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Acoustic studies of hydrated nickel sulphate in aqueous ascorbic acid systems

Shashi Kant, Sunil Kumar, Manish Kumar, Vikas Bharti

Department of Chemistry, Himachal Pradesh University, Summer Hill, Shimla, India

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ABSTRACT

The ultrasonic velocities (U), densities (ρ) and viscosity (η) for hydrated nickel sulphate ($NiSO_4.6H_2O$) in 2, 4 and 6% aqueous ascorbic acid has been measured at a frequency of 3 MHz and temperatures 298.15, 303.15, 308.15, and 313.15 K. Various thermodynamic and acoustic parameters such as adiabatic compressibility (β), intermolecular free length (Lf), relative association (RA), specific acoustic impedance (Z), molar compressibility (W), Rao's molar sound function (R), relaxation time (τ), and Gibb's free energy of activation (ΔG^*) has been calculated. The results have been interpreted in terms of ion-ion and ion-solvent interactions which determine the chemical structure of solute and solvent molecules in the system.

Keywords: Ultrasonic velocity, Adiabatic compressibility, Relaxation time, Ascorbic acid, Hydrated Nickel sulphate.

INTRODUCTION

Acoustic parameters are useful in elucidating the nature of ion-solvent and ion-ion interactions, thereby predicting the structural changes occurring in the solutions. Ion-ion and ion-solvent interactions are of prime importance to understand reaction rates and chemical equilibria, especially involving dissolved electrolytes [1]. As acoustic methods provide a better insight into nature and strength of molecular interactions in the liquid mixture, these are helpful to study chemical processes, chemical synthesis and contribute to the fields of medicine, agriculture, engineering, defense, industry etc. [2-14]. The behavior of nickel sulphate has been studied in aqueous medium in recent times [1]. Multi-component solutions similar to bio-fluids are the work systems of current interest, as they can be utilized for better understanding of biological processes [15-22]. This prompted us to study hydrated nickel sulphate ($NiSO_4.6H_2O$) in 2, 4 and 6% aqueous ascorbic acid, keeping in consideration its importance in biological systems.

The study was carried out at four different temperatures 303.15, 308.15, 313.15 and 318.15 K by measuring density and sound velocity of the solutions. The measured values of density and sound velocity were then used to derive various thermodynamic and acoustic parameters which are then used to interpret ion-ion and ion-solvent interactions.

MATERIALS AND METHODS

Double distilled water with specific conductance in the range of 0.1×10^{-6} to 1.0×10^{-6} $\Omega^{-1}cm^{-1}$ was used for preparation of the solutions. Nickel sulphate and ascorbic acid (AR grade) were dried under vacuum for 24 hours before use. Water of crystallization was estimated by the standard method [23]. All the solutions were prepared by weight and conversions of molality to molarity were done by using the standard expression [24]. The concentration range for nickel sulphate was 0.01m to 0.12m and solvents were 2, 4 and 6% aqueous ascorbic acid. The density and velocity of sound were measured with the help of DSA (Density and Sound Analyser) 5000, Antor Paar, GmbH, Garz, Austria. All measurements were made at 303.15, 308.15, 313.15 and 318.15K. Repeatability of the instrument for density is 1.0×10^{-6} gcm^{-3}, for sound velocity 1.0×10^{-2} ms^{-1} and for temperature is 1.0×10^{-3} K.

RESULTS AND DISCUSSION

Various acoustic parameters were obtained for hydrated nickel sulphate in 2, 4 and 6% aqueous ascorbic acid, using ultrasonic velocity and density data (Table-1).

Adiabatic compressibility (β), defined as $\beta = (-1/V_m)(\partial V_m/\partial P)_S$ was obtained using the Newton-Laplace equation (1) [25]:

$$\beta = 1/U^2\rho \qquad (1)$$

Where ρ is the density and U is the ultrasonic velocity of solutions. There is a limiting adiabatic compressibility for every solvent which decreases with the increase in concentration of the solution. A gradual and almost linear decrease in adiabatic compressibility (Table-1) was observed as concentration of solute was increased showing presence of prominent ion-solvent interactions [26]. Also adiabatic compressibility has decreased with increase in temperature and ascorbic acid content of the solvent (Fig.1, Fig.2).

