ULTRASONIC INVESTIGATIONS OF LACTOSE IN AQUEOUS SOLUTION OF TETRAALKYLAMMONIUM BROMIDE SALTS AT 303.15, 308.15 AND 313.15K

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Abstract: Densities (ρ) and Ultrasonic velocity (U) for different molal concentration of lactose in aqueous solutions of tetra alkylammonium bromide salts in 0.0M, 0.05M and 0.1M at various temperature (303.15, 308.15 and 313.15 K) were measured at atmospheric pressure. From these experimental results, different derived parameters such as adiabatic compressibility (β), change in adiabatic compressibility (Δβ), relative change in adiabatic compressibility ($\Delta\beta/\beta_0$), relative association (Ra) and acoustic impedance (Z) have been calculated. The results are interpreted in terms of solute-solvent and solute-solute interactions in these systems. It has been observed that there exist strong solute-solvent interactions in these systems, which increase with increase in lactose concentration as well as with tetra alkylammonium bromide salts.

Key words: lactose, adiabatic compressibility, acoustic impedance, relative association, solute-solvent interactions.

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Introduction:

Ultrasonic investigation in aqueous solutions of electrolytes and non-electrolytes with saccharides provide useful information in understanding the behaviour of liquid systems. Intermolecular and intramolecular association, complex formation, and related structural changes affect the compressibility of the system which in turn produces corresponding variation in the ultrasonic velocity. During the last two decades, ultrasonic studies have been carried out to investigate hydration of proteins and saccharide etc., through volumetric and ultrasonic measurements, since these properties are sensitive to the degree and nature of hydration [1]. Physico-chemical study on the transport properties of electrolytes has been utilized to investigate the solvation and association behaviour of ions in different solvent media [2]. Sugars and polyols are well known stabilizing agents of proteins/enzymes in their native state owing to their ability to enhance the structure of water [3]. Moreover sugars are not only biologically important compounds but also typical non-electrolyte with hydrophilic hydroxyl group, which are capable of hydrogen bonding [4].Most biochemical processes occur in aqueous media which involve phenomena like volume change and hydration of molecules. Therefore, studies on the physicochemical properties of biomolecules like vitamins, amino acids, sugars and drugs in aqueous solution provide useful information, which is important to understand the complex mechanism of molecular interactions [5].

Compressibility is a fundamental and inherent property of liquids and sensitive measure of solute-solvent interactions which can be used to monitor solute hydration in aqueous solutions [6]. The physiological actions of biological important molecules like proteins, saccharides, biosurfactants, etc., are made possible due to various kinds of interactions which they undergo with the different metabolites present in the living body [7]. Thermodynamic properties of electrolytes in saccharide solutions are important not only for studying the nature of interactions of saccharides with ions, but also providing significant information on the stereo-structure of saccharide molecules and the action mechanism of their biological activity [8]. In order to understand these interactions, a systematic knowledge of solution behavior of such compounds may prove to be highly beneficial [7]. In this paper, we report the densities (ρ) and ultrasonic speeds (u) of Lactose in aqueous solutions of Tetraethylammonium bromide, Tetraporpylammonium bromide and Tetrabutylammonium bromide (0.0M, 0.05M and 0.1M) at temperatures 303.15, 308.15 and 313.15K. Using the experimentally measured ρ, η and u data, adiabatic compressibility (β), change in adiabatic compressibility (Δβ),relative change in adiabatic compressibility ($\Delta\beta/\beta_0$), relative association (R_a) and acoustic impedance (Z), have been calculated. All these parameters are discussed in terms of solute-solute and solute-solvent interactions occurring in these systems and the structure making/ breaking tendency of the solute in the given solvent.

Experimental technique:

Lactose and the quaternary ammonium bromide salts of high purity used in the present studies were purchased from S.D.Fines chemicals and E-Merk (India). These chemicals were used as such without further purification. The density and ultrasonic velocity at different molality of lactose in quaternary ammonium bromide salts were measured at 303.15, 308.15and 313.15 K. The weight of the sample was measured using an electronic digital balance with a precision of ±0.0001 g (Model: -SHIMADZU AX200). An ultrasonic interferometer having the frequency 3MHz (MITTAL ENTERPRISES, NEW DELHI, MODEL F–81) with an overall accuracy of ± 2ms^{-1} has been used for velocity measurement. The density was determined using a specific gravity bottle by relative measurement method with a reproducibility of ± 0.01 kg.m^{-3}.An Ostwald's viscometer (10 ml) was used for the viscosity measurement. Efflux time was determined using a digital chronometer within ± 0.01s.

An electronic digital constant temperature bath (RAAGA Industries) has been used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature.

Theory and calculation:

The various acoustical parameters such as adiabatic compressibility (β), change in adiabatic compressibility (Δβ), relative change in adiabatic compressibility ($\Delta\beta/\beta_0$), relative association (R_a) and acoustic impedance (Z) have been calculated from the experimental data using following equations:

Adiabatic compressibility (β) is given by [9]

$$\beta = \frac{1}{U^2\rho} \qquad \text{... (1)}$$

$$\Delta\beta = \beta_0 - \beta \qquad \text{... (2)}$$

Relative change in adiabatic compressibility is determined by $\Delta\beta/\beta_0$

$$Z = U\rho \qquad \text{... (3)}$$

The relative association is calculated using the relation [10]

$$R_a = \left[\frac{\rho}{\rho o}\right]\left[\frac{Uo}{U}\right]^{1/3} \qquad \text{... (4)}$$

Where, ρ & ρ_o are the densities of the solution and solvent U and U_o are the ultrasonic velocities of the solution and solvent respectively.

