

# Sound Velocities and Micellar Behaviour Studies of Dodecyltrimethylammonium Bromide in Aqueous Solutions at 295.15, 302.15 and 309.15 K

Md. Harun-Al-Rashid<sup>1</sup>, Tania Tofaz<sup>1</sup>, M. M. Islam<sup>2</sup>, T. K. Biswas<sup>2</sup>

<sup>1</sup>Department of Chemistry, Begum Rokeya University, Rangpur-5400, Bangladesh

<sup>2</sup>Department of Chemistry, University of Rajshahi, Rajshahi-6205, Bangladesh

Correspondence: Md. Harun-Al-Rashid, Department of Chemistry, Begum Rokeya University, Rangpur-5400, Bangladesh. Tel: 880-191-534-2452, E-mail: harun.brur.bd@gmail.com

Received: October 21, 2015 Accepted: October 22, 2015 Online Published: October 30, 2015

doi:10.5539/ijc.v7n2p213

URL: <http://dx.doi.org/10.5539/ijc.v7n2p213>

## Abstract

Speed of sound,  $u$ , of dodecyltrimethylammonium bromide (DTAB) aqueous solutions has been determined at 295.15, 302.15 and 309.15 K. The speeds of sound,  $u$ , data have been used to estimate isentropic compressibility,  $K_s$ , apparent molal isentropic compressibility,  $\phi_{k(s)}$ , acoustic impedance,  $Z$ , molal sound of speed,  $R_m$ , and relative association,  $R_A$ , of DTAB in aqueous solution. These parameters have been interpreted in terms of solute-solute and solute-solvent interactions and micellar behavior of DTAB.

**Keywords:** surfactant, critical micelle concentration, isentropic compressibility, solute-solute and solute-solvent interactions

## 1. Introduction

Surfactants are amphiphilic molecules consisting of polar (hydrophilic) and non-polar (hydrophobic) parts. Due to their dual character (hydrophilic and hydrophobic) surfactants are employed in different industrial applications (Herzog et al., 1998). In recent years, there has been growing interest in the interactions between protein and surfactant due to their many applications in biosciences, foods and cosmetics, drug delivery, detergency and biotechnological processes. Using various numbers of tools and techniques, these interactions have been studied and published in the past few years (Goddard et al., 1993, Brash et al., 1995, Sonesson et al., 2008, Mehta et al., 2008, Verdes et al., 2008, Mehta et al., 2009). Cationic surfactants are widely used in industry and have a great bearing on our day-to-day life (Rosen, 1989). The dodecyltrimethylammonium bromide (DTAB) is bio-degradable, non-toxic and is widely available material at low costs. Survey of available literature reveals that no serious attempt has been made to study the micellization phenomenon of dodecyltrimethylammonium bromide (DTAB) surfactant in aqueous solvent. The aim of the present work is to analyze the influence of the presence of dodecyltrimethylammonium bromide (DTAB) aqueous solutions upon several physical properties that allow the characterization of this kind of solutions.

Studies on sound velocities of surfactants solutions are of great use in characterizing the structure and properties of solutions. Also compressibility is a powerful thermodynamic parameter for elucidating the behaviour of a solute in a solvent. (Chalikian et al., 1994). Various types of interactions exist between the solutes in solutions, and these solute-solute and solute-solvent interactions are of current interest in all branches of chemistry. These interactions provide a better understanding of the nature of the solute and solvent, i.e., whether the solute modifies or distorts the structure of the solvent. In the present work, an attempt has been made to provide an interpretation of solute-solvent and solute-solute interactions prevailing in the studied cationic surfactant dodecyltrimethylammonium bromide (DTAB) aqueous solutions as well as findings the micellar behaviour at the temperatures  $T=(295.15$  to  $309.15)$  K.

