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Molecular Interactions and Aggregation Behavior of Cloxacillin Sodium in Water and Aqueous NaCl Solutions through Volumetric and Ultrasonic Sound Velocity Studies at 298.15 K

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ABSTRACT

Molecular interactions and aggregation behavior of cloxacillin sodium (CloNa) in water and aqueous NaCl solutions was studied through volumetric and ultrasonic sound velocity method at 298.15 K and atmospheric pressure. Density, ρ, and ultrasonic sound velocity, u, of cloxacillin sodium in water and aqueous NaCl solutions were measured with a digital density and ultrasonic sound velocity analyzer (DSA 5000, Anton Paar, Austria). And from the investigational data, apparent molar volume, φ_v, adiabatic compressibility, φ_S, apparent molar adiabatic compressibility, φ_k, limiting apparent molar volume, φ_v^0, limiting apparent molar adiabatic compressibility, φ_k^0, and aggregation concentration, CMC of cloxacillin sodium was calculated. The outcomes indication exhibits very significant evidence about the interactions among solute-solvent-co-solute (solute-solute, solute-solvent) and aggregation behavior in the aqueous environment and this result would be useful for the drug action in human body with pharmacological applications.

Keywords: Cloxacillin sodium, NaCl, apparent molar volume, adiabatic compressibility, apparent molar adiabatic compressibility, solute-solvent, solute-solute interactions, aggregation.

INTRODUCTION

In pharmacokinetics, drug is a very essential chemical material that used for the treatment of various disease such as therapy, prevention/diagnosis, develop mental well-being[1] etc. To recognize the drug action in human body at the molecular level, the physicochemical properties of drugs are very important[2]. By studying the physicochemical properties the binding possibility of biologically active molecules in presence of several solvents or co-solutes can be well examine[3].

Cloxacillin sodium (CloNa) is a sodium salt of cloxaclliin and semi synthetic beta-lactamase penicillin antibiotic having remarkable antibacterial actions. Cloxacillin Sodium (CloNa) shows[4–6] noticeable vulnerability to acid and base reagents, several nucleophiles, oxidizing agents, metal ions, and many solvents and is broadly used in the treatment of blood poisoning, parenchyma of skin infection, pneumonia, etc.[7].

Volumetric[2,8–12] and ultrasonic sound velocity[13,14] technique is very convenient and reliable tool to well investigation in case of drugs, amino acids, polymers etc analysis. Thermodynamic properties such as, φ_v, φ_k and aggregation concentration of any solution are very crucial parameter to recognize the solute–solvent and solute–solute interactions[15].

For cloxacillin Sodium (CloNa), there are some reports on the stability of solid CloNa, degradation[16], interactions with protein[17], Taboada[18] examined aggregation behavior of CloNa in aqueous and aqueous NaCl with light-scattering, NMR techniques. However, there is still a lack of knowledge about the interactions among solute-solvent-co-solute and aggregation behavior of CloNa in water or electrolyte solvents. In this background, this work presents a study on the solute-solvent, solute-solute interactions and aggregation behavior or properties of cloxacillin sodium (CloNa) in water and aqueous NaCl solutions through volumetric and ultrasonic velocity method. In this study, NaCl (co-solute) is taken because it is the chief electrolyte in the extracellular fluid19 in human body rather than other ions, such as Ca^+, K^+, Mg^+, $HPO4^-$, and $HCO3^-$. The range of Na+ in human body is ~92% in case of over-all positive ions and Cl^- is ~68 % for negative ions.

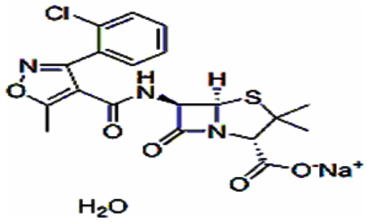


Fig. 1. Structure of Cloxacillin Sodium (CloNa)

To understand the complete interactions of CloNa with electrolytes, density, ρ, and ultrasonic sound velocity, *u* of CloNa in water and NaCl aqueous solutions was measured at 298.15 K and atmospheric pressure. From density and ultrasonic sound velocity data, apparent molar volumes, ϕ_V, adiabatic compressibility, β_S, apparent molar adiabatic compressibility, ϕ_K, their parallel limiting parameters, and aggregation concentrations (CMC) were calculated. The tremendous results are represented in our paper.