Results and discussions:

From the Figs 1-2, it is found that the density and ultrasonic velocity increase with increase in molal concentration of saccharide as well as with electrolyte content. However, the values of density decrease with increase in temperature in all the systems studied, whereas the ultrasonic velocity increases with rise in temperature. In the present case, the increase in density and ultrasonic velocity in these solutions may be attributed to the cohesion brought about by the ionic hydration. When saccharide is dissolved in aqueous electrolytic solutions, the water molecules are attracted strongly by the electrostatic forces, which introduce a greater cohesion in the solution. Thus, cohesion increases with increase of saccharide concentration in the solutions [11].

The values of density have been decreased with rise in temperature, which happens because of decrease in the extent of different intermolecular interactions. The increase in temperature may have caused increase in the kinetic energy of molecules and ions present in the solution, which in turn decreases the solute-solvent interactions [12]. Conversely the values of ultrasonic velocity increases with increase in temperature.The decrease in density and increase in velocity with rise in temperature indicate the decrease in intermolecular forces due to increase in thermal energy of the system [13]. The increase in density (ρ) and ultrasonic velocity (U) with concentration suggests that the cohesive forces increased significantly, which is due to powerful molecular interactions [14].

Fig. 3 shows that the adiabatic compressibility values significantly decrease as the function of molal concentration of lactose as well as aqueous electrolytic solutions and the same decrease with rise in temperature in all the systems. The increasing electrostrictive compression of water around the molecules results in a large decrease in the compressibility of solutions. The decrease in compressibility implies that there is an enhanced molecular association in the above system upon increase in solute content, as the new entities formed due to molecular association become compact and less compressible [1].

Further, the decrease in values of β with rise in concentration suggests the significant structural rearrangement in neighbouring atmosphere of the ion. As the temperature increases the system becomes less compressible due to the structural changes of water [15]. Tables 1-2 reveal that the values of change in adiabatic compressibility (Δβ) and relative change in adiabatic compressibility ($\Delta\beta/\beta_0$) are negative and increase with increase in molal concentration of saccharide whereas, with increase in temperature, these values are found to decrease in all the systems studied. The increase in $\Delta\beta/\beta_0$ values with increase in solute concentration may be attributed to an increase in the incompressible part in the solution.

The variation of the change and relative change in compressibility values with temperature may be attributed to thermal rupture of water structure [16]. The acoustic impedance (Z) increases with increase in the concentration of lactose and electrolyte content as well as with increase in temperature in all the ternary systems under investigation (Table 4). In the case of saccharides, the increase in acoustic impedance is the complex ratio of effective sound pressure at a point to the effective particle velocity at that point [15].

Acoustic impedance increases with increasing concentration of solute and temperature indicating that molecular interactions are associative in nature [17]. The acoustic impedance (Z) is the impedance offered to sound wave by the components of the mixtures whereas the relative association (R_a) is the measure of extent of association of the components in the medium [10]. The relative association (Ra) is influenced by two factors:

i) Breaking up of the associated solvent molecules on addition of solute in it.

ii)The solvation of solute molecule.

From the Table 3, it can be seen that the values of relative association (R_a) increase with increasing molal concentration of lactose with different temperatures. The increasing R_a depends on the solvation of solute molecules and breaking up of the solvent structures by the addition of lactose. Hence, the increase of R_a with concentration suggests that solvation of solutes is effective over breaking of the solvent structures [15].The increasing trend in Z and R_a suggests the strengthening of interaction among the component.

The interaction may be solute-solute or solute-solvent or solvent-solvent type, it is peculiar to note that these two parameters depend on density [18].

Conclusion:

In the present work, the volumetric and compressibility properties of lactose in aqueous solutions of tetralkylammonium bromide are reported at T = (303.15, 308.15, and 313.15K). The trends observed for the increase in density and ultrasonic velocity values have been attributed to increase in hydrophilic-hydrophilic and hydrophilic-ionic interactions with increase in electrolytes concentration.

The temperature variation indicates that the strength of intermolecular interaction decreases with rise in temperature. The decrease in adiabatic compressibility values as the function of molal concentration of lactose as well as aqueous electrolytic solutions in all the systems reveal that the increasing electrostrictive compression of water around the molecules results in a large decrease in the compressibility of solutions. The increase in R_a and Z values with increase in the concentration of the solute indicates the solvation of the solute molecule which further confirms the solute-solvent interactions in these systems. In the light of the above discussion, it may be concluded that there exist a powerful molecular interactions in the systems studied. Both the solute-solute interactions and solute-solvent interactions are existing in these systems.

