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ACOUSTIC STUDIES ON SOLUTE-SOLVENT INTERACTIONS OF L-ARGININE AT ISOELECTRIC pH

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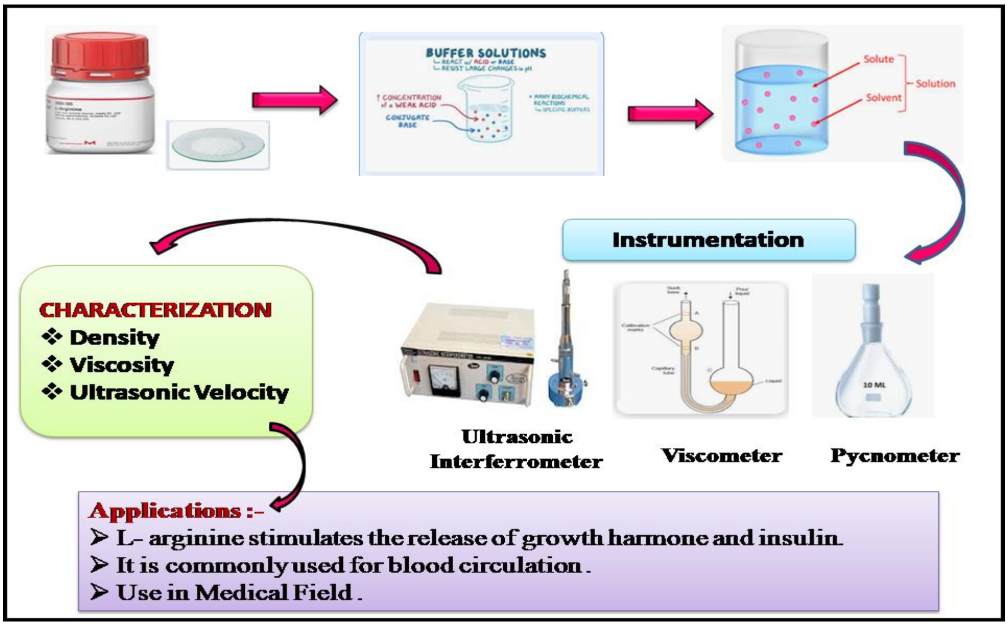
ABSTRACT:

Density (ϱ), Sound velocity (U) & Viscosity (η) measurements of L-Arginine at isoelectric pH are carried out at variable temperature [293.15 K to 313.15 K] with the interval of 5k and in aqueous solution at concentration ranges (0.02m to 0.10m). Acoustical parameters like Apparent molal compressibility (Φk), Limiting apparent molal compressibility (Φ°k), Apparent molal volume (Φ_v), limiting apparent molal volume (Φ°_v), Adiabatic compressibility (β), Acoustic impedance (Z), constants S_k and S_v, Viscosity coefficients *B* and *A* and Hydration number (η_H) have been calculated. These parameters help to gain supportive data concerning the existence of special kinds of intermolecular connections of solute-solvent system in the solutions at isoelectric pH. The outcomes are also attributed in the illumination of possible molecular exchanges in studied solvent system.

Keywords:- *L-Arginine, Ultrasonic velocity, Apparent molal volume, Viscosity coefficient constant, Jone Doles equation etc.*

GRAPHICAL ABSTRACT:-



Applications :-
- ➢ L- arginine stimulates the release of growth harmone and insulin.
- ➢ It is commonly used for blood circulation.
- ➢ Use in Medical Field.

INTRODUCTION:

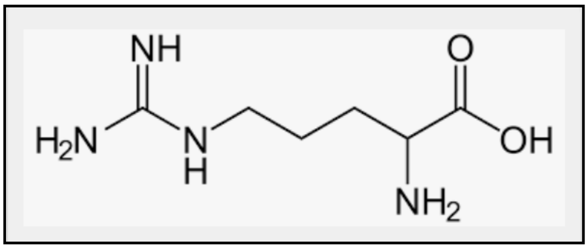
According to Liu, H., Lin, R., & Zhang, H. (2004), Proteins are very important group of macro-molecules which are fundamental in living cells, Amino acids are the important ingredients of proteins**.** But because of composite nature, their immediate structural investigation is to some extent complicated. So, the suitable approach is to learn such compounds is explained by Ali, A., Sabir, S., & Tariq, M. (2007). L-Arginine is naturally visible in fish, meat, dairy and poultry. It is necessary for making proteins and is commonly used for blood circulation has been studied by Hoseini, S. M., Ahmad Khan, M., Yousefi, M., & Costas,

