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Ultrasonic investigations of cadmium sulphate in aqueous mannitol

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ABSTRACT

The ultrasonic velocities (U), densities (ρ) and viscosity (η) for cadmium sulphate ($CdSO_4$) has been measured in aqueous mannitol systems 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 (L_f), relative association (R_A), 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, Mannitol, Cadmium sulphate.

INTRODUCTION

Carbohydrates are present abundantly in living nature and have been the subject of numerous investigations because of their abilities to preserve biosystems such as cells, vaccines, or therapeutic proteins employed in the food, pharmaceuticals, and cosmetics industries[1-6]. Carbohydrates and metal cations coexist in biological fluids, and the interactions between carbohydrates and metal cations are a subject of current interest because of the importance of carbohydrate-metal complexes in chemistry and biology. The thermodynamic and transport properties of carbohydrate-electrolyte solutions are more frequently required by chemists and engineers, including density, viscosity, adiabatic compressibility, relaxation time, intermolecular free length and Gibb's free energy of activation. Thermodynamic parameters for various 1:1 and 2:2 electrolytes including $CdSO_4$ in aqueous as well as in carbohydrate solutions at different temperatures were reported [7-10]. However, the interactions of carbohydrates with 2:2 electrolytes in terms of acoustic functions has been attempted by ultrasonic investigations of $CdSO_4$ in aqueous mannitol solutions. The measurement of the ultrasonic velocity and absorption of ultrasonic waves give deep insight into solution properties. The ultrasonic waves suffer dispersion as they pass through the liquid medium disturbing the equilibrium between molecules. The velocity of ultrasonic waves in solution can be interpreted for ion-ion and ion-solvent interactions which determine the chemical structure of solute and solvent molecules. In present study Jacobson's model and Shaaf's model have been extensively used to explain and interpret results [11,12].

MATERIALS AND METHODS

Mannitol and Cadmium Sulphate (Anala R) were dried under vacuum for 24 h before use. Double distilled water with specific conductance in the range of 0.1×10^{-6} to $1.0\times10^{-6}\Omega^{-1}cm^{-1}$ was used for preparation of the solutions. All the solutions were prepared by weight method. A ultrasonic interferometer F-81 model (Mittal Enterprises, Delhi) having frequeny 3MHz and accuracy ±0.01% was used for the measurement of ultrasonic velocity in solutions. The ultrasonic velocity measurements for various concentrations of cadmium sulphate in 5%, 15% and 25% aqueous mannitol were recorded at temperatures 298.15, 303.15, 308.15, and 313.15 K. The density and viscosity of mixtures were measured by usual methods [13, 14].

RESULTS AND DISCUSSION

The density, viscosity and ultrasonic velocity for cadmium sulphate ($CdSO_4$), was measured in 5, 15 and 25 % aqueous mannitol systems as solvent at 303.15, 308.15, 313.15 and 318.15 K and are presented in table 1. Ultrasonic velocity increases with increase in molality, as well as with temperature. This variation in ultrasonic velocity throws light on structural changes occurring in present system. The increase in ultrasonic velocity in these solutions may be attributed to the cohesion brought about by the ionic solvation. Molecular association is thus responsible for the observed increase in ultrasonic velocity in the present systems. The increase in velocity with temperature indicates decrease in intermolecular forces due to increase in thermal energy of the system. Similar works has been reported by Palani et al [15].