EXPERIMENTAL

Materials

CloNa and sodium chloride were used in this study and collected from Beximco Pharmaceuticals Limited (Bangladesh), and Loba Chemie Pvt. Ltd. (India) respectively. The chemicals specifications are given below in Table 1 that used in this investigation.

Density and Ultrasonic Sound Velocity Measurement

Each solution prepared instantly with highly purified water. Because water was redistilled and degassed with specific conductance of $<10^{-6}$ S (cm^{-1}). By weighing, solution was prepared in molality using a balance (B204-S, Mettler Toledo, Switzerland) having a precision of ± 0.0001 g. Molality uncertainty for solutions is up to $\pm 2 \cdot 10^{-5}$(mol kg^{-1}) . An analyzer (DSA 5000, Anton Paar, Austria) was used to examine density, ρ, and ultrasonic sound velocity, u, of the solutions. A density check carried out at 20°C with a) distilled and degassed water, b) dry air. For each measurement, the density meter calibrated with redistilled and gas free water in the investigational temperature area. Temperature sensitivity was controlled by a built-in Peltier method to $\pm 1 \cdot 10^{-3}$ K. The instrument sensitivity for density with respect to an accuracy is $1 \cdot 10^{-3}$(kg m^{-3}) and for ultrasonic sound velocity $1 \cdot 10^{-2}$(m s^{-1}). The density and ultrasonic sound velocity uncertainty was obtained $\pm 5 \cdot 10^{-3}$ kg m^{-3} & $\pm 5 \cdot 10^{-2}$ m s^{-1}, respectively.

Table 1: Chemicals Specifications

Chemical title	Molar mass/(kg•mol⁻¹)	Claritydeclared byprovider	Origin
Cloxacillin sodium(CloNa)	0.457864	mass fraction,≥ 0.990	Beximco Pharmaceuticals Limited (Bangladesh)
NaCl	0.058442	mass fraction,≥ 0.995	Loba Chemie Pvt. Ltd(India)

RESULTS AND DISCUSSION

The measured density ρ & ultrasonic sound velocity u of CloNa (as a function of molality) in water and aqueous sodium chloride solution at different concentration are taken in Table 2. The apparent molar volume, φ_V, adiabatic compressibility, β_S and apparent molar adiabatic compressibility, φ_K calculated from measured density ρ and ultrasonic sound velocity u using equation (1), (2) & (3) are shown in Table 2.

$$\phi_V = \frac{(\rho_0 - \rho)}{m\rho\rho_0} + \frac{M_2}{\rho} \quad (1)$$

$$\beta_S = \frac{1}{\rho u^2} \quad (2)$$

$$\phi_K = \frac{\beta_S\rho_0 - \rho\beta_S^0}{m\rho\rho_0} + \frac{\beta_S M_2}{\rho} \quad (3)$$

where

m = molality, ρ = density of solution, β_S = adiabatic compressibility, ρ_0 = density of solvent, β_S^0 = adiabatic compressibility and $M2$ = molar mass.

The reported standard combined uncertainties in m, φ_V, φ_K, φ_v^0, φ_k^0 and CMC were calculated based on first order Taylor series approximation with the help of their equivalent error equations of the following type,

$$\Delta Y = \sqrt{\left(\frac{\partial y}{\partial x_1}\Delta x_1\right)^2 + \cdots + \left(\frac{\partial y}{\partial x_n}\Delta x_n\right)^2} \quad (4)$$

where,

$\Delta x_1, \Delta x_2 \ldots \Delta x_n$ = standard errors,

$x_1, x_2 \ldots x_n$ =independent variables

The changes of apparent molar volume, φ_V and apparent molar adiabatic compressibilities, φ_k of CloNa with respect to molality at 298.15K are shown in Fig. 2(a) and 2(b). The values of φ_V, φ_k are increased with respect to CloNa concentration, indicate the strong solute-solute interaction. At a certain point the increasing rate is changed and shows the break point. To investigate the properties i.e. solute-solvent, solute-solute interaction and aggregation behavior, φ_v^0, φ_k^0 and CMC are very useful and these parameters can be obtained by regression of φ_V and φ_k vs molality data to piece-wise linear model of the following type:

$$\left.\begin{array}{ll} y = y_1^0 + b_1 m & when\ m < X \\ y = y^0 + b_1 X + b_2(m - X) & when\ m \geq X \end{array}\right\} \quad (5)$$

where

$y = \varphi_V$ or φ_K,

m = molality,

X = CMC,

y^0 = limiting parameter at infinite dilution φ_v^0or φ_k^0,

b= slopes Sv and Sk,

Subscripts 1 = for pre-CMC region,

Subscripts 2= for post-CMC region,

Fitting parameters with their standard uncertainties are presented in Table 3.

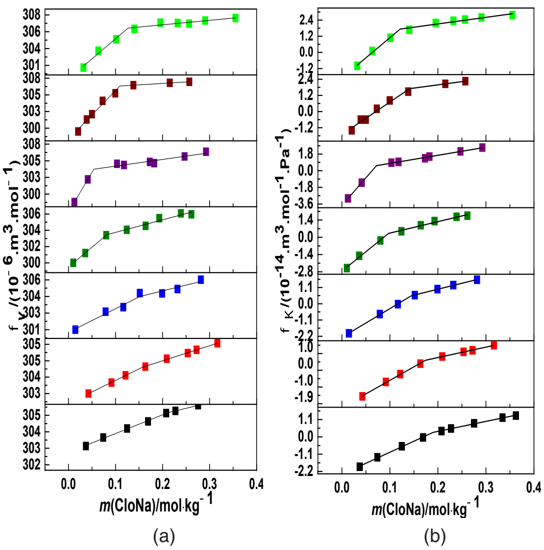
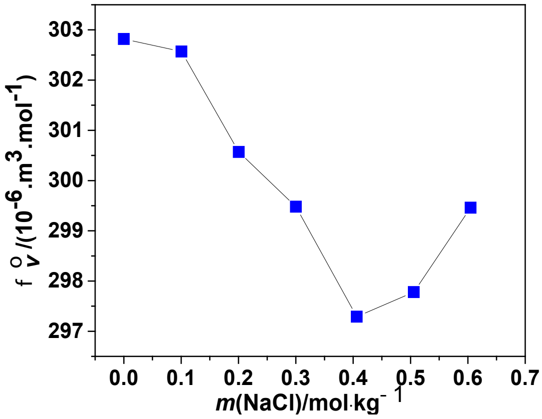


Fig. 2. The variation of (a) φ_v and (b) φ_k values of CloNa in m = (■-0.000, ■-0.10049, ■-0.0586; ■-0.30012, ■-0.40623, ■-0.50572, ■-0.60480) M aqueous NaCl solutions at 298.15 K

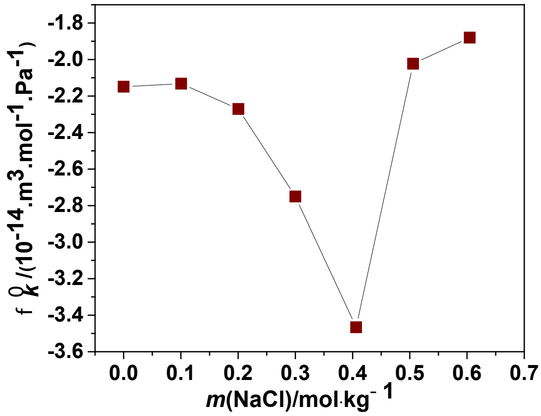
Solute-Solvent Interactions

The calculated φ_v^0, & φ_k^0 indicates the interactions between solute and solvent molecules (solute-solvent). The changes of φ_v^0 of CloNa with respect to NaCl molality at various temperatures are shown in Fig. 3(c). The values of φ_v^0 & φ_k^0 are goes to minima with the concentration of sodium chloride that is sign for the presence of strong interactions among solute, solvent and co-solute in the mixtures.

