

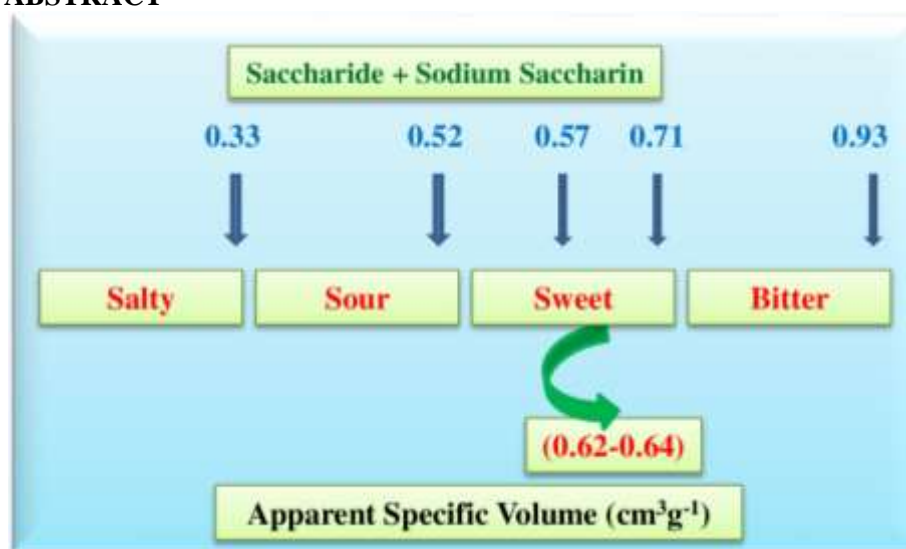
VOLUMETRIC INTERACTIONS BETWEEN MONOSACCHARIDES (D(-)-RIBOSE, D(+)-GALACTOSE) AND SODIUM SACCHARIN AT T = 308.15 K

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ABSTRACT



The interactions between monosaccharides (D(-)-ribose and D(+)-galactose) and sodium saccharin were studied in terms of concentration and temperature using a volumetric approach. The concentrations of monosaccharides and aqueous sodium saccharin are expressed as: (0.04 to 0.20), and (0.05, 0.15, and 0.3) m (mol.kg⁻¹) respectively. The densities were reported at a temperature of 308.15 K. The experimental density (ρ) values were utilized to compute the apparent molar volume (V_{ϕ}), limiting apparent molar volume (V_{ϕ}^0), and Masson's coefficient (S_V). The data was further analysed in terms of transfer volumes at infinite dilution ($\Delta_{\text{trs}}V_{\phi}^0$), volumetric interaction coefficients (V_{ABB} using McMillan–Mayer theory). The result shows positive V_{AB} (doublet interaction) values, indicating the presence of solute-cosolute interactions. The apparent specific volume (ASV) shows that monosaccharides in aqueous sodium saccharin retain their sweetness.

Keywords: Monosaccharide, Apparent specific volume, Partial molar volume, Sodium saccharin.

INTRODUCTION

Recent research has explored into the use of thermodynamic properties in determining the type and strength of intermolecular interactions in ternary systems. Carbohydrates (saccharides) are the primary energy source for humans, playing a significant role in regulating biological processes.¹⁻³ Many researchers have demonstrated the biological relevance of the properties of aqueous solutions containing saccharides due to the hydrophilic hydroxyl group (-OH) responsible for their hydrating capability.⁴⁻⁷ Sugar is crucial in confectionery and medicine, not only for sweetness but also for enhancing flavor, texture, and stability, with wide applications in various fields.⁸⁻¹¹ Recent studies have

focused on investigating the impact of mixing saccharides with metal ions and their effects on interactions between solutes and solvents, utilizing both experimental and theoretical approaches.¹²⁻¹⁶

A range of approved artificial sweeteners¹⁷⁻²² are accessible as substitutes for natural sweeteners in the food and pharmaceutical sectors. Saccharin, a common artificial sweetener, is widely employed as a food additive globally. The blending of sweeteners²³⁻²⁵ has broad applications, including reducing quantity, cost, and flavor. Understanding molecular interactions is valuable in studying how sweeteners interact with both solvents (saccharide-water) and co-solutes (saccharide-saccharin) and their impact on taste.