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Table 1

Values of change in Adiabatic compressibility of Lactose in aqueous solution of Tetraethylammonium bromide (TEABr),Tetrapropylammoniumbromide (TPABr) and Tetrabutylammonium bromide (TBABr) at 303.15, 308.15,313.15K

Molality m/(mol. kg^{-1})	$-\Delta\beta/ (\times10^{-12}\ m^2\ N^{-1})$								
	0.0M			0.05M			0.1M		
	Temperature (K)								
	303.15	308.15	313.15	303.15	308.15	313.15	303.15	308.15	313.15
System I: Lactose +Aqueous TEABr									
0.02	6.12	5.95	4.31	3.99	3.79	3.1	4.25	3.32	2.97
0.04	12.07	8.81	7.55	8.04	7.24	5.27	7.03	5.95	5.59
0.06	13.68	10.30	9.80	10.31	9.31	7.87	8.94	8.08	8.41
0.08	16.97	13.74	12.69	13.32	11.27	10.45	11.68	10.63	10.90
0.10	19.09	15.89	14.22	15.02	13.32	11.60	13.01	12.47	12.65
System II: Lactose +Aqueous TPABr									
0.02	6.12	5.95	4.31	3.99	3.62	3.41	3.91	4.24	3.42
0.04	12.07	8.81	7.55	8.04	6.69	6.24	7.08	7.05	6.48
0.06	13.68	10.30	9.80	10.31	8.64	8.43	8.87	9.12	8.88
0.08	16.97	13.74	12.69	13.32	11.45	10.73	12.22	11.06	11.88
0.10	19.09	15.89	14.22	15.02	13.49	12.15	13.47	13.28	13.41
System III: Lactose +Aqueous TBABr									
0.02	6.12	5.95	4.31	3.27	3.43	3.29	4.32	3.46	2.90
0.04	12.07	8.81	7.55	6.77	6.17	5.93	9.14	7.65	6.72
0.06	13.68	10.30	9.80	10.49	8.90	7.90	11.24	9.56	8.60
0.08	16.97	13.74	12.69	13.36	11.05	10.82	14.53	13.11	11.79
0.10	19.09	15.89	14.22	15.38	13.62	12.52	16.54	15.50	14.33

Table 2

Values of relative change in Adiabatic compressibility ($\Delta\beta/\beta_0$) of Lactose in aqueous solution of Tetraethylammonium bromide (TEABr),Tetrapropylammoniumbromide (TPABr) and Tetrabutylammonium bromide (TBABr) at 303.15, 308.15,313.15K

Molality m/(mol.kg^{-1})	$-\Delta\beta/\beta_0$ ($\times10^{-3}$)								
	0.0M			0.05M			0.1M		
	Temperature(K)								
	303.15	308.15	313.15	303.15	308.15	313.15	303.15	308.15	313.15
System I: Lactose +Aqueous TEABr									
0.02	13.85	13.62	9.99	9.13	8.74	7.23	9.90	7.77	6.98
0.04	27.32	20.18	17.50	18.40	16.70	12.29	16.65	13.92	13.13
0.06	30.97	23.59	22.71	23.60	21.47	18.36	20.83	18.91	19.76
0.08	38.42	31.47	29.41	30.49	26.00	24.38	27.21	24.88	25.61
0.10	43.22	36.39	32.96	34.38	30.73	27.06	30.31	29.18	29.73
System II: Lactose +Aqueous TPABr									
0.02	13.85	13.62	9.99	8.61	8.46	8.03	9.14	10.03	8.12
0.04	27.32	20.18	17.50	16.84	15.64	14.70	16.55	16.67	15.40
0.06	30.97	23.59	22.71	20.20	20.20	19.86	20.23	21.51	21.10
0.08	38.42	31.47	29.41	27.40	26.77	25.29	28.57	26.16	28.23
0.10	43.22	36.39	32.96	33.27	31.54	28.63	31.49	31.41	31.87
System III: Lactose +Aqueous TBABr									
0.02	13.85	13.62	9.99	7.66	8.09	7.82	10.26	8.28	6.96
0.04	27.32	20.18	17.50	15.64	14.55	14.10	21.72	18.31	16.14
0.06	30.97	23.59	22.71	24.60	20.99	18.78	26.71	22.88	20.66
0.08	38.42	31.47	29.41	31.33	27.22	25.73	34.53	31.37	28.32
0.10	43.22	36.39	32.96	36.07	32.33	29.77	39.30	37.09	34.43

Table 3

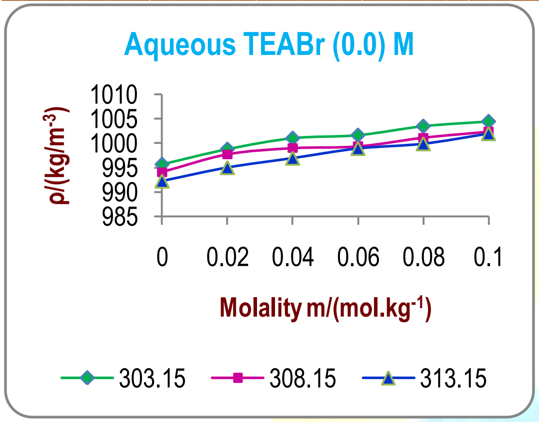
Values of Relative association (Ra) of Lactose in aqueous solution of Tetraethylammonium bromide (TEABr),Tetrapropylammoniumbromide (TPABr) and Tetrabutylammonium bromide (TBABr) at 303.15, 308.15,313.15K

