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VOLUMETRIC STUDY OF SACCHARIDE INTERACTIONS (D-ARABINOSE, D-XYLOSE, AND D-GALACTOSE) IN SODIUM SACCHARIN AT 298.15 K

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ABSTRACT

Volumetric investigations are effective for attributing the interactions between saccharides and sodium saccharin in aqueous medium. The densities with the help of bicapillary pycnometer were measured for monosaccharide (D(-)-arabinose, D(+)galactose and D(+)-Xylose) (0.04 to 0.20) *m* (mol.kg^{-1}) in water and aqueous sodium saccharin solution with molality, *m* = (0.05, 0.15 and 0.3) at 298.15 K. Experimental data were used to calculate the partial molar volume V_ϕ^0 and apparent molar volume V_ϕ. The corresponding transfer volumes $\Delta_{trs}V_\phi^0$ were calculated and showed a positive and increasing trend with concentration for saccharides from water to aqueous medium of sodium saccharin. With the help of McMillan-Mayer theory, the interaction coefficients (V_{AB}) and (V_{ABB}) were calculated. In the presence of sodium saccharin, significant interactions between a hydrophilic group of saccharides and the sodium ion have been reported. Using apparent specific volume values, a sweet taste range was identified for saccharides in the presence of sodium saccharin.

Keywords: Density, Apparent and Partial molar volume, Saccharides, Sodium saccharin.

1. INTRODUCTION

Saccharides are sugar-containing organic compounds. Saccharide works as an energy source for the human body, supplying energy to functioning muscles and the central nervous system [1]. Saccharides play an essential part in the metabolism of living organisms in biological and physiological cycles [2, 3]. Many researches on the characteristics of aqueous saccharide solutions have been conducted, revealing that these are biochemically important non-electrolytes. Saccharides have a hydrophilic hydroxyl group (-OH), which is responsible for their hydration properties [4, 5]. The thermodynamic research on saccharides in aqueous solutions have enormous implications in a variety of fields, including science, medicine, and catalysis. The interactions between metal ions and blended electrolyte saccharides have been extensively studied both experimentally and theoretically [6-16]. For solute-solvent and solute-solute interactions, the research of metal ions in aqueous medium containing saccharides is important.

In India, FSSAI (Food Safety and Standards Authority of India), a regulatory body, recognized a variety of artificial sweeteners [17-21] as food additives with widespread commercial use in the food and pharmaceutical industries. Instead of fatty sugars, the food industry uses low-calorie artificial sweeteners such as Aspartame, Acesulfame K, Sodium saccharin, and Sucralose [22, 23]. Blending sweeteners [24-26], is a standard procedure in the food and pharmaceutical industries. The principal reasons for combining sugars with artificial sweeteners are to reduce utilization, improve taste, and save expenses. Sugars form molecular interactions with receptors via water molecules that surround them. As a result, a study of saccharide-water and saccharide-cosolute interpretation is important.

Here, within a concentration range of (0.04 to 0.20) *m*, the densities of monosaccharides (D-galactose, D-arabinose and D-xylose) in water as solvent and (0.05, 0.15, and 0.3) *m* artificial sweetener sodium saccharin (cosolute) were determined experimentally at 298.15 K. Structural interactions (fig.1) in aqueous medium have been studied using partial molar volumes, transfer volume, interaction coefficients, apparent specific volume and apparent molar volumes.

