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Research Article

Molecular Interactions in DMF Solutions of some Pyrazole Schiff Bases at 308.15 K

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Abstract

The density, ultrasonic velocity and viscosity of solutions of pyrazole Schiff bases in dimethyl formamide have been measured at 308.15 K. From the experimental data, various acoustical parameters such as specific Impedance, isentropic compressibility, Rao's molar sound function, Van der Waals constant, relaxation strength, intermolecular free length, internal pressure, solvation number, relative association etc. have been evaluated, which helps in understanding the molecular interactions occurring in these solutions.

Keywords acoustical parameters, DMF, pyrazole schiff bases, ultrasonic study

Introduction

Ultrasonic velocity measurements have been used to study the nature of molecular interactions in various pure liquids [1-3], liquid mixtures [4-10] and in solutions [11-17]. However, little work has been done for some organic compound solutions [18-21], especially Schiff bases [22-25].

In present paper, acoustical properties of some pyrazole Schiff bases were studied in DMF over entire concentration range at 308.15K. These properties were interpreted in terms of molecular interactions occurring in the solution.

Material and Methods

The AR grade DMF was purified by the reported method [26]. The Schiff bases were synthesized in the laboratory and were recrystallized before use. The common structure of synthesized Schiff bases and their IUPAC names are given in Figure 1.

The densities, ultrasonic velocity and viscosity of pure solvents and their solutions were measured at 308.15 K using pycnometer, ultrasonic interferometer operating at 2 MHz and Ubbelohde viscometer respectively. The accuracy of density, ultrasonic velocity and viscosity are ± 0.0001 g/cm^3, ± 0.1% cm/sec and 0.05% respectively. The uncertainty of temperature is ±0.1 K and that of concentration is 0.0001 moles/dm^3.

Results and Discussion

Table 1 shows the experimental data of density, ultrasonic velocity and viscosity at 308.15 K. Using these experimental data, various acoustical parameters were calculated using equations reported earlier [27]. Some of these parameters are reported in Table 2.

Figure 2 shows the variation of ultrasonic velocity (U) increases with concentration for all the compounds. It is observed that ultrasonic velocity

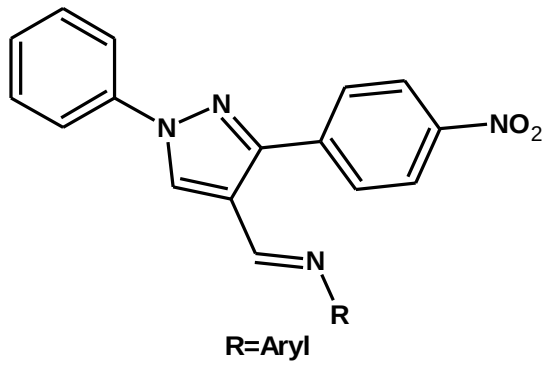

Figure 1: General structure of pyrazole Schiff bases

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JPA-1 : 3-(4-nitrophenyl)-5-phenyl-4H-pyrazole-4-carbaldehydephenylhydrazone

JPA-2 : 4-methyl-N-{(1E)-[3-(4-nitrophenyl)-5-phenyl-4H-pyrazol-4-yl] methylene} aniline

JPA-3 : 4-nitro-N-{(1E)-[3-(4-nitrophenyl)-5-phenyl-4H-pyrazol-4-yl] methylene} aniline

JPA-4 : 4-methoxy-N-{(1E)-[3-(4-nitrophenyl)-5-phenyl-4H-pyrazol-4-yl] methylene} aniline

JPA-5 : 3-chloro-4-fluoro-N-{(1E)-[3-(4-nitrophenyl)-5-phenyl-4H-pyrazol-4-yl] methylene} aniline

JPA-6 : 4-chloro-N-{(1E)-[3-(4-nitrophenyl)-5-phenyl-4H-pyrazol-4-yl] methylene} aniline

JPA-7 : 4-fluoro-N-{(1E)-[3-(4-nitrophenyl)-5-phenyl-4H-pyrazol-4-yl] methylene} aniline

JPA-8: 2,5-dichloro-N-{(1E)-[3-(4-nitrophenyl)-5-phenyl-4H-pyrazol-4-yl] methylene}aniline

JPA-9 : 2-methyl-N-{(1E)-[3-(4-nitrophenyl)-5-phenyl-4H-pyrazol 4-yl] methylene} aniline

Table 1: The density (ρ), ultrasonic velocity (U) and viscosity (η) of pyrazole Schiff bases in DMF at 308.15K.