## 2. Experimental

The surfactant used in this study was dodecyltrimethylammonium bromide,  $\text{CH}_3(\text{CH}_2)_{11}\text{N}^+(\text{CH}_3)_2\text{CH}_3\text{Br}$  (molar purity  $\geq 98\%$ , Fluka AG, Switzerland). Supplied distilled water was redistilled and deionized by passing through two ion exchange columns. The deionized water was distilled again in alkaline  $\text{KMnO}_4$  medium and used for

preparation of solution. Conductivity of this deionized water was found to be about  $4.00 \mu\text{-S}$ . The electric balance with an accuracy of  $\pm 0.0001\text{g}$  was used for weighting.

Sound velocities of DTAB in aqueous solutions were measured using high precision vibrating tube digital density meter (DSA-5000, Anton Paar, Austria). The accuracies in sound velocity was found to be  $0.1\text{m}\cdot\text{s}^{-1}$  respectively. The method is based on the principle of time lapse measurement for certain number of oscillations of a vibrating U-shaped sample tube filled with the sample liquid. The temperature of the sample tube is controlled by two integrated in-built  $100 \Omega$  Pt resistance thermometers to a level of highest accuracy and traceable to national standards. The temperature of the sample tube is controlled to  $\pm 0.001\text{K}$ . The conductivity measurements were carried out on a Laboratory Conductivity Meter (Model 4310 Jenway), which based on the resistance of the solution in a conductivity cell.

### 3. Results and Discussion

#### 3.1 Speed of Sound

Speed of sound,  $u$ , and densities,  $\rho$ , of DTAB in water have been measured at five equidistance temperatures ranging from 295.15 K to 309.15 K and given in Table 1. The variation of sound velocities of DTAB in aqueous solutions against molality is graphically shown in Figure 1. The increase or decrease in speed of sound depends on the structure properties of solute (Pandey et al., 1997). The values of sound velocities are found to increase with increases in temperature and solute concentration. The rising trend in the speed of sound is due to cohesion brought about by hydration. The water molecules are attached to solute by electrostatic effect which brings about the shrinkage in the volume of solvent caused by ionic portion of solute is increased in solvent in comparison to that of water. This implies that the system under consideration behaves like structure maker but up to a specific concentration. Such an increase may be attributed to an increase in the intermolecular interaction between the water and DTAB.

Table 1. Densities of water,  $\rho^0$ , aqueous DTAB solutions,  $\rho$ , and Speed of sound of water,  $u_0$ , aqueous DTAB solutions,  $u$ , at T= (295.15, 302.15 and 309.15) K

$m$ /mol·kg <sup>-1</sup>	295.15 K		302.15 K		309.15 K	
	$\rho$ /g·cm <sup>-3</sup>	$u$ /cm·s <sup>-1</sup>	$\rho$ /g·cm <sup>-3</sup>	$u$ /cm·s <sup>-1</sup>	$\rho$ /g·cm <sup>-3</sup>	$u$ /cm·s <sup>-1</sup>
0.00000	0.997837	1488.23	0.996000	1506.51	0.993735	1521.39
0.00102	0.997861	1488.32	0.996023	1506.68	0.993756	1521.57
0.00300	0.997905	1488.77	0.996064	1507.08	0.993793	1521.92
0.00502	0.997946	1489.26	0.996103	1507.52	0.993829	1522.29
0.00697	0.997984	1489.71	0.996138	1507.90	0.993861	1522.62
0.00895	0.998022	1490.20	0.996175	1508.31	0.993895	1522.96
0.01093	0.998061	1490.76	0.996212	1508.71	0.993928	1523.31
0.01311	0.998105	1491.33	0.996254	1509.26	0.993965	1523.76
0.01493	0.998141	1491.72	0.996288	1509.63	0.993996	1524.12
0.01692	0.998181	1491.78	0.996318	1509.68	0.994028	1524.22
0.01846	0.998206	1491.81	0.996342	1509.70	0.994049	1524.24
0.02101	0.998242	1491.87	0.996375	1509.73	0.994080	1524.27
0.02312	0.998275	1491.93	0.996404	1509.75	0.994108	1524.28
0.02501	0.998304	1491.97	0.996431	1509.77	0.994132	1524.30
0.02703	0.998331	1492.01	0.996456	1509.80	0.994153	1524.29
0.02893	0.998357	1492.03	0.996474	1509.82	0.994172	1524.28
0.03112	0.998379	1492.09	0.996489	1509.89	0.994188	1524.35