According to the configuration of CloNa (Fig. 1), the probable interactions of CloNa with NaCl are(a) two types ion–ion interactions such as i) COO–(CloNa) with Na^+(NaCl) ii) Na^+(CloNa) with Cl^- (NaCl), (b) Ion–hydrophilic interactions of Na^+(NaCl) with the hydrophilic groups (O, F, N, S and Cl) of CloNa, and(c) Ion–hydrophobic interactions of Na^+(NaCl) with the hydrophobic groups (alkyl groups, benzene ring) of CloNa. The co-sphere overlap model[20] recommends that (a) and (b) type interactions gives positive contribution to φ_v^0, however type (c) gives an opposite contribution to φ_v^0, i.e. negative. The decrease of φ_v^0values with respect to increase in NaCl concentration suggests that at the beginning the ion– hydrophobic interaction exceeds over the ion– hydrophilic and ion–ion. After that ion–hydrophilic, ion–ion interaction exceeds owing to the increasing interactions with excess co-solute (NaCl) molecules. The variations of φ_k^0 of CloNa against the concentration of NaCl are shown in Fig. 3(d). The φ_k^0 values are more negative at the minima with the rising of NaCl concentration, indicates water clustering occurrences by the increasing of hydrogen bond. Allowing the Frank and Wen[21] model, around the ions (low-charge density) water molecules are randomly distributed and bulk water molecules arranged orderly. The water molecules are more compressible as a monomeric form. The O, F, N, S and Cl (hydrophilic groups) of FluNa, have weak partial charges and (b) type interaction gives monomeric water molecules to bulk due to relaxation, where they form cluster water molecules and gives a negative role to φ_k^0. This is also supported well-accommodated aromatic rings[22] in electrolyte solution instead of pure water. After minima, the magnitude of φ_k^0 increase i.e, compressibility increases due to dominate (a) type interactions.



(c)



(d)

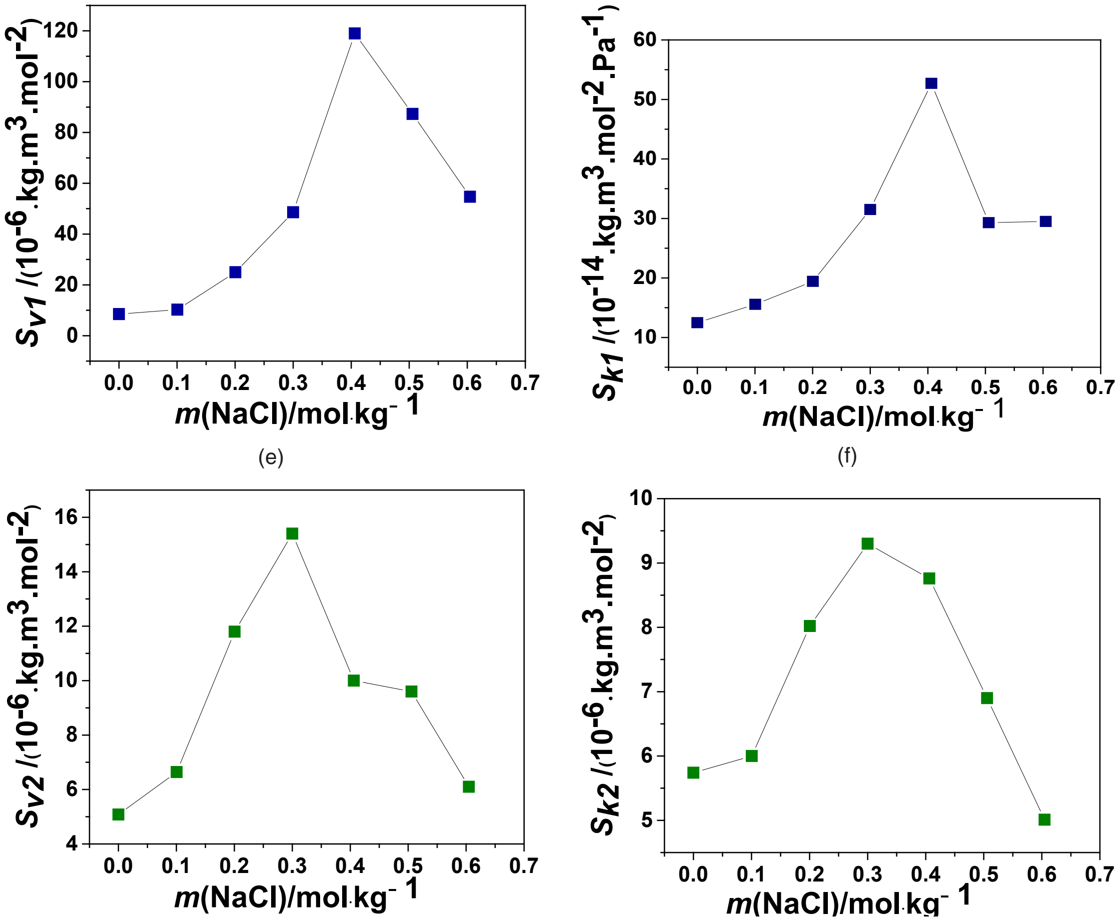
Fig. 3. The variation of φ_v^0 (c) and φ_k^0 (d) of CloNa with respect to molality of NaCl at 298.15K