Adiabatic compressibility (β), defined as $\beta = (-1/V_m)(\partial V_m/\partial P)_S$ was obtained using the Newton-Laplace equation (1) [16],

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

where ρ is the density U is the ultrasonic velocity of solutions. Table 2 show the concentration dependences of the adiabatic compressibility for cadmium sulphate in 5, 10 and 15% aqueous mannitol. A gradual and almost linear decrease in adiabatic compressibility was observed with increase in concentration of solute as well as solvent. Due to electrostriction, the solvent molecules within the primary solvation shell of electrolytic solution are rendered incompressible moreover increasing concentration of ions results in more solvent molecules to engage in incompressible solvation spheres thereby decreasing the adiabatic compressibility [17]. Further with increase in temperature, hydrogen bonding among solvent-solvent molecules decreases and thus there are more free solvent molecules available for solvation of electrolyte ions which is confirmed by decreasing values of β with increase in temperature. This decrease in adiabatic compressibility with solute concentration as well as with temperature shows presence of prominent ion-solvent interactions in the systems and these interactions increase with increase in solute concentration and with increase in temperature also. At corresponding molality, adiabatic compressibility values of cadmium sulphate are highest in 5% mannitol and least in 25% mannitol following the order as 5% > 15% > 25%. This shows that there are more ion-solvent interactions in 5% mannitol and these interactions increases with increase in solute concentration also.

Intermolecular free length (L_f), for cadmium sulphate was obtained from adiabatic compressibility (β) using equation (2) [18]:

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

Where K is the temperature dependent constant (= (93.875 + 0.375T) × 10^{-8}) [19]. Ultrasonic velocity (U) increases and intermolecular free length (L_f) decreases with increase in concentration of mannitol as well with increase in solvent concentration. In general U and L_f have been reported to vary as the inverse of each other as in the present systems [20, 21]. The decrease in the value of L_f with the increase in solvent concentration indicates the presence of significant ion-solvent interaction between solute and solvent molecules due to which the structural arrangement in the neighbourhood of constituent ions is considerably affected [22]. Order of ion-solvent interactions in different solvents is as 5% > 15% > 25% which is similar to that obtained by adiabatic compressibility. However, the relative magnitude of L_f values for cadmium sulphate increase with the increase in temperature. As temperature increases there is an increase of intermolecular distance, thereby increasing the distance between surfaces of two molecules and thus increasing L_f values.

To obtain a firm impact of interactions in solutions, relative association (R_A) was calculated by following relation (3) [23]:

$$R_A = (\rho/\rho_0)(U_0/U)^{1/3} \qquad (3)$$

where ρ_0 and U_0 are the density and ultrasonic velocity of solvent respectively. Relative association is influenced by two factors (i) breaking up of the associated solvent molecules on addition of solute in it and (ii) the solvation of solute molecules. The former leads to decrease and later to increase of relative association. In the present study, the values of R_A increases with increase in solute concentration as well as with increase in temperature showing significant ion-solvent interactions which increases with increase in solute concentration [24].

Specific acoustic impedance was obtained from relation (4) [25]:

$$Z = U\rho \qquad (4)$$

The mathematical relations for specific acoustic impedance ($Z = U\rho$) and adiabatic compressibility ($\beta = 1/U^2\rho$) show that they must exhibit opposite behaviour and the behaviour is observed in all the studied systems. Specific acoustic impedance (Z) varies directly with temperature because both the speed of sound and the mass density of solutions are highly temperature dependent. Further with increase in concentration of solute, density (ρ) of solution increases which leads to increase in specific acoustic impedance (Z) values with increase in concentration. Relative values of Specific acoustic impedance (Z) for corresponding concentrations at particular temperatures increases with increase in solvent concentration.

Table 1: Ultrasonic velocity (u), density (ρ), and viscosity (η) for cadmium sulphate in 5, 15 and 25% aqueous mannitol at different temperatures