\*Uncertainty value of speed of sound is  $\pm 0.1\text{m}\cdot\text{s}^{-1}$

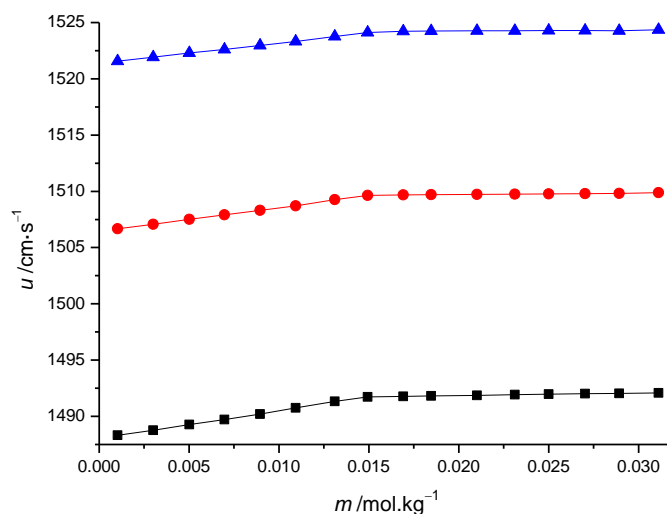


Figure 1. Plots of speed of sound versus molality of aqueous DTAB solutions at studied temperatures: (■) 295.15 K, (●) 302.15 K, (▲) 309.15 K

### 3.2 Isentropic Compressibility

Density,  $\rho$ , and Speed of sound,  $u$ , measurements are combined to calculate isentropic compressibility,  $K_s$ , using the Laplace equation (Pandey et al., 2000)

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

The isentropic compressibility,  $K_{(s)}$ , against molality of DTAB are shown in Table 2 and the graphical representations  $K_{(s)}$  versus  $m$  data are shown in figure 2. The value of isotropic compressibility decreases with increases of concentration. The decrease in isentropic compressibility is due to the increase in electrostriction compression of solvent around the molecules at which results in a large decrease in the compressibility of solvent (Riyazuddin et al., 2009)

The apparent molar isentropic compressibility,  $\varphi_{k(s)}$ , is calculated from speed of sound measurements,

$$\varphi_{k(s)} = \frac{10^3(K_s \rho^0 - K_s^0 \rho)}{m \rho_0 \rho} + K_s M / \rho \quad (2)$$

Where  $K_s$  and  $K_s^0$  are the isentropic compressibility of the solution and solvent, and  $\rho$  and  $\rho_0$  is the density of the solution and solvent, respectively and  $M$  is the molar mass. The apparent molal isentropic compressibility,  $\varphi_{k(s)}$ , against molality of DTAB is shown in Table 2. Table 2 reveals that isentropic compressibility decreases but apparent molal isentropic compressibility increase with the increase in molality of DTAB. The ionic part of DTAB holds the water molecule through hydrophilic hydration, while the hydrophobic moiety of DTAB holds the water molecule through hydrophobic hydration. The hydrophilic hydration shell of ionic group of DTAB is controlled predominantly by electrostatic interactions between water molecules and the ionic group of DTAB. Strong electrostatic force between ionic group of DTAB and water causes electrostriction. The hydrophobic moiety of DTAB forms hydration shell around it through hydrophobic hydration. Thus hydrophilic as well as hydrophobic solute-solvent interactions determine the hydration sphere of DTAB. As the concentration of DTAB increases, the solute-solute interaction relaxes water molecules from both hydrophilic and hydrophobic hydration zone and hydrophobic zone to the bulk render the solution more compact and imparts a decrease in molality of DTAB.

