

Investigations to Explore Molecular Interactions and Sweetness Response of Polyhydroxy Compounds with Amino Acids in Aqueous Systems

Muhammad Asghar Jamal,* Uzma Shahazidy, Fatimah A. Al-Saeed,* Khalid M. Al Syaad, Majid Muneer, Ikram Ahmed, and Ahmed Ezzat Ahmed



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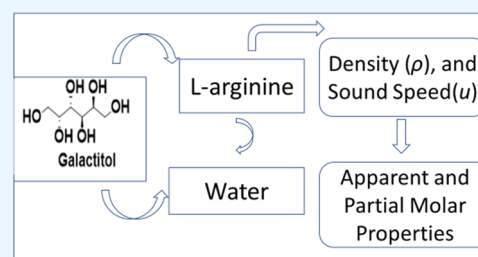


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ABSTRACT: In conjunction with the development of people's living standards, the modern world demands good-quality food such as sweets, candies, chocolates, diet drinks, beverages, and so on, but because of obesity and other health issues people concentrate more on sugar-free or low-calorie products. Polyols are such a kind of food with desirable qualities, and they play a role in controlling the blood glucose level in diabetic patients. The density (ρ) and sound speed (u) of sugar alcohol in water and in (0.02, 0.04, and 0.06) mol kg⁻¹ L-arginine solutions at different temperatures (293.15–318.15 K) and atmospheric pressure were measured by using Anton Paar DSA5000M. Experimental density and sound velocity data were further used to compute volumetric and acoustic parameters such as apparent molar volume (Φ_V), partial molar volume (Φ_V^0), compressibility (Φ_k^0), expansibility (Φ_E^0), and so on. The positive trends of apparent molar volume (Φ_V), and partial molar volume Φ_V^0 , values indicate strong hydrophilic interactions in ternary solutions. These interactions give a complete picture about solvation behavior, the effect of temperature, and hydrogen bonding present among (galactitol + L-arginine) mixtures. The apparent specific volume values were calculated, and it was found that these values of the investigated mixtures lie on the borderline with the reported values of sweeteners. This study may offer a new vision in elucidation of mechanistic modifications between sugar alcohol, amino acid, and their mode of interactions.



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.^{1,2} 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.¹ Sugar alcohol as galactitol is a sweetener, and its sweetness behavior is about 50–60% like the sweetness of sucrose.³ 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.⁴

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.⁵ 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.⁶

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,^{7,8} 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.⁹

In continuation of our earlier studies on sugar alcohol/sweeteners in aqueous system,¹⁰ 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

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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.¹¹ 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 ρ and sound velocity u of galactitol in water and in aqueous L-arginine solutions (0.02, 0.04, and 0.06) 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.¹² 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 radiated 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 ρ 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

Table 2. Comparisons of Experimental Densities (ρ^0) and Sound Velocities (u^0) of Pure Water with Literature Values at Different Temperatures

T/K	experimental work		literature values	
	ρ^0 /g cm ⁻³	u^0 /m s ⁻¹	ρ^0 /g cm ⁻³	u^0 /m s ⁻¹
293.15	0.998203	1482.64	0.998202, ³	1483.35, ³
			0.998220 ²	1483.10 ²
298.15	0.997037	1497.65	0.997748, ³	1497.12, ³
			0.997100 ²	1497.00 ²
303.15	0.995675	1509.10	0.995241, ¹	1509.51, ³
			0.994058 ¹⁷	1509.40 ²
308.15	0.994149	1519.56	0.994258, ³	1520.17, ³
			0.994058 ²	1519.83 ²
313.15	0.992240	1529.25	0.992570, ³	1529.89, ³
			0.992210 ²	1528.80 ⁵
318.15	0.990583	1537.49	0.990356, ³	1537.11, ³
			0.990201 ²⁰	1537.21 ¹⁹

replaced the solvent molecules. Thus, the structure become more compact and rigged than aqueous solutions.¹³ 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) mol kg⁻¹ of L-arginine, which indicates the fact that water molecules develop hydrogen bonds not only with galactitol but also with of 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.¹⁴ 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.¹⁵

3.2. Apparent Molar and Partial Molar Volume. The apparent molar volume Φ_V is significant to determine the molecular interactions of the galactitol in water and aqueous L-arginine solutions at different temperatures.^{10,16}

The apparent molar volume Φ_V has been calculated through the experimental density values using the following equation:¹⁶

$$\Phi_V = \frac{1000(\rho^0 - \rho)}{m\rho^0\rho} + \frac{M}{\rho} \quad (1)$$

where M , m , ρ , and ρ^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 Φ_V values are presented at different temperatures in Table S1 of the Supporting Information. The partial molar volume Φ_V^0 was obtained by a regression analysis based on the least squares method through the following equation:¹⁶

$$\Phi_V = \Phi_V^0 + S_V m \quad (2)$$

Table 3. Densities (ρ) and Sound Velocities (u) of Galactitol in Water and in Aqueous L-Arginine Solutions at Various Molal Concentrations of Galactitol (m) and Temperatures^a

