1. INTRODUCTION

Polyhydroxy compounds like carbohydrates and their derivatives (sugar alcohols) carry out numerous functions in biological procedures such as body growth, apoptosis, metabolic regulation, and cryo-preservation. Polyhydroxy compounds have received excessive consideration because of their preservation abilities in biological systems for preservation of vaccines and use in protein therapy, especially their role in the pharmaceutical and cosmetics industries. In particular, their existence may improve the antioxidant capacity of compounds to some degree. Sugar alcohol as galactitol is a sweetener, and its sweetness behavior is about 50–60% like the sweetness of sucrose. The interactions of sugar alcohol with model molecules of proteins at different temperatures play a significant role in understanding the nature of action of bioactive molecules and the thermodynamic behavior of the biochemical process in the living organisms.

Because proteins are particularly macromolecules, it is very difficult to directly study proteins, where amino acids are very appropriate model compounds of proteins. Amino acids are essential chemicals in the life cycle. Therefore, studies on sugar alcohol–amino acid interactions are very significant in food technology, immunology, biosynthesis, pharmacology, and medicines. However, thermo-physical studies of the interactions between sugar alcohol and amino acids in solutions are rare. The transport properties of amino acids in mixed aqueous solutions are affected by variations in solute concentration and temperature.

The amino acid used in the present study is L-arginine. L-Arginine is a nonessential amino acid and the building block of protein, and it has vast applications such as helping in controlling the blood pressure in the body and preventing swelling in the gastrointestinal area in newborn babies. Literature analysis revealed the thermodynamic studies of binary mixtures of sugar alcohol, but ternary systems of sugar alcohol with amino acid in aqueous systems are very complex and rare. These thermodynamic parameters are also affected by concentration and temperature variations.

In continuation of our earlier studies on sugar alcohol/sweeteners in aqueous systems, in the present study density and sound velocity for galactitol in aqueous amino acids (L-arginine) solutions at different temperatures (293.15–318.15 K) have
been measured. Using measured data, acoustic and volumetric parameters such as isentropic compressibility, specific molar volume, partial molar volume, expansibility, intermolecular free length, and transfer volume of galactitol from water to arginine solutions were computed. These parameters have been used to deliberate molecular interactions occurring in solutions and the structure-making/breaking tendency of the components in working solutions. The objective of the present work is to lift the primary knowledge related to amino acid–sugar alcohol interactions in aqueous solutions, which is still oblivious.

2. MATERIALS AND METHODS

2.1. Materials. Galactitol (Sigma-Aldrich CAS no. 608-66-2) and L-arginine (Merck CAS no. 74-79-3) were procured from Sigma-Aldrich and Merck. The doubly distilled and deionized water was obtained from Sigma-Aldrich (CAS no. 7732-18-5) and used in solution preparation. All chemicals used in the present work are given in Table 1. Furthermore, all apparatus used in this work was completely cleaned and dried before using it.

Table 1. Investigated Chemicals with Specification

| name of chemical | source          | CAS no. | purity (%) | purity method |
|------------------|-----------------|---------|------------|---------------|
| galactitol       | Sigma-Aldrich   | 608-66-2| 99         | crystallization|
| L-arginine       | Merck           | 74-79-3 | 99         | crystallization|

2.2. Methods. The density \( \rho \) and sound velocity \( u \) of galactitol in water and in aqueous \( L \)-arginine solutions (0.02, 0.04, and 0.06) \( \text{mol kg}^{-1} \) have been measured in the temperature range of 293.15–318.15 K and at a pressure of 101 kPa using DSA 5000M. The calibration of DSA 5000M was made from density and speed of sound data of pure water and air at 293.15 K according to the instruction manual. The density of a sample was calculated by determining the frequency of the U-tube, first injected water and then with instigated sample solution. The principle of measuring density was based on the circulation time method. The investigated sample was placed just like a sandwich between two piezoelectric ultrasonic transducers: one emitted sound waves over the sample-occupied cavity (3 MHz frequency), while the second transducer received emitted waves from the first one. Sound speed measurements have been performed through dividing the traveled distance between the source and the collector and the calculated transmission time for sound velocity 0.5 m s\(^{-1}\), having accuracy 0.1 m s\(^{-1}\). The Sartorius electronic digital weight balance (model no. SAR CP2245, USA) with an accuracy of ±0.001 mg has been used for preparation of solutions.

3. RESULTS AND DISCUSSION

3.1. Density and Sound Velocity. The density and sound velocity for pure water and along with literature values at various temperatures are listed in Table 2, which showed that measured values are comparable with literature values.

The measured values of density \( \rho \) and sound velocity \( u \) for galactitol in water and in aqueous \( L \)-arginine solutions at various temperatures are reported in Table 3.

Schemes S1 and S2 for galactitol in water and in aqueous \( L \)-arginine solutions are reported in the Supporting Information. In aqueous galactitol solution, the solvent molecules surround the solute molecules. As a result, solute–solvent interaction occurred, but the addition of \( L \)-arginine in aqueous solution replaced the solvent molecules. Thus, the structure become more compact and rigged than aqueous solutions. The presence of solute–solute and solute–solvent interactions leads to a change in density and sound velocity values.