The variation of  $\varphi_{k(s)}$  with  $m$  of DTAB exhibits that at lower concentration zone  $\varphi_{k(s)}$  change rapidly after that  $\varphi_{k(s)}$  shows very slow changing trend with the increase in  $m$  of DTAB. This may be attributed to the CMC conformation of DTAB. After the micelle formation the solution becomes less compressible. The slow change after CMC reveals that the relaxation of water molecules from hydration spheres to the bulk due to interactions between solute and solvent.

Table 2. Isentropic compressibility,  $K_s$ , and Apparent molal isentropic compressibility,  $\varphi_{k(s)}$ , of aqueous DTAB solutions at T= (295.15, 302.15 and 309.15) K

$m$ /mol·kg <sup>-1</sup>	295.15 K		302.15 K		309.15 K	
	$K_s \times 10^{-7}$ /bar <sup>-1</sup>	$\varphi_{k(s)}$ /cm <sup>3</sup> ·bar <sup>-1</sup> ·mol <sup>-1</sup>	$K_s \times 10^{-7}$ /bar <sup>-1</sup>	$\varphi_{k(s)}$ /cm <sup>3</sup> ·bar <sup>-1</sup> ·mol <sup>-1</sup>	$K_s \times 10^{-7}$ /bar <sup>-1</sup>	$\varphi_{k(s)}$ /cm <sup>3</sup> ·bar <sup>-1</sup> ·mol <sup>-1</sup>
0.00102	4.5242	6.4473E-05	4.4227	4.3467E-01	4.3465	4.2815E-01
0.00300	4.5212	9.4670E-06	4.4202	1.4802E-01	4.3443	1.4581E-01
0.00502	4.5181	-4.9716E-06	4.4174	8.8455E-02	4.3420	8.7144E-02
0.00697	4.5152	-8.8266E-06	4.4150	6.3726E-02	4.3400	6.2787E-02
0.00895	4.5120	-1.3272E-05	4.4125	4.9651E-02	4.3379	4.8924E-02
0.01093	4.5085	-1.9942E-05	4.4100	4.0628E-02	4.3358	4.0037E-02
0.01311	4.5048	-2.2996E-05	4.4066	3.3878E-02	4.3331	3.3390E-02
0.01493	4.5023	-2.1282E-05	4.4043	2.9747E-02	4.3309	2.9319E-02
0.01692	4.5017	-6.7124E-06	4.4038	2.6259E-02	4.3302	2.5879E-02
0.01846	4.5015	3.2511E-06	4.4036	2.4074E-02	4.3300	2.3726E-02
0.02101	4.5009	1.6406E-05	4.4033	2.1172E-02	4.3297	2.0866E-02
0.02312	4.5004	2.4718E-05	4.4031	1.9251E-02	4.3295	1.8973E-02
0.02501	4.5000	3.1323E-05	4.4028	1.7805E-02	4.3293	1.7548E-02
0.02703	4.4997	3.7574E-05	4.4025	1.6481E-02	4.3292	1.6244E-02
0.02893	4.4994	4.2999E-05	4.4023	1.5406E-02	4.3292	1.5185E-02
0.03112	4.4990	4.7928E-05	4.4019	1.4330E-02	4.3287	1.4125E-02

