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## DENSITY, VISCOSITY AND SPEED OF SOUND IN SOLUTIONS OF SOME IMIDAZOLINONE DERIVATIVES IN DMSO AT 308.15 K

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### ABSTRACT

Density, ultrasonic velocity and viscosity of imidazolinone derivatives have been measured in dimethyl sulfoxide (DMSO) at 308.15 K over the wide range of concentration. From these experimental data, various acoustical parameters such as acoustic impedance ( $Z$ ) isentropic compressibility ( $\kappa_s$ ), intermolecular length ( $L_f$ ) relaxation strength ( $r$ ), Rao's molar sound function ( $R_m$ ), Vander Waal's constant ( $b$ ), molar compressibility ( $W$ ), internal pressure ( $\pi$ ), free volume ( $V_f$ ), relative association ( $R_A$ ), solvation number ( $S_n$ ), apparent molar volume ( $\Phi_v$ ), apparent molar compressibility ( $\Phi_k$ ) etc. have been evaluated to study the molecular interactions occurring in these solutions. It is observed that both solute-solute and solute-solvent interactions exist in the studied systems.

**Key Words:** Imidazolinone Derivatives, Density, Viscosity, Speed of Sound, DMSO,

### INTRODUCTION

Ultrasonic investigations find extensive applications in characterizing aspects of thermodynamics and physico chemical behavior of solutions of organic compounds (Varma *et al.*, 1991; Hakin *et al.*, 1994; Rajkotia and Parsania, 1998; Naidu and Prasad, 2005; Guler *et al.*, 2007; Kushare *et al.*, 2007). The measurement of ultrasonic speed enables to accurate determination of some useful parameters, which are highly sensitive to molecular interactions (Ogawa *et al.*, 1984; Mishra *et al.*, 1997; Meshram *et al.*, 2001; Kirnapure *et al.*, 2007) In the last few years, our investigation groups have carried out some studies on acoustical properties of organic compounds in various solvents (Baluja and Parsania, 1995; Baluja, 2002; Baluja and Oza, 2003; Baluja and Shah, 2004; Baluja *et al.*, 2005; Baluja and Shah, 2006). In continuation of these investigations, in the present study, we have selected some imidazolinone derivatives.

Literature survey shows that imidazolinone derivatives have broad biological spectrum. Many imidazolinone compounds are known to be potent herbicides (Forlani *et al.*, 1995; Ladner 2006). Some of imidazolinone derivatives are known to have anti-convulsant (Josh *et al.*, 2003), antimicrobial (Szymaoska *et al.*, 2002), CNS depressant activity (Heindel *et al.*, 2006) and antihypertensive activity (Ismail *et al.*, 2000). So, it was of our interest to study the interactions of these compounds in different solvents in different concentrations, which may be useful for their applications.

Thus, the present paper reports acoustical properties of imidazolinone derivatives in DMSO over a wide concentration range at 308.15K. The results are interpreted in terms of molecular interactions occurring in the solution.

### MATERIALS AND METHODS

#### Theory

Various acoustical parameters were calculated using following standard equations using experimental data of density, viscosity and ultrasound velocity of pure solvent and solutions.

Acoustic impedance ( $Z$ ):  $Z = \rho U$  .....(1)

Where  $\rho$  is the density and  $U$  is the ultrasonic velocity of solution.

Isentropic compressibility ( $\kappa_s$ ):  $\kappa_s = 1/ (U^2\rho)$  .....(2)

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Intermolecular length ( $L_f$ ):  $L_f = K_J \kappa_S^{1/2}$  .....(3)

Where  $K_J$  is Jacobson constant ( $= 2.0965 \times 10^{-6}$ ).

Relaxation Strength ( $r$ ):  $r = 1 - (U/U_\infty)^2$  .....(4)

Where  $U_\infty = 1600$  m/s.

Rao's molar sound function ( $R_m$ ):  $R_m = (M/\rho)U^{1/3}$  .....(5)

Where  $M$  is the molecular weight of solution.

Van der Waal's Constant ( $b$ ):  $b = (M/\rho) (1-RT/MU^2 (\sqrt{1+MU^2/3RT}-1))$ .....(6)

Where  $R$  is gas constant and  $T$  is absolute temperature.

Molar Compressibility ( $W$ ):  $W = (M/\rho) \kappa_S^{-1/7}$  .....(7)

Internal pressure ( $\pi$ ):  $\pi = P_f RT [K\eta/U]^{1/2} \rho^{2/3} / M^{7/6}$  .....(8)

Where  $P_f$  is the packing factor ( $= 2$ ) and  $K$  is a constant ( $= 4.28 \times 10^9$ ) and  $\eta$  is viscosity of solutions

Free volume ( $V_f$ ):  $V_f = [MU/K\eta]^{3/2}$  .....(9)

Relative Association ( $R_A$ ):  $R_A = \rho/\rho_0 (U_0/U)^{1/3}$  .....(10)

Where  $\rho$  and  $\rho_0$  are the densities of solutions and solvent respectively.

Solvation number ( $S_n$ ):  $S_n = M_2/M_1 [1 - \kappa_S / \kappa_{S1}] [(100 - X) / X]$  .....(11)

Where  $X$  is the number of grams of solute in 100 gm of the solution.  $M_1$  and  $M_2$  are the molecular weights and  $\kappa_{S1}$  and  $\kappa_S$  are isentropic compressibility of solvent and solute respectively.