Intermolecular free length (L_f) was obtained from adiabatic compressibility (β) using equation (2) [27]:

$$L_f = K\beta^{1/2} \qquad (2)$$

Where K is the temperature dependent constant (= $(93.875 + 0.375T) \times 10^{-8}$) [28]. It is clear from Table-1 that ultrasonic velocity (U) has increased whereas intermolecular free length (L_f) has decreased with increase in concentration for nickel sulphate in all the solvent systems. In general U and L_f have been reported to vary as the inverse of each other as in the present systems [29-30]. U increases steadily with increase in concentration and temperature, which suggests that ion-solvent interactions increase [31]. Ultrasonic velocity has increased with the increase in ascorbic acid content of the solvent indicating thereby stronger ion-solvent interactions. As temperature increases there is loosening of intermolecular forces due to thermal agitation of the molecules and thus increasing L_f values (Fig.3). Similarly L_f has decreased with increase in the ascorbic content of the solvent (Fig.4) which indicates increase in ion-solvent interactions with increase in ascorbic acid content of the solvent as predicted by ultrasonic velocity data.

Specific acoustic impedance (Z) and Molar compressibility (W) were also calculated using relations (3) [32]:

$$Z = U\rho \qquad (3)$$

Where M is the apparent molecular weight of the solution and can be calculated according to the following equation:

$$M = M_1W_1 + M_2W_2 \qquad (4)$$

Where W_1 and W_2 are weight fractions of solvent and solute, respectively. M_1 and M_2 are molecular weights of solvent and solute respectively. When an acoustic wave propagates through a medium, the pressure is different at different particles of the medium. The ratio of instantaneous pressure excess at any particle of the medium to the instantaneous velocity of that particle is termed as specific acoustic impedance (Z) of that medium [31]. Specific acoustic impedance (Z) has increased with rise in temperature, concentration and ascorbic acid content of the solvent (Fig.5, Fig.6) showing strong ion-solvent interactions [7, 10].

Another parameter studied was solvation number (S_N), obtained by relation (5) [33-34]:

$$S_N = (n_1/n_2)(1-\beta/\beta_0) \qquad (5)$$

Where n_1 and n_2 are the numbers of moles of the solvent and solute; β and β_0 are compressibility coefficients of the solution and the pure solvent respectively. To explain solvation number of solute molecules, Frank and Wen model of solute-solvent interactions [14, 35] has been used which pictures three different solvent-structure regions around the solute molecule. In the present case, solvation number has decreased with increase in concentration and likely to attain the primary solvation in pure crystalline state [31]. Considerably high solvation numbers (Table-1) obtained especially at low solute concentrations and at low temperatures indicate either strong electrostriction around the sulphate ions and/or the range of the cation electrostrictive interaction, which is longer than the primary sheath of solvation [36]. Solvation numbers has decreased with increase in solute concentration as well as with increase in temperature for nickel sulphate in all the studied solvents (Fig.7).

The dispersion of ultrasonic waves in system contains information about the characteristic time of relaxation process that causes the dispersion. The relaxation time was calculated as (7) [37-39]:

$$\tau = (4\eta/3\rho U^2) \quad (7)$$

Where η is viscosity.

The relaxation time decreases with increase in temperature and its temperature dependence is used to calculate Gibb's free energy of activation for relaxation process (8) [31, 40]:

$$\Delta G^* = kT \ln(kT\tau/h) \quad (8)$$

Where k is Boltzmann constant, T, the absolute temperature and h is Planck's constant.

The values of ΔG^* have slightly increased with rise in temperature and ascorbic acid content of the solvent (Fig.8, Fig.9). Nearly constant values of ΔG^* with varying concentration suggest that the rearrangement of molecules in solution are characteristic of physical properties of solute only [39].