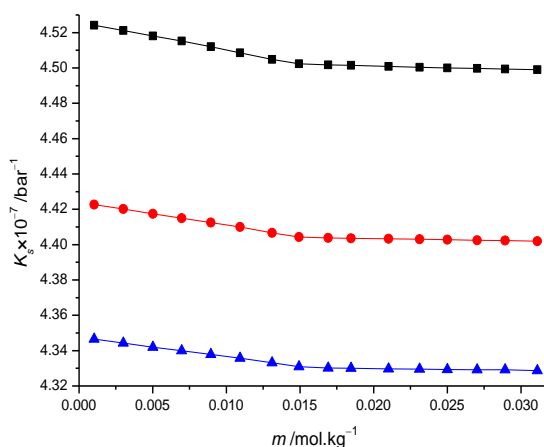


Figure 2. Plots of Isentropic compressibility versus molality of aqueous DTAB solutions at studied temperatures: (■) 295.15 K, (●) 302.15 K, (▲) 309.15 K

### 3.3 Acoustic Impedance

Acoustic impedance is defined as (Pandey et al., 2000):

$$Z = u\rho \quad (3)$$

The acoustic impedances,  $Z$ , of aqueous DTAB solutions as a function of concentration are presented in Table 3. The variations of  $Z$  with the DTAB in aqueous solutions are shown in Figure 3. The figures demonstrate that acoustic impedance increases with the increase in molality. Acoustic impedance is a measure of resistance to the propagation of sound wave through any medium. The increase in  $Z$  with the molality indicates that as concentration increases the sound wave has to face more resistance to flow. As the concentration increases the

solute-solute interactions also increase. The positive acoustic impedance is therefore an evidential parameter for solute-solvent and solute-solute interaction. The  $Z$  values indicate that there exists strong solute-solute along with solute-solvent interaction in aqueous solutions (Kannappan et al., 2008).

Table 3. Acoustic impedance,  $Z$ , of aqueous DTAB solutions at  $T = (295.15, 302.15 \text{ and } 309.15) \text{ K}$

$m$ /mol·kg <sup>-1</sup>	$Z$ /g·cm <sup>-2</sup> ·s <sup>-1</sup>		
	295.15 K	302.15 K	309.15 K
0.00102	1485.14	1500.69	1512.07
0.00300	1485.65	1501.15	1512.47
0.00502	1486.20	1501.65	1512.90
0.00697	1486.71	1502.08	1513.27
0.00895	1487.25	1502.54	1513.66
0.01093	1487.87	1503.00	1514.06
0.01311	1488.50	1503.61	1514.56
0.01493	1488.95	1504.03	1514.97
0.01692	1489.07	1504.12	1515.12
0.01846	1489.13	1504.18	1515.17
0.02101	1489.25	1504.26	1515.25
0.02312	1489.36	1504.32	1515.30
0.02501	1489.44	1504.38	1515.36
0.02703	1489.52	1504.45	1515.38
0.02893	1489.58	1504.50	1515.40
0.03112	1489.67	1504.59	1515.49

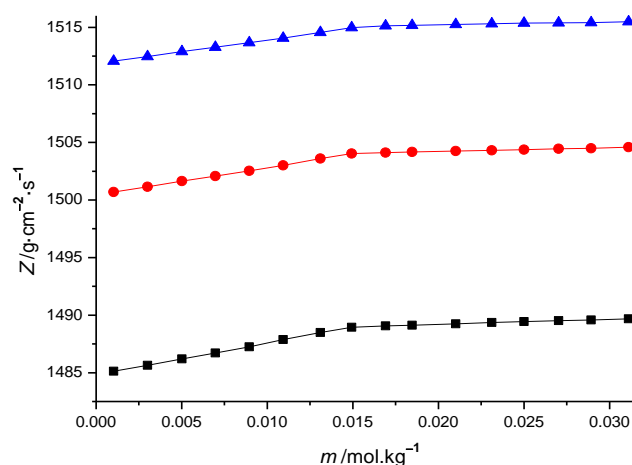


Figure 3. Plots of acoustic impedance versus molality of aqueous DTAB solutions at studied temperatures: (■) 295.15 K, (●) 302.15 K, (▲) 309.15 K