Figure 1 represents the linear relationship between density versus molality for the binary system of galactitol in water. The addition of \( L \)-arginine to aqueous galactitol solution is performed. The density values increase with the rise in the molal concentration (0.02, 0.04, and 0.06) \( \text{mol kg}^{-1} \) of \( L \)-arginine, which indicates the fact that water molecules develop hydrogen bonds not only with galactitol but also with \( L \)-arginine molecules that make the structure compact and it became more prominent at large concentrations of \( L \)-arginine at the same temperatures.

However, there is a decline in density data with the rise in the temperature, because of the increase in kinetic energy of system molecules that govern structure binding energy of solvent molecules and make them less dense. The kinetic energies of solution molecules increase with the increase in temperature, and the solution becomes less dense. At the same time, sound waves pass over the solutions more comfortably at high temperature. Moreover, at high concentration of both galactitol and \( L \)-arginine in aqueous systems at constant temperature, molecular interactions are produced, which reduce the movement of free ions or molecules in solutions; as a result, interference decreased in the way of moving sound waves from solutions and it increased the sound velocity.

3.2. Apparent Molar and Partial Molar Volume. The apparent molar volume \( \Omega_V \) is significant to determine the molecular interactions of the galactitol in water and aqueous \( L \)-arginine solutions at different temperatures.

The apparent molar volume \( \Omega_V \) has been calculated through the experimental density values using the following equation:

\[
\Omega_V = \frac{1000(\rho^0 - \rho)}{m \rho^0} + \frac{M}{\rho} 
\]

where \( M, m, \rho, \) and \( \rho^0 \) are the molar mass of the solute, molality of solutions, density of solution, and density of the solvent, respectively. The calculated apparent molar volume \( \Omega_V \) values are presented at different temperatures in Table S1 of the Supporting Information. The partial molar volume \( \Omega_{V}^0 \) was obtained by a regression analysis based on the least squares method through the following equation:

\[
\Omega_V = \Omega_{V}^0 + S_V m
\]
| $m$/mol kg$^{-1}$ | 293.15 K | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K |
|-----------------|----------|----------|----------|----------|----------|----------|
| Galactitol + Water |          |          |          |          |          |          |
| 0.01226         | 1.000241 | 0.999056 | 0.997243 | 0.996139 | 0.994205 | 0.992321 |
| 0.05033         | 1.001369 | 1.000175 | 0.998352 | 0.997241 | 0.995293 | 0.993404 |
| 0.07495         | 1.002889 | 1.001682 | 0.999847 | 0.998726 | 0.996760 | 0.994865 |
| 0.09028         | 1.003826 | 1.002612 | 1.000768 | 0.999643 | 0.997665 | 0.995756 |
| 0.11480         | 1.005310 | 1.004084 | 1.002228 | 1.001093 | 0.999100 | 0.997179 |
| 0.13342         | 1.006843 | 1.005511 | 1.003323 | 1.002184 | 1.000176 | 0.998239 |
| 0.15304         | 1.007583 | 1.006341 | 1.004464 | 1.003320 | 1.001298 | 0.999339 |
| 0.17528         | 1.008884 | 1.007634 | 1.005744 | 1.004590 | 1.002556 | 1.000584 |
| 0.19501         | 1.010019 | 1.008764 | 1.006862 | 1.005701 | 1.003654 | 1.001661 |
| Galactitol + 0.02 mol kg$^{-1}$-Arginine |          |          |          |          |          |          |
| 0.03403         | 1.001502 | 1.000287 | 0.998867 | 0.997312 | 0.994428 | 0.993462 |
| 0.05273         | 1.002662 | 1.001438 | 1.000004 | 0.998441 | 0.995444 | 0.994555 |
| 0.07689         | 1.004146 | 1.002911 | 1.001460 | 0.999888 | 0.996975 | 0.995956 |
| 0.09895         | 1.005485 | 1.004242 | 1.002774 | 1.001197 | 0.998269 | 0.997224 |
| 0.11849         | 1.006659 | 1.005409 | 1.003926 | 1.002346 | 0.999407 | 0.998338 |
| 0.13555         | 1.007675 | 1.006419 | 1.004922 | 1.003341 | 1.000391 | 0.999303 |
| 0.15470         | 1.008805 | 1.007542 | 1.006030 | 1.004449 | 1.001490 | 1.000377 |
| 0.17757         | 1.010138 | 1.008867 | 1.007338 | 1.005759 | 1.002788 | 1.001647 |
| 0.19572         | 1.011181 | 1.009909 | 1.008363 | 1.006789 | 1.003807 | 1.002646 |
| Galactitol + 0.04 mol kg$^{-1}$-Arginine |          |          |          |          |          |          |
| 0.03403         | 1.001631 | 1.000508 | 0.998936 | 0.997312 | 0.994428 | 0.993462 |
| 0.05273         | 1.002788 | 1.001655 | 1.000696 | 0.998577 | 0.995636 | 0.994574 |
| 0.07689         | 1.004267 | 1.003123 | 1.002147 | 1.001019 | 0.997961 | 0.996868 |
| 0.09895         | 1.005602 | 1.004455 | 1.003277 | 1.002323 | 0.999253 | 0.998128 |
| 0.11849         | 1.006773 | 1.005613 | 1.004876 | 1.003469 | 1.000384 | 0.999336 |
| 0.13555         | 1.007784 | 1.006619 | 1.005870 | 1.004461 | 1.001365 | 1.000195 |
| 0.15470         | 1.008909 | 1.007738 | 1.006978 | 1.005566 | 1.002459 | 1.001264 |
| 0.17757         | 1.010237 | 1.009058 | 1.008284 | 1.006871 | 1.003752 | 1.002529 |
| 0.19572         | 1.011276 | 1.010095 | 1.009311 | 1.007899 | 1.004769 | 1.003525 |
| Galactitol + 0.06 mol kg$^{-1}$-Arginine |          |          |          |          |          |          |
| 0.03403         | 1.004063 | 1.002964 | 1.001515 | 0.999903 | 0.996953 | 0.994934 |
| 0.05273         | 1.005209 | 1.004102 | 1.002638 | 1.001020 | 0.998054 | 0.996011 |
| 0.07689         | 1.006676 | 1.005558 | 1.004082 | 1.002451 | 0.999464 | 0.997392 |
| 0.09895         | 1.008082 | 1.006873 | 1.005385 | 1.003745 | 1.000739 | 0.998641 |
| 0.11849         | 1.009157 | 1.008028 | 1.006528 | 1.004881 | 1.001858 | 0.999739 |
| 0.13555         | 1.010174 | 1.009026 | 1.007517 | 1.005863 | 1.002827 | 1.000689 |
| 0.15470         | 1.011296 | 1.010135 | 1.008616 | 1.006957 | 1.003904 | 1.001749 |
| 0.17757         | 1.012623 | 1.011444 | 1.009991 | 1.008249 | 1.005175 | 1.003002 |
| 0.19572         | 1.013666 | 1.012471 | 1.010927 | 1.009264 | 1.006172 | 1.003988 |