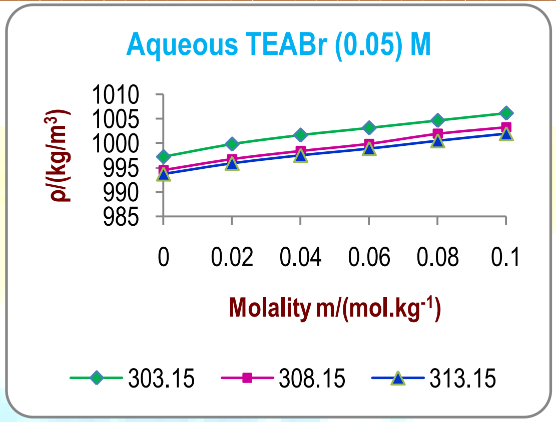
Molality m/(mol. kg^{-1})	Ra								
	0.0M			0.05M			0.1M		
	Temperature (K)								
	303.15	308.15	313.15	303.15	308.15	313.15	303.15	308.15	313.15
System I: Lactose +Aqueous TEABr									
0.02	1.0013	1.0019	1.0015	1.0015	1.0011	1.0013	1.0026	1.0028	1.0024
0.04	1.0016	1.0023	1.0025	1.0020	1.0017	1.0024	1.0047	1.0057	1.0058
0.06	1.0017	1.0026	1.0039	1.0029	1.0027	1.0030	1.0076	1.0071	1.0060
0.08	1.0025	1.0029	1.0039	1.0035	1.0042	1.0038	1.0088	1.0096	1.0079
0.10	1.0028	1.0035	1.0057	1.0045	1.0050	1.0050	1.0116	1.0103	1.0080
System II: Lactose +Aqueous TPABr									
0.02	1.0013	1.0019	1.0015	1.0027	1.0025	1.0021	1.0042	1.0026	1.0023
0.04	1.0016	1.0023	1.0025	1.0053	1.0055	1.0056	1.0067	1.0055	1.0056
0.06	1.0017	1.0026	1.0039	1.0075	1.0074	1.0065	1.0085	1.0077	1.0067
0.08	1.0025	1.0029	1.0039	1.0093	1.0086	1.0076	1.0093	1.0108	1.0074
0.10	1.0028	1.0035	1.0057	1.0102	1.0090	1.0080	1.0104	1.0108	1.0083
System III: Lactose +Aqueous TBABr									
0.02	1.0013	1.0019	1.0015	1.0047	1.0040	1.0039	1.0035	1.0051	1.0053
0.04	1.0016	1.0023	1.0025	1.0084	1.0077	1.0077	1.0054	1.0069	1.0074
0.06	1.0017	1.0026	1.0039	1.0090	1.0109	1.0095	1.0069	1.0076	1.0074
0.08	1.0025	1.0029	1.0039	1.0091	1.0142	1.0110	1.0077	1.0084	1.0087
0.10	1.0028	1.0035	1.0057	1.0109	1.0160	1.0115	1.0093	1.0095	1.0099

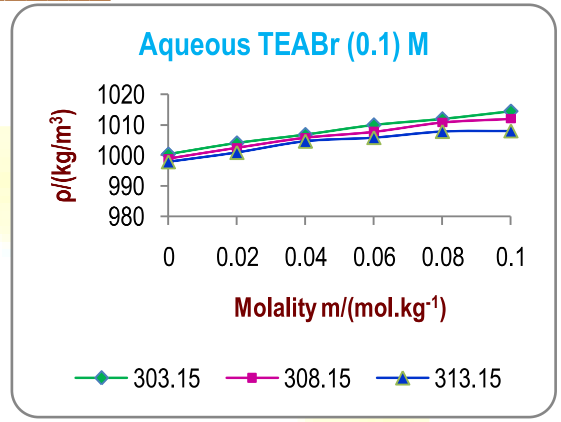
Table 4

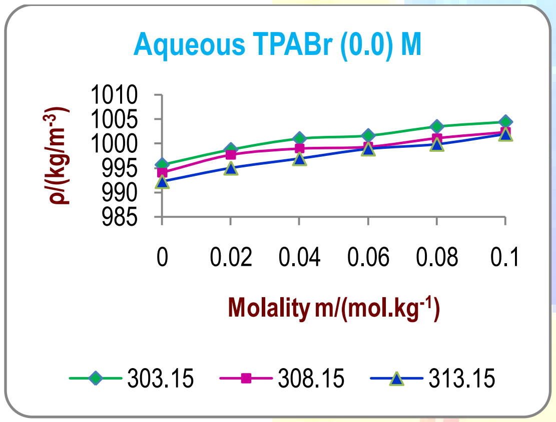
Values of Acoustic impedance (Z) of Lactose in aqueous solution of Tetraethylammonium bromide (TEABr),Tetrapropylammoniumbromide (TPABr) and Tetrabutylammonium bromide (TBABr) at 303.15, 308.15,313.15K