	Molality (mol kg^{-1})	u (m s^{-1})	ρ (kg m^{-3})	η x 10^{-2} (Poise)
		5% Mannitol		
298.15K	**U_0 = 1524.2**	**ρ_0 = 1.014**	**η_0 = 1.065**	
	0.01	1523.8	1.021	1.102
	0.02	1528.4	1.027	1.123
	0.04	1531.8	1.040	1.159
	0.06	1532.6	1.052	1.191
	0.08	1537.0	1.064	1.220
	0.10	1540.8	1.075	1.247
303.15K	**U_0 = 1533.4**	**ρ_0 = 1.011**	**η_0 = 0.952**	
	0.01	1534.0	1.019	0.987
	0.02	1537.8	1.026	1.006
	0.04	1542.2	1.071	1.038
	0.06	1545.8	1.050	1.064
	0.08	1542.4	1.061	1.089
	0.10	1548.4	1.071	1.112
308.15K	**U_0 = 1541.2**	**ρ_0 = 1.009**	**η_0 = 0.853**	
	0.01	1543.6	1.017	0.879
	0.02	1547.4	1.024	0.895
	0.04	1549.8	1.037	0.924
	0.06	1552.4	1.048	0.950
	0.08	1554.2	1.057	0.977
	0.10	1558.6	1.067	1.001
313.15K	**U_0 = 1550.0**	**ρ_0 = 1.005**	**η_0 = 0.748**	
	0.01	1551.4	1.014	0.761
	0.02	1555.0	1.022	0.773
	0.04	1560.8	1.035	0.800
	0.06	1563.6	1.046	0.827
	0.08	1566.6	1.054	0.855
	0.10	1568.8	1.062	0.881
		15% Mannitol		
298.15K	**U_0 = 1556.0**	**ρ_0 = 1.050**	**η_0 = 1.432**	
	0.01	1559.8	1.052	1.479
	0.02	1562.0	1.059	1.501
	0.04	1565.6	1.071	1.535
	0.06	1567.6	1.080	1.564
	0.08	1570.4	1.089	1.588
	0.10	1573.6	1.097	1.612
303.15K	**U_0 = 1565.6**	**ρ_0 = 1.041**	**$_0$ = 1.258**	
	0.01	1569.8	1.049	1.268
	0.02	1571.2	1.055	1.299
	0.04	1574.2	1.067	1.322
	0.06	1577.0	1.077	1.342
	0.08	1579.6	1.085	1.360
	0.10	1583.4	1.093	1.376
308.15K	**U_0 = 1574.4**	**ρ_0 = 1.037**	**η_0 = 1.113**	
	0.01	1578.8	1.045	1.142

	0.02	1580.8	1.051	1.160
	0.04	1584.4	1.064	1.188
	0.06	1585.6	1.074	1.215
	0.08	1589.2	1.082	1.240
	0.10	1593.8	1.090	1.264
313.15K	**U_0 = 1582.6**	**ρ_0 = 1.032**	**η_0 = 0.933**	
	0.01	1587.8	1.042	0.984
	0.02	1588.4	1.050	1.011
	0.04	1590.2	1.062	1.053
	0.06	1593.0	1.072	1.090
	0.08	1595.2	1.079	1.123
	0.10	1601.2	1.085	1.153
		25%mannitol		
298.15K	**U_0 = 1591.2**	**ρ_0 = 1.074**	**η_0 = 1.786**	
	0.01	1594.2	1.080	1.832
	0.02	1597.2	1.087	1.854
	0.04	1600.8	1.098	1.892
	0.06	1604.6	1.109	1.923
	0.08	1608.2	1.119	1.953
	0.10	1610.6	1.128	1.957
303.15K	**U_0 = 1600.8**	**ρ_0 = 1.069**	**η_0 = 1.565**	
	0.01	1604.6	1.077	1.630
	0.02	1607.2	1.083	1.662
	0.04	1610.0	1.095	1.712
	0.06	1614.8	1.106	1.758
	0.08	1617.8	1.116	1.792
	0.10	1620.2	1.124	1.838
308.15K	**U_0 = 1611.2**	**ρ_0 = 1.065**	**η_0 = 1.369**	
	0.01	1611.6	1.073	1.440
	0.02	1614.0	1.081	1.477
	0.04	1617.4	1.093	1.538
	0.06	1622.8	1.104	1.590
	0.08	1626.0	1.113	1.639
	0.1	1626.2	1.121	1.684
313.15K	**U_0 = 1616.8**	**ρ_0 = 1.061**	**η_0 = 1.103**	
	0.01	1619.6	1.070	1.156
	0.02	1622.0	1.078	1.186
	0.04	1624.6	1.091	1.236
	0.06	1626.4	1.102	1.282
	0.08	1632.4	1.110	1.324
	0.10	1633.8	1.117	1.365