$m/\text{mol kg}^{-1}$	$\rho/\text{g cm}^{-3}$					
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K
	Galactitol + Water					
0.03226	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	1.006423	1.005188	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 L-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	0.995544	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 L-Arginine					
0.03403	1.001631	1.000508	0.999836	0.998452	0.995424	0.994387
0.05273	1.002788	1.001655	1.000968	0.999577	0.996536	0.995474
0.07689	1.004267	1.003123	1.002417	1.001019	0.997961	0.996868
0.09895	1.005602	1.00445	1.003727	1.002323	0.99925	0.998128
0.11849	1.006773	1.005613	1.004876	1.003469	1.000384	0.999236
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 L-Arginine					
0.03403	1.004063	1.002964	1.00151	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	1.008002	1.006873	1.005385	1.003745	1.000739	0.998641
0.11849	1.009167	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	1.00991	1.008249	1.005175	1.003002
0.19572	1.013666	1.012471	1.010927	1.009264	1.006172	1.003988
	$u/\text{m s}^{-1}$					
$m/\text{mol kg}^{-1}$	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K
	Galactitol + Water					
0.03226	1488.80	1503.39	1515.82	1525.39	1535.1	1542.95
0.05033	1492.21	1506.88	1519.33	1528.62	1538.34	1546.19
0.07495	1496.86	1511.60	1524.06	1532.97	1542.69	1550.55
0.09028	1499.66	1514.52	1526.99	1535.65	1545.37	1553.25
0.11480	1504.21	1519.15	1531.62	1539.87	1549.6	1557.51
0.13342	1507.59	1522.61	1535.09	1543.01	1552.75	1560.68
0.15304	1511.12	1526.23	1538.74	1546.30	1556.04	1564.01
0.17528	1515.13	1530.27	1542.79	1549.91	1559.71	1567.71
0.19501	1518.59	1533.78	1546.34	1553.04	1562.91	1570.96
	Galactitol + 0.02 mol kg ⁻¹ L-Arginine					
0.03403	1485.80	1504.73	1517.08	1527.64	1536.61	1544.52
0.05273	1486.89	1508.40	1520.71	1531.21	1540.13	1548.02
0.07689	1488.31	1513.09	1525.32	1535.72	1544.59	1552.45
0.09895	1489.62	1517.34	1529.48	1539.75	1548.58	1556.41
0.11849	1490.79	1521.04	1533.11	1543.24	1552.07	1559.84

Table 3. continued

$m/\text{mol kg}^{-1}$	$u/\text{m s}^{-1}$					
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K
Galactitol + 0.02 mol kg ⁻¹ 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
Galactitol + 0.04 mol kg ⁻¹ 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
0.09895	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
Galactitol + 0.06 mol kg ⁻¹ 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
0.09895	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	1524.04	1536.95	1549.19	1559.37	1566.89	1574.89
0.19572	1527.41	1540.43	1552.73	1562.72	1569.91	1577.87

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

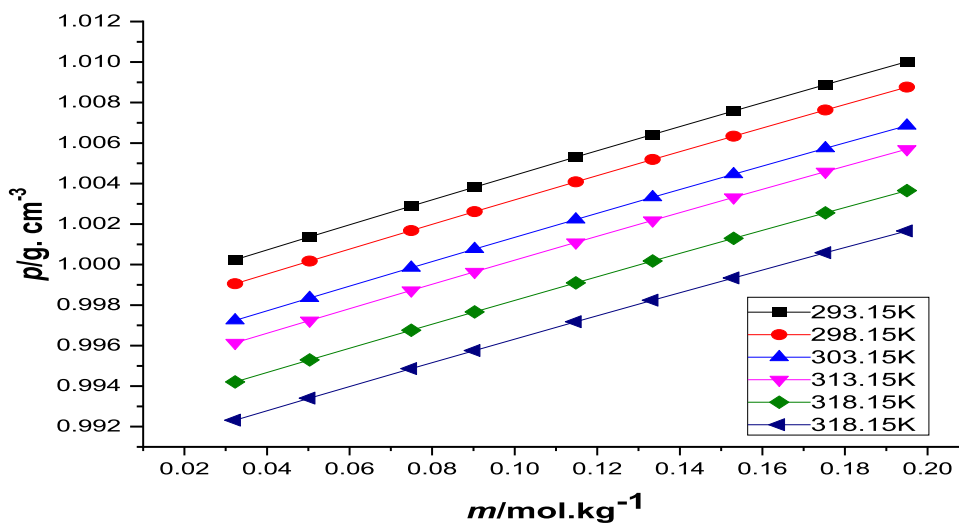


Figure 1. Variation of experimental density of galactitol in water corresponding to molality at different temperatures.

In eq 2, S_V is the experimental slope, which shows the strength of solute–solute interactions and is influenced by a number of factors.¹¹ The computed results of apparent molar volume Φ_V , partial molar volume Φ_V^0 , and experimental slope S_V for the investigated mixture (galactitol in aqueous L-arginine) at various temperatures are reported in Table 4.