Apparent Molar Volume ( $\phi_V$ ):  $\phi_V = [M/\rho] - [(1000\{\rho - \rho_0\}) / (\rho C)]$ .....(12)

$C$  is the concentration of the solution in molarity.

Apparent Molar Compressibility ( $\phi_k$ ):  $\phi_k = [(\rho_0 \kappa_S - \rho \kappa_{S1}) (1000/C\rho_0)] + [\kappa_{S1} M_2/\rho_0]$

..... (13)

Where  $M_2$  is the molecular weight of the solute.

### Experimental

The DMSO used in the present work were of AR grade and were purified according to the standard procedure (Riddick *et al.*, 1986). The purity of DMSO was checked by GC-MS and was found to be more than 99.97%. The compounds were recrystallized before use. The purity of compounds was checked by CHN analysis, IR, NMR and mass spectral data. The structures of all the compounds are given in Figure 1 along with their codes.

The solutions of all the synthesized compounds in DMSO were prepared in the concentration range 0.01 to 0.1M. The Mettler Toledo balance (Model No. AB 204-S) was used for the preparation of solutions.

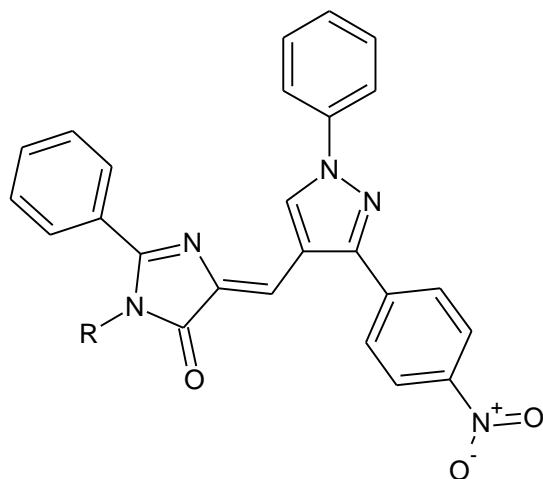
The computation of ultrasonic and thermodynamic properties requires the measurements of ultrasonic velocity, viscosity and density.

The densities ( $\rho$ ) of pure solvents and their solutions were measured by using a single capillary pycnometer, made of borosil glass having a bulb capacity of 10 ml. The ultrasonic velocity ( $U$ ) of pure solvents and their solutions were measured by using single crystal variable path ultrasonic interferometer operating at 2 MHz. The accuracy of density and velocity are  $\pm 0.0001$  g/cm<sup>3</sup> and  $\pm 0.1\%$  cm/sec respectively. The viscosity ( $\eta$ ) of pure solvents and solutions were measured by an Ubbelohde viscometer with an accuracy of 0.05%. All the measurements were carried out at 308.15 K. The temperature was maintained by digital controller NOVA thermostatic bath (Model: NV-8550-E). The uncertainty of temperature is  $\pm 0.1$  K and that of concentration is 0.0001 moles /dm<sup>3</sup>.

### RESULTS AND DISCUSSION

Table 1 shows density, viscosity and some other acoustical parameters of pure solvent and solutions of imidazolinone derivatives at 308.15K. Figure 2 shows the variation of ultrasonic velocity  $U$  with concentration. It is observed that density, viscosity and ultrasonic velocity ( $U$ ) increases with concentration for all the compounds. The ultrasonic velocity depends on intermolecular free length ( $L_f$ ). The velocity increases with decreases in  $L_f$  or vice versa (Koul *et al.*, 1985). Table 1 show that  $L_f$

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**Figure 1: General structure of Imidazolinone derivative**

Where R is:

PAIM-1: 4-CH<sub>3</sub>-C<sub>6</sub>H<sub>4</sub>-

PAIM-2: 4-OCH<sub>3</sub>-C<sub>6</sub>H<sub>4</sub>-

PAIM-3: 2-OCH<sub>3</sub>-C<sub>6</sub>H<sub>4</sub>-

PAIM-4: C<sub>10</sub>H<sub>7</sub>-

PAIM-5: 4-F-C<sub>6</sub>H<sub>4</sub>-

PAIM-6: 4-Cl-C<sub>6</sub>H<sub>4</sub>-

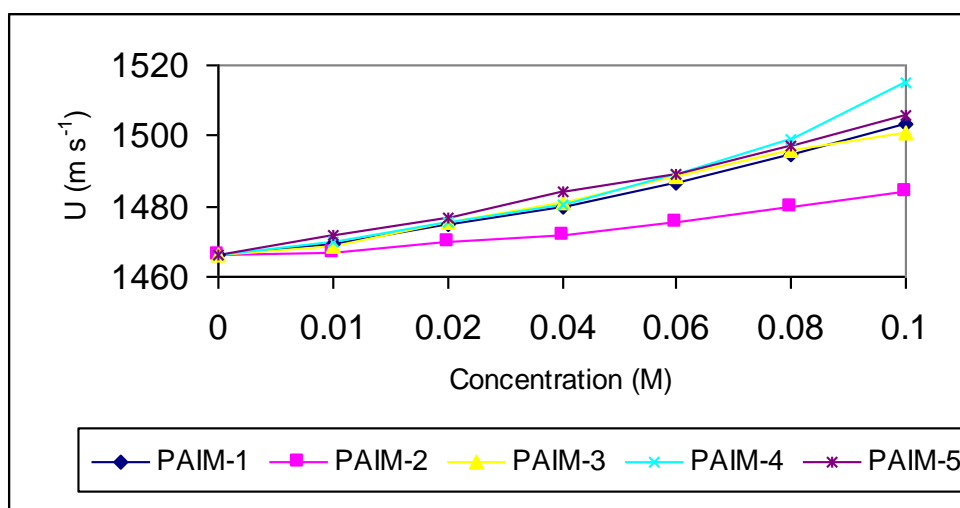
PAIM-7: C<sub>6</sub>H<sub>5</sub>-

PAIM-8: 3-Cl-4-F-C<sub>6</sub>H<sub>3</sub>-

PAIM-9: 2,5-di-Cl-C<sub>6</sub>H<sub>3</sub>-

decreases continuously which suggests that there is strong interaction between DMSO and compound molecules (Abraham *et al.*, 2000).

This is further supported by isentropic compressibility ( $\kappa_s$ ), which are also observed to decrease with concentration for all the compounds (as shown in Figure 3).



**Figure 2: Variation of ultrasonic velocity (U) with concentration of imidazolinone derivatives in DMSO at 308.15K.**

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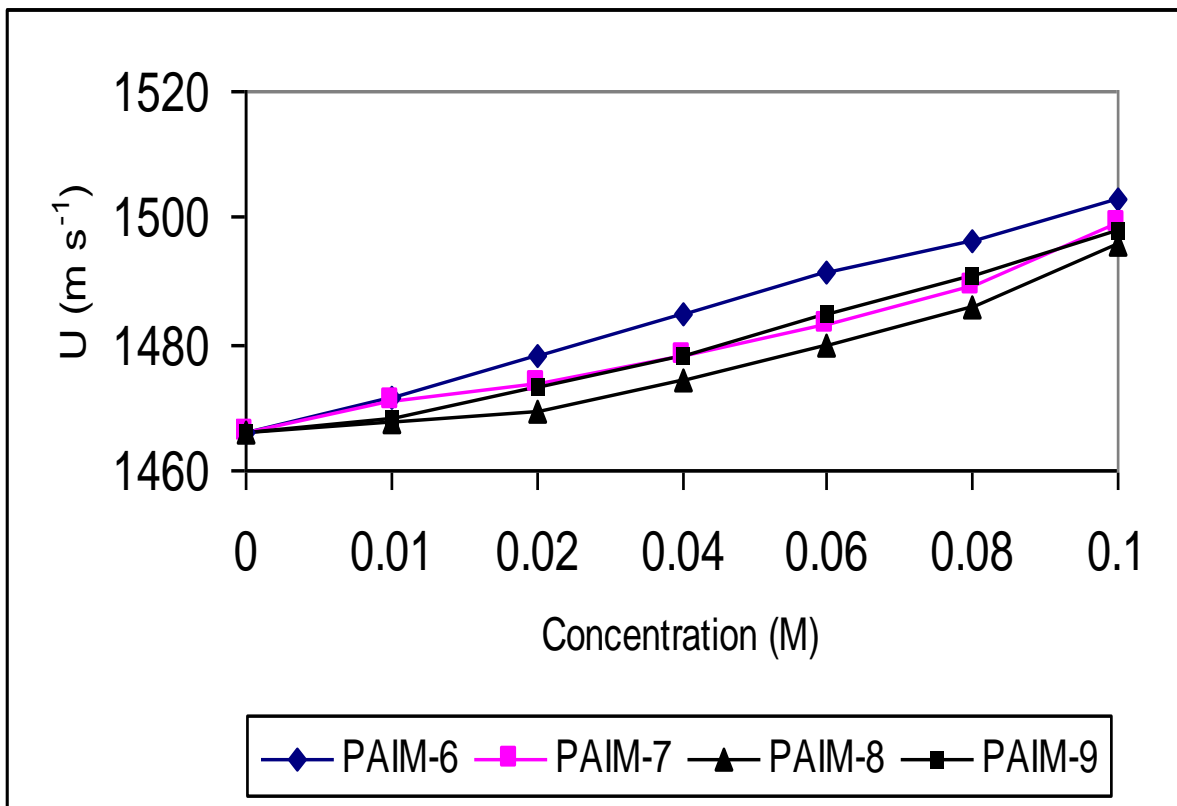
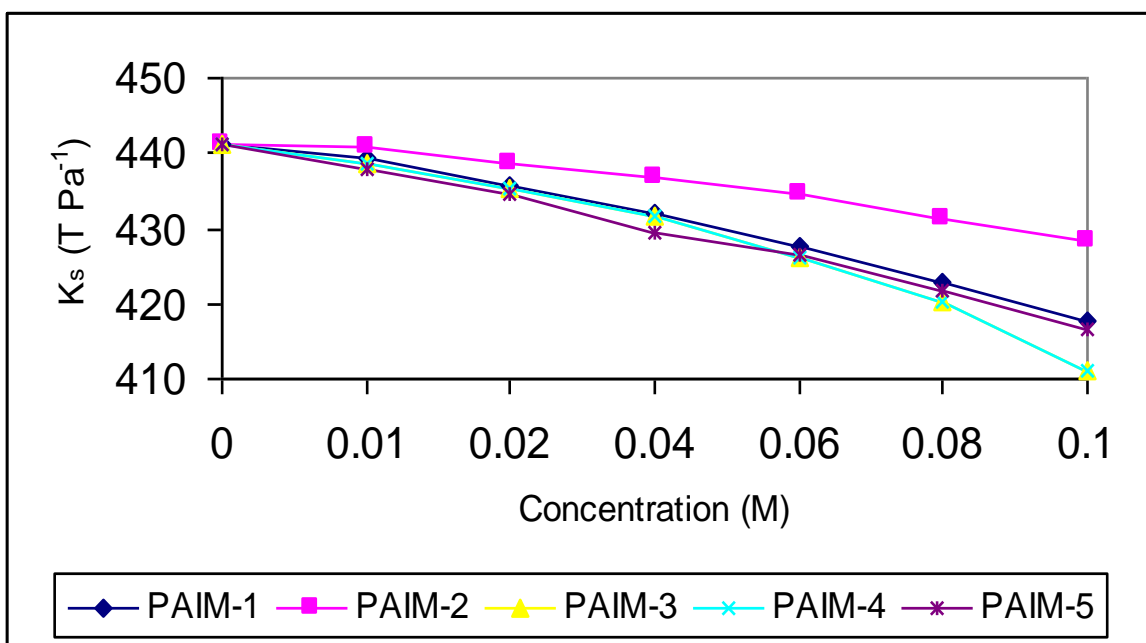