Table 2: Adiabatic compressibility (β), inter molecular free length (L_f), relative association (R_A) specific acoustic impedance (Z), molar compressibility (W) and Rao's molar sound function (R) for cadmium sulphate in 5, 15 and 25% aqueous mannitol at different temperatures.

Molality (mol kg^{-1})	**β x 10^{-10} (m^2 N^{-1})**	**L_f x 10^{-11} (m)**	**R_A**	**Z x 10^6 (kg m^{-2}s^{-1})**	**W x 10^{-3} (m^3 mol^{-1}). (m^{2}N^{-1})**	**R x 10^{-4} (m s^{-1}). (m^{3}mol^{-1})**
			5% Mannitol			
298.15K						
0.01	4.2222	4.2264	1.0090	1.5543	0.407	2.1425
0.02	4.1561	4.1931	1.0179	1.5743	0.407	2.1396
0.04	4.0979	4.1637	1.0270	1.5931	0. 409	2.1520

0.06	4.0546	4.1416	1.0367	1.6092	0. 412	2.1629
0.08	3.9934	4.1102	1.0455	1.6292	0. 415	2.1752
0.10	3.9002	4.0620	1.0644	1.6641	0. 414	2.1669
303.15K						
0.01	4.1704	4.2386	1.0088	1.5631	0. 408	2.1494
0.02	4.1215	4.2137	1.0149	1.5778	0. 409	2.1524
0.04	4.0506	4.1773	1.0258	1.6008	0. 411	2.1610
0.06	3.9857	4.1437	1.0368	1.6231	0. 413	2.1691
0.08	3.9506	4.1254	1.0514	1.6411	0. 414	2.1696
0.10	3.8944	4.0960	1.0570	1.6583	0. 418	2.1887
308.15K						
0.01	4.1268	4.2545	1.0074	1.5698	0. 409	2.1581
0.02	4.0784	4.2295	1.0135	1.5845	0. 410	2.1611
0.04	4.0149	4.1964	1.0258	1.6071	0. 412	2.1666
0.06	3.9594	4.1673	1.0361	1.6269	0. 414	2.1763
0.08	3.9018	4.1369	1.0486	1.6490	0. 416	2.1813
0.10	3.8436	4.1059	1.0575	1.6693	0. 419	2.1935
313.15K						
0.01	4.0975	4.2773	1.0086	1.5731	0. 411	2.1681
0.02	4.0466	4.2507	1.0158	1.5892	0. 411	2.1688
0.04	3.9661	4.2082	1.0275	1.6154	0. 413	2.1759
0.06	3.9104	4.1785	1.0378	1.6355	0. 416	2.1857
0.08	3.8954	4.1705	1.0371	1.6387	0. 422	2.2184
0.10	3.8080	4.1235	1.0574	1.6739	0. 421	2.2065
15% Mannitol						
298.15K						
0.01	3.9070	4.0655	1.0011	1.6409	0. 440	2.3117
0.02	3.8703	4.0464	1.0073	1.6542	0. 441	2.3145
0.04	3.8093	4.0144	1.0179	1.6768	0. 444	2.3241
0.06	3.7680	3.9925	1.0260	1.6930	0. 447	2.3392
0.08	3.7235	3.9689	1.0340	1.7102	0. 450	2.3544
0.10	3.6813	3.9464	1.0409	1.7262	0. 454	2.3717
303.15K						
0.01	3.8684	4.0823	1.0068	1.6467	0. 442	2.3232
0.02	3.8396	4.0670	1.0122	1.6576	0. 443	2.3278
0.04	3.7819	4.0364	1.0231	1.6797	0. 446	2.3371
0.06	3.7335	4.0105	1.0321	1.6984	0. 449	2.3504
0.08	3.6938	3.9891	1.0392	1.7139	0. 453	2.3677
0.10	3.6492	3.9649	1.0460	1.7307	0. 456	2.3853
308.15K						
0.01	3.8391	4.1035	1.0068	1.6498	0. 444	2.3366
0.02	3.8075	4.0866	1.0121	1.6614	0. 445	2.3415
0.04	3.7439	4.0523	1.0239	1.6858	0. 448	2.3487
0.06	3.7035	4.0304	1.0332	1.7029	0. 451	2.3612
0.08	3.6594	4.0063	1.0401	1.7195	0. 454	2.3790
0.10	3.6116	3.9801	1.0468	1.7372	0. 458	2.3971
313.15K						
0.01	3.8066	4.1227	1.0086	1.6545	0. 446	2.3477
0.02	3.7748	4.1054	1.0162	1.6678	0. 446	2.3474
0.04	3.7237	4.0775	1.0274	1.6888	0. 449	2.3560
0.06	3.6760	4.0513	1.0365	1.7077	0. 452	2.3693
0.08	3.6421	4.0326	1.0428	1.7212	0. 456	2.3886
0.10	3.5948	4.0064	1.0473	1.7373	0. 461	2.4119