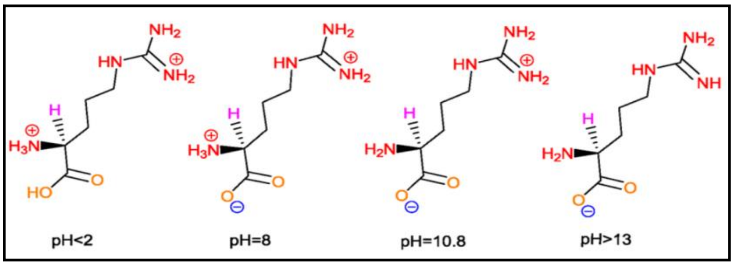
B. (2020). L-Arginine also provokes the discharge of hormone, insulin & extra substances in the body. It may be synthesized in a lab and used as supplements is cleared by Oh, H. S., Oh, S. K., Lee, J. S., Wu, C., & Lee, S. J. (2017) . The perceptive of drug-receptor connections in biological systems have been a issue of investigation. Structural arrangement of the protein or amino acid depends on pH of the medium. it may exist as cation, anion or zwitter ion decided by pH of the biological system.

Rooney, D., Jacquemin, J., & Gardas, R. (2009) suggested that ,the thermophysical characteristics of fluid systems are strictly linked to the molecular exchanges. Ultrasonic revisions of liquids enable to realize the molecular exchanges and structural activities of solute-solvent system .

Ultrasonic studies provide broad application in characterizing thermodynamic and physicochemical performance of given systems is explore by Bera, S., Mondal, D., Martin, J. T., & Singh, M. (2015) . Now a day's acoustical study of aqueous solutions helpful to denote different interaction in fluids systems. The spectroscopic techniques are also used to determine structural and functional association of bio-molecules. The methods are frequently consistent and therefore providing a matching information about the studied bio-molecular structure [Saudagar, P., & Tripathi, T. (Eds.). (2022)] Along with the techniques used, the measurement of sound velocity is simple to appreciate the character of molecular systems. Here the values of density (ρ), sound velocity (U) and viscosity (η), have been measured for L-Arginine of different molal composition, at different temperature and at isoelectric pH. The intend of this investigation is acoustic analysis of L- Arginine in isoelectric buffer system and results are correlated with electrostatic intermolecular interactions

MATERIALS AND METHODS :





Structure of L-Arginine at different pH

L-Arginine from E-Merck (CAS No.74-79-3) having purity 99.9% used without further purification. Distilled water is used to prepare fresh solution of L- Arginine. Molal solution of L-Arginine is prepared of concentration ranges from 0.02 to 0.10 molal with precision ± 0.001 g on an electronic balance (M- AJO20, aiwa). The density was work out using a pycnometer with an accuracy of ± 1 x 10^{-2} kgm^{-3}. An ostwald's viscometer (10 cm^3) was used to measure viscosity. An ultrasound interferometer (Mittal-Entp. Model: F-81) of frequency 2-MHz is employ to measure ultrasonic velocity of the solution with an overall accuracy of ± 0.1%. Constant temperature thermostat (Raaga Inds.) is used to maintain the desire temperature with accuracy ± 0.1K. and buffers solution of pH 10.8 (isoelectric point) is prepared by using NH_4Cl and NH_4OH (Basic buffer). This buffer system used as stock solvent to prepare experimental solution.

Calculations of various types of acoustical and thermodynamical parameters using experimentally measured parameters.