TABLE – 1 ADIABATIC COMPRESSIBILITY (β), INTERMOLECULAR FREE LENGTH (L_f), SPECIFIC ACOUSTIC IMPEDANCE (Z), SOLVATION NUMBER (S_N) AND GIBB'S FREE ENERGY OF ACTIVATION (ΔG^*) FOR NICKEL SULPHATE IN DIFFERENT SOLVENTS AT DIFFERENT TEMPERATURES

$C \times 10^2$ (mol lit^{-1})	ρ (g cm^{-3})	U (m s^{-1})	L_f (10^{-11}m)	β (10^{-10}Pa^{-1})	Z (10^6 kg m^{-2}s^{-1})	S_N	ΔG^* (10^{-21} J mol^{-1})
$NiSO_4.6H_2O$ in 2% aqueous ascorbic acid							
T= 303.15K	**β_0 = 4.3500**			**ρ_0 = 1.003581gcm^{-3}**			**U_0 = 1513.49ms^{-1}**
1.0026	1.005195	1514.53	4.3207	4.3336	1.5230	20.6166	4.5904
2.0028	1.006664	1515.44	4.3153	4.3227	1.5260	17.1371	4.5993
3.9955	1.009373	1517.16	4.3039	4.2999	1.5321	15.6945	4.6113
5.9765	1.011797	1518.90	4.2932	4.2786	1.5378	14.9274	4.6204
7.9448	1.013987	1520.50	4.2831	4.2584	1.5431	14.3584	4.6297
9.8998	1.016003	1522.17	4.2739	4.2400	1.5480	13.7912	4.6390
11.8407	1.017846	1523.67	4.2641	4.2206	1.5529	13.5234	4.6456
T = 308.15K	**β_0 = 4.2985**			**ρ_0 = 1.001902gcm^{-3}**			**U_0 = 1523.80ms^{-1}**
1.0008	1.003401	1524.79	4.3343	4.2831	1.5306	19.5719	4.2470
1.9990	1.004774	1525.65	4.3291	4.2729	1.5335	16.2738	4.2556
3.9873	1.007312	1527.29	4.3183	4.2515	1.5393	14.9314	4.2683
5.9638	1.009637	1528.91	4.3082	4.2316	1.5447	14.1586	4.2789
7.9273	1.011748	1530.47	4.2983	4.2122	1.5498	13.6980	4.2882
9.8777	1.013731	1532.06	4.2893	4.1947	1.5546	13.1795	4.2984
11.8136	1.015519	1533.46	4.2797	4.1758	1.5595	12.9745	4.3065
T = 313.15K	**β_0 = 4.2572**			**ρ_0 = 1.000031gcm^{-3}**			**U_0 = 1532.61ms^{-1}**
0.9988	1.001429	1533.54	4.3524	4.2427	1.5364	18.6189	3.8925
1.9950	1.002733	1534.33	4.3475	4.2331	1.5391	15.4265	3.9014
3.9787	1.005142	1535.91	4.3372	4.2130	1.5446	14.1664	3.9143
5.9503	1.007349	1537.44	4.3274	4.1940	1.5498	13.5037	3.9257
7.9089	1.009406	1538.93	4.3177	4.1752	1.5549	13.1324	3.9358
9.8547	1.011368	1540.45	4.3090	4.1585	1.5595	12.6515	3.9463
11.7857	1.013119	1541.77	4.2996	4.1403	1.5643	12.4811	3.9559
T = 318.15K	**β_0 = 4.2258**			**ρ_0 = 0.997980gcm^{-3}**			**U_0 = 1539.88ms^{-1}**
0.9967	0.999308	1540.74	4.3749	4.2116	1.5404	18.2723	3.5998
1.9906	1.000552	1541.53	4.3702	4.2024	1.5430	15.0863	3.6087
3.9698	1.002873	1543.04	4.3602	4.1833	1.5483	13.6957	3.6212
5.9368	1.005064	1544.49	4.3506	4.1648	1.5535	13.1138	3.6324