25%mannitol

298.15K						
0.01	3.6433	3.9259	1.0060	1.7217	0. 483	2.5320
0.02	3.6062	3.9059	1.0119	1.7362	0. 485	2.5359
0.04	3.5540	3.8775	1.0213	1.7577	0. 488	2.5492
0.06	3.5021	3.8491	1.0307	1.7795	0. 491	2.5625
0.08	3.4553	3.8233	1.0393	1.7996	0. 494	2.5777
0.10	3.4175	3.8023	1.0471	1.8168	0. 498	2.5943
303.15K						
0.01	3.6062	3.9415	1.0067	1.7282	0. 485	2.5445
0.02	3.5746	3.9242	1.0117	1.7406	0. 487	2.5505
0.04	3.5232	3.8959	1.0224	1.7629	0. 490	2.5611
0.06	3.4674	3.8649	1.0316	1.7860	0. 493	2.5747
0.08	3.4236	3.8404	1.0403	1.8055	0. 496	2.5893
0.10	3.3892	3.8211	1.0472	1.8211	0. 500	2.6086
308.15K						
0.01	3.5883	3.9672	1.0074	1.7292	0. 488	2.5577
0.02	3.5511	3.9466	1.0144	1.7447	0. 488	2.5588
0.04	3.4974	3.9166	1.0250	1.7678	0. 491	2.5697
0.06	3.4395	3.8841	1.0341	1.7916	0. 494	2.5838
0.08	3.3983	3.8608	1.0419	1.8097	0. 498	2.6011
0.1	3.3732	3.8465	1.0493	1.8230	0. 502	2.6189
313.15K						
0.01	3.5629	3.9885	1.0079	1.7330	0. 489	2.5691
0.02	3.5260	3.9678	1.0149	1.7485	0. 490	2.5702
0.04	3.4728	3.9378	1.0266	1.7724	0. 493	2.5782
0.06	3.4305	3.9138	1.0366	1.7923	0. 496	2.5904
0.08	3.3808	3.8853	1.0428	1.8120	0. 500	2.6115
0.10	3.3539	3.8698	1.0491	1.8250	0. 504	2.6323

Rao’s molar sound function (R) and Molar compressibility (W) were also calculated using relations (5) and (6) [26, 27]:

$$R = (M/\rho)U^{1/3} \qquad (5)$$

$$W = (M/\rho)\beta^{-1/7} \qquad (6)$$

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

$$M = M_1W_1 + M_2W_2 \qquad (7)$$

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. Rao's constant and Wada's constant are independent of temperature and the variations of these constants with concentration of solute is linear.