Apparent Molal Volume (Øv), Partial molal volume (Φ_v^o) and S_v:-

$$\Phi v = \frac{M}{\rho o} + \frac{1000\,(\rho o - \rho)}{m\rho o} \text{ -------------- } (1)$$

Where ,Φ_v - apparent molal volume , m-molality, ρ- solution density ,ρ_o - pure solvent density , M- mol.wt. If graph is plotted among Φ_v and $m^{1/2}$,relation obtained is called Messon's relation. This relation is ensuring from least square fit method.

$$\Phi_v = \Phi^0_v + S_v \, . \, m^{1/2} \text{ ----------------------}(2)$$

Where, Φ^0_v is the Partial molal volume and S_v is a slope. The plot of Φ^0_v against ($\sqrt{m}$) were establish to be linear

Viscosity Coefficient constant B and A:-

The viscosity coefficients B & A for the L-Arginine solutions were intended from the Jone -Doles equation.

$(\eta\ /\ \eta_o)\ - 1 = A + B\ .\ m^{1/2}$ -------(3)

$(\eta\ /\ \eta_o)$ - relative viscosity, m - molality, A and B are viscosity coefficients constants.

Hydration Number (η_H) :-

$$n_H = \frac{n1}{n2}\{1 - \frac{\beta}{\beta o}\}$$

Where n_1 :- No. of solvent moles

n_2:- No. of solute moles

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

Acoustic impendance $(Z) = \rho\ U$

Relative adiabatic compressibility

$(\Delta\beta\ /\beta o) = (\beta - \beta o)/\beta o$

The apparent molal compressibility $(\Phi k) = \frac{1000}{m\rho\rho o}(\rho o\beta - \rho\beta o) + \frac{\beta o M}{\rho o}$

Where $\beta o, \beta$ - Adiabatic compressibility of solvent and solution. $\rho o, \rho$ - Density of solvent and solution respectively

Limiting apparent molal compressibility or partial molal compressibility $(\Phi°k)$

$(\Phi k) = (\Phi° k) + S_k\ m^{1/2}$

RESULT AND DISCUSSION:

Temp. (K)	**Conc. mol Kg⁻¹**	**ρ × 10³ (kg /m³) Mean dev = ± 0.00150 Std.dev.= ± 0.00189**	**η × 10⁻³ (N s m⁻²) Mean dev.= ± 0.09844 Std.dev.= ± 0.12392**	**U m s⁻¹**
293.15 k	Pure Solvent	0.9952	0.956666	1487
	0.02	0.9964	0.968216	1491
	0.04	0.9975	0.975387	1495
	0.06	0.9985	0.983876	1499
	0.08	0.9994	0.99228	1503
	0.1	1.0003	0.999196	1506
298.15k	Pure Solvent	0.9947	0.883599	1496
	0.02	0.9958	0.887477	1500.66
	0.04	0.9969	0.891362	1505
	0.06	0.9979	0.903883	1509
	0.08	0.998802	0.911972	1513.5
	0.1	0.9998	0.909971	1518
303.15 k	Pure Solvent	0.994	0.7931	1506
	0.02	0.9951	0.798209	1510
	0.04	0.99613	0.800505	1514
	0.06	0.997101	0.804055	1518
	0.08	0.9981	0.809184	1522
	0.1	0.9991	0.810079	1525.5
308.15 k	Pure Solvent	0.9925	0.72077	1516
	0.02	0.99358	0.724256	1520
	0.04	0.9946	0.727821	1524
	0.06	0.995601	0.731318	1528
	0.08	0.9966	0.736203	1532
	0.1	0.9976	0.742559	1536
313.15 k	Pure Solvent	0.9907	0.659825	1524
	0.02	0.99172	0.661845	1528
	0.04	0.9927	0.666577	1532
	0.06	0.9937	0.671318	1536
	0.08	0.99468	0.680138	1540
	0.1	0.99567	0.674027	1543