Solute-Solute Interactions

The interactions of solute with solute (solute-solute) in terms of slope S_v and S_k are displayed in Fig. 4 with respect to NaCl molality at 298.15 K. The slopes S_{v1}, S_{k1} (pre- CMC) and S_{v2}, S_{k2} (post-CMC) goes to the maxima. It is clearly declaring that S_{v1}, S_{k1} provide information about the interactions between CloNa molecules, while S_{v2}, S_{k2} deliver about the micelle CloNa interactions. The S_{v1}, S_{k1}, S_{v2} and S_{k2} values show strong interactions among the CloNa-CloNa molecules and micelle-micelle. And increasing trend with the NaCl concentration may be due to decreasing electrostatic repulsion among their head group charge. But for decreasing trend, the exact interpretation is difficult. However, in case of pre-CMC slopes there may be increase electrostatic repulsion among their head group charge due to excess co-solute (NaCl) molecule interaction and for post-CMC slopes transition happen from spherical micelles to other geometrical shapes[23-25].

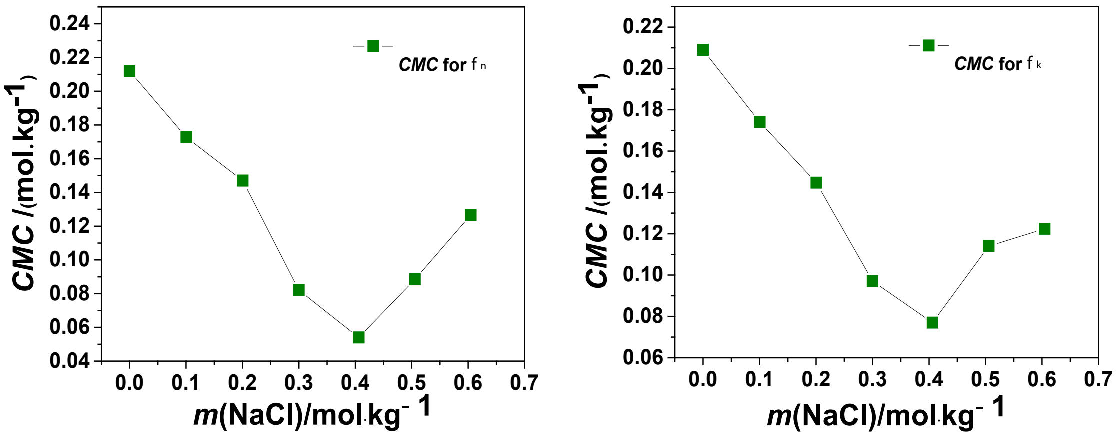
Aggregation Behaviors

The piecewise linear regression model is used for determining CMC point as shown in equation 5. X is the critical micelle concentration. The variation of CMC values of CloNa are presented in Fig. 5 with respect to NaCl molality. Previously CMC values reported by light scattering[18,26] and NMR[18] techniques in water and aqueous NaCl solutions exhibits a near agreement with these CMC values. It is observed from Fig. 5(i) and 5(j) that the CMC values attained from φ_V-m, φ_k-m data show similar action to NaCl concentration. The gradually decreasing CMC values acquire a minimum value and then increasing with the increasing NaCl concentration. The decreasing trend is owing to the decreasing of electrostatic repulsion between the polar head groups (O, F, N, S and Cl) in presence of NaCl. And after that at the higher concentration of NaCl, the transition of spherical micelle[25] to cylindrical micelle is responsible for increasing CMC values. Because at higher salt concentrations, solute molecules are packed compactly and they successfully adopt a more cylindrical shape and cylindrical shape contain additional solute molecules rather than spherical that is form at upper solute concentrations.



(g) (h)

Fig. 4. The variation of slopes of CloNa with respect to NaCl molality at 298.15K. The figure (e), (f) represent pre-CMC and (g), (h) represent post-CMC slopes. The presented slopes are achieved from φ_v and φ_k



(i) (j)

Fig. 5. The CMC of CloNa attained from (i) φ_v and (j) φ_k with respect to NaCl concentration at T = 298.15K

Table 2: The Density, ρ, and Ultrasonic Sound Velocity, u, Apparent Molar Volume, φ_v , Adiabatic compressibility, β_s, Apparent molar Adiabatic compressibility, φ_k of CloNa in Water and Aqueous NaCl Solutions at 298.15 K and Atmospheric Pressurea