7.8907	1.007081	1545.92	4.3410	4.1466	1.5584	12.7757	3.6429
9.8314	1.008978	1547.37	4.3327	4.1307	1.5629	12.2683	3.6550
11.7585	1.010783	1548.61	4.3232	4.1126	1.5677	12.1719	3.6646
$NiSO_4.6H_2O$ in 4% aqueous ascorbic acid							
T = 303.15K	**β_0 = 4.2883**			**ρ_0 = 1.011738gcm^{-3}**			**U_0 = 1518.18 ms^{-1}**
1.0107	1.013374	1519.30	4.2915	4.2751	1.5396	16.5279	4.6347
2.0193	1.014957	1520.58	4.2845	4.2612	1.5433	16.9137	4.6392
4.0298	1.018035	1522.55	4.2725	4.2373	1.5500	15.9103	4.6460
6.0308	1.020988	1524.22	4.2617	4.2158	1.5562	15.0843	4.6536
8.0221	1.023844	1526.19	4.2502	4.1932	1.5626	14.8426	4.6575
10.0036	1.026655	1528.64	4.2376	4.1684	1.5694	14.9809	4.6575
11.9749	1.029381	1530.16	4.2278	4.1491	1.5751	14.4922	4.6620
T = 308.15K	**β_0 = 4.2401**			**ρ_0 = 1.009996gcm^{-3}**			**U_0 = 1528.10ms^{-1}**
1.0089	1.011581	1529.09	4.3063	4.2280	1.5468	15.3145	4.2829
2.0157	1.013127	1530.37	4.2995	4.2145	1.5505	16.1904	4.2866
4.0223	1.016136	1532.20	4.2880	4.1920	1.5569	15.2042	4.2949
6.0194	1.019051	1534.02	4.2767	4.1701	1.5632	14.7476	4.3006
8.0068	1.021898	1535.90	4.2655	4.1483	1.5695	14.5015	4.3048
9.9846	1.024703	1538.16	4.2534	4.1248	1.5762	14.5701	4.3064
11.9523	1.027441	1539.82	4.2432	4.1049	1.5821	14.2320	4.3110
T = 313.15K	**β_0 = 4.2017**			**ρ_0 = 1.008067gcm^{-3}**			**U_0 = 1536.54ms^{-1}**
1.0070	1.009618	1537.47	4.3254	4.1902	1.5523	14.6958	3.9337
2.0117	1.011133	1538.58	4.3190	4.1778	1.5557	15.1958	3.9380
4.0142	1.014105	1540.41	4.3076	4.1557	1.5621	14.6521	3.9452
6.0073	1.017010	1542.41	4.2959	4.1331	1.5686	14.5717	3.9506
7.9907	1.019838	1544.20	4.2849	4.1121	1.5748	14.2767	3.9558
9.9646	1.022656	1546.16	4.2736	4.0904	1.5812	14.1902	3.9593
11.9285	1.025398	1548.25	4.2621	4.0684	1.5876	14.1563	3.9618
T = 318.15K	**β_0 = 4.1726**			**ρ_0 = 1.005962gcm^{-3}**			**U_0 = 1543.50ms^{-1}**
1.0048	1.007480	1544.28	4.3492	4.1621	1.5558	13.4712	3.6467
2.0074	1.008969	1545.48	4.3426	4.1495	1.5593	14.8183	3.6497
4.0055	1.011898	1547.27	4.3313	4.1279	1.5657	14.3339	3.6569
5.9941	1.014765	1549.35	4.3193	4.1052	1.5722	14.4139	3.6610
7.9730	1.017583	1551.22	4.3081	4.0840	1.5785	14.2173	3.6660
9.9421	1.020343	1552.95	4.2975	4.0639	1.5845	13.9560	3.6716
11.9018	1.023098	1554.92	4.2863	4.0427	1.5908	13.8985	3.6744
$NiSO_4.6H_2O$ in 6% aqueous ascorbic acid							
T = 303.15K	**β_0 = 4.2257**			**ρ_0 = 1.019974gcm^{-3}**			**U_0 = 1523.19ms^{-1}**
1.0190	1.021688	1524.14	4.2604	4.2134	1.5572	15.3575	4.6906
2.0360	1.023352	1525.69	4.2526	4.1980	1.5613	17.2528	4.6932
4.0636	1.026582	1527.41	4.2411	4.1754	1.5680	15.6630	4.7020
6.0823	1.029701	1529.45	4.2291	4.1516	1.5749	15.3645	4.7067
8.0921	1.032782	1531.00	4.2185	4.1309	1.5812	14.7516	4.7121
10.0923	1.035760	1532.73	4.2077	4.1097	1.5875	14.4345	4.7166
12.0830	1.038673	1534.67	4.1964	4.0878	1.5940	14.2968	4.7192
T = 308.15K	**β_0 = 4.1805**			**ρ_0 = 1.018173gcm^{-3}**			**U_0 = 1532.77ms^{-1}**
1.0172	1.019843	1533.73	4.2759	4.1684	1.5642	15.1745	4.3303
2.0323	1.021477	1535.26	4.2682	4.1534	1.5682	16.9914	4.3327
4.0560	1.024667	1536.93	4.2569	4.1315	1.5748	15.3871	4.3410
6.0709	1.027768	1538.86	4.2452	4.1087	1.5816	15.0341	4.3465
8.0766	1.030805	1540.37	4.2348	4.0886	1.5878	14.4399	4.3531
10.0730	1.033781	1542.04	4.2241	4.0680	1.5941	14.1415	4.3581
12.0602	1.036720	1543.91	4.2130	4.0466	1.6006	14.0219	4.3606
T = 313.15K	**β_0 = 4.1449**			**ρ_0 = 1.016186gcm^{-3}**			**U_0 = 1540.84ms^{-1}**
1.0152	1.017824	1541.82	4.2958	4.1329	1.5693	15.1281	3.9878
2.0282	1.019429	1543.33	4.2882	4.1184	1.5733	16.8073	3.9897
4.0478	1.022583	1545.04	4.2768	4.0966	1.5799	15.3104	3.9980