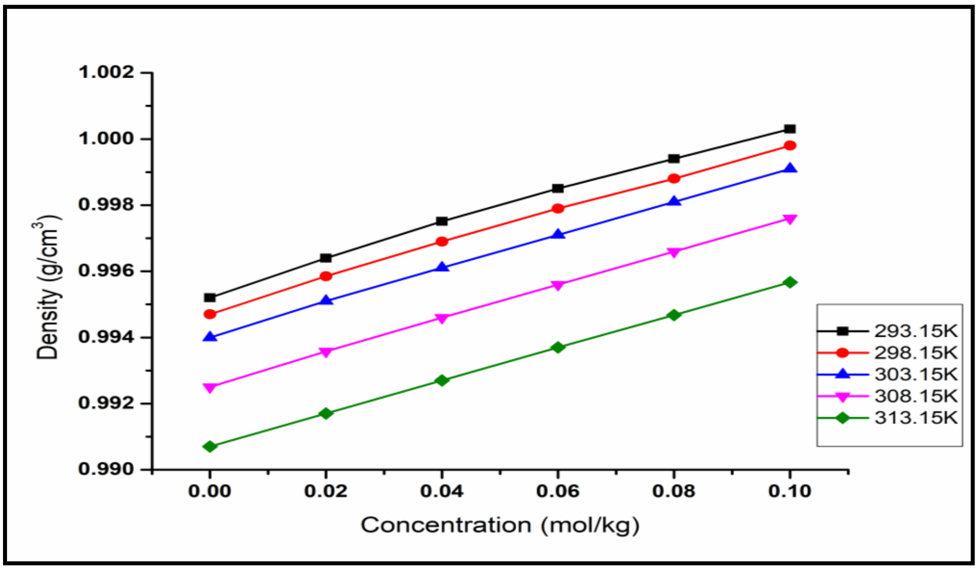


Fig-1. *Density Versus Concentration*

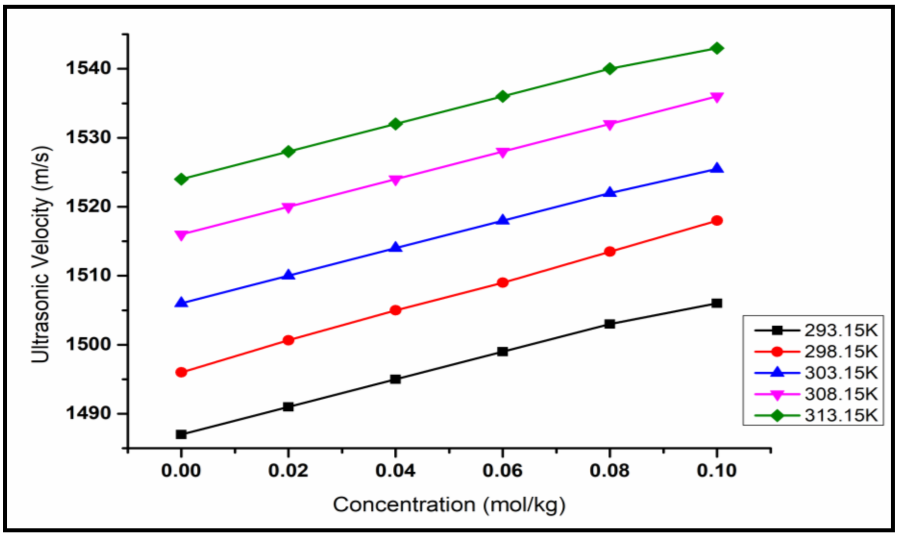


Fig-2.*Ultrasonic Velocity Versus concentration*

In the present investigation, density of the L-Arginine solution at isoelectric pH (pH=10.8) were work out from 293.15 K - 313.15 K. It was found that density of solution reduces with raise in temperature and it gain with enhance in molality as depicted in fig.1.

Sound velocity is an essential experimental factor. It provides information regarding extent of molecular interface, stiffness of medium & is greatly exaggerated by the molality & temperature has been studied by Sarvazyan, A. P. (1991). Ultrasound velocity gain with raise in temperature and with concentration in aqueous medium. The enlarge in ultrasound velocity in any solution indicates the bigger association and stiffness with the molecules of solution . The greater involvement may be due to different types of weak electrostatic forces such as hydrogen bonding, dipole - dipole, ion-dipole, dispersion forces and Van-der Walls forces is cleared by Yadav, M., & Yadav, H. S. (Eds.). (2021). The strong association in the present work is due to intermolecular hydrogen bonding among the amino group of L-Arginine and water molecules as shown in Fig.-2

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Table-1. Apparent molal volume (Φ_V), partial molal volume ($\Phi_v{}^o$) and S_v, of L- Arginine at different molality and temperature are given below.