m/mol·kg^{-1}	ρ/kg m^{-3}	u/m s^{-1}	φ_V/10^{-6} m^3 mol^{-1}	β_s/10^{-10}. Pa^{-1}	φ_K/10^{-14} m^3 mol^{-1} Pa^{-1}
			Cloxa+water		
0.00000	994.069	1519.98	-	4.3542±0.0006	-
0.03709	999.78	1525.15	303.10±0.20	4.3000±0.0006	-1.70±0.20
0.07391	1005.293	1529.84	303.49±0.08	4.2503±0.0006	-1.20±0.10
0.12465	1012.67	1535.36	303.90±0.06	4.1890±0.0005	-0.60±0.06
0.16916	1018.936	1539.36	304.22±0.04	4.1416±0.0005	-0.04±0.05
0.20779	1024.205	1542.20	304.60±0.04	4.1052±0.0005	0.45±0.04
0.22741	1026.844	1543.90	304.70±0.03	4.0856±0.0005	0.57±0.04
0.27564	1033.193	1547.89	304.96±0.00	0.0000±0.0000	0.84±0.00
0.33466	1040.711	1552.10	305.23±0.02	3.9887±0.0005	1.19±0.02
0.36240	1044.138	1553.97	305.40±0.02	3.9660±0.0005	1.34±0.02
			Cloxa+water+NaCl(0.10049m)		
0.00000	998.077	1525.87	-	4.3033±0.0006	-
0.04265	1004.609	1531.81	303.00±0.20	4.2422±0.0006	-1.50±0.20
0.09145	1011.840	1537.39	303.50±0.10	4.1814±0.0005	-0.67±0.09
0.12129	1016.131	1540.28	303.83±0.07	4.1481±0.0005	-0.22±0.07
0.16344	1022.045	1543.73	304.23±0.05	4.1057±0.0005	0.38±0.05
0.20903	1028.260	1547.37	304.59±0.04	4.0617±0.0005	0.79±0.04
0.25362	1034.172	1550.95	304.85±0.03	4.0199±0.0005	1.06±0.03
0.27238	1036.599	1552.44	305.00±0.03	4.0028±0.0005	1.15±0.03
0.31669	1042.225	1555.38	305.30±0.03	3.9661±0.0005	1.44±0.03
			Cloxa+water+NaCl(0.20058m)		
0.00000	1002.042	1531.63	-	4.2541±0.0006	-
0.01466	1004.329	1533.79	300.90±0.40	4.2325±0.0006	-2.00±0.60
0.07899	1013.985	1541.70	302.74±0.10	4.1492±0.0005	-0.70±0.10
0.11657	1019.421	1545.22	303.20±0.06	4.1083±0.0005	-0.03±0.07
0.15175	1024.219	1548.05	304.64±0.05	4.0742±0.0005	0.58±0.05
0.19914	1030.754	1551.77	304.61±0.04	4.0289±0.0005	0.99±0.04
0.23176	1035.045	1554.16	305.06±0.03	3.9999±0.0005	1.26±0.03
0.28156	1041.314	1557.65	306.02±0.03	3.9580±0.0005	1.62±0.03
			Cloxa+water+NaCl(0.30012m)		
0.01027	1005.835	1537.12	-	4.2078±0.0005	-
0.03577	1007.442	1538.76	300.00±0.80	4.1922±0.0005	-2.50±0.70
0.08030	1011.349	1542.23	301.20±0.00	0.0000±0.0000	-1.50±0.00
0.12389	1017.875	1547.00	303.40±0.10	4.1051±0.0005	-0.26±0.09
0.16407	1024.089	1550.85	304.06±0.07	4.0600±0.0005	0.48±0.06
0.19320	1029.646	1554.01	304.55±0.05	4.0217±0.0005	0.97±0.04
0.23903	1033.459	1556.09	305.49±0.04	3.9961±0.0005	1.31±0.04
0.26144	1039.426	1559.37	306.08±0.03	3.9565±0.0005	1.66±0.03
0.01027	1042.369	1561.01	305.97±0.03	3.9370±0.0005	1.75±0.03
			Cloxa+water+NaCl(0.40623m)		
0.00000	1009.984	1543.16	-	4.1578±0.0005	-
0.01265	1011.97	1545.25	298.80±0.60	4.1384±0.0005	-2.80±0.60
0.04111	1016.243	1549.00	302.20±0.20	4.1011±0.0005	-1.30±0.20
0.10324	1025.172	1554.15	304.54±0.07	4.0385±0.0005	0.86±0.07
0.11808	1027.298	1555.49	304.37±0.06	4.0232±0.0005	0.96±0.06
0.17294	1034.861	1560.17	304.82±0.04	3.9699±0.0005	1.34±0.04
0.18248	1036.192	1560.43	304.64±0.04	3.9634±0.0005	1.53±0.04
0.24694	1044.550	1564.40	305.65±0.03	3.9118±0.0005	2.09±0.03
0.29255	1050.206	1566.47	306.35±0.03	3.8804±0.0005	2.50±0.03

