Molecular Interaction Dynamics of L-Arginine in 2% aqueous D-Fructose: A Comprehensive Insight of Volumetric, Acoustic and Viscometric properties

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Abstract

Present study investigates the interaction of L-Arginine in an aqueous D-fructose medium through ultrasonic velocity(U), density(ρ) and viscosity(η) measurements for the range of 293.15K to 313.15 K. Results obtained through experimental data is used to analyze some thermos-acoustic, volumetric and viscometrical parameters such as free length, Gibb's free energy, molar volume, Rao's constant, compressibility, Wada's constant and relaxation time of ternary mixture of L-Arginine and 2% D-Fructose in the concentration range of 0.01M to 0.1 M. Due to the unique structure of L-Arginine it allows to get involved in various chemical reactions. There are significant intermolecular and intra-molecular interactions in presence of co-solute, D-Fructose. The study throws light on ion-ion interactions in the complex behavior of L-Arginine and also the effect on heat-denaturation temperature of protein due to aqueous fructose in a ternary mixture.

Keywords: L-Arginine, 2% aqueous D-fructose solutions, Solute-solvent interaction, Solute-solute interaction and Ternary mixture.
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Introduction

In the last few decades, ultrasonic study of binary, ternary mixtures are used to analyze many thermodynamic parameters of the mixtures. Such studies have relevance in biochemical, pharmaceutical, geological nature of bio-fluid, etc [1].

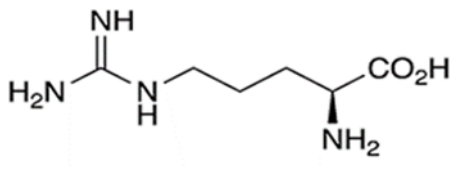

Figure 1: Molecular arrangements of L-arginine

Arginine is one of the amino acids and commonly found in the zwitterion form in the humanoid. L-arginine can be found in many natural sources and available in many food contents. The use of such food contents is varied in different societies. Biochemistry of L-arginine can be understood from its molecular structure. Refer figure 1. It has (-NH_2) and a (-COOH) attached to a central carbon atom and hence is always referred to as alpha-amino acid. It possesses a side chain that contains a Guanidine group (–NH–C (=NH)–NH_2). The dipolar ion form of molecules has an equal number of +ve and -ve functional groups (NH_3^+ and -COO^-) having ionized alanine zero charge termed as zwitterion. It holds significant importance in glycolysis and gluconeogenesis [2] showing varied functional behaviours viz. protein synthesis, energy fuel, urea synthesis, glucose-alanine cycle and homo synthesis [3].

During protein synthesis, a long peptide chain forms through peptide bonds between amino and carboxyl groups of zwitterions. A peptide is essentially a covalent bond formed between the amino group of one zwitterion and carboxylic group of another. L-alanine zwitterion is a common constituent of most proteins. L-alanine is an integral component of virtually entire proteins and peptides

produced in the human body [4]. Proteins are fundamental to human survival, playing a critical role in a wide array of biological functions due to their complex structures and diverse roles [5-6]. The correct folding and molecular assembly of proteins are crucial for their proper function, while misfolding or aggregation can lead to dysfunction and disease. Since amino acids are the basic building blocks of proteins and exhibit chirality, most natural proteins are composed of L-amino acids. These amino acids form specific structural motifs, including α-helices and β-sheets, which are essential for the protein's overall structure. The complexity of protein architecture is further enhanced by the interactions between side chains and the backbone, contributing to the stability and functionality of the protein [7]. These interactions have their strengths and weaknesses, in these protein-protein interactions, specifically the stacking of polypeptide chains, plays a crucial role in determining the protein structures. These forces play a crucial role in determining the size and compactness of the internal space of protein. However, the forces are not equally strong. External environmental conditions can induce the rupture of chemical internal bonds, resulting in enlargement of protein internal cavities, consequently protein structure and function undergo varying degree of alteration.

Currently, most protein research focuses on composite systems, integrating multiple proteins. In comparison to naturally occurring proteins, group [8]. Recently, chemical modification has been recognized to alter various chemical, physical properties. The functional properties of composite systems which include proteins as the main component are becoming important in the field of medicine delivery using nanotechnology. The composites with a combination of protein molecules and nanoparticles enhance the stability and content delivery of the medicine [9].