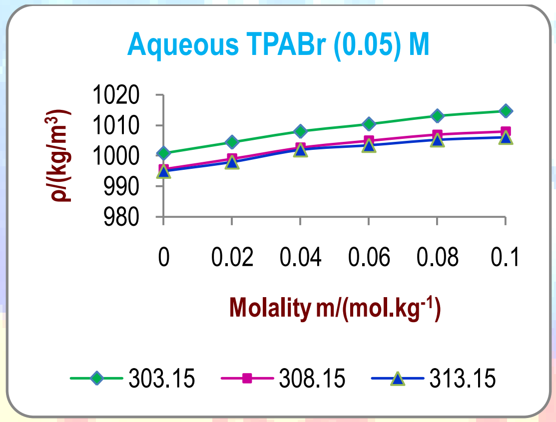
Molality in $mol.kg^{-1}$	$Zx10^6$ Kg m^{-2} s^{-1}								
	0.0M			0.05M			0.1M		
	Temperature (K)								
	303.15	308.15	313.15	303.15	308.15	313.15	303.15	308.15	313.15
System I: Lactose +Aqueous TEABr									
0.00	1.5014	1.5089	1.5166	1.5110	1.5147	1.5226	1.5268	1.5291	1.5314
0.02	1.5143	1.5221	1.5263	1.5199	1.5231	1.5298	1.5372	1.5378	1.5392
0.04	1.5264	1.5282	1.5336	1.5285	1.5305	1.5350	1.5444	1.5452	1.5469
0.06	1.5298	1.5314	1.5392	1.5337	1.5354	1.5408	1.5503	1.5506	1.5530
0.08	1.5371	1.5387	1.5453	1.5403	1.5405	1.5467	1.5569	1.5577	1.5592
0.10	1.5417	1.5436	1.5497	1.5445	1.5453	1.5500	1.5617	1.5620	1.5627
System I: Lactose +Aqueous TPABr									
0.00	1.5014	1.5089	1.5166	1.5228	1.5259	1.5314	1.5303.15	1.5376	1.5402
0.02	1.5143	1.5221	1.5263	1.5321	1.5350	1.5399	1.5416	1.5481	1.5490
0.04	1.5264	1.5282	1.5336	1.5412	1.5434	1.5481	1.5494	1.5560	1.5577
0.06	1.5298	1.5314	1.5392	1.5457	1.5488	1.5533	1.5544	1.5619	1.5636
0.08	1.5371	1.5387	1.5453	1.5534	1.5555	1.5590	1.5621	1.5704	1.5706
0.10	1.5417	1.5436	1.5497	1.5593	1.5601	1.5624	1.5655	1.5731	1.5747
System III: Lactose +Aqueous TBABr									
0.00	1.5014	1.5089	1.5166	1.5332	1.5339	1.5393	1.5456	1.5496	1.5521
0.02	1.5143	1.5221	1.5263	1.5431	1.5437	1.5488	1.5570	1.5604	1.5623
0.04	1.5264	1.5282	1.5336	1.5527	1.5519	1.5570	1.5687	1.5708	1.5716
0.06	1.5298	1.5314	1.5392	1.5612	1.5599	1.5624	1.5743	1.5754	1.5756
0.08	1.5371	1.5387	1.5453	1.5675	1.5677	1.5698	1.5821	1.5841	1.5843
0.10	1.5417	1.5436	1.5497	1.5726	1.5734	1.5750	1.5877	1.5900	1.5902

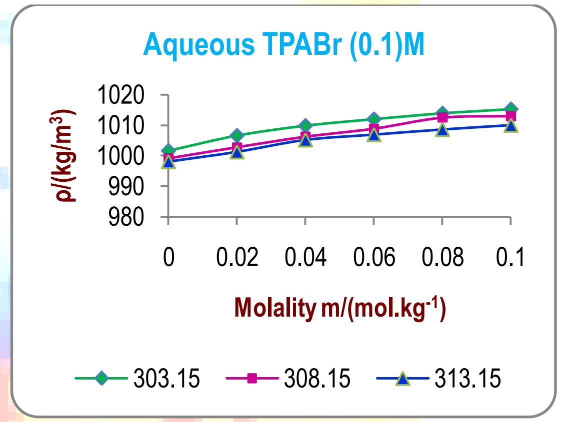

Aqueous TEABr (0.0) M
ρ/(kg/m-3)
1010
1005
1000
995
990
985
0 0.02 0.04 0.06 0.08 0.1
Molality m/(mol.kg-1)
303.15 308.15 313.15


Aqueous TEABr (0.05) M
ρ/(kg/m3)
1010
1005
1000
995
990
985
0 0.02 0.04 0.06 0.08 0.1
Molality m/(mol.kg-1)
303.15 308.15 313.15


Aqueous TEABr (0.1) M
ρ/(kg/m3)
1020
1010
1000
990
980
0 0.02 0.04 0.06 0.08 0.1
Molality m/(mol.kg-1)
303.15 308.15 313.15


Aqueous TPABr (0.0) M
ρ/(kg/m-3)
1010
1005
1000
995
990
985
0 0.02 0.04 0.06 0.08 0.1
Molality m/(mol.kg-1)
303.15 308.15 313.15


Aqueous TPABr (0.05) M
ρ/(kg/m3)
1020
1010
1000
990
980
0 0.02 0.04 0.06 0.08 0.1
Molality m/(mol.kg-1)
303.15 308.15 313.15


Aqueous TPABr (0.1)M
ρ/(kg/m3)
1020
1010
1000
990
980
0 0.02 0.04 0.06 0.08 0.1
Molality m/(mol.kg-1)
303.15 308.15 313.15