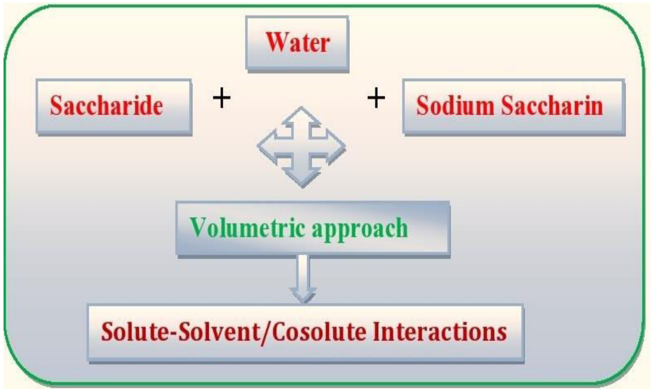

Fig. 1: Structural Interactions

2. MATERIAL AND METHODS

2.1. Chemicals

The sugars D(+)-Xylose, D(+)-Galactose and D(-)-Arabinose as well as artificial sweetener sodium saccharin, were purchased from Sigma with a maximum purity of 99.0 % and used directly as received from suppliers.

To prepare the solution, we used freshly prepared triply distilled water and an airtight stoppered glass bottle. Using an analytical Dhona balance (uncertainty in accuracy $\pm 1\times10^{-4}$ g), all solutions were prepared on a weight-by-weight basis. Water was used as a solvent for a binary sugar system, whereas stock solutions of sodium saccharin (0.05, 0.15, and 0.3) *m* were used for ternary systems. In binary and ternary mixtures, saccharide has been used as the solute.

2.2. Methods

At the temperature studied, a bi-capillary Pycnometer [27, 28] was used to measure the densities of both binary and ternary systems. The pycnometer was immersed vertically in a glass-walled water bath, and the temperature was kept constant at ±0.01 K by using a dimmer. Pure organic liquids such as acetophenone, carbon tetrachloride, and ethyl acetate were used to calibrate the Pycnometer at 298.15
K. The findings were compared to the reported values, and they were found to be in good agreement. Water density at 298.15 K was obtained from the literature [29]. Using the same procedures, the density of aqueous sugar solutions in water and sodium saccharin was determined. The experimental uncertainties in the density measurements were 0.000204g.cm^{-3}.

3. RESULTS AND DISCUSSION

3.1. Apparent molar volume

At 298.15K, the apparent molar volumes of D(+)-Xylose, D(-)-Arabinose, and D(+)-Galactose in solvent and co-solute were calculated using the equation below. [5,30]

$$V_\phi = M/\rho - (\rho - \rho_o)/m\rho\rho_o) \qquad 1$$

Where ρ denotes the solution's density and *m* denotes the molality of the solution. Furthermore, M stands for the solute's molar mass, and V_ϕ stands for the apparent molar volume.

The densities (ρ) and apparent molar volumes (V_ϕ)of D(+)-Galactose, D(+)-Xylose, and D(-)-Arabinose, in water at 298.15 K are shown in Table 1. Additionally, Additionally, table 2 shows the ρ and V_ϕ of saccharides in (0.05, 0.15, and 0.3) *m* aqueous sodium saccharin. The measured density and apparent molar volume are shown to be concentration dependent and to vary linearly with solute and cosolute concentrations.

When a solute is added to a solvent to form a solution, there may be a volume change due to solute-solvent interaction. The V_ϕ, data of binary (saccharide + water) and ternary (saccharide + water + sodium saccharin) systems are correlated with the molality using the least square fit approach and Masson's equation [31,32].

Table 1: Densities (ρ/kg.m^{-3}), and apparent molar volumes (V_ϕ.10^6/m^3.mol^{-1}) of monosaccharides in water at m(mol.kg^{-1})

D(-)-Arabinose			D(+)-Xylose			D(+)-Galactose		
M (mol.kg^{-1})	ρ	V_ϕ	*M* (mol.kg^{-1})	ρ	V_ϕ	*M* (mol.kg^{-1})	ρ	V_ϕ
0.0000	997.047		0.0000	997.047		0.0000	997.047	
0.0432	999.49	93.52	0.0431	999.39	95.65	0.0430	1000.03	110.52
0.0814	1001.62	93.62	0.0810	1001.43	95.67	0.0814	1002.66	110.69
0.1213	1003.82	93.74	0.1215	1003.57	95.71	0.1202	1005.28	110.87
0.1612	1006.00	93.87	0.1619	1005.68	95.73	0.1610	1008.00	111.05
0.2107	1008.67	94.00	0.2136	1008.35	95.76	0.2120	1011.34	111.28