The plots of Φ_V against molality are presented in Figures 2–5, which reveal that an increase in Φ_V values with the rise in temperature indicates that greater interactions occur in

solutions. A positive increase in Φ_V values leads to the formation of cavities in the structure.¹⁷

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.¹⁸ The apparent molar volume Φ_V values of mixtures give understanding about molecular interactions existing among the particles of solution mixtures. Greater values of Φ_V at high concentration and

Table 4. Partial Molar Volumes (ϕ_V^0) and Experimental Slopes (S_V) of Galactitol in Water and in Aqueous L-Arginine Solutions at Various Molal Concentrations of L-Arginine (m_{Arg}) and Temperatures^a

$m_{\text{Arg}}/\text{mol kg}^{-1}$	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K
	$\phi_V^0/\text{cm}^3 \text{mol}^{-1}$					
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
	$S_V/\text{kg cm}^3 \text{mol}^{-1}$					
0.00	7.256	6.667	6.282	6.154	5.967	4.006
0.02	5.555	4.987	4.046	3.276	2.687	2.119
0.04	5.514	4.534	4.032	3.058	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 (ϕ_V^0), and temperature (T) are ±0.00003 mol kg⁻¹, ±0.03043 cm³ mol⁻¹, and ±0.02 K, respectively.

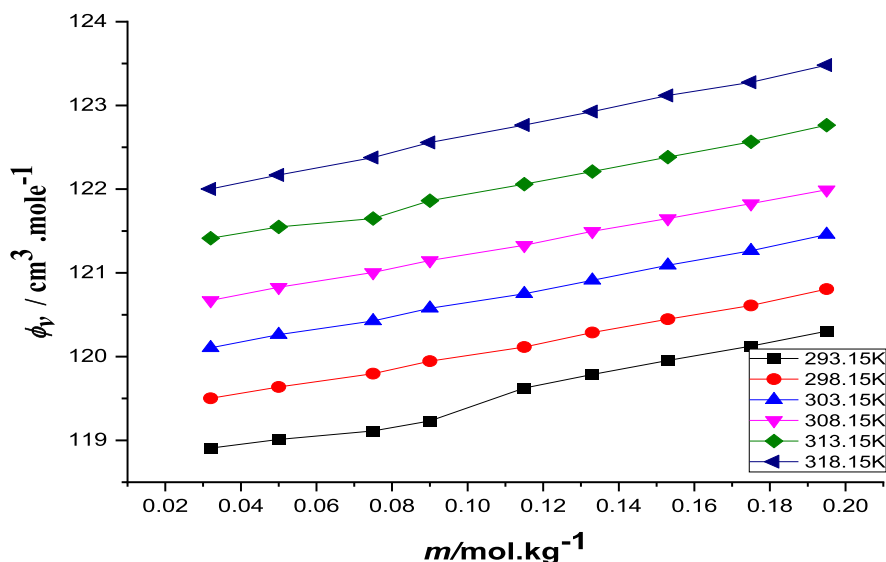


Figure 2. Plot between apparent molar volume (ϕ_V) and molality (m) of galactitol in water at different temperatures.

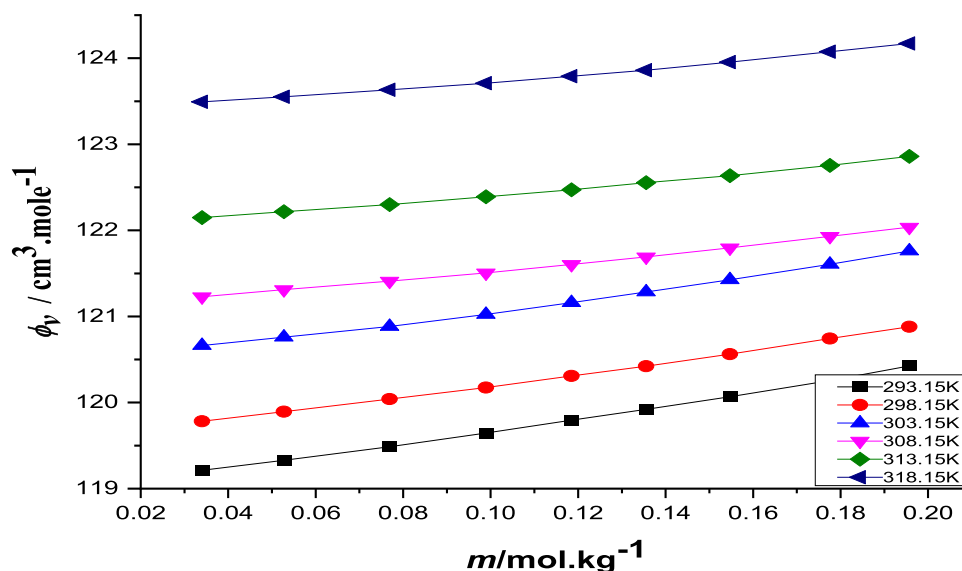


Figure 3. Plot between apparent molar volume (ϕ_V) and molality (m) of galactitol in 0.02 mol kg⁻¹L-arginine at different temperatures.