Conc. (M)	Density g.cm-3	Velocity x 10-5 cm.s-1	Viscosity x 103 poise	Density g.cm-3	Velocity x 10-5 cm.s-1	Viscosity x 103 poise	Density g.cm-3	Velocity x 10-5 cm.s-1	Viscosity x 103 poise
	JPA-1			JPA-2			JPA-3		
0.00	0.9344	1.4308	6.0630	0.9344	1.4308	6.0630	0.9344	1.4308	6.0630
0.01	0.9363	1.4324	6.1019	0.9361	1.4336	6.4353	0.9362	1.4312	6.1603
0.02	0.9371	1.4344	6.1958	0.9373	1.4352	6.5899	0.9373	1.4328	6.2274
0.04	0.9390	1.4356	6.3185	0.9386	1.4364	6.7569	0.9400	1.4336	6.3480
0.06	0.9407	1.4372	6.4403	0.9399	1.4384	6.8963	0.9430	1.4352	6.5514
0.08	0.9425	1.4380	6.5120	0.9426	1.4404	7.0549	0.9447	1.4376	6.6725
0.10	0.9447	1.4408	6.6373	0.9449	1.4436	7.1745	0.9474	1.4392	6.8831
	JPA-4			JPA-5			JPA-6		
0.01	0.9363	1.4312	6.2496	0.9359	1.4316	6.1212	0.9356	1.4344	6.2305
0.02	0.9376	1.4320	6.3015	0.9370	1.4324	6.1648	0.9379	1.4356	6.4363
0.04	0.9398	1.4320	6.4409	0.9383	1.4328	6.2538	0.9395	1.4376	6.5825
0.06	0.9413	1.4324	6.5258	0.9407	1.4344	6.4311	0.9415	1.4396	6.7085
0.08	0.9424	1.4340	6.6387	0.9430	1.4352	6.7413	0.9421	1.4384	6.8218
0.10	0.9444	1.4352	6.7865	0.9435	1.4376	7.0426	0.9446	1.4368	6.9063
	JPA-7			JPA-8			JPA-9		
0.01	0.9357	1.4332	6.1706	0.9367	1.4340	6.2219	0.9365	1.4352	6.3115
0.02	0.9369	1.4344	6.2187	0.9379	1.4348	6.4021	0.9378	1.4364	6.4568
0.04	0.9377	1.4356	6.2832	0.9399	1.4352	6.5861	0.9396	1.4388	6.6395
0.06	0.9397	1.4384	6.3848	0.9418	1.4360	6.8059	0.9412	1.4408	6.8533
0.08	0.9422	1.4404	6.5176	0.9428	1.4372	6.9961	0.9426	1.4432	7.0412
0.10	0.9446	1.4436	6.6924	0.9441	1.4388	7.3976	0.9441	1.4456	7.2769

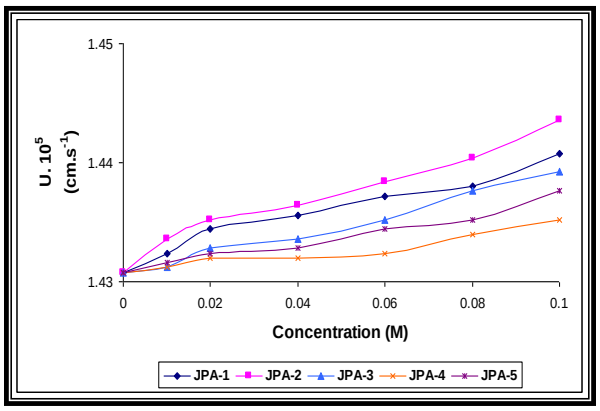
increases with concentration for all the compounds except JPA-6 in DMF solvent. In JPA-6, velocity increases up to 0.06M and then it decreases.