Concentration (mol/kg)	√ ***m***	**Φv (m³ mol⁻¹) L- Arginine**				
		293.15k	298.15k	303.15k	308.15k	313.15k
0.02	0.1414	114.7508	119.8351	119.9195	120.1008	124.3565
0.04	0.2	117.2629	119.8351	121.1771	122.6082	125.3587
0.06	0.2449	119.7749	121.5107	123.248	123.4381	125.3659
0.08	0.2828	122.287	123.578	123.6922	123.8791	125.6182
0.10	0.3162	123.7942	123.8564	123.9437	124.131	125.6687
$\Phi_v{}^o$ (m^3 mol^{-1})		106.9	115.4	116.5	117.5	123.6
Sv (m^3 $Kg^{1/2}$ $mol^{-3/2}$)		53.14	26.53	24.84	22.43	6.986

The variation of Φ_v with a molality is given in following figure.

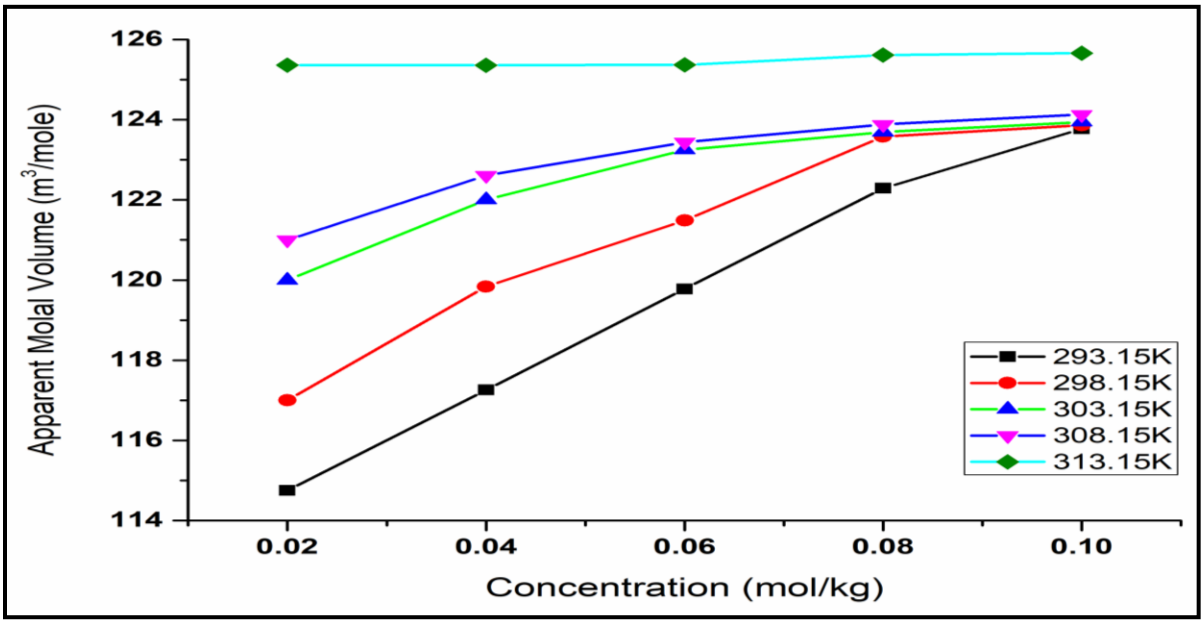

Fig-3. Apparent molal volume Versus Concentration.

Table-1 represents that the positive Φ_v values for the L-Arginine solution at isoelectric pH is signal of strong hydrophilic exchanges of solute. As the concentration of L-Arginine increases, the increase in Φ_v values indicates ionic interactions govern over ion-solvent interactions at higher concentrations [Zolkiflee, N. F., Affandi, M. M. M., & Majeed, A. B. A. (2020)]. The L-Arginine molecules tend to aggregate or associate, foremost to a drop in hydration .The structural organization of water around the solute is disrupted, causing a net expansion effect. This suggests weakening of ion-solvent relations and strengthening of ionic interactions as concentration rises. The prominent increase in Φ_v values of L-Arginine solution with concentration might be attributed to augmented hydrophobic scenery of alkyl side chain in L-Arginine is explained by Arsule, A. D., Sawale, R. T., Kalyankar, T. M., & Deosarkar, S. D. (2020).