Table 3. Densities ($\rho$) and Sound Velocities ($u$) of Galactitol in Water and in Aqueous $L$-Arginine Solutions at Various Molal Concentrations of Galactitol ($m$) and Temperatures $^\circ$C
In eq 2, \( S \) is the experimental slope, which shows the strength of solute–solute interactions and is influenced by a number of factors. The computed results of apparent molar volume \( \Omega_V \), partial molar volume \( \Omega^0_V \), and experimental slope \( S \) for the investigated mixture (galactitol in aqueous L-arginine) at various temperatures are reported in Table 4.

The plots of \( \Omega_V \) against molality in Figures 2–5 reveal that an increase in \( \Omega_V \) values with the rise in temperature indicates that greater interactions occur in solutions. A positive increase in \( \Omega_V \) values leads to the formation of cavities in the structure. At low temperature (293.15 K), galactitol molecules do not avail greater space to adjust themselves with solvent molecules, whereas at high temperature (318.15 K) cavities have greater space for the accommodation of solute components to adequate in water molecules through a better approach.

Table 3. continued

\[
\begin{array}{ccccccccc}
\text{m/mol kg}^{-1} & 293.15 \text{ K} & 298.15 \text{ K} & 303.15 \text{ K} & 308.15 \text{ K} & 313.15 \text{ K} & 318.15 \text{ K} \\
\hline
\text{Galactitol + 0.02 mol kg}^{-1}\text{-L-Arginine} \\
0.13555 & 1491.82 & 1524.23 & 1536.21 & 1546.26 & 1555.02 & 1562.77 \\
0.15470 & 1492.98 & 1527.80 & 1539.68 & 1549.57 & 1558.29 & 1565.98 \\
0.17757 & 1494.38 & 1532.00 & 1543.78 & 1553.44 & 1562.09 & 1569.73 \\
0.19572 & 1495.51 & 1535.25 & 1546.97 & 1556.43 & 1565.01 & 1572.63 \\
\hline
\text{Galactitol + 0.04 mol kg}^{-1}\text{-L-Arginine} \\
0.03403 & 1492.54 & 1506.32 & 1519.77 & 1529.36 & 1538.25 & 1548.07 \\
0.05273 & 1496.32 & 1510.06 & 1523.52 & 1533.06 & 1541.90 & 1551.71 \\
0.07689 & 1501.15 & 1514.84 & 1528.31 & 1537.78 & 1546.53 & 1556.33 \\
0.09985 & 1505.50 & 1519.14 & 1532.62 & 1542.02 & 1550.68 & 1560.46 \\
0.11849 & 1509.31 & 1522.93 & 1536.40 & 1545.73 & 1554.27 & 1564.04 \\
0.13555 & 1512.55 & 1526.17 & 1539.65 & 1548.91 & 1557.35 & 1567.11 \\
0.15470 & 1516.15 & 1529.78 & 1543.26 & 1552.43 & 1560.72 & 1570.43 \\
0.17757 & 1520.40 & 1534.06 & 1547.47 & 1556.59 & 1564.63 & 1574.33 \\
0.19572 & 1523.69 & 1537.35 & 1550.78 & 1559.85 & 1567.59 & 1577.27 \\
\hline
\text{Galactitol + 0.06 mol kg}^{-1}\text{-L-Arginine} \\
0.03403 & 1495.34 & 1508.11 & 1520.32 & 1530.94 & 1539.9 & 1548.13 \\
0.05273 & 1499.23 & 1511.96 & 1524.16 & 1534.77 & 1543.67 & 1551.89 \\
0.07689 & 1504.19 & 1516.9 & 1529.09 & 1539.67 & 1548.45 & 1556.63 \\
0.09985 & 1508.67 & 1521.38 & 1533.56 & 1544.09 & 1552.72 & 1560.85 \\
0.11849 & 1512.58 & 1525.32 & 1537.49 & 1547.96 & 1556.40 & 1564.49 \\
0.13555 & 1515.94 & 1528.72 & 1540.90 & 1551.31 & 1559.53 & 1567.58 \\
0.15470 & 1519.66 & 1532.49 & 1544.68 & 1555.01 & 1562.91 & 1570.95 \\
0.17757 & 1523.69 & 1537.35 & 1550.78 & 1559.85 & 1567.59 & 1577.27 \\
0.19572 & 1527.41 & 1540.43 & 1552.73 & 1562.72 & 1569.91 & 1577.87 \\
\end{array}
\]