Brief research in the Web of Science database resulted in a set of approximately 100 records for the years 2018-2023. Key studies on intermolecular free lengths are documented in various journal articles Thermodynamic parameters provide valuable insights into hydration behaviour of solutes and their interactions with solvents [10-14]. Geochemical research temperature and pressure conditions [15]. Biology, medicine, catalysis and environmental studies intersect to address pressing issues in sustainability and human health [16-17], the study explores the molecular interactions between electrolytes and saccharides in various solutions. Extensive research has investigated the thermodynamic properties of amino acids in aqueous solutions containing saccharides [18-26]. In our preceding research, we have explored data on thermodynamic behaviour of amino acid-citrate salt solutions [27-31]. Citrate salts, which play a vital role in biology and industry, have diverse applications in food, pharmaceutical and cosmetic sectors as well as various biological processes [32-33].

Our research aims to conduct a systematic study on ultrasonic physical parameters of L-arginine in 2% aqueous D-fructose mixtures at temperatures in the range 293.15K, 298.15K, 303.15K, 308.15K and 313.15k along with thermodynamic parameters from these experimental data. The results will be put in terms of solute-solvent interaction, solute-solute, and solute-solvent in the presence of co-solute (2% D-fructose) and ion-ion interactions that occur in these ternary systems.

Experimental

L-arginine and D-fructose with AR grade with a minimum of 99.9% were used from E-Merck, Germany chemicals. They were used without further purification. The stock solutions of L-Arginine in 2% D-Fructose were prepared with double-distilled water (Model number SSA-DDAQ-15 All Quartz Distillation), and their concentrations were measured gravimetrically. Before creating the solutions, the water used for experimentation was degassed and deionized using distillation. The ternary aqueous solutions were prepared (Metter Toledo Balances & Scales) with a precision of ± 0.0001 g, and the two stock solutions and water were weighed in an appropriate proportion according to the desired concentration range 0.01 to 0.1 molal using mass burettes as per table 1.

Table 1: Specification of chemicals.

Name of chemical	Molar mass g.mol-1	Source	CAS number	Mass purity
L-Arginine	174.2	Sigma-Aldrich	74-79-3	≥ 99%
D-Fructose	180.16	Sigma-Aldrich	57-48-7	≥ 99%

In order to make a 0.01 to 0.1 molality solution, the two chemicals are taken in a clean and dry conical flask with a stopper. Necessary amounts of water and glucose were added. The necessary quantities of amino acids for a certain molality were dissolved, and a similar process has been used for other amino acids with varying molarities. Viscosity measurements were done (Ostwald Viscometer standards) for double-distilled water for flow time. Ultrasonic interferometer calibrated with double distilled water for sound velocity measurement (Considering Madder's Velocity), and the error factor was found to be 0.2 m/s. The velocity was measured using an ultrasonic interferometer operated at 3MHz, (Mittal's Interferometer Enterprises, New Delhi) with an overall precision of ±3 ms^{-1}. Adiabatic Densities were measured using a digital densitometer

(Metter Toledo). Temperatures were controlled to ± 0.002 K at intervals of 293.15 ≤ T/K ≤ 313.15K. Calibration of the densitometer about ± 0.0001g/ml. A layered measuring cylinder made of steel is used to hold the liquid mixture. At the proper temperature, it has been filled using an electronically controlled constant temperature bath. The temperature is accurate to within ± 0.1.

Equations

Following equations are used to calculate the derived parameters-

Adiabatic compressibility (β) is determined by using the following equation

$$\beta = \frac{1}{U^2\rho} \quad (1)$$

Relaxation time (τ) is calculated using the relation

$$\tau = \frac{4\eta}{3\rho U^2} \quad (2)$$

Molar volume (V_m) is volume occupied by a substance per unit amount of substance given by

$$V_m = \frac{M}{\rho} \quad (3)$$

Rao's constant (R_a) is given by a relation

$$R_a = \frac{M}{\rho}(u)^{\frac{1}{3}} \quad (4)$$

Free length defined as the distance between the substances of molecules in the liquid phase. Free length (L_f) is calculated using the relation

$$L_f = K_T\beta^{1/2} \quad (5)$$

K_T – Jacobson's Constant Values are taken from the book by Prof. Baldev Raj Wada's constant (W) is calculated by the following equation

$$w = (\frac{M_{eff.}}{\rho})\beta^{-1/7} \quad (6)$$

Where, $M_{eff.} = M +$ Weight of solute (7)

Where, $M = x_1M_1 + x_2M_2$ is the average molecular weight of solvent, here M_1 and M_2 are the molecular weights and x_1 and x_2 are the mole fraction of solvent components (fructose + water).