6.0584	1.025661	1546.84	4.2655	4.0748	1.5865	14.8132	4.0038
8.0599	1.028676	1548.32	4.2551	4.0551	1.5927	14.2337	4.0101
10.0524	1.031659	1549.89	4.2447	4.0352	1.5990	13.9122	4.0165
12.0357	1.034612	1551.73	4.2336	4.0141	1.6054	13.8184	4.0192
T = 318.15K	**β_0 = 4.1182**			**ρ_0 = 1.014029gcm^{-3}**		**U_0 = 1547.47ms^{-1}**	
1.0130	1.015643	1548.50	4.3198	4.1062	1.5727	15.3321	3.6894
2.0238	1.017228	1549.99	4.3123	4.0919	1.5767	16.7770	3.6911
4.0389	1.020346	1551.75	4.3008	4.0701	1.5833	15.3295	3.6981
6.0451	1.023410	1553.40	4.2898	4.0493	1.5898	14.6448	3.7049
8.0422	1.026410	1554.86	4.2795	4.0299	1.5959	14.0810	3.7117
10.0300	1.029367	1556.35	4.2693	4.0107	1.6021	13.7240	3.7182
12.0087	1.032294	1558.19	4.2582	3.9898	1.6085	13.6502	3.7224

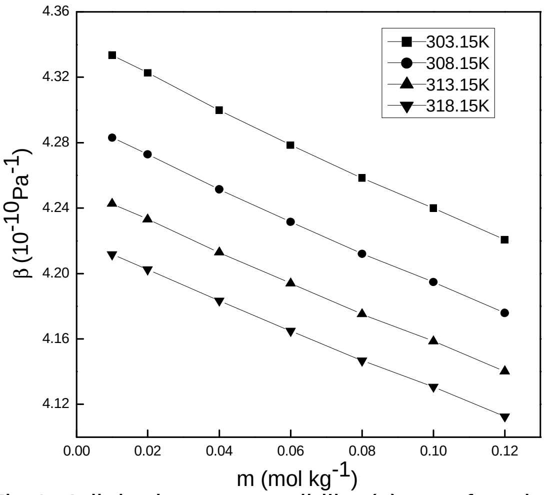

Fig.1: Adiabatic compressibility (β) as a function of molality (m) for $NiSO_4.6H_2O$ in 2% aqueous ascorbic acid at different temperatures.

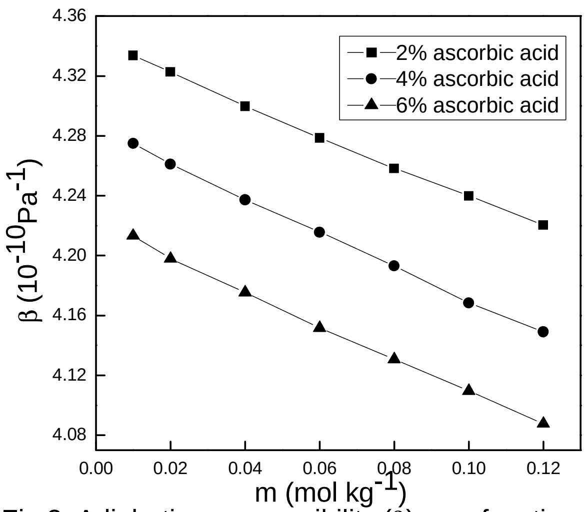

Fig.2: Adiabatic compressibility (β) as a function of molality (m) for $NiSO_4.6H_2O$ in aqueous solvents containing different percent of ascorbic acid at 303.15 K.