Moreover, the values of Φ_v raise with boost in the molality of L-Arginine solution as solute-solvent interaction replace solute-solute interactions and the solute become less hydrated is suggested by Mou, I. A. (2018) . It may be due to the more number of solvent molecules are available initially. Hence salvation or hydration of solute molecule will be greater. As the molality is amplified, few solvent molecules are available and diminish the hydration process of solute molecule. Hence at higher concentration solute-solute interaction would be more. Apparent molal volume increases with increasing temperature. The boost of heat weakens the connection of molecules of water from the terminal zwitter-ions of L-Arginine , flee a few water molecules into the bulk and thus leading to growth in volume. In support, to this the values of L - Arginine, may be due to the larger alkyl chain ensuing in less approving interaction with the solvent. The Φ_v values for L-Arginine solution are good obedience with studied literature values and are reported in Table-1.

If Sv is positive, the ions acts as a structure-maker i.e, enhances the hydrogen-bonded structure of water. The positive & lesser values of Sv compared to Φ_v propose that the ions contributes positively to structuring of the solvent, but its effect is weaker than what the Φ_V magnitude alone indicates. There is moderate solute-solvent interaction, but not very strong. The solute is a weak structure-maker. It means solute molecules have greater affinity among themselves than the affinity toward the solvent molecules or ions. Such solute molecules are called kosmotropes. These solute molecules show weaker interactions with solvent molecules. They form weaker bonds (secondary forces) with solvent than the bond (H- bonding or Van der Waals'forces) present in solvent itself.

Table-2. Relative viscosities (η/η₀) for L-Arginine solution at different concentrations and temperatures are given below

Concentration (mol /Kg)	√ ***m***	**(η / η₀) for L-Arginine**				
		293.15k	298.15k	303.15k	308.15k	313.15k
0.02	0.1414	1.012178	1.004391	1.00644	1.004918	1.003082
0.04	0.2	1.01958	1.008786	1.00926	1.009782	1.010234
0.06	0.2449	1.028442	1.022955	1.01381	1.014634	1.017417
0.08	0.2828	1.037227	1.03211	1.02017	1.021412	1.030765
0.10	0.3162	1.044453	1.029846	1.0212	1.030124	1.021494

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The viscosity data for L-Arginine solution at isoelectric pH are represented in Table- (2). Normally, the viscosities follow expected order as they augment with rising L-Arginine molality and reduce with a rise of hotness of the solution.

Table -3. Viscosity coefficient constant *B* & *A* for L-Arginine solution.

L-Arginine	**$(\eta / \eta_o-1)/\sqrt{m}$ versus $\sqrt{m}$** ***B* = (Unit:- $dm^3\ mol^{-1}$)** ***A* = (Unit :- $dm^{3/2}\ mol^{-1/2}$)**	**$B \times 10^3 / \Phi^o_v$**
293.15k	*B* = 0.326 *A* = 0.037	3.087
298.15k	*B* =0.464 *A* = -0.034	4.021
303.15k	*B* = 0.155 *A* = 0.02	1.377
308.15k	*B* = 0.334 *A* = -0.016	2.876
313.15k	*B* = 0.366 *A* = -0.022	2.969

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If B - coefficient is positive but smaller in value. The solute still has structure-making tendency (since B > 0). But because the value is small, the solute–solvent interactions are weak to moderate. This means the solute only induces a slight ordering in the water structure and it is a weak structure-maker. Hydration is present, but not very strong. Smaller positive *B* coefficient suggests that the ions behave as a weak structure maker, with limited hydration & moderate solute-solvent exchanges.

The sign of (dB/dT) is another signal of structure strengthening and structure weakening ability of the ions that for L-Arginine is negative therefore this is classified as structure supporter in aqueous medium at isoelectric point. (kosmotrope). If (dB/dT) is negative as temperature increases, *B* decreases. This means solute-solvent interactions (hydration) weaken with ascend in temperature. The structured hydration shell around the solute breaks down, since hydrogen bonds in water become less stable at higher temperature. Hence, the solute behaves more like a structure-maker at lower temperatures, but its structure-making ability reduces as temperature rises.