Table 2: Densities (ρ/kg.m^{-3}) and apparent molar volumes ($V_\phi.10^6$/m^3.mol^{-1}) of monosaccharides in aqueous sodium saccharin (0.05, 0.15, and 0.3) *m* at 298.15 K and atmospheric pressure

m(mol.kg^{-1})	ρ	$V_\phi.10^6$	*m*(mol.kg^{-1})	ρ	$V_\phi.10^6$	*m*(mol.kg^{-1})	ρ	$V_\phi.10^6$
	0.05			*0.15*			*0.3*	
D(-)-Arabinose + Sodium saccharin								
0.0000	1001.68		0.0000	1007.64		0.0000	1021.51	
0.0407	1003.92	93.82	0.0395	1009.81	93.95	0.0412	1023.75	94.07
0.0807	1006.13	94.06	0.0776	1011.90	94.17	0.0800	1025.85	94.25
0.1201	1008.26	94.29	0.1161	1013.97	94.41	0.1199	1027.98	94.47
0.1595	1010.37	94.49	0.1565	1016.12	94.65	0.1627	1030.22	94.67
0.1996	1012.48	94.71	0.1994	1018.36	94.87	0.2098	1032.65	94.92
D(+)-Xylose+ Sodium saccharin								
0.0000	1001.68		0.0000	1008.24		0.0000	1021.51	
0.0391	1003.75	96.02	0.0405	1010.37	96.54	0.0436	1023.76	96.60
0.0794	1005.88	96.10	0.0800	1012.43	96.68	0.0817	1025.73	96.76
0.1199	1008.03	96.18	0.1199	1014.50	96.81	0.1233	1027.84	96.91
0.1599	1010.11	96.25	0.1598	1016.53	96.95	0.1642	1029.89	97.08
0.1998	1012.17	96.33	0.1998	1018.54	97.08	0.2098	1032.14	97.26
D(+)-Galactose + Sodium saccharin								
0.0000	1001.68		0.0000	1009.28		0.0000	1021.79	
0.0380	1004.30	110.89	0.0435	1012.26	111.02	0.0432	1024.71	111.30
0.0796	1007.13	111.06	0.0818	1014.85	111.14	0.0812	1027.25	111.44
0.1205	1009.87	111.17	0.1215	1017.49	111.28	0.1218	1029.91	111.55
0.1604	1012.51	111.35	0.1604	1020.04	111.45	0.1616	1032.50	111.67
0.2085	1015.64	111.56	0.2098	1023.24	111.63	0.2082	1035.49	111.78

$$V_\phi = V_\phi^0 + S_v.m \qquad 2$$

Where V_ϕ^0 and S_v represent experimental intercept and slope values of the apparent molar volume, V_ϕ as a result of molalities *m*, The intercept of equation 2 is the partial molar volume, V_ϕ^0 and the slope describing the solute-solute interaction, S_v.

Table 3 summarizes these results at 298.15 K, whereas fig. 2, 3, and 4 show the variation of V_ϕ of saccharides in water and in (0.05, 0.15, and 0.3) *m* sodium saccharin, respectively. The literature figures of $V_\phi.10^6$ (m^3.mol^{-1}) for arabinose at the studied temperature are 93.2 [4], 93.23 [33], 93.43 [34], 91.9 [35], 93.3 [36], 93.7 [37], 94.0 [38], and 93.43 [39], but the observed value is 93.39. V_ϕ^0 values for xylose and galactose observed experimentally at 298.15 K are 95.62 and 110.33, respectively. Similarly, the reported values of xylose in water at same temperature are 95.4 [4], 95.82 [33], 95.68 [34], 94.8 [35], 95.88 [36], 95.4 [37], 95.60 [38], and 95.68 [39].