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.¹⁷

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,

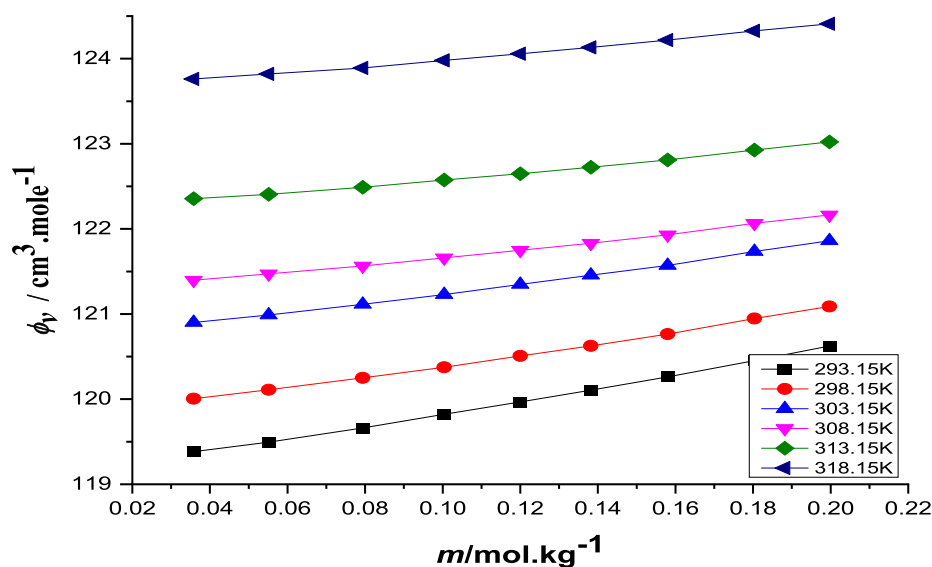


Figure 4. Plot between apparent molar volume (ϕ_v) and molality (m) of galactitol in 0.04 mol kg^{-1} L-arginine at different temperatures.

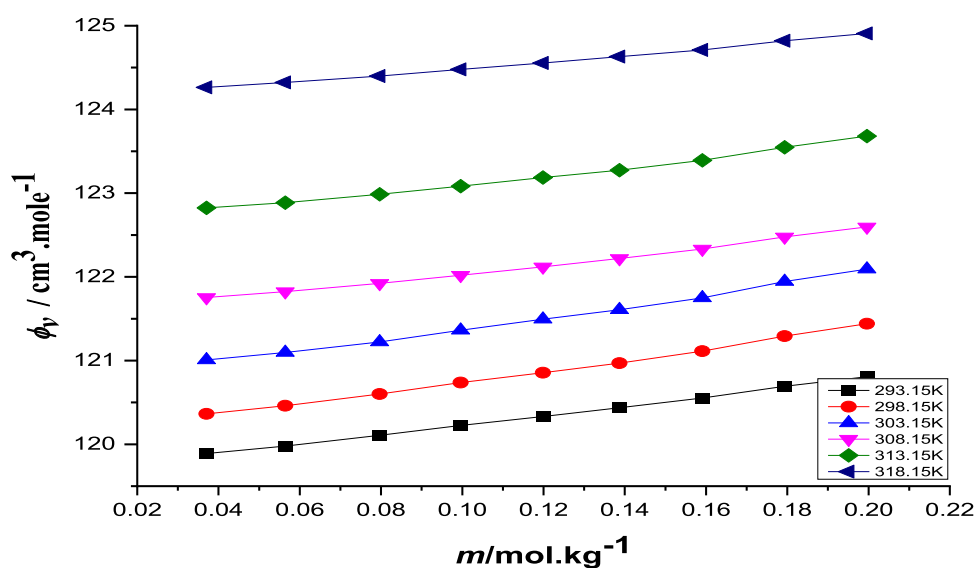


Figure 5. Plot between apparent molar volume (ϕ_v) and molality (m) of galactitol in 0.06 mol kg^{-1} L-arginine at different temperatures.

hence generating the gap among solution molecules that leads to positive change in apparent molar volume.¹⁹ The trends of ϕ_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 ϕ_v values, and values are also increased with the rise in temperature.²⁰

Commonly the following types of molecular interactions in the solutions of galactitol in water and in aqueous L-arginine are supposed: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.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 ϕ_v and partial molar volume ϕ_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.¹⁰

Table 5. Partial Molar Transfer Volume ($\Delta_t\phi_V^0$) of Galactitol in Water and in Aqueous L-Arginine Solutions at Various Molal Concentrations of L-Arginine (m_{Arg}) and Temperatures