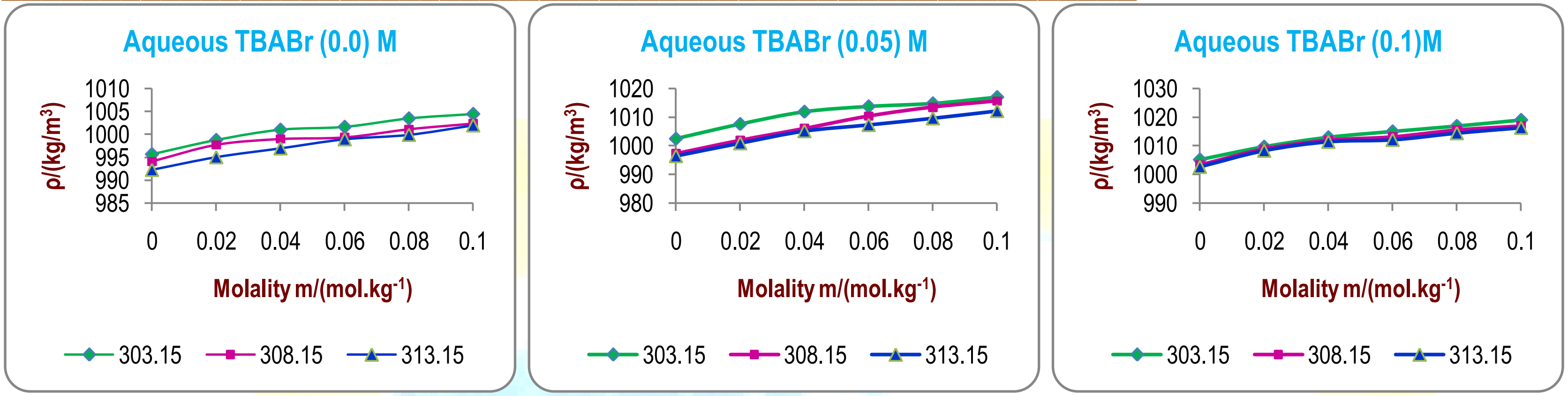
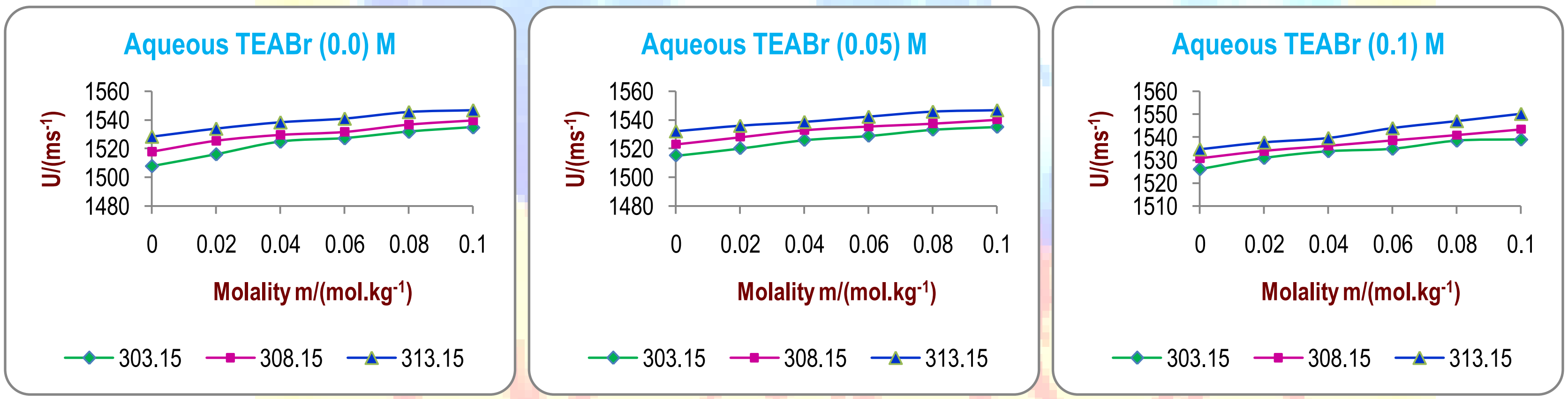


Fig.1 Values of Density (ρ) of Lactose in aqueous solution of Tetraethylammonium bromide (TEABr),Tetrapropylammoniumbromide (TPABr) and Tetrabutylammonium bromide (TBABr) at 303.15, 308.15,313.15K