			Cloxa+water+NaCl(0.50572m)		
0.00000	1013.645	1548.47	-	4.1144±0.0005	-
0.02042	1016.819	1551.41	299.50±0.40	4.0860±0.0005	-1.50±0.30
0.03887	1019.579	1553.68	301.30±0.20	4.0631±0.0005	-0.80±0.20
0.04994	1021.205	1555.04	302.10±0.10	4.0495±0.0005	-0.60±0.10
0.07263	1024.417	1557.18	304.10±0.10	4.0257±0.0005	0.20±0.10
0.09956	1028.18	1559.41	305.24±0.07	3.9995±0.0005	0.82±0.07
0.13802	1033.397	1562.24	306.45±0.05	3.9649±0.0005	1.46±0.05
0.21541	1043.699	1567.77	306.81±0.03	3.8982±0.0005	2.06±0.03
0.25704	1049.04	1570.61	306.96±0.03	3.8643±0.0005	2.26±0.03
			Cloxa+water+NaCl(0.60480m)		
0.00000	1017.54	1554.11	-	4.0690±0.0005	-
0.03186	1022.407	1558.55	301.00±0.50	4.0266±0.0005	-1.00±0.20
0.06342	1026.985	1561.87	303.30±0.20	3.9916±0.0005	0.10±0.10
0.10141	1032.314	1564.92	304.84±0.10	3.9555±0.0005	1.06±0.07
0.14013	1037.516	1567.70	306.28±0.07	3.9217±0.0005	1.68±0.05
0.19528	1044.759	1571.46	307.14±0.05	3.8759±0.0005	2.19±0.04
0.23221	1049.57	1574.03	307.09±0.04	3.8456±0.0005	2.35±0.03
0.25649	1052.689	1575.68	307.01±0.03	3.8262±0.0005	2.44±0.03
0.29076	1056.876	1577.87	307.42±0.02	3.8004±0.0005	2.60±0.03
0.35519	1064.60	1582.11	307.77±0.02	3.7527±0.0005	2.80±0.02