Results and Discussion

Physical Parameters: In the recent work, values of density, velocity, and viscosity are used to calculate derived thermodynamics for the mixture of L-arginine in 2% aqueous D-fructose mixtures at varied temperatures and concentrations. Table 2 to 5 indicate the data for temperature range 293.15 K to 313.15 K. Determination of density, viscosity, and velocity is a valuable tool to determine the properties of liquid structure due to the link between liquid state and macroscopic properties. [34].

The results can be discussed as follows-

Density, Viscosity, and Ultrasonic Velocity:

Experimentally evaluated parameters like density and viscosity are plotted as a function of concentration at constant temperature. It can be observed from figures 2 and 3 that there is a drop in density and viscosity with the rise in temperature. This is the general observation in almost all liquids and liquid mixtures. The effect of the quantity of solute present in the system on density and viscosity is being studied by many researchers at different temperatures [35].

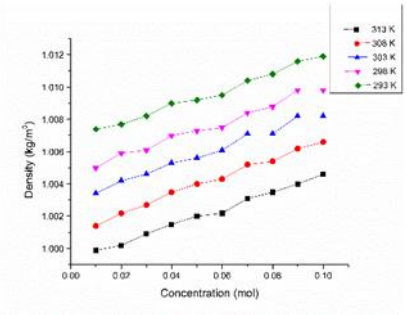

Figure 2: Variation of Density with respect to concentration.

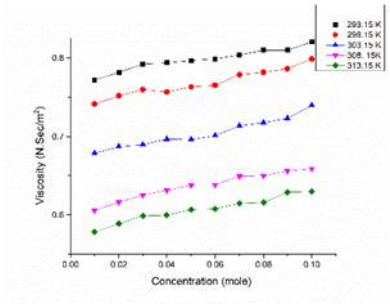

Figure 3: Variation of Viscosity with respect to concentration.

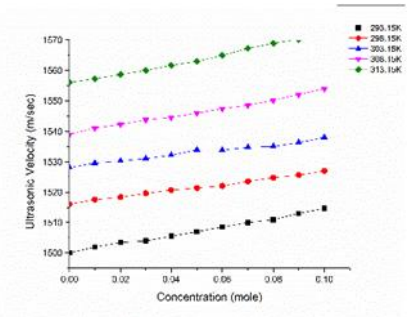

Figure 4: Variation of ultrasonic velocity with respect to concentration.

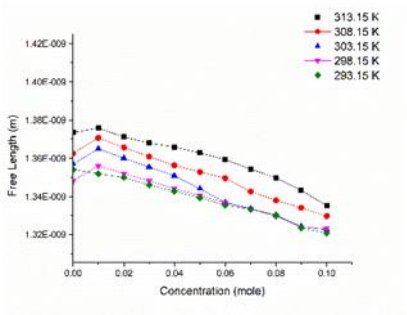

Figure 5: Variation of free length with respect to concentration.

However, in the ternary mixture, density decreases with the increase in temperature [36]. From Figure 3, viscosity decreases with temperature.

This is similar to the behaviour of density as it decreases with the rise in temperature. This is an indication of a stable structure and weak solute-solvent interaction [37]. With the rise in temperature, density and viscosity decrease. This trend reveals the decrease in molecular interaction in the mixture [38].