$m_{\text{Arg}}/\text{mol kg}^{-1}$	$\Delta_t\phi_V^0/\text{cm}^3 \text{ mol}^{-1}$					
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K
0.02	0.406	0.431	0.583	0.734	0.764	1.541
0.04	0.554	0.557	0.814	0.902	0.97	1.786
0.06	1.095	0.909	0.916	1.253	1.432	2.286

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 ϕ_V^0 can be explained through hydrogen bonding among solutions 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 $\Delta_t\phi_V^0$ explains solute–solvent interactions in binary and ternary solutions. It is calculated using eq 3:

$$\Delta_t\phi_V^0 = \phi_V^0(\text{L-arginine}) - \phi_V^0(\text{water}) \quad (3)$$

The transfer volume $\Delta_t\phi_V^0$ 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 $\Delta_t\phi_V^0$ shows the impact of solute–solute and solute–solvent interactions. 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

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 ϕ_V^0 at different temperatures:^{17,27}

$$\phi_V^0 = a + bT + cT^2 \quad (4)$$

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

The partial molar expansibility ϕ_E^0 is determined as follows:

Table 6. Fitting Coefficients of Equation 4 of Galactitol in Water and Galactitol in Aqueous L-Arginine Solution

$m_{\text{Arg}}/\text{mol kg}^{-1}$	$a/(\text{cm}^3 \text{ mol}^{-1})$	$b/(\text{cm}^3 \text{ mol}^{-1})$	$c/(\text{cm}^3 \text{ mol}^{-1})$
0.00	121.54	−0.1391	0.0004
0.02	334.66	−1.5708	0.0028
0.04	330.03	−1.5412	0.0028
0.06	501.72	−2.6651	0.0046

$$\phi_E^0 = \left(\frac{\partial \phi_V^0}{\partial T} \right)_p = b + 2cT \quad (5)$$

The partial molar expansibility ϕ_E^0 values obtained from eq 5 are listed in Table 7. From Table 7, positive ϕ_E^0 value increases

Table 7. Partial Molar Expansibility (ϕ_E^0) and Temperature Derivatives of Partial Molar Expansibility $[(\partial \phi_E^0 / \partial T)_p]$ for Galactitol in Water and in Aqueous L-Arginine Solutions at Different Temperatures

T/K	$\phi_E^0/\text{cm}^3 \text{ mol}^{-1} \text{ K}^{-1}$	$(\partial \phi_E^0 / \partial T)_p/\text{cm}^3 \text{ mol}^{-1} \text{ K}^{-2}$
Galactitol + Water		
293.15	0.09542	0.0008
298.15	0.09942	
303.15	0.10342	
308.15	0.10742	
313.15	0.11142	
318.15	0.11542	
Galactitol + 0.02 mol kg ^{−1} L-Arginine		
293.15	0.07084	0.0056
298.15	0.09884	
303.15	0.12684	
308.15	0.15484	
313.15	0.18284	
318.15	0.21084	
Galactitol + 0.04 mol kg ^{−1} L-Arginine		
293.15	0.10044	0.0075
298.15	0.12844	
303.15	0.15644	
308.15	0.18444	
313.15	0.21244	
318.15	0.24044	
Galactitol + 0.06 mol kg ^{−1} L-Arginine		
293.15	0.03188	0.0092
298.15	0.07788	
303.15	0.12388	
308.15	0.16988	
313.15	0.21588	
318.15	0.26188	

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.²⁸

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

$$\left(\frac{\partial \phi_E^0}{\partial T} \right)_p = \left(\frac{\partial^2 \phi_V^0}{\partial T^2} \right)_p = 2c \quad (6)$$

Table 8. Apparent Specific Volumes (ASVs) for Galactitol in Water and in Aqueous L-Arginine Solutions at Various Molal Concentrations of Galactitol (*m*) and Temperatures^a