\(^{a}\text{m}\) is the molality of galactitol. The standard uncertainties of molality \((m)\), density \((\rho)\), sound velocity \((u)\), and temperature \((T)\) are ±0.00003 mol kg\(^{-1}\), ±0.000006 g cm\(^{-3}\), ±0.029 m s\(^{-1}\), and 0.02 K, respectively. The expanded uncertainties \((k=2)\) in \(\rho\) and \(u\) are ±1.32 \times 10^{-6} \text{ g cm}^{-3} and ±0.058 m s\(^{-1}\) respectively.

Figure 1. Variation of experimental density of galactitol in water corresponding to molality at different temperatures.

In eq 2, \( S \) is the experimental slope, which shows the strength of solute–solute interactions and is influenced by a number of factors. The computed results of apparent molar volume \( \Omega_V \), partial molar volume \( \Omega^0_V \), and experimental slope \( S \) for the investigated mixture (galactitol in aqueous L-arginine) at various temperatures are reported in Table 4.

The plots of \( \Omega_V \) against molality in Figures 2–5 reveal that an increase in \( \Omega_V \) values with the rise in temperature indicates that greater interactions occur in solutions. A positive increase in \( \Omega_V \) values leads to the formation of cavities in the structure. At low temperature (293.15 K), galactitol molecules do not avail greater space to adjust themselves with solvent molecules, whereas at high temperature (318.15 K) cavities have greater space for the accommodation of solute components to adequate in water molecules through a better approach. The apparent molar volume \( \Omega_V \) values of mixtures give understanding about molecular interactions existing among the particles of solution mixtures. Greater values of \( \Omega_V \) at high concentration and...
temperature also suggest for greater molecular interactions in solution mixtures, while at lower temperature solute molecules have less space to take in themselves with solvent molecules. Similarly, at higher concentration of solute, more solute—solvent interactions are observed because the solvent molecules do not directly attach to the solute surface through hydrogen bonding,

Table 4. Partial Molar Volumes ($\bar{V}_i$) and Experimental Slopes ($S_\bar{V}$) of Galactitol in Water and in Aqueous L-Arginine ($m_{Arg}$) and Temperatures

| $m_{Arg}$/mol kg$^{-1}$ | 293.15 K | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K |
|------------------------|-----------|-----------|-----------|-----------|-----------|-----------|
| 0.00                   | 118.608 ± 0.045 | 119.275 ± 0.002 | 119.926 ± 0.008 | 120.378 ± 0.034 | 121.287 ± 0.005 | 121.871 ± 0.01 |
| 0.02                   | 119.014 ± 0.008 | 119.606 ± 0.017 | 120.509 ± 0.008 | 121.112 ± 0.005 | 122.051 ± 0.007 | 123.412 ± 0.004 |
| 0.04                   | 119.162 ± 0.006 | 119.832 ± 0.008 | 120.740 ± 0.009 | 121.280 ± 0.058 | 122.257 ± 0.017 | 123.657 ± 0.009 |
| 0.06                   | 119.703 ± 0.012 | 120.184 ± 0.019 | 120.842 ± 0.022 | 121.631 ± 0.001 | 122.719 ± 0.016 | 124.157 ± 0.009 |
| 0.00                   | 7.256      | 6.667      | 6.282      | 6.154      | 5.967      | 4.006      |
| 0.02                   | 5.555      | 4.987      | 4.046      | 3.376      | 2.687      | 2.119      |
| 0.04                   | 5.514      | 4.534      | 4.032      | 3.436      | 2.366      | 2.060      |
| 0.06                   | 4.786      | 4.494      | 3.925      | 2.875      | 2.314      | 2.996      |

a$m_{Arg}$ is the molality of L-arginine. The standard uncertainties of molality ($m$), partial molar volume ($\bar{V}_i$), and temperature ($T$) are ±0.00003 mol kg$^{-1}$, ±0.03043 cm$^3$ mol$^{-1}$, and ±0.02 K, respectively.