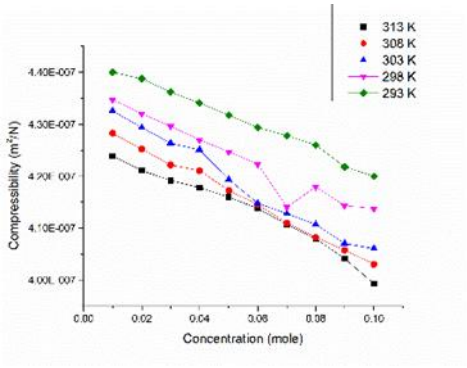

Figure 6: Variation of compressibility with respect concentration.

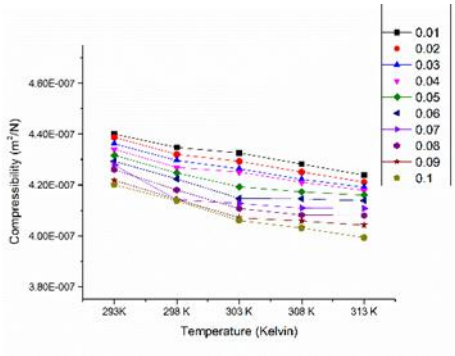

Figure 7: Variation of compressibility with respect to temperature.

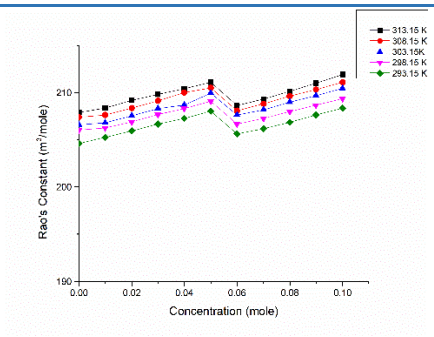

Figure 8: Variation of molar sound values velocity (Rao's constant) with respect to concentration.

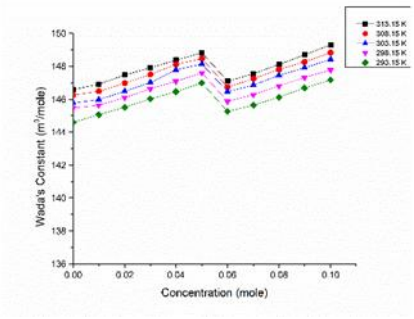

Figure 9: Variation of Molar compressibility (Wada's Constant) respect to concentration.

Perusal of Figure 3 shows a rise in the values. L-arginine and D-fructose are both polar in nature. As per, Jacobson's theory, there are two types of force existing in a liquid mixture, *viz*.,

(i) Physical forces due to dispersion.

(ii) Chemical forces due to dipole-dipole interactions [39, 40].

Being polar in nature, L-arginine and Aqueous D-fructose, dipole-dipole interactions are more prominent in the given mixture. Figure 4 indicates the variation of velocity with respect to concentration.

Figure 5 shows a graph of free length versus concentration. The least value of free length in the concentration range of 0.07 m to 0.1 m indicates the compact structure of the system.

Strong molecular contraction is the result of components that are much closer in the mixture [41-44]. Free length is the deciding factor for velocity. With the decrease in free length the ultrasonic velocity decreases. [45].

There exists a typical close packing due to the increasing strength of interaction across the components of the mixture. As the strength of the mixture increases, there exists a definite close packing.

Table 2 Experimental Readings for Arginine+ 2% D- Fructose at 293.15K.

Conc. (mole)	Density (ρ) (kg/m^3)	Viscosity (η) (N. Sec/m^2)	Ultra. Velo. (U) (m/sec)	1) Adi. Comp. (β) (m^2/N)	13) Relx. Time (τ) (Sec.)	Molar Vol. (V) m^3/mole)	14)Rao Const. (R) (m^3/mole)	15)Wada Const. (W) m^3/mole)	16)Free Length (L_f) (m)
Pure	1.007	0.78	1500	4.41355E-07	4.59009E-07	17.87488	204.6162	144.5874044	1.354E-09
0.01	1.0074	0.781251	1502	4.40006E-07	4.5834E-07	17.92352	205.2642	145.0442809	1.35193E-09
0.02	1.0077	0.781484	1504	4.38705E-07	4.57122E-07	17.97385	205.932	145.5131143	1.34993E-09
0.03	1.0082	0.796809	1508	4.36165E-07	4.63387E-07	18.02087	206.6535	146.0148633	1.34602E-09
0.04	1.009	0.797576	1511	4.3409E-07	4.61627E-07	18.06211	207.2637	146.448748	1.34281E-09
0.05	1.0092	0.791381	1515	4.31715E-07	4.55535E-07	18.114	208.0424	146.9846197	1.33913E-09
0.06	1.0095	0.798645	1519	4.29317E-07	4.57163E-07	17.88623	205.6071	145.2519445	1.33541E-09
0.07	1.0104	0.803268	1521	4.27807E-07	4.58192E-07	17.92582	206.1526	145.646733	1.33306E-09
0.08	1.0108	0.809388	1524	4.25956E-07	4.59685E-07	17.97452	206.8484	146.1328737	1.33017E-09
0.09	1.0116	0.809623	1531	4.21736E-07	4.55263E-07	18.01569	207.6392	146.6760851	1.32357E-09
0.1	1.0119	0.769748	1534	4.19964E-07	4.31022E-07	18.06567	208.3511	147.1715274	1.32078E-09