<i>m</i> /mol kg ⁻¹	ASV/cm ³ g ⁻¹					
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K
	Galactitol + Water					
0.03226	0.6523	0.6559	0.6594	0.6618	0.6668	0.6697
0.05033	0.6531	0.6566	0.6601	0.6626	0.6675	0.6704
0.07495	0.6542	0.6576	0.6611	0.6636	0.6685	0.6712
0.09028	0.6549	0.6583	0.6618	0.6644	0.6691	0.6719
0.11480	0.6560	0.6593	0.6628	0.6652	0.6700	0.6729
0.13342	0.6569	0.6602	0.6636	0.6660	0.6708	0.6737
0.15304	0.6579	0.6610	0.6646	0.6669	0.6716	0.6748
0.17528	0.6590	0.6620	0.6656	0.6680	0.6726	0.6761
0.19501	0.6601	0.6630	0.6661	0.6690	0.6737	0.6775
	Galactitol + 0.02 mol kg ⁻¹ L-Arginine					
0.03403	0.6544	0.6575	0.6623	0.6654	0.6705	0.6778
0.05273	0.6550	0.6581	0.6628	0.6659	0.6708	0.6782
0.07689	0.6559	0.6589	0.6635	0.6664	0.6713	0.6786
0.09895	0.6567	0.6596	0.6643	0.6669	0.6718	0.6790
0.11849	0.6576	0.6604	0.6650	0.6675	0.6722	0.6795
0.13555	0.6582	0.6610	0.6657	0.6680	0.6727	0.6799
0.15470	0.6590	0.6618	0.6665	0.6685	0.6731	0.6804
0.17757	0.6601	0.6628	0.6675	0.6693	0.6738	0.6810
0.19572	0.6610	0.6635	0.6683	0.6699	0.6744	0.6816
	Galactitol + 0.04 mol kg ⁻¹ L-Arginine					
0.03403	0.6553	0.6587	0.6636	0.6664	0.6716	0.6793
0.05273	0.6559	0.6593	0.6641	0.6668	0.6719	0.6796
0.07689	0.6568	0.6600	0.6648	0.6673	0.6723	0.6800
0.09895	0.6577	0.6607	0.6654	0.6678	0.6728	0.6805
0.11849	0.6585	0.6615	0.6661	0.6683	0.6732	0.6809
0.13555	0.6593	0.6621	0.6662	0.6687	0.6736	0.6814
0.15470	0.6601	0.6629	0.6673	0.6693	0.6741	0.6818
0.17757	0.6612	0.6639	0.6682	0.6700	0.6747	0.6827
0.19572	0.6621	0.6647	0.6689	0.6706	0.6753	0.6829
	Galactitol + 0.06 mol kg ⁻¹ L-Arginine					
0.03403	0.6581	0.6607	0.6642	0.6683	0.6742	0.6821
0.05273	0.6586	0.6612	0.6647	0.6687	0.6745	0.6824
0.07689	0.6593	0.6620	0.6654	0.6692	0.6752	0.6828
0.09895	0.6599	0.6627	0.6662	0.6698	0.6756	0.6833
0.11849	0.6605	0.6634	0.6669	0.6703	0.6762	0.6837
0.13555	0.6611	0.6640	0.6675	0.6709	0.6767	0.6841
0.15470	0.6617	0.6648	0.6683	0.6715	0.6773	0.6845
0.17757	0.6625	0.6658	0.6694	0.6723	0.6782	0.6851
0.19572	0.6631	0.6666	0.6702	0.6729	0.6789	0.6856

^a*m* is the molality of aqueous galactitol. The standard uncertainties of molality (*m*), ASV, and temperature (*T*) are ± 0.00003 mol kg⁻¹, ± 0.0005 cm³ g⁻¹, and ± 0.02 K, respectively.

The values obtained for $(\partial^2\phi_V^0/\partial T^2)_p$ 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.^{30,31} 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

$(\partial^2\phi_V^0/\partial T^2)_p$ 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.³²

$$ASV = \frac{\phi_V}{M} \quad (7)$$

where ASV, ϕ_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 (0.51–0.71) cm³ g⁻¹, and the results obtained in such a range are considered as sweeteners.^{16,33}

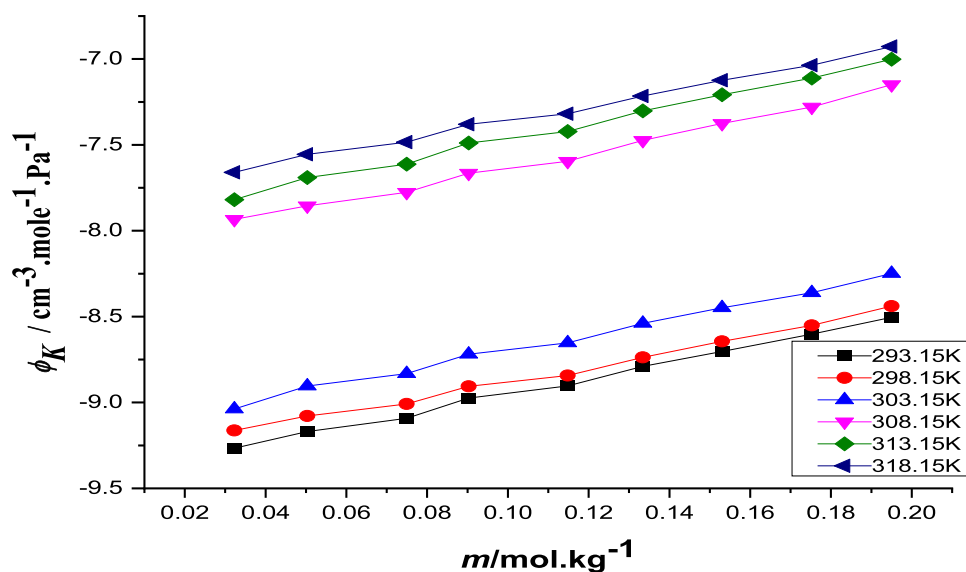


Figure 6. Plot between apparent molar isentropic compressibility (ϕ_k) and molality (m) of galactitol in aqueous solution at different temperatures.