Continuing earlier research^{26,27} conducted at 298.15 K, this study extends the investigation to a temperature of 308.15 K. The research involves the determination of the density of monosaccharides (ranging from 0.04 to 0.20 mol.kg⁻¹) in aqueous solutions containing sodium saccharin at concentrations of 0.05, 0.15, and 0.3 molal. The study utilizes partial molar volumes, transfer volumes at infinite dilution, interaction coefficients, and apparent specific volumes to determine the interactions between monosaccharides and aqueous solutions of sodium saccharin.

MATERIALS AND METHODS

D(-)-ribose, D(+)-galactose, and sodium saccharin, all with a maximum purity of 99.0% from Sigma, were used.

Aqueous solutions of the solute and cosolute were prepared Using freshly prepared triply distilled water. In both binary and ternary solutions, water was employed as the solvent, monosaccharides were used as the solute, and sodium saccharin (with molalities of 0.05, 0.15, and 0.3) was prepared as a stock solution for future use. Measurements were taken using an analytical Dghona balance with an accuracy uncertainty of 0.0001 g, employing a weight-by-weight (w/w) scale.

A bi-capillary Pycnometer²⁸⁻³² was used to measure the densities (ρ) monosaccharide in water and aqueous sodium saccharin at 308.15 K. A glass-walled water bath with a constant temperature of 0.01 K was used to submerge the pycnometer in a vertical position. The water density at the studied temperature were taken from literature.^{33,34} Using organic solvents, the Pycnometer was calibrated at temperatures of $T = 298.15$ K, and 308.15 K. The results were in good agreement with the reported values when they were compared. The experimental variability in density measurements was 0.204 Kg.m⁻³. The same technique was used to investigate the density of the systems under study.

RESULTS AND DISCUSSION

Apparent molar volume

At $T = 308.15$ K, the apparent molar volumes (V_ϕ) of the studied monosaccharides in water and sodium saccharin were calculated using equation.^{35,36}

$$V_\phi = M/\rho - [1000(\rho - \rho_o)/(m\rho\rho_o)] \quad (1)$$

Where, ρ - density of the solution, m – molality of the solution, and M - molar mass of a solute.

The apparent molar volumes V_ϕ and densities (ρ) of monosaccharides (D(-)-ribose, D(+)-galactose) in water and an aqueous sodium saccharin ($m = 0.05, 0.15, \text{ and } 0.3$) at $T = 308.15$ K are shown in Table 1 and 2. The findings reveal that density and apparent molar volume, varies linearly with solute and cosolute concentrations. Using the least-square fit approach and Masson's equation,³⁷ the V_ϕ values of binary and ternary mixtures are correlated with molality as shown below:

$$V_\phi = V_\phi^0 + S_v \cdot \sqrt{m} \quad (2)$$

The equation (2) is in the form of a straight line, $y = mx + c$, with intercept V_ϕ^0 (partial molar volume at infinite dilution) and slope S_v (Masson's coefficient). As a result of molalities m , the apparent molar volume given by V_ϕ . The above equation for solutes as a nonelectrolyte is given as,^{38,39}

$$V_\phi = V_\phi^0 + S_v \cdot m \quad (3)$$

The results are summarized in Table 3, while the variations of V_{ϕ} of monosaccharide in water and $m = (0.05, 0.15, \text{ and } 0.3)$ in sodium saccharin are represented in Fig. 1, and 2. Table 3 shows the partial molar volume (V_{ϕ}^0) of D(-)-ribose and D(+)-galactose calculated at 308.15 K, from the present study as $(96.161 \text{ and } 110.950) \cdot 10^{-6}$ respectively. Banipal et al.^{39,40} reported $(96.18) \cdot 10^{-6}$. Another group of authors⁴¹ were reported $(96.24) \cdot 10^{-6}$. Similarly, at the same temperature, Chauhan and Kumar⁴² suggest the value $(95.01) \cdot 10^{-6}$. For D(+)-galactose, authors suggest that the V_{ϕ}^0 at 308.15 K is $(110.92, \text{ and } 110.84) \cdot 10^{-6}$, respectively. Banipal et al.⁴³ reported $(110.96) \cdot 10^{-6}$ at 308.15 K. The calculated values show good agreement with those reported in the literature.