Figure 2. Plot between apparent molar volume ($\bar{V}_i$) and molality ($m$) of galactitol in water at different temperatures.

Figure 3. Plot between apparent molar volume ($\bar{V}_i$) and molality ($m$) of galactitol in 0.02 mol kg$^{-1}$ L-arginine at different temperatures.
hence generating the gap among solution molecules that leads to positive change in apparent molar volume.\textsuperscript{19} The trends of $\Omega_V$ in solutions of galactitol in aqueous $L$-arginine and water ($0.06$ mol kg$^{-1}$-$L$-Arg > 0.04 mol kg$^{-1}$-$L$-Arg > 0.02 mol kg$^{-1}$-$L$-Arg > water) are noted.

Similar trends have been reported in aqueous carbohydrate solutions, and strong ongoing interactions were observed because of the presence of various force components present among solutions. The temperature also has slight effect on the $\Omega_V$ values, and values are also increased with the rise in temperature.\textsuperscript{20}

Commonly the following types of molecular interactions in the solutions of galactitol in water and in aqueous $L$-arginine are supposed:\textsuperscript{21}

- Hydrophilic–hydrophobic molecular interactions among the hydrophilic part of galactitol molecules and the hydrophobic part of $L$-arginine molecules.
- Hydrophobic–hydrophobic molecular interactions among the galactitol molecules and nonpolar components of $L$-arginine molecules.
- Ion–ion molecular interactions among ($-OH$) groups of galactitol molecules and functional group of $L$-arginine molecules.
- Ion-hydrophilic molecular interactions among hydrophilic components galactitol molecules and ion of $L$-arginine molecules.

All these molecular interactions can affect the stability of the solvent structure in solutions and have been subjected to the molalities and kind of ions existing in solutions.\textsuperscript{22} All $S_V$ values reported in Table 4 are positive, which show weak solute–solvent interactions in aqueous $L$-arginine solutions. The apparent molar volume $\Omega_V$ and partial molar volume $\Omega_V^0$ values indicate a linear relationship with rising temperature and concentration of $L$-arginine (amino acid) that illustrates strong solute–solvent interactions due to hydration of solvent molecules around the solute molecules in the solution mixture.\textsuperscript{10}
All these interactions are caused because of hydrogen bonding among hydroxyl groups of galactitol, water molecules, and anionic parts of l-arginine and amino group. In this research work, molecular interactions in aqueous galactitol and aqueous l-arginine systems can be described in the terms of solvation behavior and sweetness response. 

Table 4 showed that positive values of partial molar volume, which indicate more dominance of solute–solute interactions in aqueous l-arginine solutions than galactitol in water. 10 This increasing value of partial molar volume \( \Omega^0_T \) can be explained through hydrogen bonding among solution components, in which l-arginine molecules replaced the water molecules. As a result, solute–solute interactions became dominant, and this behavior of galactitol leads to the structure-making behavior. 23

### 3.3. Partial Molar Transfer Volume

The partial molar transfer volume \( \Delta \Omega^0_T \) explains solute–solvent interactions in binary and ternary solutions. It is calculated using eq 3:

\[
\Delta \Omega^0_T = \Omega^0_T(\text{l-arginine}) - \Omega^0_T(\text{water})
\]

The transfer volume \( \Delta \Omega^0_T \) values for the galactitol in water and in aqueous l-arginine are observed and reported in Table 5. All values are positive and can be explained in the light of the cosphere overlapping model. 24

According to the overlapping model, when two and more than two molecules come in contact with one another in solution, alteration in overlapping occurred; as a result, a change in their thermodynamic values occurred, which was termed as partial molar transfer volume. 25 The positive trend of partial molar transfer volume \( \Delta \Omega^0_T \) shows the impact of solute–solute and solute-solvent interactions. 26 Table 5 indicates that by increasing the concentration of l-arginine in aqueous solutions, more and more l-arginine molecules replaced the water molecules as a result greater solute–solute interactions. 26

| \( m_{\text{arg}} \) mol kg\(^{-1} \) | \( \Delta \Omega^0_T/\text{cm}^3 \text{ mol}^{-1} \) | \( T/\text{K} \) |
|---|---|---|
| 0.02 | 0.406 | 293.15 298.15 303.15 308.15 313.15 318.15 |
| 0.04 | 0.554 | 0.431 0.754 0.764 1.541 |
| 0.06 | 1.095 | 0.577 0.902 1.786 |

The positive trend of partial molar transfer volume indicates that by increasing the concentration of l-arginine in aqueous solutions, more and more l-arginine molecules replaced the water molecules as a result greater solute–solute interactions. 26

### 3.4. Partial Molar Expansibility and Hepler’s Constant

The partial molar expansibility is used to probe the molecular associations in various systems and can be explained in terms of structure-making or structure-breaking ability of solute components in solutions, and the following equation is used to express the variation of \( \Omega^0_T \) at different temperatures: 17

\[
\Omega^0_T = a + bT + cT^2
\]

where \( a, b, \) and \( c \) are fitting coefficients of eq 4 and are reported in Table 6.