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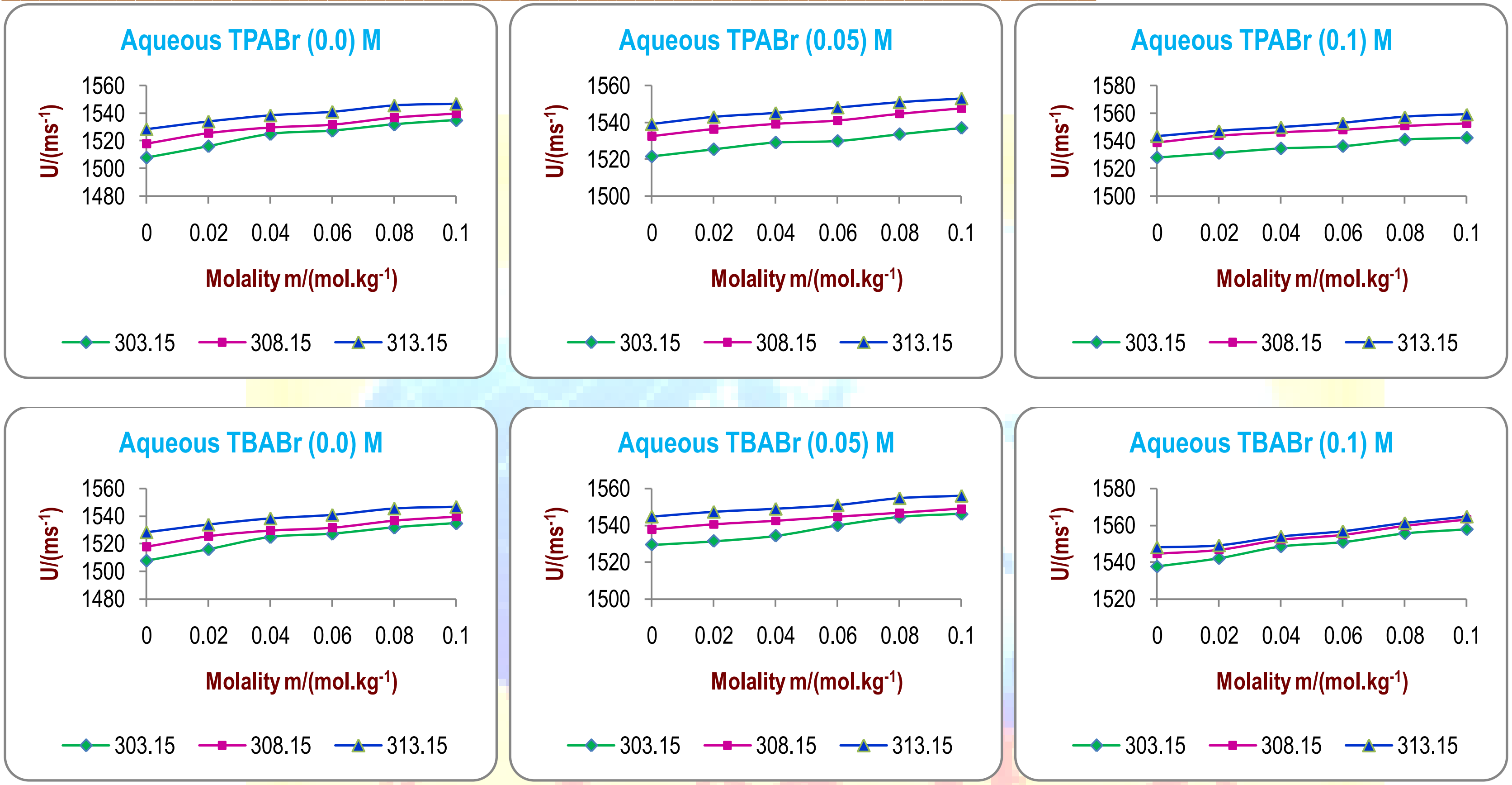

Fig.2 Values of Ultrasonic velocity (U) of Lactose in aqueous solution of Tetraethylammonium bromide (TEABr),Tetrapropylammoniumbromide (TPABr) and Tetrabutylammonium bromide (TBABr) at 303.15, 308.15,313.15K

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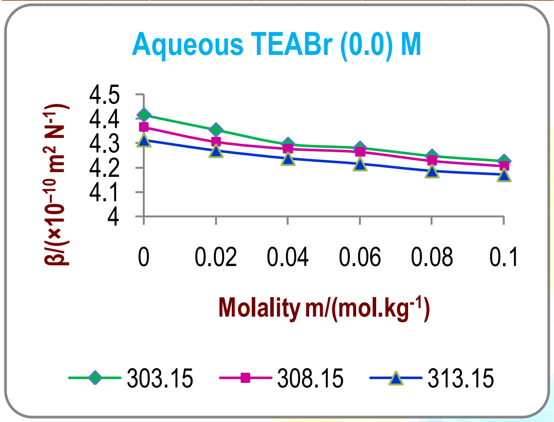