The positive V_{ϕ}^0 values offer significant solute-solvent interactions for a molality $m = (0.05, 0.15 \text{ and } 0.3)$ monosaccharide-sodium saccharin system.^{44,45} Additionally, the Masson's coefficient (S_v) is smaller than the value for V_{ϕ}^0 . It indicates that interactions among solute-solute are much weaker than those between solutes and solvents.

Partial Molar Volume of Transfer at infinite dilution ($\Delta_{\text{trs}}V_{\phi}^0$)

The $\Delta_{\text{trs}}V_{\phi}^0$ were obtained from V_{ϕ}^0 of monosaccharides in aqueous sodium saccharin to V_{ϕ}^0 in water by using the following equation.⁴⁰

$$\Delta_{\text{trs}}V_{\phi}^0 = V_{\phi}^0(\text{in aqueous solutions}) - V_{\phi}^0(\text{in water}) \quad (4)$$

The V_{ϕ}^0 values shown in Table 3 can be used to calculate the $\Delta_{\text{trs}}V_{\phi}^0$. It is frequently used to study solute-cosolute interactions in aqueous solutions. The positive values of $\Delta_{\text{trs}}V_{\phi}^0$ were obtained and increased with cosolute concentration, indicating solute-solvent interactions. The $\Delta_{\text{trs}}V_{\phi}^0$ variations are plotted against the molalities, m , of sodium saccharin for the studied temperature in Fig. 3.

The following two categories can be used to classify the potential interactions between solute and cosolute in the present study:

I) Hydrophilic-ionic interactions between sodium saccharin's Na^+ ion and monosaccharide hydrophilic groups ($-\text{C}=\text{O}$, $-\text{OH}$, and $-\text{O}-$).

II) There are hydrophobic-ionic interactions between monosaccharide and sodium saccharin's Na^+ ion hydrophobic groups.

The co-sphere overlap model⁴⁶ states that type (I) interactions positively affect the $\Delta_{\text{trs}}V_{\phi}^0$ while type (II) interactions negatively affect it. Similar findings were reported by Kumar et al.⁴⁷ The obtained positive $\Delta_{\text{trs}}V_{\phi}^0$ values for the present study indicate type-I interaction in the studied systems.

Table 1. Density ($\rho/\text{kg}\cdot\text{m}^{-3}$), and apparent molar volumes ($V_{\phi}/\text{m}^3\cdot\text{mol}^{-1}$) monosaccharides in water at 308.15 K.

D(-)-ribose			D(+)-Galactose		
m (mol.kg ⁻¹)	ρ	V_{ϕ}	m (mol.kg ⁻¹)	ρ	V_{ϕ}
0.0000	994.032		0.0000	994.032	
0.0431	996.36	96.20	0.0430	996.99	111.18
0.0814	998.40	96.25	0.0814	999.60	111.37
0.1208	1000.49	96.31	0.1202	1002.20	111.56
0.1611	1002.60	96.35	0.1610	1004.89	111.79
0.2076	1005.02	96.39	0.2120	1008.19	112.04

Table 2. Density ($\rho/\text{kg.m}^{-3}$), and apparent molar volumes ($V_{\phi}/\text{m}^3.\text{mol}^{-1}$) of monosaccharides in aqueous sodium saccharin (0.05, 0.15, and 0.3) m at 308.15 K and atmospheric pressure.