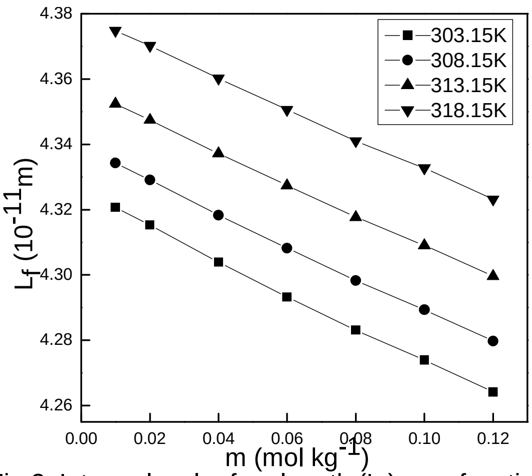

Fig.3: Intermolecular free length (L_f) as a function of molality (m) for $NiSO_4.6H_2O$ in 2% percent aqueous ascorbic acid at different temperatures.

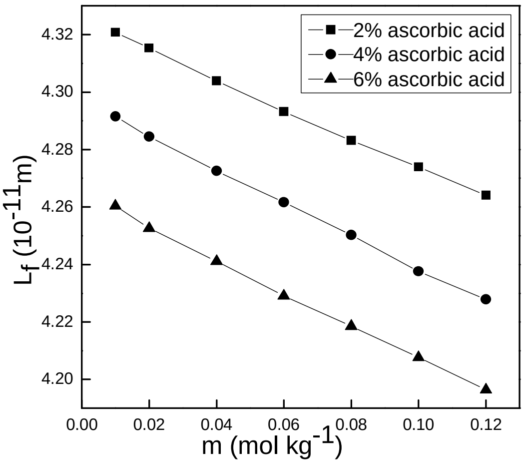

Fig.4: Intermolecular free length (L_f) as a function of molality (m) for $NiSO_4.6H_2O$ in aqueous solvents containing different percent of ascorbic acid at 303.15 K.

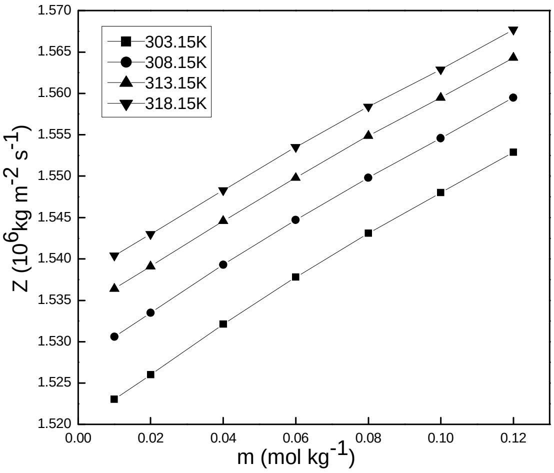

Fig.5: Specific acoustic impedance (Z) as a function of molality (m) for $NiSO_4.6H_2O$ in 2% aqueous percent of ascorbic acid at different temperatures.

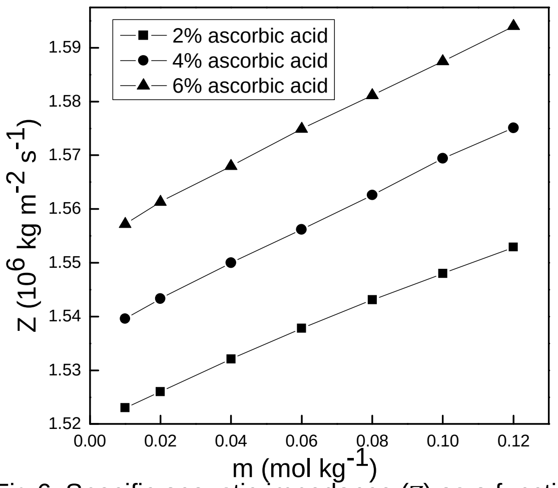

Fig.6: Specific acoustic impedance (Z) as a function of molality (m) for $NiSO_4.6H_2O$ in aqueous solvents containing different percent of ascorbic acid at 303.15 K.