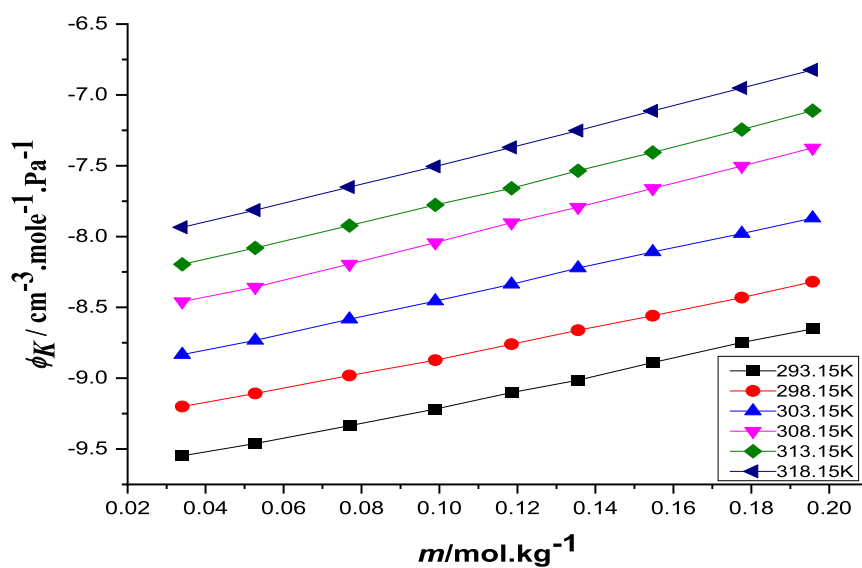


Figure 7. Plot between apparent molar isentropic compressibility (ϕ_k) and molality (m) of galactitol in 0.02 mol kg⁻¹ L-arginine at different temperatures.

On the other hand, aqueous galactitol solution has an ASV range of (0.65 – 0.67) mol g⁻¹ but aqueous L-arginine systems have ranges (0.65 – 0.68) 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 ϕ_k provides understanding of intermolecular collaborations in solutions and is calculated by the following equation:^{36,10}

$$\phi_k = \frac{1000(\rho^0 \beta_s^0 - \rho \beta_s)}{m \rho^0} + \frac{\beta_s M}{\rho} \quad (8)$$

where β_s stands for compressibility of solutions and β_s^0 for water, respectively and β_s is determined by the following equation:

$$\beta_s = \frac{1}{u^2 \rho} \quad (9)$$

Calculated results of ϕ_k for aqueous galactitol and aqueous L-arginine solutions are reported in supplementary Table S2. The ϕ_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 ϕ_k , the apparent molar compressibility at infinite dilution is known as partial molar compressibility ϕ_k^0 and is calculated as³⁹

$$\phi_k^0 = \phi_k + S_k m \quad (10)$$

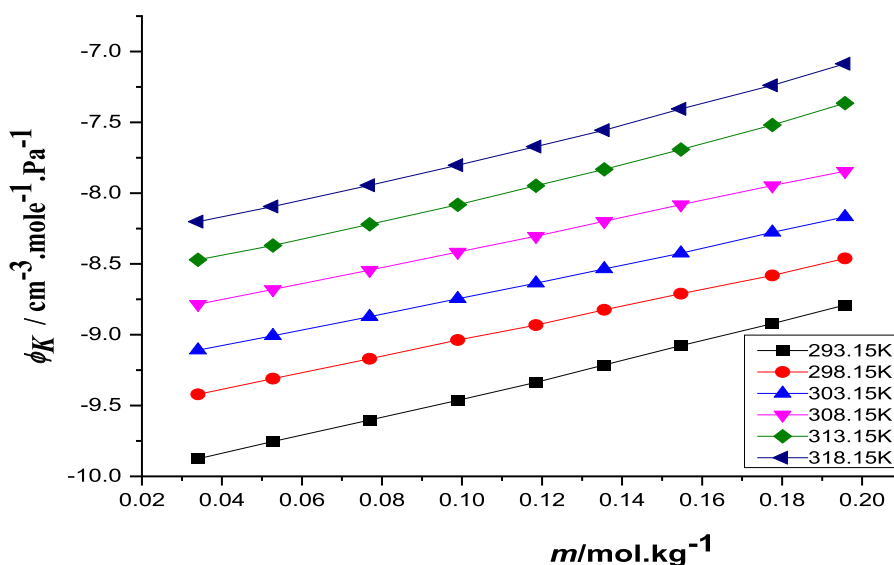


Figure 8. Plot between apparent molar isentropic compressibility (ϕ_K) and molality (m) of galactitol in 0.04 mol kg⁻¹ L-arginine at different temperatures.