### 3.4 Relative Association

The relative association is defined as a measure of the extent of interaction between the component molecules in a real mixture relative to that in an ideal one (Frank et al., 1957):

$$R_A = \left( \frac{\rho}{\rho_0} \right) \left( \frac{u_0}{u} \right) \quad (4)$$

where  $\rho$ ,  $\rho_0$  and  $u$ ,  $u_0$  are densities and ultrasonic speeds of the mixture and the solvent respectively. The relative associations,  $R_A$ , values of DTAB are shown in Table 4. The graphical representations  $R_A$  versus  $m$  data are shown

in Figure 4.  $R_A$  influenced by two factors (i) the breaking up of solvent structure on addition of solute to it; and (ii) the salvation of solutes that are simultaneously present. Decrease in  $R_A$ , is due to former and letter results in increase of  $R_A$ . The relative association,  $R_A$  for DTAB in aqueous shown slow initial decrease in molality then at a certain molality the  $R_A$  increases with the increase in molality which indicates that solvation of DTAB predominates over the breaking of the solvent structure. The positive relative association,  $R_A$  demonstrates that solute-solvent interaction causes association of DTAB, which increases with DTAB concentration.

Table 4. Relative association,  $R_A$ , of aqueous DTAB solutions at T= (295.15, 302.15 and 309.15) K

$m$ /mol·kg <sup>-1</sup>	$R_A$		
	295.15 K	302.15 K	309.15 K
0.00102	0.999964	0.999910	0.999903
0.00300	0.999706	0.999686	0.999710
0.00502	0.999418	0.999433	0.999503
0.00697	0.999154	0.999217	0.999319
0.00895	0.998863	0.998982	0.999130
0.01093	0.998527	0.998755	0.998934
0.01311	0.998189	0.998432	0.998676
0.01493	0.997964	0.998222	0.998471
0.01692	0.997964	0.998219	0.998438
0.01846	0.997969	0.998230	0.998446
0.02101	0.997965	0.998243	0.998457
0.02312	0.997958	0.998259	0.998479
0.02501	0.997960	0.998273	0.998490
0.02703	0.997960	0.998278	0.998517
0.02893	0.997973	0.998283	0.998543
0.03112	0.998022	0.998317	0.998579

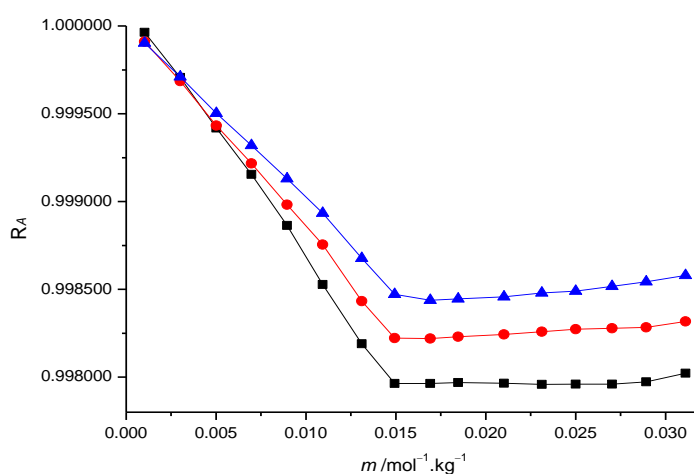


Figure 4. Plots of relative association versus molality of aqueous DTAB solutions at studied temperatures: (■) 295.15 K, (●) 302.15 K, (▲) 309.15 K

### 3.5 Molar Sound Velocity

Molal sound velocity indicates the presence of solvent-solvent interactions. It can be represented as (Frank et al., 1957):

$$R_m = Vu^{1/3} \quad (5)$$

where  $V$  and  $u$  are the molar volume and speed of sound, respectively. The molar sound velocity,  $R_m$ , shows an asymptotic increase with the increase in molality. The values of  $R_m$  are presented in Table 5 and the variations of  $R_m$  against molality are shown in Figure 5. The increase in  $R_m$  values with molality is evident of the structure of solution due to solute-solute and solute-solvent intersections. The asymptotic nature reveals that the reinforcement of structure of solution approaches to attain a maximum value