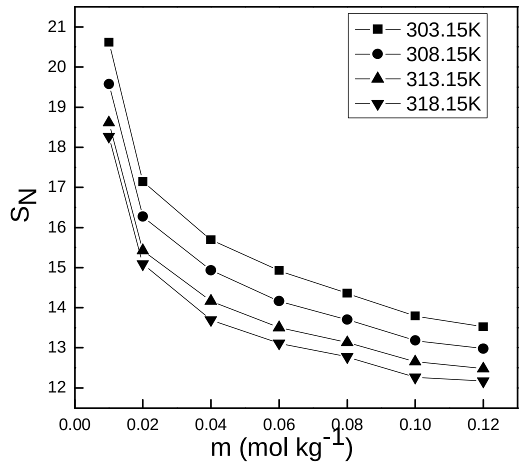

Fig.7: Solvation number (S_N) as a function of molality (m) for $NiSO_4.6H_2O$ in 2% aqueous percent of ascorbic acid at different temperatures.

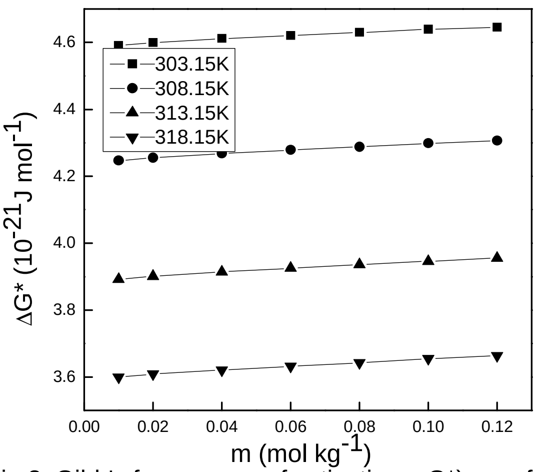

Fig.8: Gibb's free energy of activation (ΔG^*) as a function of molality (m) for $NiSO_4.6H_2O$ in 2% aqueous ascorbic acid at different temperatures.

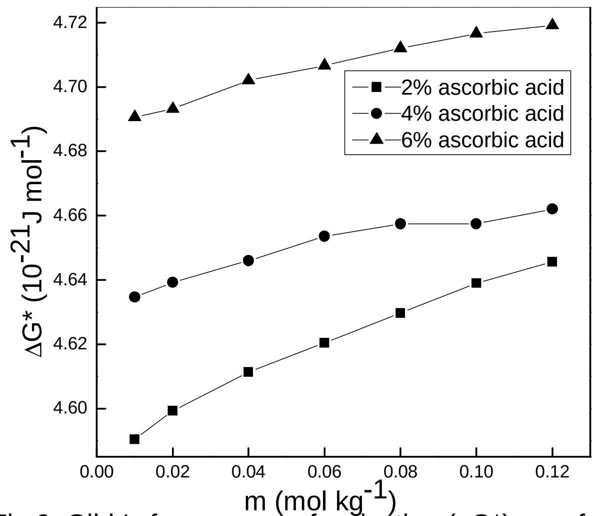

Fig.9: Gibb's free energy of activation (ΔG*) as a function of molality (m) for $NiSO_4.6H_2O$ in aqueous solvents containing different percent of ascorbic acid at 303.15 K.

CONCLUSION

The decrease in adiabatic compressibility (β) and increase in intermolecular free length (L_f) show presence of ion-solvent interactions for nickel sulphate in different solvents. Acoustic parameters such as specific acoustic impedance, molar compressibility, solvation number were reported. Characteristic time of relaxation was obtained and used to get Gibb's free energy of activation for relaxation process. Introduction of ascorbic acid to aqueous medium contribute to strengthening of ion-solvent interactions. All these parameters indicate that there is increase in ion-solvent interactions with increase in temperature, increase in concentration of the solution and ascorbic acid content of the solvent.

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