The intermolecular free path length (L_f) decreases continuously as listed in Table 2. Thus, variation of L_f is reverse of ultrasonic velocity. The ultrasonic velocity increases with decrease in L_f or vice versa. The decrease of L_f is due to decrease in the distance between compound and solvent molecules. When distance between molecules decreases, velocity increases. Thus, the decrease in L_f and increase in velocity suggests strong interaction between solvent and compound molecules.

The compound-solvent interaction is further confirmed by isentropic compressibility (κ_s). Figure 3 shows the variation of isentropic compressibility with concentration. It is observed that isentropic compressibility also decrease with concentration for

Table 2: Variation of acoustical parameters with concentration of pyrazole Schiff bases in DMF at 308.15K.

	Conc. (M)	Z .10-5 g.cm-2	L_f (Ao)	W.10-3 cm-1.dyn-1	R_m.10-3 cm-8/3.s-1/3	b cm3.mol-1	r	RA	π	V_f (cm3)
JPA-1	0.00	1.3369	0.4397	2.3022	4.0912	78.2194	0.2003	1.0000	441.2994	0.2558
	0.01	1.3412	0.4388	2.3390	4.1557	79.4226	0.1985	1.0017	434.2123	0.2605
	0.02	1.3442	0.4380	2.3783	4.2252	80.7131	0.1963	1.0020	428.9072	0.2617
	0.04	1.3480	0.4372	2.4544	4.3593	83.2524	0.1949	1.0038	417.1632	0.2673
	0.06	1.3520	0.4363	2.5307	4.4939	85.7905	0.1931	1.0052	406.0701	0.2730
	0.08	1.3553	0.4356	2.6058	4.6261	88.2992	0.1922	1.0070	394.3380	0.2814
	0.10	1.3611	0.4343	2.6803	4.7573	90.7441	0.1891	1.0087	384.8030	0.2867
JPA-2	0.01	1.3420	0.4385	2.3395	4.1569	79.4240	0.1972	1.0012	445.7689	0.2407
	0.02	1.3452	0.4377	2.3773	4.2234	80.6638	0.1954	1.0021	442.4826	0.2386
	0.04	1.3482	0.4370	2.4539	4.3588	83.2273	0.1940	1.0032	431.5182	0.2417
	0.06	1.3520	0.4361	2.5305	4.4944	85.7763	0.1918	1.0041	420.2862	0.2463
	0.08	1.3577	0.4349	2.6031	4.6216	88.1639	0.1895	1.0065	410.8155	0.2496
	0.10	1.3641	0.4334	2.6766	4.7511	90.5668	0.1859	1.0082	400.5557	0.2552
JPA-3	0.01	1.3399	0.4392	2.3422	4.1613	79.5527	0.1999	1.0018	435.6594	0.2571
	0.02	1.3430	0.4384	2.3842	4.2354	80.9378	0.1981	1.0026	428.8030	0.2604
	0.04	1.3476	0.4375	2.4652	4.3776	83.6401	0.1972	1.0053	415.9454	0.2672
	0.06	1.3534	0.4364	2.5450	4.5174	86.2800	0.1954	1.0082	406.6346	0.2688
	0.08	1.3581	0.4352	2.6278	4.6636	89.0231	0.1927	1.0094	394.9750	0.2755
	0.10	1.3635	0.4341	2.7068	4.8022	91.6338	0.1909	1.0119	387.0913	0.2762
JPA-4	0.01	1.3400	0.4391	2.3418	4.1604	79.5358	0.1999	1.0019	438.8903	0.2515
	0.02	1.3426	0.4386	2.3826	4.2323	80.8944	0.1990	1.0031	431.6658	0.2556