Table 5. Molal sound velocities,  $R_m$ , of aqueous DTAB solutions at T= (295.15, 302.15 and 309.15) K

$m$ /mol·kg <sup>-1</sup>	$R_m$ /cm <sup>3</sup> ·mol <sup>-1</sup> ·(cm·s <sup>-1</sup> ) <sup>1/3</sup>		
	295.15 K	302.15 K	309.15 K
0.00102	206.01	207.24	208.39
0.00300	206.14	207.37	208.52
0.00502	206.28	207.50	208.65
0.00697	206.41	207.63	208.78
0.00895	206.54	207.76	208.91
0.01093	206.68	207.89	209.04
0.01311	206.83	208.04	209.18
0.01493	206.94	208.16	209.30
0.01692	207.06	208.27	209.42
0.01846	207.15	208.36	209.51
0.02101	207.29	208.51	209.66
0.02312	207.42	208.63	209.78
0.02501	207.52	208.74	209.89
0.02703	207.64	208.86	210.01
0.02893	207.75	208.97	210.12
0.03112	207.88	209.10	210.25

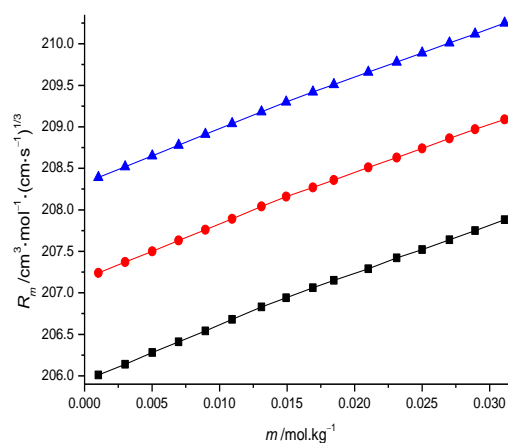


Figure 5. Plots of molar sound velocity versus molality of aqueous DTAB solutions at studied temperatures:

(■) 295.15 K, (●) 302.15 K, (▲) 309.15 K

### 3.6 Micellar Behaviour

Conductivity technique has been found to be highly useful for studying the association behaviour of various system (Fujiwara et al., 1997, Moulik et al., 1996). The concentration dependence of specific conductivity of aqueous solutions of DTAB is shown in Figure 6. and the data are presented in Table 6. The specific conductance increases with increasing DTAB concentration and shows a sharp break in its value where micelle starts to form and is determined by extrapolating the specific conductivity data in the pre-micellar region to intersect with a

straight line drawn through the data in the micellar region. The estimated CMC value thus obtained for DTAB in aqueous is  $1.52 \times 10^{-2} \text{ mol}\cdot\text{kg}^{-1}$  at 302 K, in good agreement with the values reported earlier in literature (Benrra et al., 2003, Jones et al., 1972, Markarian et al., 2005) as well as satisfying our different experimental CMC values. The CMC (critical micelle concentration) of aqueous solution of Dodecyltrimethylammonium bromide (DTAB) at different temperatures using different methods (sound velocities, isentropic compressibility, acoustic impedance and relative association) were found from sharp braking point, presented in Table 7.

Table 6. Conductance,  $G$ , Specific conductance,  $K$ , and Critical Micelle Concentration (CMC),  $m_{\text{cmc}}$ , of aqueous DTAB solutions as a function of molality,  $m$ , at temperature 302.15 K

$m$ / $\text{mol}\cdot\text{kg}^{-1}$	$G/\mu\text{S}$	$C/\text{m}^{-1}$	$K/\mu\text{S}\cdot\text{m}^{-1}$	$m_{\text{cmc}}/\text{mol}\cdot\text{kg}^{-1}$
0.00102	106.86		240.02	
0.00300	297.26		667.68	
0.00502	472.26		1060.74	
0.00697	635.26		1426.86	
0.00895	809.26		1817.68	
0.01093	990.26		2224.22	
0.01311	1183.26		2657.72	
0.01493	1339.26		3008.11	0.0152±0.00017
0.01692	1410.26	2.2461	3167.58	
0.01846	1445.26		3246.20	
0.02101	1514.26		3401.18	
0.02312	1565.26		3515.73	
0.02501	1607.26		3610.07	
0.02703	1653.26		3713.39	
0.02893	1697.26		3812.22	
0.03112	1756.26		3944.74	

\* ± Indicate standard error.