Figure 3: Variation of isentropic compressibility ( $\kappa_s$ ) with concentration of imidazolinone derivatives in DMSO at 308.15K.



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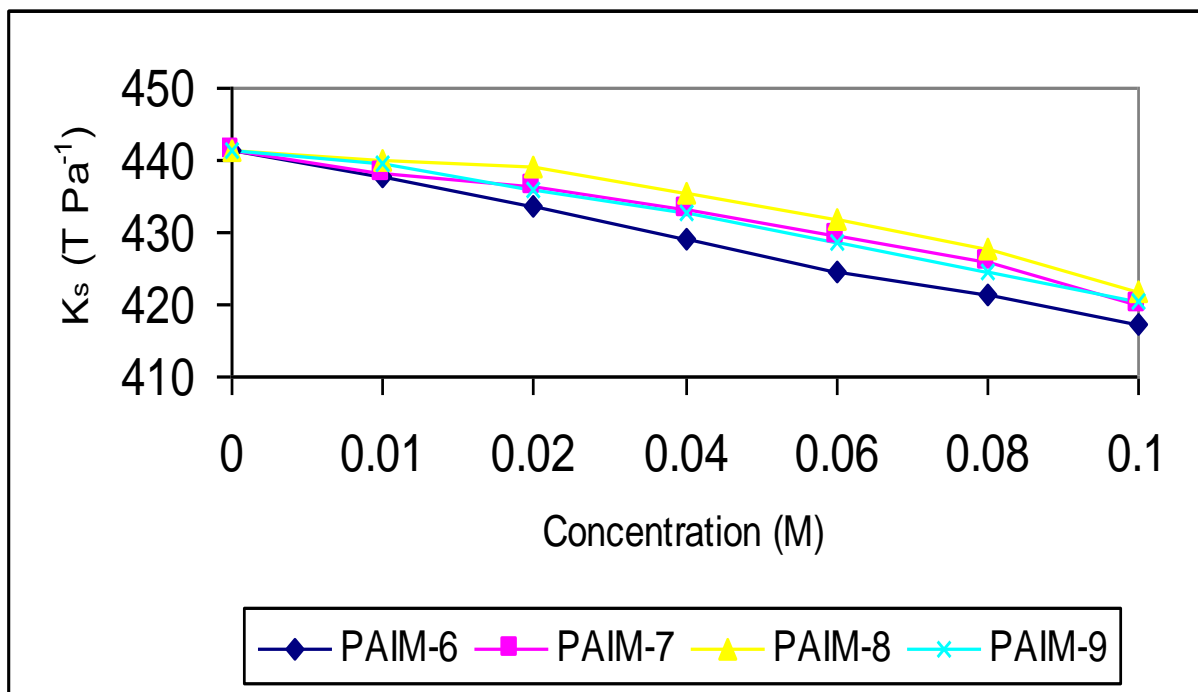
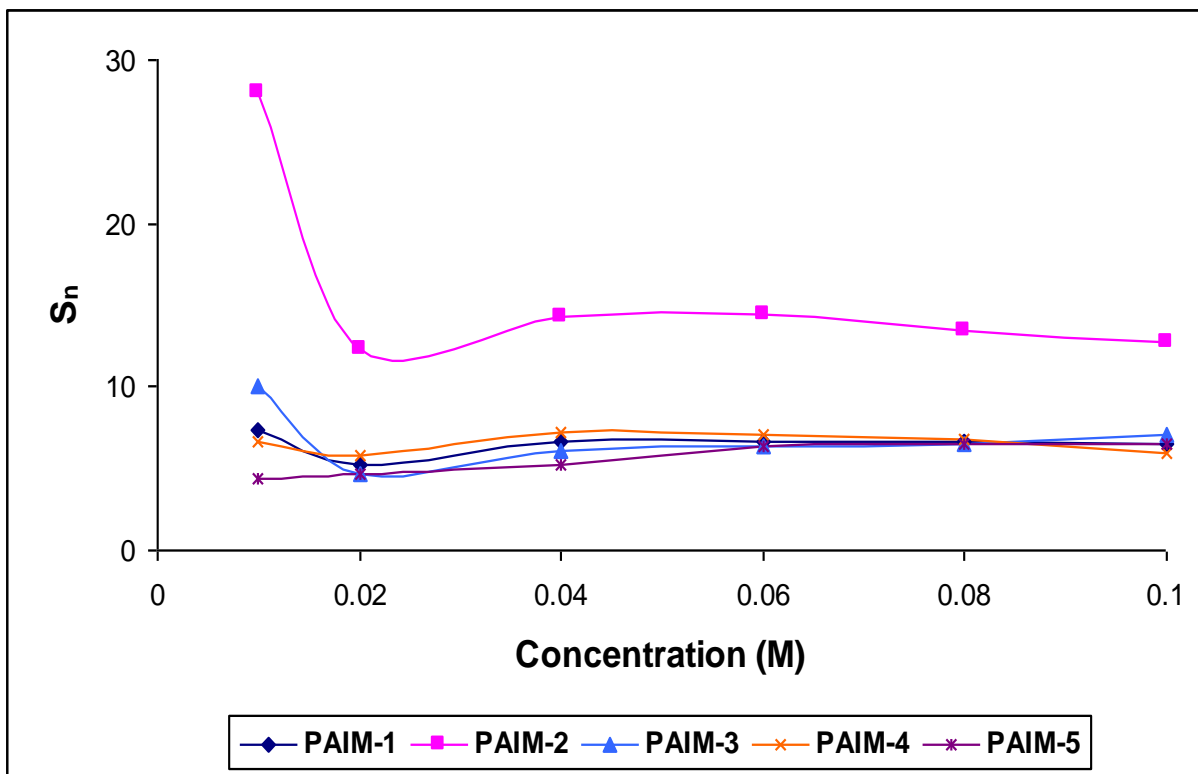