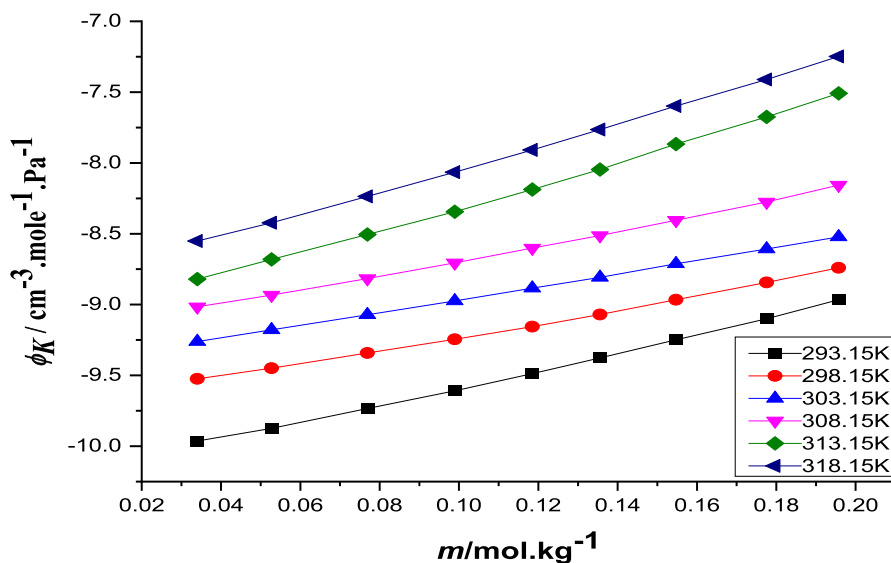


Figure 9. Plot between apparent molar isentropic compressibility (ϕ_K) and molality (m) of galactitol in 0.06 mol kg⁻¹ L-arginine at different temperatures.

Table 9. Partial Molar Compressibilities (ϕ_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_{Arg}) and Temperatures^a

$m_{\text{arg}}/\text{mol kg}^{-1}$	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15 K
	$\phi_K^0/\text{cm}^3 \text{mol}^{-1} \text{Pa}^{-1}$					
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
	$S_K/\text{kg cm}^3 \text{mol}^{-2}$					
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_{Arg} is the molality of aqueous L-arginine. The standard uncertainties of molality (m), partial molar isentropic compressibility (ϕ_K^0) are ± 0.00003 mol kg⁻¹, ± 0.015 cm³ mol⁻¹ Pa⁻¹, and ± 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)^a

$m/\text{mol kg}^{-1}$	$L_f/10^{-7} \text{ m}$					
	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

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

where Φ_k^0 stands for partial molar compressibility, S_k stands for the experimental slope, and m is molality of solution. Calculated data of Φ_k^0 are stated in Table 9. Negative trends of Φ_k^0 describe that molecular interactions became weak in the ternary mixture of in 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.⁴⁰

$$L_f = k\sqrt{\beta} \quad (11)$$

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

make the structure more complex and packed and as a result L_f values are decreased.^{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_f for aqueous sugar alcohol and L-arginine solutions in water show that by rising concentrations of solute, cosolute, and temperature, the L_f values decline, which indicate the strong molecular interactions such as solute–solute and solute–solvent molecules. This decreasing trend of L_f with concentration and temperature makes the structure compact.⁴³ A decreasing trend in L_f values also indicates the close stuffing of molecules and structure-making behavior.¹

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–solvent 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 Φ_V and partial molar volume Φ_V^0 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 $\Delta_t\Phi_V^0$ 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 sweetness improved in the presence of L-arginine and the solvation behavior of aqueous galactitol also enriched in the existence of L-arginine than aqueous sugar alcohol solution. Positive trends of Φ_E^0 indicated the structure-promoting response of galactitol in L-arginine solutions.

■ ASSOCIATED CONTENT

SI 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 (Φ_V), and Table S2 for apparent molar isentropic compressibility (Φ_k) (PDF)

■ AUTHOR INFORMATION

Corresponding Authors

Muhammad Asghar Jamal – Department of Chemistry, Government College University Faisalabad, Faisalabad 38000, Pakistan; orcid.org/0000-0001-5785-1832; Email: asgharjamal@gcuf.edu.pk, m.asgharjamal@gmail.com

Fatimah A. Al-Saeed – Department of Biology, College of Science, King Khalid University, Abha 61413, Saudi Arabia; Research Center for Advanced Materials Science (RCAMS), King Khalid University, Abha 61413, Saudi Arabia; Email: falsaed@kku.edu.sa

Authors

Uzma Shahazidy – Department of Chemistry, Government College University Faisalabad, Faisalabad 38000, Pakistan
Khalid M. Al Syaad – Department of Biology, Faculty of Science, King Khalid University, Abha 61413, Saudi Arabia
Majid Muneer – Department of Chemistry, Government College University Faisalabad, Faisalabad 38000, Pakistan
Ikram Ahmed – Department of Chemistry, University of Sahiwal, Sahiwal 54000, Pakistan
Ahmed Ezzat Ahmed – Department of Biology, Faculty of Science, King Khalid University, Abha 61413, Saudi Arabia; Department of Theriogenology, Faculty of Veterinary Medicine, South Valley University, 83523 Qena, Egypt

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acsomega.2c04102>

Notes

The authors declare no competing financial interest.

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