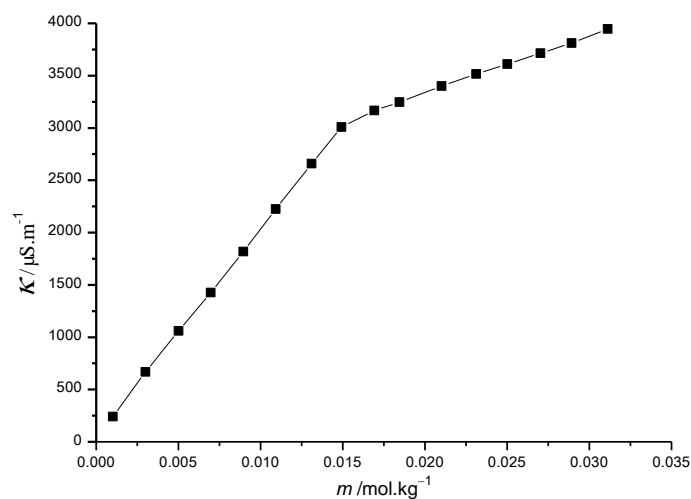


Figure 6. Plot of Specific Conductance versus molality of aqueous DTAB solutions at 302.15 K temperature



Table 7. Critical Micelle Concentration (CMC),  $m_{\text{cmc}}$ , of aqueous solution of DTAB at T= (295.15, 302.15 and 309.15) K using different methods.

T/K	Sound velocity, $m_{\text{cmc}}/\text{mol}\cdot\text{kg}^{-1}$	Isentropic compressibility, $m_{\text{cmc}}/\text{mol}\cdot\text{kg}^{-1}$	Acoustic impedance $m_{\text{cmc}}/\text{mol}\cdot\text{kg}^{-1}$	Relative association, $m_{\text{cmc}}/\text{mol}\cdot\text{kg}^{-1}$
295.15	0.01507±0.00039	0.01511±0.00040	0.01517±0.000429	0.01497±0.00036
302.15	0.01524±0.00029	0.01527±0.00031	0.01529±0.000346	0.01516±0.00025
309.15	0.01583±0.00039	0.01585±0.00041	0.01590±0.000462	0.01575±0.00033

\* ± Indicates standard error.

#### 4. Conclusions

In summary, we have determined speed of sound of DTAB in aqueous solutions at 295.15, 302.15 and 309.15 K. The isentropic compressibility,  $K_s$ , apparent molal isentropic compressibility,  $\phi_{k(s)}$ , acoustic impedance, Z, molal sound of speed,  $R_m$ , and relative association,  $R_A$ , have been determined and reported. These results confirmed that strong solute-solute, solute-solvent interactions occur in the reported system. From the analysis of the experimental data, it is found that the micellar behaviour of DTAB in aqueous solution is temperature dependent. The critical micelle concentration of DTAB in aqueous at different temperatures from this study are of the order:  $m_{\text{cmc}}(309.15) > m_{\text{cmc}}(302.15) > m_{\text{cmc}}(295.15)$ . This may be due to a decrease in the hydrophobic character of molecule DTAB in aqueous with the increase in temperature.

#### Acknowledgements

The authors gratefully acknowledge to the Department of Chemistry, University of Rajshahi, Bangladesh for providing necessary facilities and help when needed for the work.

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