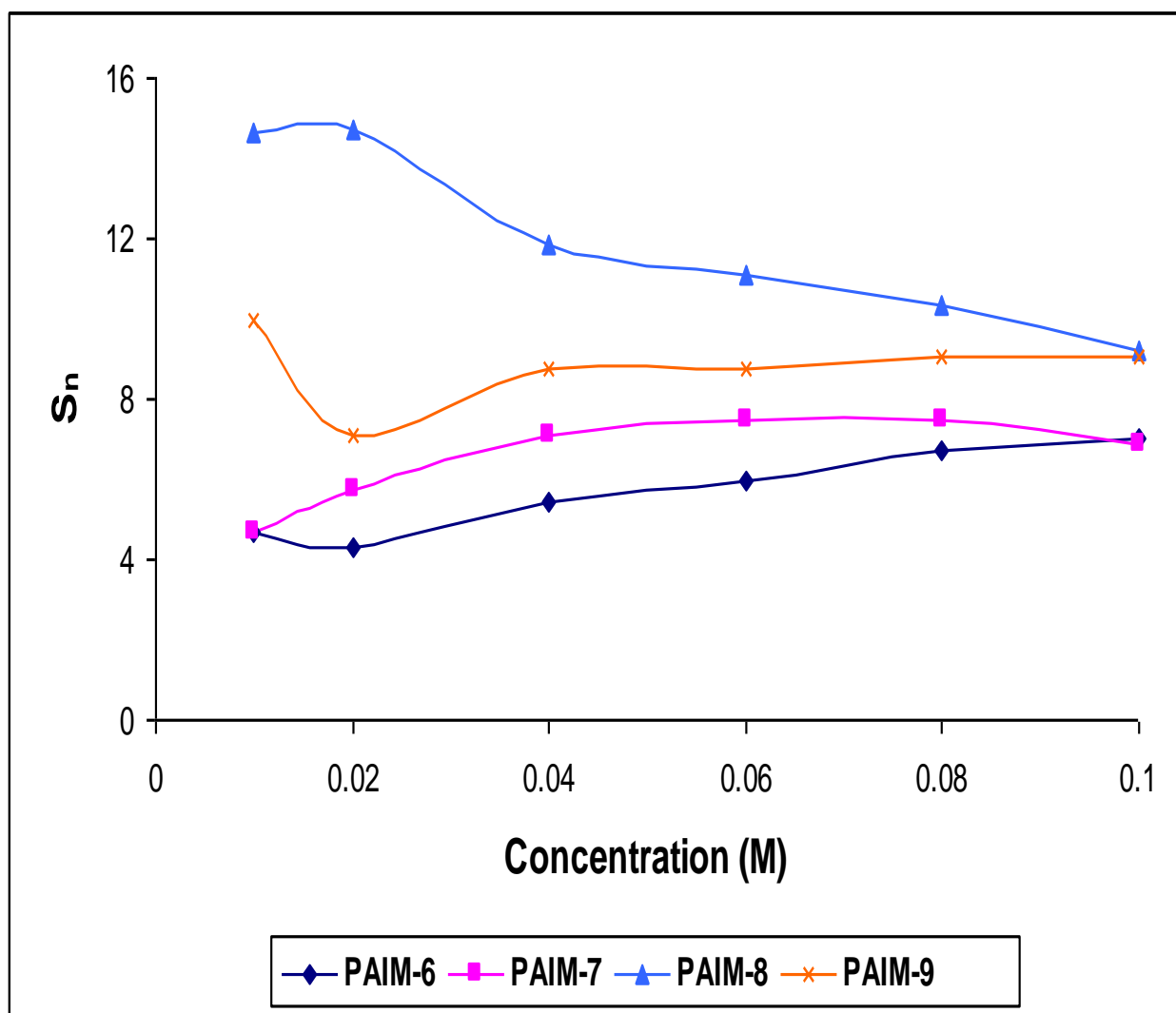
Figure 3 4: Variation of solvation number ( $S_n$ ) with concentration of imidazolinone derivatives in DMSO at 308.15K.