Table 3: Experimental Readings for Arginine+ 2% D- Fructose at 298.15K.

Conc. (mole)	Density (ρ) (kg/m^3)	Viscosity (η) (N. Sec/m^2)	Ultr. Velo. (U) (m/sec)	1) Adi. Compr. (β) (m^2/N)	13)Relx. Time (τ) (Sec.)	Molar Vol. (V) m^3/mole)	14)Rao Const. (R) (m^3/mole)	15)Wada Const. (W) m^3/mole)	16)Free Length (L_f) (m)
Pure	1.0049	0.75	1522	4.29584E-07	4.29584E-07	17.91223	206.0414	145.4501763	1.34808E-09
0.01	1.005	0.750778	1513	4.34666E-07	4.35117E-07	17.96632	206.2554	145.6444492	1.35603E-09
0.02	1.0059	0.75145	1517	4.3199E-07	4.32826E-07	18.00601	206.8932	146.0950803	1.35185E-09
0.03	1.0061	0.739348	1521	4.29636E-07	4.23534E-07	18.05848	207.6782	146.6352396	1.34816E-09
0.04	1.007	0.745506	1525	4.27003E-07	4.24444E-07	18.09798	208.3148	147.0850911	1.34403E-09
0.05	1.0073	0.745953	1529	4.24645E-07	4.22354E-07	18.14817	209.0749	147.6096573	1.34031E-09
0.06	1.0075	0.732029	1533	4.22348E-07	4.12228E-07	17.92174	206.6462	145.880949	1.33668E-09
0.07	1.0084	0.778636	1237	6.48079E-07	6.72823E-07	17.96137	192.81	137.5285118	1.65579E-09
0.08	1.0088	0.781222	1540	4.17978E-07	4.35378E-07	18.01015	207.9813	146.8186188	1.32975E-09
0.09	1.0098	0.776155	1546	4.14329E-07	4.28778E-07	18.0478	208.6864	147.309946	1.32393E-09
0.1	1.0098	0.768462	1547	4.13794E-07	4.2398E-07	18.10324	209.3725	147.7897337	1.32307E-09

Table 4: Experimental Readings for Arginine+ 2% D- Fructose at 303.15K.

Conc. (mole)	Density (ρ) (kg/m^3)	Viscosity (η)(N. Sec/m^2)	Ultra Velo. (U) (m/sec)	1) Adi. Comp. (β) (m^2/N)	13) Relx. Time (τ) (Sec.)	Molar Vol. (V) (m^3/mole)	14)Rao Const. (R) (m^3/mole)	15)Wada Const. (W) m^3/mole)	16)Free Length (L_f) (m)
Pure	1.0034	0.69	1527	4.27413E-07	3.9322E-07	17.93901	206.5752	145.7730666	1.35696E-09
0.01	1.0034	0.68829	1518	4.32496E-07	3.96911E-07	17.99497	206.8116	145.9810336	1.36501E-09
0.02	1.0042	0.688839	1523	4.29319E-07	3.94309E-07	18.0365	207.5163	146.472147	1.35998E-09
0.03	1.0046	0.687116	1528	4.26344E-07	3.90597E-07	18.08545	208.3069	147.0156319	1.35526E-09
0.04	1.0053	0.686311	1238	6.49027E-07	5.93912E-07	18.12859	194.6574	138.7798504	1.67215E-09
0.05	1.0056	0.694916	1540	4.19308E-07	3.88512E-07	18.17885	209.9294	148.1265956	1.34403E-09
0.06	1.0061	0.700751	1548	4.14779E-07	3.87542E-07	17.94668	207.6065	146.4618253	1.33676E-09
0.07	1.0071	0.733135	1551	4.12766E-07	4.03484E-07	17.98456	208.1791	146.8730525	1.33351E-09
0.08	1.0071	0.717282	1555	4.10645E-07	3.92731E-07	18.04055	209.0066	147.4387824	1.33008E-09
0.09	1.0082	0.722682	1561	4.0705E-07	3.92224E-07	18.07644	209.6914	147.917814	1.32424E-09
0.1	1.0082	0.709553	1563	4.06009E-07	3.84113E-07	18.13197	210.4253	148.4264564	1.32255E-09