m (mol.kg^{-1})	ρ 0.05	$V_{\phi}.10^6$	m (mol.kg^{-1})	ρ 0.15	$V_{\phi}.10^6$	m (mol.kg^{-1})	ρ 0.3	$V_{\phi}.10^6$
D(-)-ribose + Sodium saccharin								
0.0370	1000.55	96.62	0.0399	1008.02	96.91	0.0385	1020.18	97.05
0.0798	1002.81	96.70	0.0798	1010.10	97.02	0.0809	1022.36	97.17
0.1199	1004.90	96.79	0.1287	1012.60	97.25	0.1236	1024.53	97.31
0.1598	1006.96	96.88	0.1663	1014.50	97.37	0.1674	1026.71	97.46
0.1998	1009.01	96.93	0.2079	1016.58	97.52	0.2013	1028.38	97.59
D(+)-galactose + Sodium saccharin								
0.0380	1001.16	111.64	0.0435	1009.02	111.84	0.0432	1021.06	112.35
0.0796	1003.96	111.87	0.0818	1011.57	112.02	0.0812	1023.56	112.47
0.1205	1006.67	112.07	0.1215	1014.19	112.17	0.1218	1026.19	112.64
0.1604	1009.28	112.25	0.1604	1016.72	112.30	0.1616	1028.73	112.82
0.2085	1012.37	112.46	0.2098	1019.87	112.54	0.2082	1031.65	113.02

Table 3. Monosaccharide (V_{ϕ}^0), (S_v), and ASV in water and in (0.05, 0.15, and 0.3) m aqueous sodium saccharin at 298.15 K.

Systems	Parameters		
	$V_{\phi}^0.10^{-6}$ ($\text{m}^3.\text{mol}^{-1}$)	$S_v.10^{-6}$ ($\text{m}^3.\text{kg}.\text{mol}^{-2}$)	ASV. 10^{-6} ($\text{m}^3.\text{kg}^{-1}$)
D(-)-ribose + Water	96.161	1.14	0.641
D(+)-galactose + Water	110.950	5.17	0.616
D(-)-ribose + 0.05 m Sodium saccharin	96.548	1.981	0.643
D(-)-ribose + 0.15 m Sodium saccharin	96.752	3.721	0.644
D(-)-ribose + 0.3 m Sodium saccharin	96.908	3.332	0.645
D(+)-galactose + 0.05 m Sodium saccharin	111.477	4.80	0.619
D(+)-galactose + 0.15 m Sodium saccharin	111.671	4.09	0.620
D(+)-galactose + 0.3 m Sodium saccharin	112.150	2.73	0.622

Table 4. Transfer volume at infinite dilution, ($\Delta_{\text{trs}}V_{\phi}^0$) from binary systems (monosaccharide + water) to ternary systems (monosaccharide + water + sodium saccharin) at 308.15 K.

monosaccharides	Molalities (mol.kg^{-1}) of Sodium saccharin		
	0.05	0.15	0.30
	$(\Delta_{\text{trs}}V_{\phi}^0).10^{-6}(\text{m}^3.\text{mol}^{-1})$		
D(-)-ribose	0.3870	0.5902	0.7467
D(+)-galactose	0.526	0.720	1.199