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The decrease of  $\kappa_s$  with increasing concentration might be due to aggregation of solvent molecules around solute molecules thereby indicating the presence of solute-solvent interactions (Jayakumar *et al.*, 1998; Eyring and Kincaid, 1932; Suryanaraana and Kuppaswamy, 1981; Nath and Rashmi, 1990; Ali and Nain, 1997; Roy *et al.*, 2007). The decrease of relaxation strength ( $r$ ) and increase of acoustical impedance ( $Z$ ) in Table 1 further confirms the solute-solvent interactions in these systems (Syal *et al.*, 1995).

Properties like Rao's molar sound function ( $R_m$ ), molar compressibility ( $W$ ) and Van der Waal's constant ( $b$ ) are observed to increase linearly (correlation coefficient,  $\gamma = 0.9999 - 1.0000$ ) with concentration for all the compounds. The linear variation of these acoustical properties indicates absence of complex formation (Wanchoo, 1987).



The internal pressure ( $\pi$ ) is the results of forces of attraction and repulsion between the molecules in a solution. Table 1 show that internal pressure decreases with concentration, which indicates the decrease in cohesive forces (Naidu and Prasad, 2005). Although decrease in compressibility ( $\kappa_s$ ), intermolecular free length ( $L_f$ ), relaxation strength ( $r$ ) and increase of velocity ( $U$ ), viscosity ( $\eta$ ) suggest predominance of

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**Table 1: Some acoustical parameters of imidazolinone derivatives in DMSO at 308.15K.**

Conc. M	Density $10^{-3}$ $\text{kg.m}^{-3}$	Viscosity $\times 10^3$ Pas	$Z.10^{-6}$ $\text{kg.m}^{-2}$ $\text{s}^{-1}$	$L_f. 10^2$ m	r	$\pi.10^6$ Pas	$V_f.10^2$ m	$R_A$	$R_m.10^4$ $10^{1/3}$ $\text{s}^{-1/3}$ $\text{mol}^{-1}$	$W 10^3$ $\text{m}^3$ $\text{Pas}^{-8/7}$ $\text{mol}^{-1}$	$b 10^5$ $\text{m}^3 \text{mol}^{-1}$
PAIM-1											
0.00	1.0546	10.6786	1.5460	0.4040	0.1605	580.2555	0.1255	1.0000	3.9064	2.2339	7.4083
0.01	1.0548	10.8684	1.5497	0.4030	0.1568	565.9824	0.1279	0.9995	4.0197	2.2986	7.6177
0.02	1.0556	11.0201	1.5568	0.4014	0.1504	551.3063	0.1312	0.9990	4.1330	2.3631	7.8224
0.04	1.0569	11.2149	1.5638	0.3998	0.1448	522.8142	0.1389	0.9991	4.3542	2.4897	8.2323
0.06	1.0578	11.3297	1.5727	0.3977	0.1365	494.9934	0.1484	0.9983	4.5793	2.6181	8.6439
0.08	1.0585	11.5119	1.5822	0.3954	0.1272	471.1396	0.1568	0.9972	4.8063	2.7475	9.0562
0.10	1.0593	11.6860	1.5928	0.3930	0.1169	449.2968	0.1655	0.9960	5.0338	2.8770	9.4662
PAIM-2											
0.01	1.0548	11.4230	1.5472	0.4037	0.1596	579.3780	0.1187	1.0000	4.0255	2.3021	7.6329
0.02	1.0556	11.4607	1.5513	0.4028	0.1564	560.6900	0.1238	1.0001	4.1440	2.3698	7.8526
0.04	1.0568	11.7311	1.5556	0.4019	0.1536	531.5112	0.1303	1.0007	4.3790	2.5045	8.2934
0.06	1.0576	11.8204	1.5602	0.4009	0.1499	501.3955	0.1398	1.0008	4.6160	2.6400	8.7359
0.08	1.0587	11.9286	1.5665	0.3995	0.1448	474.7798	0.1493	1.0008	4.8524	2.7752	9.1741
0.10	1.0596	12.0699	1.5729	0.3980	0.1393	451.3386	0.1583	1.0006	5.0898	2.9109	9.6127
PAIM-3											
0.01	1.0547	10.9877	1.5487	0.4033	0.1577	569.1968	0.1257	0.9995	4.0194	2.2984	7.6185
0.02	1.0560	11.1123	1.5582	0.4011	0.1495	553.6098	0.1297	0.9992	4.1320	2.3627	7.8193
0.04	1.0571	11.3051	1.5654	0.3995	0.1434	524.7778	0.1374	0.9990	4.3545	2.4898	8.2306
0.06	1.0577	11.5032	1.5743	0.3973	0.1346	498.4611	0.1453	0.9979	4.5815	2.6192	8.6448
0.08	1.0585	11.7619	1.5831	0.3952	0.1262	476.1012	0.1520	0.9970	4.8072	2.7479	9.0562
0.10	1.0592	12.1373	1.5896	0.3937	0.1202	458.2775	0.1559	0.9965	5.0312	2.8758	9.4673
PAIM-4											
0.01	1.0550	10.8602	1.5509	0.4028	0.1559	562.7173	0.1290	0.9995	4.0379	2.3089	7.6508
0.02	1.0557	11.0263	1.5574	0.4012	0.1499	545.8134	0.1329	0.9990	4.1693	2.3839	7.8905
0.04	1.0572	11.2326	1.5651	0.3995	0.1439	513.1970	0.1422	0.9992	4.4263	2.5310	8.3671
0.06	1.0584	11.4098	1.5757	0.3971	0.1342	483.2012	0.1524	0.9985	4.6874	2.6799	8.8439
0.08	1.0593	11.5934	1.5877	0.3942	0.1225	456.3501	0.1629	0.9971	4.9516	2.8305	9.3216
0.10	1.0603	11.7999	1.6061	0.3899	0.1037	431.9558	0.1739	0.9945	5.2224	2.9841	9.7966
PAIM-5											