	0.04	1.3458	0.4381	2.4639	4.3752	83.6264	0.1990	1.0055	419.3375	0.2609
	0.06	1.3483	0.4376	2.5465	4.5209	86.4027	0.1985	1.0070	405.9316	0.2694
	0.08	1.3514	0.4369	2.6303	4.6692	89.2037	0.1967	1.0078	394.0167	0.2764
	0.10	1.3554	0.4360	2.7110	4.8111	91.8899	0.1954	1.0097	384.2565	0.2808
JPA-5	0.01	1.3398	0.4391	2.3484	4.1724	79.7575	0.1994	1.0014	432.9837	0.2605
	0.02	1.3422	0.4386	2.3952	4.2550	81.3214	0.1985	1.0024	424.4208	0.2661
	0.04	1.3444	0.4382	2.4899	4.4225	84.5140	0.1981	1.0037	408.3543	0.2766
	0.06	1.3493	0.4371	2.5818	4.5843	87.5745	0.1963	1.0059	396.5408	0.2813
	0.08	1.3534	0.4364	2.6728	4.7443	90.6138	0.1954	1.0082	389.5644	0.2771
	0.10	1.3564	0.4355	2.7693	4.9156	93.8323	0.1927	1.0081	381.8653	0.2744
JPA-6	0.01	1.3420	0.4383	2.3458	4.1685	79.6296	0.1963	1.0004	437.2935	0.2537
	0.02	1.3464	0.4374	2.3856	4.2379	80.9337	0.1949	1.0026	435.3944	0.2488
	0.04	1.3506	0.4364	2.4713	4.3893	83.7867	0.1927	1.0039	422.2180	0.2546
	0.06	1.3554	0.4354	2.5554	4.5377	86.5793	0.1904	1.0055	409.5225	0.2613
	0.08	1.3551	0.4356	2.6412	4.6894	89.4979	0.1918	1.0065	397.3357	0.2677
	0.10	1.3572	0.4355	2.7209	4.8288	92.1933	0.1936	1.0095	385.8920	0.2754
JPA-7	0.01	1.3410	0.4387	2.3412	4.1600	79.4903	0.1976	1.0008	436.2359	0.2564
	0.02	1.3439	0.4380	2.3796	4.2278	80.7627	0.1963	1.0018	429.4383	0.2604
	0.04	1.3462	0.4375	2.4594	4.3691	83.4388	0.1949	1.0024	415.2002	0.2699
	0.06	1.3517	0.4362	2.5366	4.5053	85.9851	0.1918	1.0039	403.2954	0.2774
	0.08	1.3571	0.4350	2.6115	4.6370	88.4563	0.1895	1.0061	393.4229	0.2823
	0.10	1.3636	0.4335	2.6867	4.7692	90.9113	0.1859	1.0079	385.2136	0.2847
JPA-8	0.01	1.3432	0.4382	2.3519	4.1786	79.8307	0.1967	1.0017	435.5128	0.2555
	0.02	1.3457	0.4377	2.4027	4.2681	81.5250	0.1958	1.0028	430.6859	0.2534
	0.04	1.3489	0.4371	2.5039	4.4465	84.9261	0.1954	1.0049	415.9884	0.2591
	0.06	1.3524	0.4364	2.6047	4.6245	88.3081	0.1945	1.0067	403.5192	0.2625
	0.08	1.3550	0.4358	2.7077	4.8067	91.7633	0.1931	1.0075	390.8352	0.2676
	0.10	1.3584	0.4350	2.8096	4.9869	95.1677	0.1913	1.0085	384.6903	0.2609
JPA-9	0.01	1.3441	0.4379	2.3394	4.1567	79.3895	0.1954	1.0012	445.7462	0.2410
	0.02	1.3471	0.4372	2.3767	4.2222	80.6194	0.1940	1.0023	442.5822	0.2387
	0.04	1.3519	0.4361	2.4526	4.3563	83.1330	0.1913	1.0037	431.3647	0.2425
	0.06	1.3561	0.4351	2.5284	4.4901	85.6468	0.1891	1.0049	421.0248	0.2457
	0.08	1.3604	0.4340	2.6045	4.6246	88.1639	0.1864	1.0059	411.1702	0.2490
	0.10	1.3648	0.4330	2.6800	4.7579	90.6548	0.1837	1.0069	400.1661	0.2554