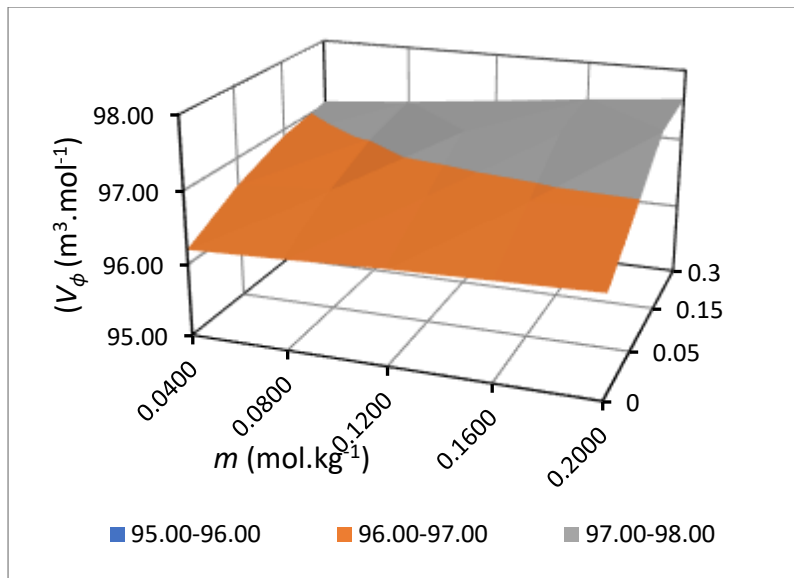


Fig. 1. Variation of V_ϕ with molality, (0.04 to 0.20) m of D(-)-ribose in water and (0.05, 0.15, 0.3) m ($\text{mol} \cdot \text{kg}^{-1}$) sodium saccharin at 308.15 K.

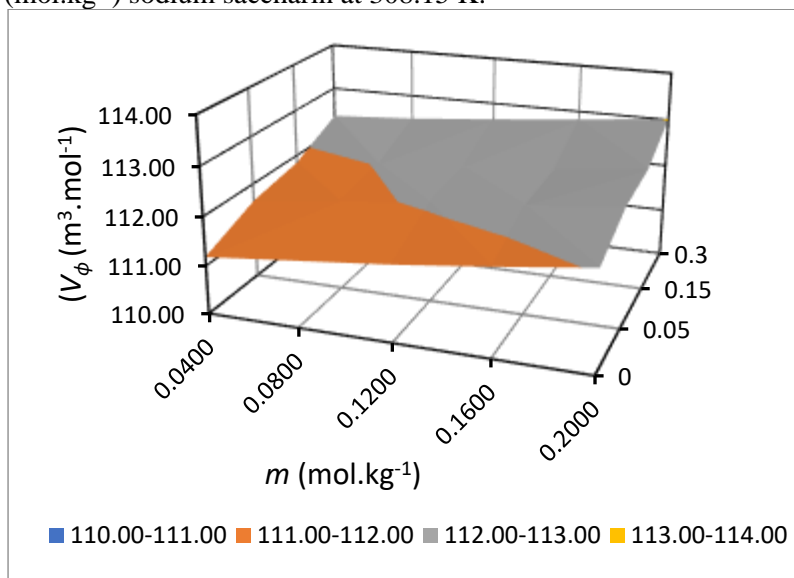


Fig. 2. Variation of V_ϕ with molality, (0.04 to 0.20) m of D(+)-galactose in water and (0.05, 0.15, 0.3) m ($\text{mol} \cdot \text{kg}^{-1}$) sodium saccharin at 308.15 K.

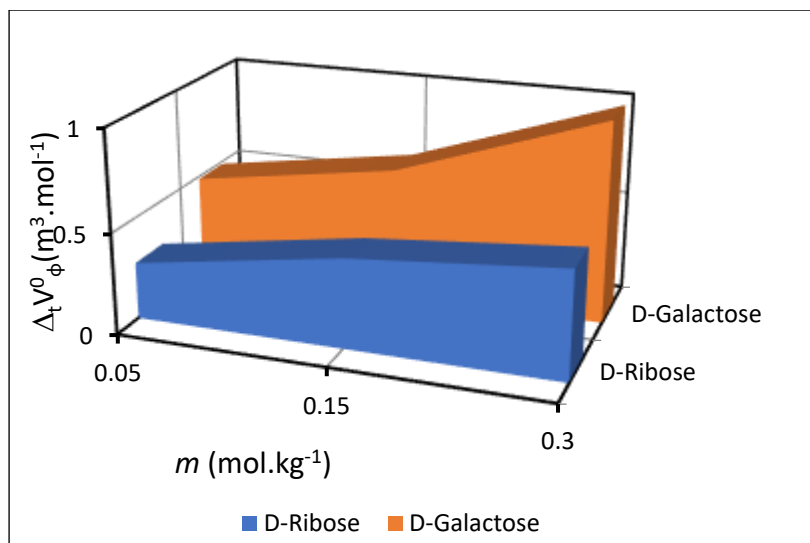


Fig. 3. Transfer volume at infinite dilution, ($\Delta_{\text{trs}}V_{\phi}^0$) of D(-)-ribose and D(+)-galactose with molalities (0.05, 0.15, 0.3) m of sodium saccharin at $T = 308.15$ K.