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0.01	1.0547	10.9877	1.5521	0.4024	0.1541	568.1955	0.1262	0.9988	4.0246	2.3012	7.6229
0.02	1.0554	11.0351	1.5582	0.4010	0.1485	550.5858	0.1314	0.9984	4.1399	2.3669	7.8327
0.04	1.0564	11.2105	1.5681	0.3986	0.1393	520.3800	0.1401	0.9976	4.3705	2.4984	8.2541
0.06	1.0572	11.3814	1.5744	0.3972	0.1337	493.7404	0.1485	0.9972	4.5987	2.6288	8.6758
0.08	1.0576	11.6108	1.5834	0.3950	0.1244	470.3091	0.1561	0.9958	4.8323	2.7618	9.1002
0.10	1.0585	11.8030	1.5937	0.3926	0.1145	448.5179	0.1646	0.9948	5.0639	2.8937	9.5185

PAIM-6

0.01	1.0548	11.0262	1.5522	0.4024	0.1541	567.9824	0.1259	0.9989	4.0318	2.3053	7.6365
0.02	1.0556	11.1080	1.5602	0.4005	0.1467	549.8325	0.1310	0.9982	4.1557	2.3759	7.8598
0.04	1.0569	11.3038	1.5693	0.3984	0.1388	518.4728	0.1399	0.9979	4.3989	2.5148	8.3069
0.06	1.0582	11.5746	1.5784	0.3964	0.1309	492.2323	0.1473	0.9976	4.6415	2.6534	8.7519
0.08	1.0594	11.7557	1.5853	0.3949	0.1253	467.2369	0.1560	0.9977	4.8821	2.7910	9.1957
0.10	1.0601	11.9872	1.5935	0.3929	0.1173	445.2233	0.1639	0.9969	5.1273	2.9308	9.6430

PAIM-7

0.01	1.0549	10.8387	1.5515	0.4026	0.1550	565.9095	0.1283	0.9992	4.0149	2.2957	7.6058
0.02	1.0556	10.9424	1.5555	0.4017	0.1518	551.4223	0.1319	0.9992	4.1200	2.3558	7.8000
0.04	1.0568	11.0514	1.5620	0.4003	0.1467	522.5358	0.1406	0.9994	4.3295	2.4757	8.1885
0.06	1.0581	11.2413	1.5694	0.3986	0.1407	498.2653	0.1479	0.9994	4.5388	2.5954	8.5743
0.08	1.0592	11.3327	1.5774	0.3968	0.1337	474.0378	0.1573	0.9991	4.7494	2.7157	8.9602
0.10	1.0596	11.4330	1.5886	0.3941	0.1220	451.3469	0.1673	0.9973	4.9682	2.8400	9.3519

PAIM-8

0.01	1.0551	10.9261	1.5485	0.4034	0.1587	564.8399	0.1275	1.0001	4.0358	2.3080	7.6510
0.02	1.0556	11.2020	1.5509	0.4029	0.1568	551.0757	0.1290	1.0002	4.1651	2.3819	7.8932
0.04	1.0567	11.4350	1.5580	0.4013	0.1508	518.3843	0.1377	1.0001	4.4246	2.5303	8.3752
0.06	1.0581	11.5872	1.5656	0.3996	0.1448	487.9734	0.1477	1.0002	4.6822	2.6777	8.8524
0.08	1.0593	11.7623	1.5741	0.3976	0.1374	461.2044	0.1576	0.9999	4.9415	2.8258	9.3291
0.10	1.0599	11.9473	1.5852	0.3950	0.1262	436.7815	0.1677	0.9984	5.2076	2.9774	9.8106