The partial molar expansibility \( \Omega^0_E \) is determined as follows:

\[
\Omega^0_E = \left( \frac{\partial \Omega^0_V}{\partial T} \right)_p = b + 2cT
\]

The partial molar expansibility \( \Omega^0_E \) values obtained from eq 5 are listed in Table 7. From Table 7, positive \( \Omega^0_E \) value increases with the rise of temperature of the investigated mixture indicate the structure-making property of solute in solution, which may occur because of the existence of more powerful molecular interactions than electrostatic forces of attraction. 29

Hepler’s proposed a statistical equation to define the affinity of solute components, which support or interrupt solvent arrangement around solute particle in the solution. It is calculated by using eq 4 in the following way: 29

\[
\left( \frac{\partial \Omega^0_E}{\partial T} \right)_p = \left( \frac{\partial^2 \Omega^0_V}{\partial T^2} \right)_p = 2c
\]
The values obtained for \( (\partial^2 \Omega_v^0 / \partial T^2)_p \) stand for Hepler’s constant and are given in Table 7. This constant describes the structure-forming or structure-deforming ability of solute in solution. Galactitol solutions have positive trends of Hepler’s constant, showing that in these solutions the galactitol acts as a structure maker. From Table 7, positive and increasing trends of partial molar expansibility indicated that at less concentration of arginine, molecules develop less interactions with surrounding water molecules via hydrogen bonding between water and solute molecules, such type solute acts as structure-making compounds. Moreover, when there are greater numbers of \( L \)-arginine molecules in solutions, then amino groups of \( L \)-arginine molecules developed greater hydrogen bonding both with water molecules and \(-OH\) groups of galactitol, so such solutions have greater positive values and show more structure-making behavior.

### 3.5. Apparent Specific Volume

The apparent specific volume (ASV) was determined by the given equation.

\[
\text{ASV} = \frac{\Omega_v}{M}
\]

\(\text{ASV} = \frac{\Omega_v}{M}\) where \(\Omega_v\), \(\Omega_v\), and \(M\) stand for apparent specific volume, apparent molar volume, and molar mass of solute (galactitol) respectively, and values obtained from eq 7 are shown in Table 8. The ASV values increase with the concentration of solute. The reported values for the sweetness range are \((0.51 \sim 0.71) \text{ cm}^3 \text{ g}^{-1}\), and the results obtained in such a range are considered as sweeteners.
On the other hand, aqueous galactitol solution has an ASV range of \((0.65 - 0.67)\) \(\text{mol g}^{-1}\) but aqueous L-arginine systems have ranges \((0.65 - 0.68)\) \(\text{mol g}^{-1}\) respectively.\(^{34,35}\) The ASV values for galactitol in the presence of L-arginine have nearly ideal sweet range values. It is also observed that the sweetness of galactitol enhanced in the presence of L-arginine.

### 3.6. Apparent and Partial Molar Isentropic Compressibility

The apparent molar isentropic compressibility \(\Omega_k\) provides understanding of intermolecular collaborations in solutions and is calculated by the following equation:\(^{36,10}\)

\[
\Omega_k = \frac{1000(\rho_0^0 \beta_s - \rho_0^P \beta_s^0)}{m \rho} + \frac{\beta M}{\rho} \quad (8)
\]

where \(\beta_s\) stands for compressibility of solutions and \(\beta_s^0\) for water, respectively and \(\beta_s^0\) is determined by the following equation:

\[
\beta_s^0 = \frac{1}{\rho} \quad (9)
\]

Calculated results of \(\Omega_k\) for aqueous galactitol and aqueous L-arginine solutions are reported in supplementary Table S2. The \(\Omega_k\) shows negative decreasing trends for all investigated solutions, which indicate that part of solutions involved by hydrate solute components is compressed lightly because of the existence of strong molecular collaborations than in bulk mixtures.\(^{37}\)

Figures 6–9 represent the negative decreasing trend due to compatibility of the structure.\(^{38}\) The investigated solutions have a negative decreasing trend with concentration and temperature for aqueous sugar alcohol and aqueous L-arginine systems because of the development of the complex arrangement in solutions.\(^{10,17}\) From measured data of \(\Omega_k\), the apparent molar compressibility at infinite dilution is known as partial molar compressibility \(\Omega_k^0\) and is calculated as\(^{39}\)

\[
\Omega_k^0 = \Omega_k + S_k m \quad (10)
\]
Figure 8. Plot between apparent molar isentropic compressibility ($\phi_K$) and molality ($m$) of galactitol in 0.04 mol kg$^{-1}$-arginine at different temperatures.

Figure 9. Plot between apparent molar isentropic compressibility ($\phi_K$) and molality ($m$) of galactitol in 0.06 mol kg$^{-1}$-arginine at different temperatures.