Volumetric Interaction Coefficients

In order to investigate volumetric interaction coefficients, Kozak et al. developed the "McMillan-Mayer theory"^{48,49} of solutions. Based on this, Krishnan et al.⁵⁰ investigated how solutes and cosolutes interacted in solvation spheres.

The transfer volume at infinite dilution is also related as:

$$\Delta_{\text{trs}}V_{\phi}^0 = 2V_{AB}m_B + 3V_{ABB}m_B^2 + \dots \quad (5)$$

A and B represent solute and co-solute, respectively. These parameters were determined by analyzing experimental data and using the least-squares approach to equation 5. The calculated values at $T = 308.15$ K, of V_{AB} for ribose and galactose are (3.108 and 3.568) $\text{m}^3 \cdot \text{mol}^{-2} \cdot \text{kg}$ respectively, whereas for V_{ABB} are (-4.186 and -3.568) $\text{m}^3 \cdot \text{mol}^3 \cdot \text{kg}^2$. The triplet interaction parameter (V_{ABB}) contributes negative values, whereas the doublet interaction parameter (V_{AB}) contributes positive values. Positive V_{AB} values reveal the strong interactions between monosaccharides (ribose and galactose) and sodium saccharin. Negative values of V_{ABB} on the other hand, indicate the absence of ribose/galactose-saccharin-saccharin interactions. Jiang and coworkers⁵¹ noted a similar observation for ternary systems at 303.15 K.

The "Group Additivity Model" of Savage and Wood⁵² examines the four types of pair interactions among electrolyte (sodium saccharin) and non-electrolyte (monosaccharide). The type III interaction between $\text{Na}^+ - \text{O}$ (hydrophilic groups of saccharide) is more dominant and contributes positively to V_{AB} . The obtained pair interaction coefficient is positive as a result of interactions between Na^+ and the hydrophilic groups of saccharides. According to both theories, interactions between solutes and cosolutes are happening.

Apparent Specific Volume (ASV)

Shamil and Birch⁵³ classified the aqueous solutions as salt, sweet, bitter, and sour with respect to taste. The ASV range suggested by Parke et al⁵⁴, for sweet molecules is from (0.51 to 0.71) $\times 10^{-6} \text{m}^3 \cdot \text{kg}^{-1}$ with an ideal value of (0.618 $\times 10^{-6} \text{m}^3 \cdot \text{kg}^{-1}$) at the center. The equation (6) is used to calculate the ASV of solutes in solvent and cosolute.

$$\text{ASV} = V_{\phi}^0 / M \quad (6)$$

Where M is the molar mass of the solute. Table 3 shows the ASV values of monosaccharide in water and sodium saccharin ($m = 0.05, 0.15, \text{ and } 0.3$) at $T = 308.15$ K. The ASV values for the studied monosaccharide ranged from (0.616 to 0.645) $\times 10^{-6} \text{m}^3 \cdot \text{kg}^{-1}$. The obtained results show that the

monosaccharides used in this study retained their sweetness when blended with sodium saccharin stock solutions.

CONCLUSION

The study includes volumetric data of monosaccharides (D(-)-ribose and D(+)-galactose) in water and aqueous sodium saccharin at $T = 308.15$ K. Thermodynamic parameters, including V_ϕ , V_ϕ^0 , $\Delta_{\text{trs}}V_\phi^0$, ASV, V_{AB} and $V_{\text{ABB}}V_{\text{ABB}}$ were calculated using the experimental data. Monosaccharide is positively transferred from water to aqueous sodium saccharin, and the transferred value increases with concentration. The observed positive values of V_{AB} suggest strong solute-cosolute interactions between them. The monosaccharides used in this study retained their sweetness $(0.616\text{--}0.645)10^{-6}.\text{m}^3.\text{kg}^{-1}$ when blended with sodium saccharin stock solutions.

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