Table 5: Experimental Readings for Arginine+ 2% D- Fructose at 308.15K.

Conc. (mole)	Density (ρ) (kg/m^3)	Viscosity (η)(N. Sec/m^2)	Ultr. Velo. (U) (m/sec)	1) Adi. C0mpr. (β) (m^2/N)	13)Relx. Time (τ) (Sec.)	Molar Vol (V) (m^3/mole)	14)Rao Const. (R) (m^3/mole)	15)Wada Const. (W) m^3/mole)	16)Free Length (L_f) (m)
Pure	1.0015	0.63	1536	4.2322E-07	3.55505E-07	17.97304	207.3729	146.2554421	1.36246E-09
0.01	1.0014	0.605463	1527	4.28267E-07	3.45733E-07	18.03091	207.6334	146.4780876	1.37055E-09
0.02	1.0022	0.605946	1532	4.25136E-07	3.4348E-07	18.07249	208.3392	146.9698602	1.36554E-09
0.03	1.0027	0.625325	1537	4.22164E-07	3.51986E-07	18.11972	209.1106	147.5016711	1.36075E-09

0.04	1.0035	0.620946	1242	6.4601E-07	5.3485E-07	18.16111	195.2163	139.1213573	1.68329E-09
0.05	1.004	0.627629	1545	4.17262E-07	3.49181E-07	18.20782	210.4913	148.4663454	1.35283E-09
0.06	1.0043	0.622196	1550	4.14451E-07	3.43826E-07	17.97884	208.0681	146.7409217	1.34827E-09
0.07	1.0052	0.649334	1556	4.10893E-07	3.55742E-07	18.01855	208.7965	147.2463193	1.34247E-09
0.08	1.0054	0.640883	1561	4.08183E-07	3.48797E-07	18.07106	209.6289	147.8149954	1.33803E-09
0.09	1.0062	0.635761	1565	4.05777E-07	3.4397E-07	18.11237	210.2875	148.2781698	1.33408E-09
0.1	1.0066	0.638831	1570	4.03036E-07	3.43296E-07	18.16079	211.0739	148.8185355	1.32957E-09

Table 6: Experimental Readings for Arginine+ 2% D- Fructose at 313.15K.