PAIM-9

0.01	1.0555	10.9900	1.5499	0.4031	0.1577	565.1664	0.1269	1.0003	4.0430	2.3122	7.6633
0.02	1.0566	11.2092	1.5566	0.4016	0.1522	548.4095	0.1302	1.0003	4.1808	2.3909	7.9158
0.04	1.0576	11.4516	1.5631	0.4001	0.1467	514.1080	0.1394	1.0001	4.4563	2.5484	8.4283
0.06	1.0587	11.6665	1.5720	0.3981	0.1388	483.1997	0.1493	0.9996	4.7329	2.7064	8.9378
0.08	1.0596	11.9465	1.5797	0.3963	0.1318	457.2147	0.1577	0.9991	5.0095	2.8644	9.4474
0.10	1.0603	12.0950	1.5883	0.3943	0.1234	431.4833	0.1689	0.9982	5.2885	3.0235	9.9577



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COMPOUND CODES	$A \cdot 10^{-2}$ $\text{TPa}^{-1} \text{m}^{-3} \text{mol}^{-1}$	$B \cdot 10^{-2}$ $\text{TPa}^{-1} \text{m}^{-9/2} \text{mol}^{-3/2}$	$\phi_k^0 \cdot 10^{-5}$ $\text{TPa}^{-1} \text{mol}^{-1}$	$S_k \cdot 10^{-5}$ $\text{TPa}^{-1} \text{m}^{-3/2} \text{mol}^{-3/2}$
PAIM-1	-1.90	1.18	0.01	1.12
PAIM-2	-0.20	3.50	1.18	1.28
PAIM-3	-2.80	1.94	-1.10	2.84
PAIM-4	-2.60	2.18	-0.33	0.30
PAIM-5	-4.00	5.58	-2.10	6.00
PAIM-6	-3.80	4.59	-2.10	5.41
PAIM-7	-3.50	6.57	-1.90	9.09
PAIM-8	-0.80	3.50	1.30	3.23
PAIM-9	-1.85	0.94	0.30	0.75

Solute-solvent interaction, the decrease in internal pressure indicates the existence of solute-solute interactions also in these systems.

The free volume ( $V_f$ ) of solute molecule at particular temperature and pressure depends on the internal pressure of liquid in which it was dissolved. The decrease in molecular association causes an increase in free volume ( $V_f$ ). Thus, free volume is an inverse function of internal pressure. It is evident from Table 2 that  $V_f$  increases with concentration for all the compounds in DMSO solutions, indicating the presence of solute-solute interactions. This suggests that both solute-solute and solute-solvent interactions exist in these systems.

Further, the existence of solute-solute and solute-solvent interactions in these solutions were confirmed by some parameters calculated from Bachem's (Bachem, 1936) and Gucker's relation (Gucker, 1993) in which isentropic compressibility and apparent molar compressibility ( $\phi_k$ ) respectively, of the solutions are fitted. These equations are:

$$\kappa_s = \kappa_s^0 + AC + BC^{3/2} \dots (14)$$

$$\phi_k = \phi_k^0 + S_k \sqrt{C} \dots (15)$$

where C is the concentration. Using these equations, values of A and  $\phi_k^0$  were evaluated from the intercept and B and  $S_k$  values are evaluated from the slope of the plots. All these values of intercept and slopes are given in Table 2.

It is evident from Table 2 that for all the compounds, A values are negative and B values are positive but low. The negative A and low B values indicate solute-solvent interactions in these systems (Nikam *et al.*, 1998). Further,  $\phi_k^0$  values are negative except for PAIM-1, PAIM-2, PAIM-8 and PAIM-9, which have low positive values. However, for all the compounds,  $S_k$  values are positive. These negative or low positive  $\phi_k^0$  values and positive  $S_k$  confirms the existence of solute-solvent interactions (Pandey *et al.*, 1987; Naidu and Prasad, 2005; Nikam and Hiray, 1996).

The interactions occurring in different solutions can also be confirmed by the solvation number ( $S_n$ ), which is measure of structure forming or structure breaking tendency of solute in a solution. Figure 3 shows that except for PAIM-8,  $S_n$  values decrease suddenly at the beginning and then increase with concentration for all other compounds. For PAIM-8, it decreases with concentration. However, for all compounds,  $S_n$  values are positive. The positive values of solvation number indicate structure forming tendency of solute in a solvent. (Pandey *et al.*, 2000). Thus, all compounds show structure forming tendency in DMSO. This further confirms the existence of solute-solvent interactions in these systems. Although, in PAIM-8 solutions  $S_n$  values decrease, values are positive. So, PAIM-8 also exhibit structure forming tendency, which decreases with concentration. Further, for some compounds, after increasing  $S_n$  values become almost constant, which may due to dominant effect of intermolecular attractions between solute molecules (i.e., solute-solute interactions) over solute-solvent interactions (Bhullar *et al.*, 1991). This suggests that structure forming tendency decreases with concentration in most of the compounds,

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which may be due to dipole-dipole interactions, steric hindrance etc. Such interactions may cause weakening between solute and solvent molecules due to which structure forming tendency also decreases. This weak association is further confirmed by low relative association value ( $R_A$ ), which is almost same for some compounds (Table 1). Results of other authors (Sanaria and Parsania, 2000) also indicated predominance of solute-solvent interactions over solute-solute interactions in some systems. However, it is observed in some cases (Desai and Parsania, 1997; Rajkotia and Parsania, 1998) that solute exhibits structure forming tendency and also solute-solute interactions.

Thus, it is concluded that in DMSO solution of studied compounds, both solute-solute and solute-solvent interactions exist. Some of the acoustical parameters suggest predominance of solute-solvent interactions in the studied systems.

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