Table-4. Adiabatic compressibility, Acoustic impedence, Apparent molal compressibility, Partial molal compressibility, Hydration number values for aqueous L-Arginine solutions at different temperatures**.**

Temp (K)	**Concentration (Mole/kg)**	**Adi Comp.(β) (Kg m^2S^1)x(10^{-7})**	**Acou.Imp.(Z)**	**App.Mol.Comp (∅k)x(10^{-5})**	**Hydration No (η_H)**
293.15	Pure	4.54	1479.862	-	-
	0.02	4.51	1485.632	-9.7	19.43477
	0.04	4.49	1491.277	-9.4	19.20124
	0.06	4.46	1496.752	-9.1	18.97057
	0.08	4.43	1502.098	-8.8	18.74268
	0.1	4.41	1506.452	-8	17.81199
298.15	Pure	4.49	1488.071	-	-
	0.02	4.46	1494.357	0.00011	21.6359
	0.04	4.43	1500.335	-0.0001	20.90579
	0.06	4.4	1505.831	-9.7	20.066
	0.08	4.37	1511.687	-9.6	20.01231
	0.1	4.34	1517.696	-9.6	19.99719
303.15	Pure	4.44	1496.964	-	-
	0.02	4.41	1502.601	-8.9	18.94374
	0.04	4.38	1508.141	-8.6	18.78744
	0.06	4.35	1513.6	-8.4	18.58566
	0.08	4.33	1519.108	-8.3	18.47989
	0.1	4.3	1524.127	-8	18.00944
308.15	Pure	4.38	1504.63	-	-
	0.02	4.36	1510.242	-8.6	18.84572
	0.04	4.33	1515.771	-8.4	18.61729
	0.06	4.3	1521.279	-8.3	18.48888
	0.08	4.28	1526.791	-8.2	18.38415
	0.1	4.25	1532.314	-8.1	18.29118
313.15	Pure	4.35	1509.827	-	-
	0.02	4.32	1515.348	-8.2	18.53266
	0.04	4.29	1520.817	-8	18.39456
	0.06	4.27	1526.323	-8	18.3156
	0.08	4.24	1531.807	-7.9	18.22311
	0.1	4.22	1536.319	-7.3	17.39775