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 (9) [28-30]:

$$\tau = (4\eta/3\rho U^2) \qquad (9)$$

where η is viscosity.

Table 3: Relaxation time (τ) and Gibb's free energy of activation (ΔG^*) for relaxation process for cadmium sulphate in 5, 15 and 25% of aqueous mannitol at different temperatures.

	$\tau \times 10^{-13}$ (s)				$\Delta G^* \times 10^{-20}$ (J mol^{-1})			
				5 % Mannitol				
Molality	**298.15 K**	**303.15 K**	**308.15 K**	**313.15 K**	**298.15 K**	**303.15 K**	**308.15 K**	**313.15 K**
0.01	6.32211	5.48821	4.83657	4.15755	5.6268	5.1990	4.8168	4.3081
0.02	6.22311	5.52828	4.86695	4.17068	5.5618	5.2294	4.8434	4.3218
0.04	6.33264	5.60605	4.94630	4.23052	5.6336	5.2879	4.9122	4.3833
0.06	6.43878	5.65437	5.01526	4.31183	5.7020	5.3238	4.9711	4.4656
0.08	6.49598	5.73629	5.08281	4.44076	5.7384	5.3839	5.0280	4.5929
0.10	6.48469	5.77415	5.12996	4.47317	5.7313	5.4114	5.0672	4.6244
				15 % Mannitol				
0.01	7.70466	6.63309	5.84567	4.99430	6.4380	5.9891	5.6200	5.1006
0.02	7.74571	6.65017	5.88898	5.08840	6.4599	5.9998	5.6514	5.1812
0.04	7.79643	6.66632	5.9304	5.22804	6.4868	6.0100	5.6812	5.2982
0.06	7.85744	6.68055	5.99962	5.34243	6.5188	6.0189	5.7306	5.3918
0.08	7.88389	6.69813	6.05029	5.45339	6.5327	6.0299	5.7664	5.4806
0.10	7.91240	6.69508	6.08684	5.52646	6.5475	6.0280	5.7920	5.5381
				25 % Mannitol				
0.01	8.89928	7.83749	6.88949	5.49158	7.0311	6.6871	6.3187	5.5108
0.02	8.91457	7.92139	6.99337	5.57575	7.0382	6.7316	6.3824	5.5765
0.04	8.96568	8.04223	7.17200	5.72321	7.0617	6.7950	6.4896	5.6893
0.06	8.97952	8.09991	7.29183	5.86395	7.0680	6.8249	6.5601	5.7943
0.08	8.99768	8.18018	7.42645	5.96830	7.0764	6.8661	6.6379	5.8705
0.10	8.99956	8.30578	7.57403	6.10408	7.0772	6.9299	6.7215	5.9677

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 (10) [31]:

$$\Delta G^* = kT \ln(kT\tau/h) \qquad (10)$$

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

The relaxation time increases with increase in concentration of solute and decreases with increase in temperature. The values of ΔG^* are almost constant with increase temperature. The constant values of ΔG^* suggests that the rearrangement of molecules in solution are characteristic of physical properties of solute only [30].

CONCLUSION

Mannitol used in aqueous system as solvent, modify the water structure into more arranged structure due to formation of hydrogen bonds. $CdSO_4$ added as solute produces free solvent molecules by breaking of hydrogen bonds of the solvent system which is inferred by increase in ultrasonic velocity and decrease in adiabatic compressibility with concentration which shows that solute-solute interactions are smaller than solute-solvent interactions. These structural modifications in solutions are further conformed by the decreasing values of intermolecular free length with concentration. The ΔG^* value were almost constant suggesting structural modifications are characteristic of physical properties of solute only.

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