Table 9. Partial Molar Compressibilities ($\phi_K^0$) and Experimental Slopes ($S_k$) for Galactitol in Water and in Aqueous L-Arginine Solutions at Various Molal Concentrations of L-Arginine ($m_{\text{Arg}}$) and Temperatures$^a$

| $m_{\text{Arg}}$/mol kg$^{-1}$ | 293.15 K | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K |
|-------------------------------|----------|----------|----------|----------|----------|----------|
| 0.00                          | -9.405 ± 0.032 | -9.270 ± 0.007 | -9.172 ± 0.029 | -8.051 ± 0.053 | -7.963 ± 0.001 | -7.775 ± 0.048 |
| 0.02                          | -9.714 ± 0.011 | -9.365 ± 0.006 | -9.041 ± 0.010 | -8.683 ± 0.011 | -8.395 ± 0.006 | -8.152 ± 0.003 |
| 0.04                          | -10.078 ± 0.006 | -9.614 ± 0.007 | -9.287 ± 0.005 | -8.976 ± 0.006 | -8.646 ± 0.005 | -8.383 ± 0.006 |
| 0.06                          | -10.132 ± 0.006 | -9.648 ± 0.004 | -9.406 ± 0.003 | -9.160 ± 0.005 | -9.040 ± 0.009 | -8.795 ± 0.006 |
| 0.00                          | 4.402 | 4.771 | 3.793 | 3.498 | 2.966 | 2.927 |
| 0.02                          | 4.636 | 4.738 | 5.953 | 4.267 | 3.901 | 3.314 |
| 0.04                          | 5.890 | 5.694 | 5.169 | 5.548 | 4.850 | 4.077 |
| 0.06                          | 4.659 | 3.478 | 4.233 | 4.054 | 4.324 | 4.087 |

$^a$m$_{\text{Arg}}$ is the molality of aqueous L-arginine. The standard uncertainties of molality ($m$), partial molar isentropic compressibility ($\phi_K^0$) are ±0.00003 mol kg$^{-1}$, ±0.015 cm$^3$ mol$^{-1}$ Pa$^{-1}$, and ±0.02 K, respectively.
where $\phi_k^0$ stands for partial molar compressibility, $S_k$ stands for the experimental slope, and $m$ is molality of solution. Calculated data of $\phi_k^0$ are stated in Table 9. Negative trends of $\phi_k^0$ describe that molecular interactions became weak in the ternary mixture of solute-solvent due to participation of L-arginine molecules as a cosolute, which replaced the water molecules from solution and increased the solute–solute interactions so, L-arginine molecules are dynamic in the solution that modified the structure.

Likewise, attractive forces, various types of molecular interactions among hydrophilic groups of L-arginine and water molecules create contact with sugar alcohol molecules and as a result form hydrophilic–hydrophilic interactions. Table 9 shows the experimental slope $S_k$ data, which provide information about solute–solute interactions.

### 3.7. Intermolecular Free Length

Ultrasonic velocity is a parameter known as intermolecular free length $L_f$ which has been determined by the use of adiabatic compressibility values of solutions. It measures the degree of intermolecular interactivity that exists in solutions. A statistical equation is used to measure the intermolecular free length:

$$L_f = k \sqrt{\beta} \tag{11}$$

where $k$ is the Jacobson constant of temperatures ($k = (93.875 + 0.375T) \times 10^{-8}$).

Results of $L_f$ are given in Table 10 that showed with the rise in temperature and concentrations of solute of solutions, the values of $L_f$ decrease as a result of strong association among solute (galactitol), solvent (water), and amino acid molecules, which

| $m$ (mol kg$^{-1}$) | 293.15 K | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K |
|---------------------|----------|----------|----------|----------|----------|----------|
| Galactitol + Water  |          |          |          |          |          |          |
| 0.032               | 1.856    | 1.839    | 1.825    | 1.819    | 1.815    | 1.809    |
| 0.050               | 1.850    | 1.833    | 1.820    | 1.810    | 1.800    | 1.790    |
| 0.074               | 1.843    | 1.826    | 1.813    | 1.805    | 1.799    | 1.795    |
| 0.090               | 1.839    | 1.822    | 1.809    | 1.800    | 1.795    | 1.789    |
| 0.114               | 1.832    | 1.815    | 1.802    | 1.799    | 1.789    | 1.785    |
| 0.133               | 1.827    | 1.810    | 1.797    | 1.795    | 1.785    | 1.779    |
| 0.153               | 1.821    | 1.805    | 1.792    | 1.789    | 1.779    | 1.775    |
| 0.175               | 1.815    | 1.799    | 1.786    | 1.785    | 1.775    | 1.769    |
| 0.195               | 1.810    | 1.794    | 1.781    | 1.770    | 1.769    | 1.765    |

| Galactitol + 0.02 mol kg$^{-1}$ L-Arginine |          |          |          |          |          |          |
| 0.034               | 1.861    | 1.844    | 1.831    | 1.820    | 1.810    | 1.802    |
| 0.052               | 1.858    | 1.836    | 1.822    | 1.811    | 1.803    | 1.795    |
| 0.076               | 1.856    | 1.830    | 1.817    | 1.806    | 1.798    | 1.790    |
| 0.098               | 1.853    | 1.823    | 1.810    | 1.799    | 1.791    | 1.783    |
| 0.118               | 1.855    | 1.817    | 1.804    | 1.793    | 1.786    | 1.778    |
| 0.135               | 1.847    | 1.812    | 1.799    | 1.788    | 1.781    | 1.773    |
| 0.154               | 1.845    | 1.807    | 1.794    | 1.784    | 1.776    | 1.769    |
| 0.177               | 1.842    | 1.802    | 1.789    | 1.779    | 1.772    | 1.764    |
| 0.195               | 1.840    | 1.796    | 1.783    | 1.773    | 1.766    | 1.759    |