Adiabatic compressibility:-

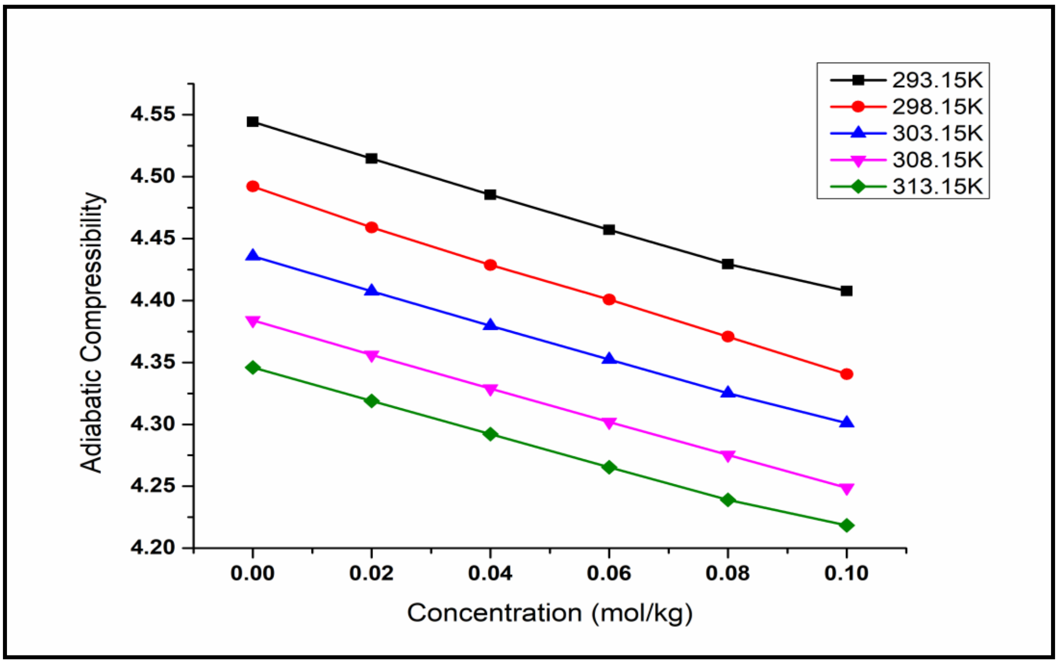

Fig-4. Adiabatic compressibility versus concentration

The adiabatic compressibility of L-Arginine solutions at the isoelectric pH decreases with increasing solute concentration**,** suggests that L-Arginine acts as a structure-maker (kosmotropic agent), enhancing water structure and reducing the medium's ability to be compressed. The raise in velocity and reduction in adiabatic compressibility with an increase in molality indicates that tightly bonded system formed by molecule. As the hotness gain, due to enlarged thermal motion of components in medium, strengthened interactive forces enhance resistance to compression. Hence, the adiabatic compressibility of the system decreases.

Acoustic Impendance:-. The sharp ascend in acoustic impendance at isoelectric pH indicates the strength of intermolecular interaction increases [Dhal, K., Singh, S., & Talukdar, M. (2023)]

Hydration Number:

Hydration number is significant factor in understanding the behavior of electrolyte in a solution. From the calculated values, it is found that with enlarge in concentration, hydration number slightly decreases. At low concentration, L-Arginine molecules are well separated, so water molecules can form complete hydration shells. As concentration increases, solute–solute interactions (ion-ion, zwitterion - zwitterion, dipole-dipole) become stronger and compete with solute-solvent interactions. This leads to overlap of hydration shells and a fall in the quantity of water molecules directly associated with each solute molecule. Therefore, the hydration number reduces with rising molality. This decreasing hydration number indicates destruction of solvent structure due to crowding effects and reduced availability of "free" water molecules. The salvation of any solute can also be selected from the values of (B/Φ^{o}_{v}) suggest the solute is hydrated or solvated. if this value is more than 2.5 , greater will be the association is suggested in their work by Shah, Shaukat A., Ratnakar B. Lanjewar, and Ashish P. Wakulkar . From table no.-3, it is clear that $(B/\Phi^{o}_{v}$) values are greater than 2.5 suggest strong association of L-Arginine with solvent at isoelectric point. This value decreases with boost in temperature indicates less salvation at higher temperature due to thermal agitation

In our experimental work, it is found that - hydration number reduces with enhance in concentration at all temperature but apparent molal volume grow with strengthen in molality

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& with boost in temperature. This may be due to, In dilute solutions, there are more solvent molecules are available for hydration, so each solute can form a full hydration shell. As concentration increases, number of solute increases and they compete for a limited solvent molecules. This reduces the number of solvent molecules available per solute, lowering the hydration number. Solute-solute interactions becoming more significant at higher concentrations and Structural changes takes place in the solvent, such as disruption of hydrogen bonding in solution. Thus, an increasing Φ_v with molality indicates that solute particles occupy more volume or cause greater expansion of the solution due to interactions. Apparent molal volume usually gains with temperature due to thermal expansion. Solute-solvent interactions can be weakening with temperature, leading to a greater effective volume of the solute. Hydrogen bonding in water breaks down at higher temperatures, leading to less tightly packed solvent molecules around the solute .

CONCLUSION :

The volumetric, viscometric & acoustical parameters derived for L -Arginine solution at isoelectric pH have been reported which are explore in terms of interactions of L-Arginine. The observed decrease in adiabatic compressibility and hydration number indicates that solute–solute interactions become more significant at higher concentrations, since water molecules are partially excluded from the hydration shell. L-Arginine has both positive and negative groups, but due to its strongly basic side chain, the molecule remains positively charged overall. When concentration increases, molecules come closer and mutual interactions (ion-ion, ion-dipole, hydrogen bonding) dominate over solute-solvent hydration. The enlarge in sound velocity and diminish in compressibility were ascribed to the creation of H- bonds among ions and solvent molecules.

From viscometric data, the minus values of dB/dT propose that L-Arginine acts as structure creator in aqueous medium at isoelectric pH but its structure – strengthening effect weakens with increasing hotness due to reduced hydration. These consequences are complementing the conclusions of dB/dT. The adiabatic compressibility decreases as the molality of L-Arginine in water/solvent increase, this indicates predominance of strong solute–solute interactions, tight packing of molecules, and reduced hydration of the solute. Smaller positive values of *B*- coefficient advise weak solute-solvent interactions and reduced hydration.

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