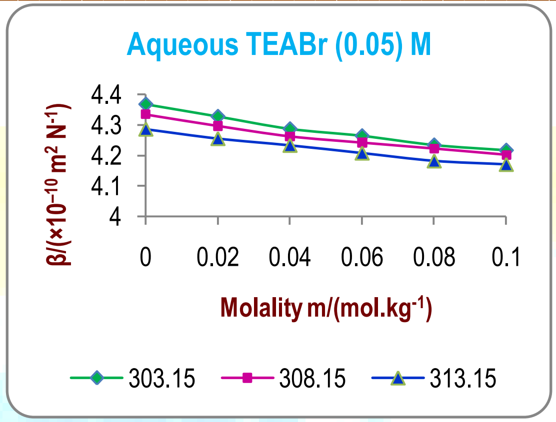

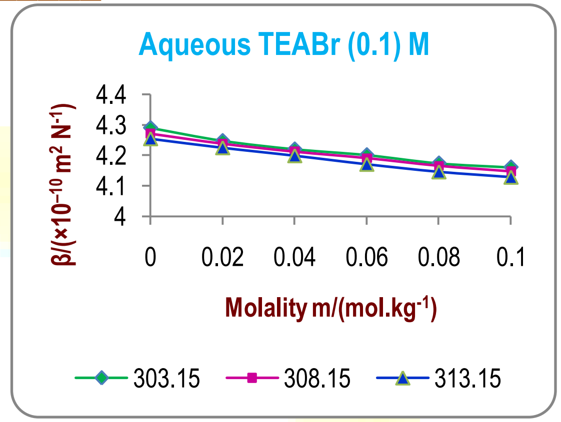

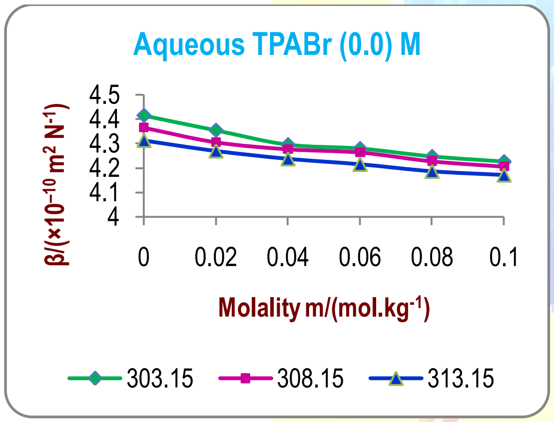

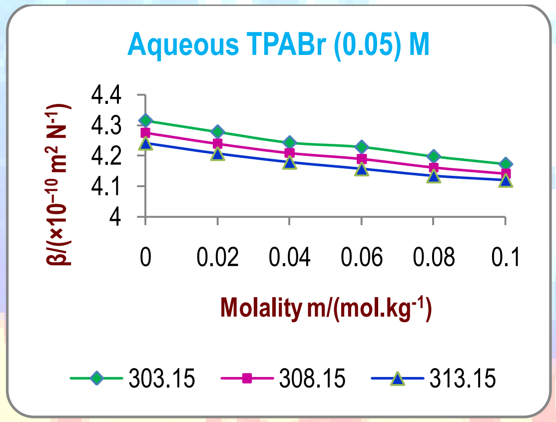

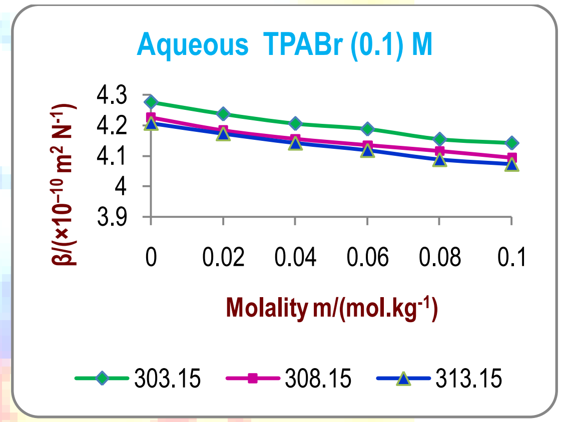

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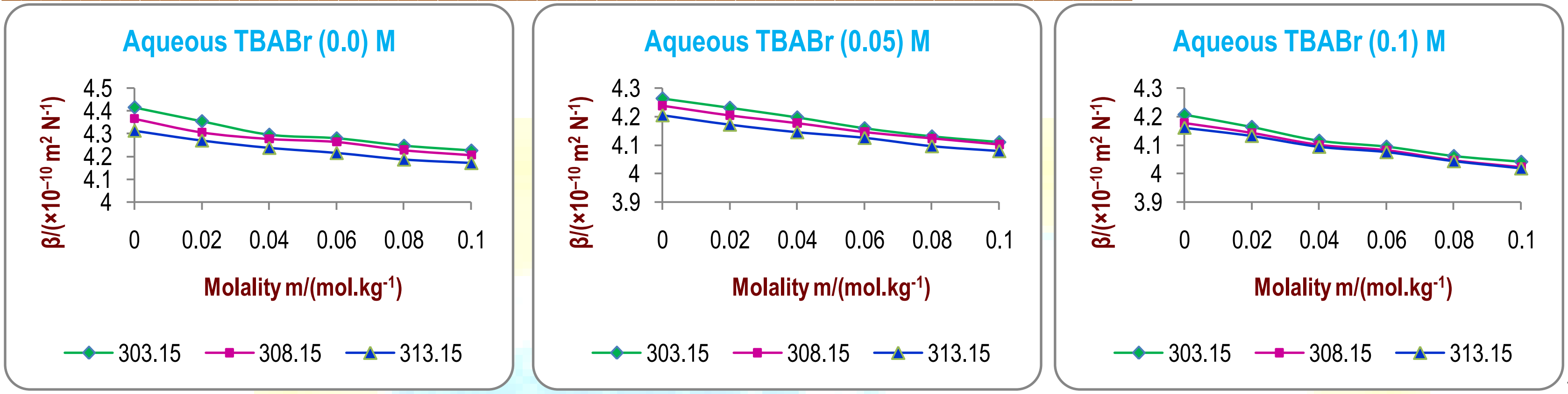

Fig.3 Values of Adiabatic compressibility (β) of Lactose in aqueous solution of Tetraethylammonium bromide (TEABr),Tetrapropylammoniumbromide (TPABr) and Tetrabutylammonium bromide (TBABr) at 303.15, 308.15,313.15K.