)	0.723C+1.3758 (0.991)
κ_S x10 ⁺¹⁰ , Pa ⁻¹	-3.5647C+5.3127 (-0.991)	-3.9378 C+5.411 (-0.989)	-4.0649C+5.487 (-0.991)
R _m x10 ⁻⁴ , m ^{10/3} s ^{1/3} mol ⁻¹	19.094C+ 9.291 (0.999)	18.657C+ 9.3304 (0.999)	18.62C+9.3218 (0.999)
b x10 ⁵ , m ³	16.378C+8.3065 (0.999)	16.005 C+ 8.3586 (0.999)	15.97C+8.3689 (0.999)
L _f x10 ¹¹ ,m	-1.6575 C+4.824 (-0.989)	-1.7918C+4.8726 (-0.988)	-1.8384 C+4.903 (-0.987)
π x10 ⁻⁸ , Pa	- 6.54C+ 5.3534 (0.983)	-5.78C+5.1948 (0.998)	-5.4923C+5.1669 (0.991)
V _f x 10 ⁷ ,m ³	11.618C ² - 1.1879C +1.2117 (0.843)	12.929C ² -1.2574C+1.278 (0.893)	16.175C ² -1.868C +1.3101 (0.827)
τ x10 ¹³ , s	10.07C +8.343 (0.981)	11.185C +7.9867 (0.993)	12.79C +7.876 (0.971)
$(\alpha/f^2)_{cl}$.10 ¹⁴ , s ² m ⁻¹	1.255C+1.2185 (0.982)	1.335C+1.1809 (0.988)	1.31C+1.1988 (0.974)

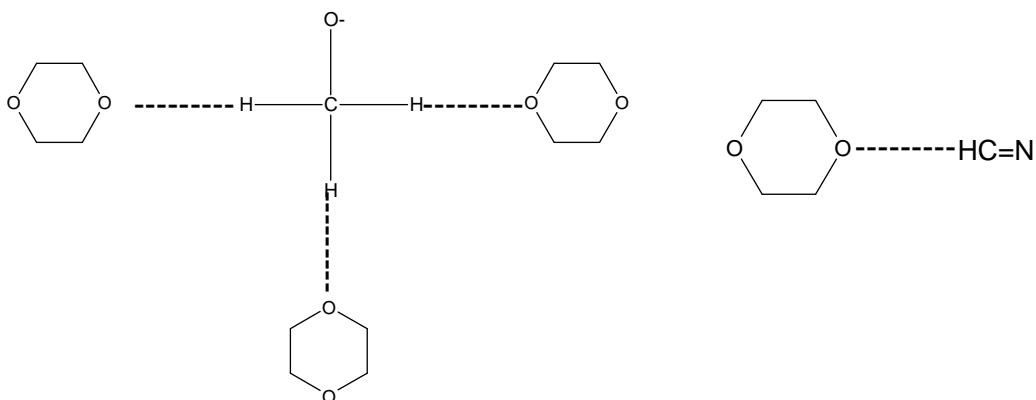
Table-4: The least squares equations and correlation coefficients for SB-2 + DO

Parameters	Least squares equation (Correlation coefficients γ)		
	303.15K	308.15K	313.15K
ρ , kg.m ⁻³	174.25C+1046 (0.996)	180 C+1042.2 (0.999)	188.42 C+1040.8 (0.995)
η , mPa s	1.9285C+1.0922 (0.999)	1.9419 C+1.0343 (0.999)	2.051 C+1.006 (0.990)
U, ms ⁻¹	369.81C+1343.3 (0.997)	370.41C+ 1337.5 (0.994)	471.0 C+1320.9 (0.985)
Z x 10 ⁻⁶ , kg.m ⁻² s ⁻¹	0.7841C+1.3899 (0.996)	0.6683C+1.3927 (0.996)	0.8232 C+1.3724 (0.993)
κ_S x10 ⁺¹⁰ , Pa ⁻¹	-3.6121C+5.295 (-0.997)	-3.7153 C+5.3701 (-0.974)	-4.6293 C+5.5023 (-0.985)
R _m x10 ⁻⁴ , m ^{10/3} s ^{1/3} mol ⁻¹	19.014C+9.301 (1)	19.32C +9.2838 (0.9938)	19.208C+ 9.2956 (0.9999)
b x10 ⁵ , m ³	16.211 C+8.3088 (1)	16.154C+8.3256 (1)	16.093 C+8.3669 (0.999)
L _f x10 ¹¹ ,m	-1.7288C+4.821 (-0.997)	-1.7425 C+4.85 (-0.996)	-2.1397 C+4.912 (-0.985)
π x10 ⁻⁸ , Pa	-7.2373 C +5.351 (-0.998)	-6.8321 C+5.205 (-0.997)	-6.4315 C+5.1568 (-0.987)
V _f x 10 ⁷ ,m ³	1.1584C+1.2765 (0.992)	1.0411C+1.3769 (0.979)	1.0315C+1.3972 (0.956)
τ x10 ¹³ , s	7.3521C +7.7286 (0.998)	7.6397 C+7.4188 (0.994)	7.4718 C+7.4026 (0.953)
$(\alpha/f^2)_{cl}$.10 ¹⁴ , s ² m ⁻¹	0.8C+1.1308 (0.997)	0.865C+1.0889 (0.988)	0.925C+1.0881 (0.961)

The plots of S_n against C for SB-1 and SB-2 in 1, 4-dioxane at different temperatures are presented in Figs.1 and 2 respectively. It is observed that S_n increased nonlinearily with C and decreased with T indicating competing solvent-solute and solute-solute interaction [36]. SB-1 has more solvation tendency than SB-2 due to different structure, nature and size of Schiff bases

Figure 1 The plots of S_n against C for SB-1 in 1,4-dioxane at 303.15, 308.15 and 313.15 KFigure 2 The plots of S_n against C for SB-2 in 1,4-dioxane at 303.15, 308.15 and 313.15 K

The lone pairs of electrons of azomethine, methoxy and 1, 4-dioxane are electronegative groups, while hydrogen of azomethine and hydrogen of methyl group are electropositive groups. The dipole-dipole interaction of opposite type led to structure formation, while of the same type led to breaking of the structure formed previously [37]. The azomethine hydrogen of SB-1 can form intermolecular hydrogen bonding with methoxy group predominant solvating tendency of SB-1 over SB-2 due to presence of methoxy group.



Various acoustical parameters and solvation number suggested existence of strong molecular interactions in the solutions. The structure of Schiff bases, their nature, temperature and concentration affected molecular interactions. SB-1 showed predominant solvating tendency in 1, 4-dioxane.

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