Conc. (mole)	Density (ρ) (kg/m^3)	Viscosity (η)(N. Sec/m^2)	Ultr. Velo. (U) (m/sec)	1 Adi. Compr. (β) (m^2/N)	13)Relx. Time (τ) (Sec.)	Molar Vol (V) (m^3/mole)	14)Rao Const. (R) (m^3/mole)	15)Wada Const. (W) (m^3/mole)	16)Free Length (L_f) (m)
Pure	0.9996	0.61	1539	4.22373E-07	3.4353E-07	18.0072	207.9022	146.5753843	1.37331E-09
0.01	0.9999	0.578226	1536	4.23898E-07	3.26811E-07	18.05796	208.3526	146.9128796	1.37578E-09
0.02	1.0002	0.578399	1541	4.21025E-07	3.24694E-07	18.10863	209.1638	147.468314	1.37112E-09
0.03	1.0009	0.598512	1544	4.19097E-07	3.34446E-07	18.1523	209.8042	147.9209297	1.36797E-09
0.04	1.0015	0.596013	1546	4.17763E-07	3.31989E-07	18.19737	210.4159	148.3557639	1.36579E-09
0.05	1.002	0.606519	1549	4.15939E-07	3.36366E-07	18.24416	211.0933	148.8302237	1.36281E-09
0.06	1.0022	0.589209	1553	4.13716E-07	3.2502E-07	18.01651	208.6386	147.0856727	1.35916E-09
0.07	1.0031	0.614538	1558	4.10696E-07	3.36518E-07	18.05628	209.3232	147.5646514	1.35419E-09
0.08	1.0035	0.607715	1563	4.0791E-07	3.30524E-07	18.10527	210.1155	148.1090237	1.34959E-09
0.09	1.004	0.628512	1570	4.0408E-07	3.38625E-07	18.15206	210.9725	148.6920743	1.34324E-09
0.1	1.0046	0.613291	1579	3.99248E-07	3.26474E-07	18.19694	211.8975	149.3161092	1.33518E-09

Figures 6 and 7 denote the variation of adiabatic compressibility with concentration and temperature, respectively. At lower concentrations, the changes in compressibility are complicated. There is a fall in compressibility with the rise in molar concentration. As the concentration of D-fructose increases in the mixture, molecular clustering breaks up, leading to increased dipole interaction.

The existence of a minimum value of compressibility for small concentrations of aqueous fructose shows the predominance of the associated nature of L-arginine. As the temperature decreases, associated groups break down increasingly, and the forces of attraction between the molecules decrease. This increases the adiabatic compressibility of this system. Figure 5 confirms the particular interactions that are apparent among the interactions of solute and solvent in the range of 0.07m to 0.1m.

Figures 8 and 9 are the curves for molar sound velocity (Rao's constant) vs. Concentration and Molar compressibility (Wada's Constant) vs. Concentration. It indicates both 'R' and 'W' have similar trends and it depends on temperatures [46]. The presence of complex formation is observed due to the nonlinear variation of 'R' and 'W' in the mixture.

Conclusions

The present investigation systematically elucidates the molecular interaction dynamics of L-arginine in 2% aqueous D-fructose solutions through precise measurements of density, ultrasonic velocity, and viscosity across a range of concentrations (0.01 M to 0.1 M) and temperatures (293.15 K to 313.15 K). The computation of various thermodynamic, acoustic, and volumetric parameters-including adiabatic compressibility, free length, relaxation time, molar volume, Rao's constant, and Wada's constant-provides deep insight into the intricate nature of solute–solvent and solute–solute interactions prevalent in these ternary systems.

The data reveal non-linear variation of these properties with concentration and temperature, indicative of significant intermolecular and intramolecular interactions modulated by the presence of D-fructose as a co-solute. The observed trends in adiabatic compressibility and free length suggest enhanced molecular association and structural modification within the solvent matrix, attributed to strong hydrogen-bonding and electrostatic interactions involving the zwitterionic center of L-arginine and the hydroxyl groups of D-fructose. The systematic decrease in viscosity and concurrent changes in relaxation time further highlight the complex interplay between cohesive and dispersive forces as the system transitions with concentration and thermal input.

These results collectively point towards the formation of a more structured and less compressible liquid system upon increasing L-arginine content, with evidence of cooperative solute–solvent association. Importantly, these findings have direct implications for understanding the biochemical behavior of amino acids in carbohydrate-rich environments,

highlighting potential modulations in protein stability, folding, and related thermodynamic properties due to fructose—a matter of significance in physiological and pharmaceutical contexts.

In summary, the volumetric, acoustic, and viscometric analyses presented herein advance the comprehension of molecular interactions in ternary aqueous systems, and provide a rigorous physicochemical framework for further explorations of biologically relevant mixtures.

Author Contribution

S. G. Rathod – Synthesis, Manuscript writing, Basic concept, Editing.
A. A. Mistry –Editing of manuscript, Validation, Supervision
Rupali Thete - Editing, Methodology, Visualization.
S. A. Shah– Editing, validation.

Data Availability

Data will be made available on reasonable request.

Declarations

Conflict of Interest: The authors declare no competing interests.

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