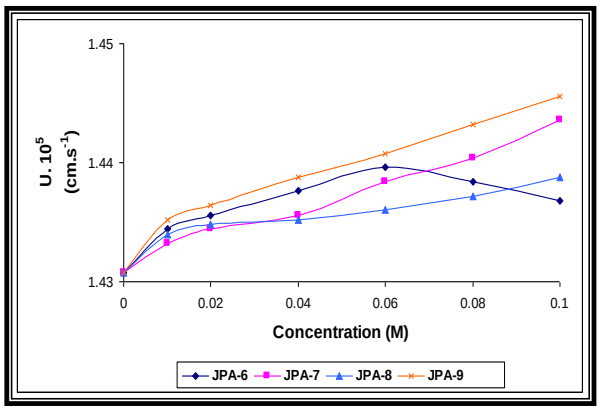
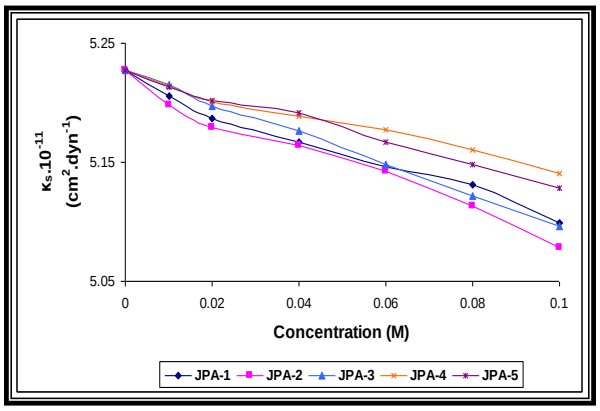


Figure 2: Variation of ultrasonic velocity (U) with concentration of pyrazole Schiff bases in DMF at 308.15 K



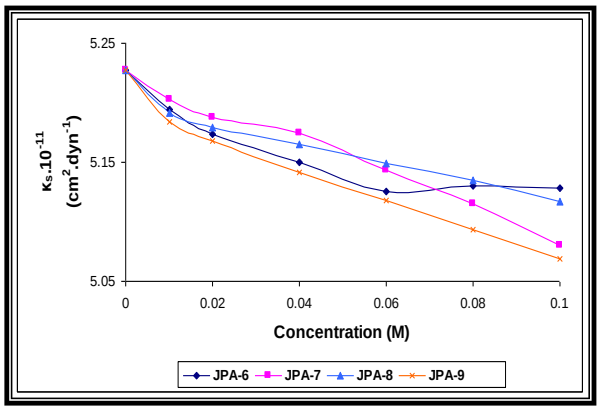
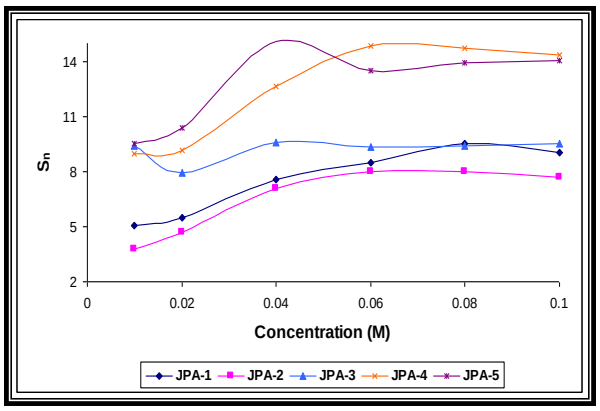


Figure 3: Variation of isentropic compressibility (κs) with concentration of pyrazole Schiff bases in DMF at 308.15 K.



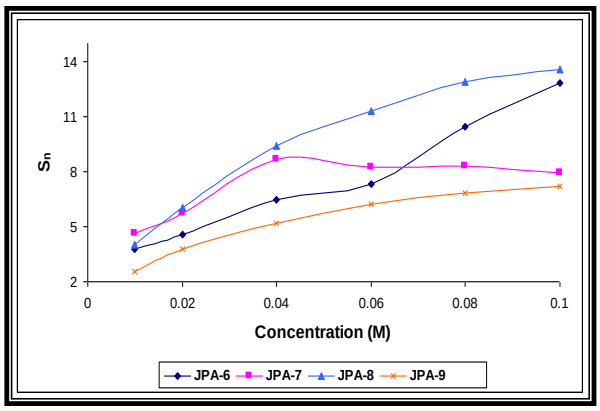


Figure 4: Variation of solvation number (S_n) with concentration of pyrazole Schiff bases in DMF at 308.15 K.