[a]Uncertainties(u) are u(ρ) = $5•10^{-3}$(kg m^{-3}), u(u) = $5•10^{-2}$(m s^{-1}) , u(T) = 0.001 K and standard combined uncertainty(uc) in molality, uc(m) ≤ $2•10^{-5}$(mol kg^{-1})

Table 3: Fitting values or parameters of eq 5 for and with respect to NaCl molality at 298.15 K and Atmospheric Pressurea

m/mol.kg^{-1} (NaCl)	φ_v^0	S_{v1}	S_{v2}	CMC φ_v	φ_k^0	S_{k1}	S_{k2}	CMC φ_k
0.00000	302.82±0.03	8.52±0.24	5.08±0.32	0.212±0.012	-2.15±0.01	12.49±0.10	5.74±0.14	0.209±0.003
0.10049	302.57±0.02	10.24±0.21	6.64±0.24	0.173±0.008	-2.13±0.04	15.56±0.38	6.00±0.43	0.174±0.006
0.20058	300.57±0.39	25.00±4.80	11.80±3.70	0.147±0.040	-2.27±0.04	19.43±0.46	8.02±0.35	0.145±0.004
0.30012	299.48±0.25	48.60±4.90	15.40±2.20	0.082±0.011	-2.75±0.11	31.50±2.20	9.30±1.00	0.097±0.008
0.40623	297.29±0.42	119.00±14.00	10.00±1.70	0.054±0.005	-3.47±0.08	52.70±2.60	8.76±0.32	0.077±0.003
0.50572	297.78±0.42	87.30±8.50	9.60±2.60	0.089±0.006	-2.02±0.09	29.30±1.50	6.90±1.10	0.114±0.006
0.60480	299.46±0.40	54.70±5.60	6.10±1.70	0.127±0.009	-1.88±0.16	29.50±2.30	5.01±0.67	0.122±0.007

[a]Units are m/(mol kg^{-1}),/(10^{-6} m^3 mol^{-1}), S_{v1} (or S_{v2})/10^{-6}(kg m^3 mol^{-2}),/10^{-14}(m^3 mol^{-1} Pa^{-1}) , S_{k1}(or S_{k2})/10^{-14}(kg m^3 mol^{-2} Pa^{-1}), and CMC/(mol kg^{-1})

CONCLUSION

This work reports experimental information about the molecular interactions and aggregation behavior of cloxacillin sodium in water and in sodium chloride solution at 298.15K. From the measurement apparent molar volume, φ_V, apparent molar adiabatic compressibility, φ_k, limiting apparent molar volume, φ_v^0 , limiting apparent molar adiabatic compressibility, φ_k^0 , experimental slopes S_v, S_k and critical micelle concentration (CMC) of cloxacillin sodium were calculated and explain in terms of solute-solute, solute-solvent interactions and aggregation behavior. The φ_v^0 decreases with the increase in NaCl concentration due to the disruption of hydrophobic hydration layer of cloxacillin sodium due to ion-hydrophobic interactions and increases for the ion-ion, ion-hydrophilic interactions. The φ_k^0 shows same trend with respect to increasing NaCl concentration which supports φ_v^0. At first critical micelle concentration were found decreasing with respect to NaCl concentration due to minimization of electrostatic repulsion among the charged head groups, and increase in CMC at higher NaCl concentration is due to shape changes of micelles from spherical to different new, i.e cylindrical rod. So, apparent molar volume, limiting apparent molar volume and CMC gives us appreciated information about solute-solvent, solute-solute interaction and aggregation actions, which is very important for our body plasma co-solute interactions. Various ultrasound parameters support the existence of strong solute-solute, solute-solvent interactions and micelle phenomena. The results obtained from these studies can thus be helpful for pharmacological application of drugs.

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Conflict of Interest

The researchers declare that they have no conflict of interest.

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