| Galactitol + 0.04 mol kg$^{-1}$ L-Arginine |          |          |          |          |          |          |
| 0.032               | 1.860    | 1.844    | 1.831    | 1.820    | 1.817    | 1.802    |
| 0.050               | 1.850    | 1.834    | 1.818    | 1.808    | 1.800    | 1.790    |
| 0.074               | 1.844    | 1.828    | 1.813    | 1.803    | 1.795    | 1.785    |
| 0.090               | 1.837    | 1.821    | 1.806    | 1.796    | 1.788    | 1.778    |
| 0.114               | 1.830    | 1.815    | 1.799    | 1.790    | 1.782    | 1.772    |
| 0.133               | 1.824    | 1.809    | 1.794    | 1.784    | 1.777    | 1.767    |
| 0.153               | 1.820    | 1.804    | 1.789    | 1.780    | 1.773    | 1.763    |
| 0.175               | 1.814    | 1.799    | 1.784    | 1.775    | 1.768    | 1.758    |
| 0.195               | 1.808    | 1.793    | 1.778    | 1.769    | 1.763    | 1.753    |

| Galactitol + 0.06 mol kg$^{-1}$ L-Arginine |          |          |          |          |          |          |
| 0.032               | 1.860    | 1.844    | 1.831    | 1.820    | 1.817    | 1.810    |
| 0.050               | 1.844    | 1.829    | 1.816    | 1.805    | 1.797    | 1.789    |
| 0.074               | 1.838    | 1.824    | 1.810    | 1.799    | 1.792    | 1.784    |
| 0.090               | 1.831    | 1.816    | 1.803    | 1.792    | 1.785    | 1.777    |
| 0.114               | 1.824    | 1.810    | 1.797    | 1.786    | 1.779    | 1.771    |
| 0.133               | 1.818    | 1.804    | 1.791    | 1.781    | 1.774    | 1.766    |
| 0.153               | 1.813    | 1.799    | 1.786    | 1.776    | 1.769    | 1.762    |
| 0.175               | 1.808    | 1.794    | 1.781    | 1.771    | 1.764    | 1.757    |
| 0.195               | 1.802    | 1.787    | 1.775    | 1.765    | 1.759    | 1.752    |

“$m$ is the molality of aqueous galactitol. The standard uncertainties of molality ($m$), intermolecular free length ($L_f$), and temperature ($T$) are $\pm 0.00003$ mol kg$^{-1}$, $\pm 0.001$ m, and $\pm 0.02$ K, respectively.”

Table 10. Intermolecular Free Lengths ($L_f$) for Galactitol in Water and in Aqueous L-Arginine Solutions at Different Temperatures and Molalities of Galactitol ($m$)
make the structure more complex and packed and as a result $L_t$ values are decreased.\textsuperscript{41,42}

The solute molecules or ions may lead to structural change around solvent molecules. These structural changes depend on the corresponding strength of hydrogen or other types of bonding in the solutions. The calculated values of $L_t$ for aqueous sugar alcohol and l-arginine solutions in water show that by rising concentrations of solute, cosolute, and temperature, the $L_t$ values decline, which indicate the strong molecular interactions such as solute–solute and solute–solute molecules. This decreasing trend of $L_t$ with concentration and temperature makes the structure compact.\textsuperscript{41} A decreasing trend in $L_t$ values also indicates the close stuffing of molecules and structure-making behavior.

4. CONCLUSIONS

In the present work, volumetric and acoustic parameters for galactitol in aqueous l-arginine solutions at different temperatures were observed and used to explore the solvation behavior and sweetness response in terms of solute–solute and solute–solvant molecular interactions, and the effect of l-arginine on aqueous galactitol solutions at different temperatures (293.15, 298.15, 303.15, 313.15, and 318.15 K) was also described. Positive trends of apparent molar volumes $\Omega_0$ and partial molar volume $\Omega_0^f$ have been observed, which were retained on increasing the concentration of both galactitol and l-arginine due to hydration of water molecules around the galactitol molecules in the solution mixture. A positive trend of $\Delta \Omega_0^f$ indicated greater strength of electrostatic forces of attractions among solutions, which were much stronger due to the presence of solute–solute interactions in ternary solutions as compared with binary systems. These forces can affect the stability of the solvent structure in solutions and have been subjected to the molalities and kinds of ions existing in solutions. ASV described that investigated galactitol lies in the sweeteners range and its solvation behavior of aqueous galactitol also enriched in the existence of l-arginine than aqueous sugar alcohol solution. Positive trends of $\Omega_0^f$ indicated the structure-promoting response of galactitol in l-arginine solutions.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c04102.

Scheme S1 for galactitol in aqueous solution, Scheme S2 for galactitol in aqueous l-arginine, Table S1 containing data for apparent molar volume ($\Omega_0$), and Table S2 for apparent molar isentropic compressibility ($\Omega_t$) (PDF)

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Notes
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