all the compounds (except JPA-6). The decrease of isentropic compressibility with increasing concentration may be due to aggregation of solvent molecules around compound molecules which again indicates the existence of compound-solvent interactions in studied solutions. The increase of acoustical impedance (Z) and decrease of relaxation strength (r) (Table 2) further confirms the solute-solvent interactions in these systems. In DMF solutions of JPA-6, the decrease of ultrasonic velocity (Figure 2) and Z and increase in κ_s and r after 0.06 M suggests that at higher concentrations, solute-solute interactions dominate.

Further, for all the compounds, Rao's molar sound function (R_m), molar compressibility (W) and Van der Waals' constant (b) are observed to increase linearly with concentration as shown in Table 2. The linear variation of these acoustical properties indicates the absence of complex formation.

The parameter internal pressure (π) is a measure of cohesive forces in solutions. It is evident from Table 2 that internal pressure decreases with concentration for all the Schiff bases. This suggests the decrease in cohesive forces. This indicates that in studied solutions, other interactions such as compound-compound and solvent-solvent interactions also exist. This is further confirmed by the increase of free volume (V_f). Thus, although some of the parameters suggest predominance of solute-solvent interactions, the variation of internal pressure and free volume indicates the existence of solute-solute and solvent-solvent interactions also in these systems.

Further, the apparent molar compressibility's (ϕ_k) of the solutions is fitted to Gucker's relation [28].

$$\phi_k = \phi^\circ_k + S_k \sqrt{C}$$

where ϕ°_k is the limiting apparent molar compressibility and S_k is interaction parameter.

The values of ϕ°_k and S_k are calculated by the least square method and are reported in Table 3.

The molecular interactions in solutions can also be suggested by Bachem's relation [29], which is:

$$\kappa_s = \kappa_s^0 + AC + BC^{3/2}$$

where A and B are constants, C is molar concentration of solutions, and κ_s and κ_s^0 are isentropic compressibilities of the solution and solvent respectively. The constants A and B have been determined from the intercept and slope of the plots $(\kappa_s - \kappa_s^0)/C$ versus $C^{1/2}$ and are given in Table 3.

Table 3 show that in both solvents A and ϕok values are negative or very low whereas B and S_k values are positive. The low or negative A and ϕok values and positive B and S_k values suggest predominance of solute-solvent interactions.

The interactions occurring in solutions can also be confirmed by the solvation number (S_n). Solvation number is a measure of structure forming or structure breaking tendency of solute in a

Table 3: Bachem's constants A, B, ϕok and S_k of pyrazole Schiff bases in DMF at 308.15K

COMPOUNDS	A X 1011 dyn-1.cm3.mol-1	B X 1011 dyn-1.cm-1/2.mol-3/2	ϕokX 108 dyn-1.mol-1	S_kX 108 dyn-1cm-3/2.mol-3/2
DMF				
JPA-1	-2.82	0.15	-1.95	10.0
JPA-2	-4.02	12.02	0.62	2.0
JPA-3	-1.12	0.66	-0.42	6.0
JPA-4	-2.02	5.01	-2.42	0.6
JPA-5	-1.82	4.30	-0.270	3.5
JPA-6	-4.00	11.6	-0.20	0.4
JPA-7	-3.25	7.41	-1.92	8.6
JPA-8	-3.65	13.36	-4.02	15.0
JPA-9	-4.28	11.58	-5.25	20.0

solution. Figure 4 shows that the variation of solvation number (S_n) with concentrations for all the compounds in the both solvents. It is obvious from the figure that solvation number increases non linearly for all the compounds. This increase in S_n is due to structure forming tendency of compounds in DMF i.e.,solute-solvent interactions predominate over solute-solute interactions. Thus, it is concluded that in DMF solutions of the studied Schiff bases of Pyrazole aldehyde, solute- solvent interactions predominates over solute- solute interaction.

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