Galactose has V_ϕ^0values are 110.2 [4], 110.65 [33], 110.29 [34, 39], 111.9 [35], 110.65 [36], 110.5 [37], 110.64 [38], and 110.24 [40] at 298.15 K reported by author. The values in the literature for the studied systems in water agree well with the experimental data. While no data for comparison of V_ϕ^0values in the presence of sodium saccharin were available. Table 3 summarizes the results of $V_\phi^0$of arabinose, xylose, and galactose in (0.05, 0.15, and 0.3) *m* sodium saccharin. D-arabinose and D-xylose have lower, V_ϕ^0 values, but D-galactose has a higher V_ϕ^0value than both monosaccharides. The V_ϕ^0values provide important information regarding the solute-solvent interactions [41].

Strong solute-solvent interactions were indicated by positive V_ϕ^0values for a saccharide-sodium saccharin-water system with molality [42]. Moreover, the solute-solute interaction variable S_v is positive but smaller than V_ϕ^0 suggesting that in all of the systems examined solute-solute interactions are smaller than solute-solvent interactions.

3.2. Transfer volume

The transfer volume of ($\Delta_{trs} V_\phi^0$) of saccharides from water to aqueous sodium saccharin was determined for the analysed systems at infinite dilution using the following relation [43].

$$\Delta_{trs}V_\phi^0 = V_\phi^0(\text{in aqueous solutions}) - V_\phi^0\ (\text{in water}) \qquad 6$$

The V_ϕ^0 values of saccharides in aqueous sodium saccharin and water, which can be used to calculate transfer volumes is seen in table 3. This method is often used to examine solute-cosolute interactions in aqueous medium. The transfer volume is positive at infinite dilution and increases with concentration [35]. The $\Delta_{trs}V_\phi^0$ values of saccharides ranging from water to aqueous sodium saccharin with molality, *m* are shown in table 4.

Table 3: At 298.15 K, monosaccharide (V_ϕ^0), (S_v) and ASV were measured in water and in (0.05, 0.15, and 0.3) *m* aqueous sodium saccharin

System	Parameters		
	$V_\phi^0.10^{-6}$ ($m^3.mol^{-1}$)	$S_v.10^{-6}$ ($m^3.kg.mol^{-2}$)	ASV.10^{-6} ($m^3.kg^{-1}$)
D(-)-Arabinose + Water	93.390	2.92	0.622
D(+)-Xylose + Water	95.623	0.66	0.637
D(+)-Galactose + Water	110.327	4.50	0.612
D(-)-Arabinose + 0.05 m Sodium saccharin	93.606	5.56	0.623
D(-)-Arabinose + 0.15 m Sodium saccharin	93.721	5.86	0.624
D(-)-Arabinose + 0.3 m Sodium saccharin	93.860	5.03	0.625
D(+)-Xylose + 0.05 m Sodium saccharin	95.944	1.95	0.639
D(+)-Xylose + 0.15 m Sodium saccharin	96.408	3.37	0.642
D(+)-Xylose + 0.3 m Sodium saccharin	96.423	4.01	0.642
D(+)-Galactose + 0.05 m Sodium saccharin	110.737	3.87	0.615
D(+)-Galactose + 0.15 m Sodium saccharin	110.841	3.76	0.615
D(+)-Galactose + 0.3 m Sodium saccharin	111.193	2.87	0.617

Table 4: Transfer volume, ($\Delta_{trs}V_\phi^0$) of monosaccharides from aqueous binary systems (saccharide + water) to ternary systems (saccharide + water + sodium saccharin) at 298.15 K

monosaccharides	Molalities ($mol.kg^{-1}$) of Sodium saccharin		
	0.05	0.15	0.30
	$\Delta_{trs}V_\phi^0.10^6$($m^3.mol^{-1}$)		
D(-)-Arabinose	0.217	0.331	0.470
D(+)-Xylose	0.321	0.464	0.800
D(+)-Galactose	0.410	0.514	0.866

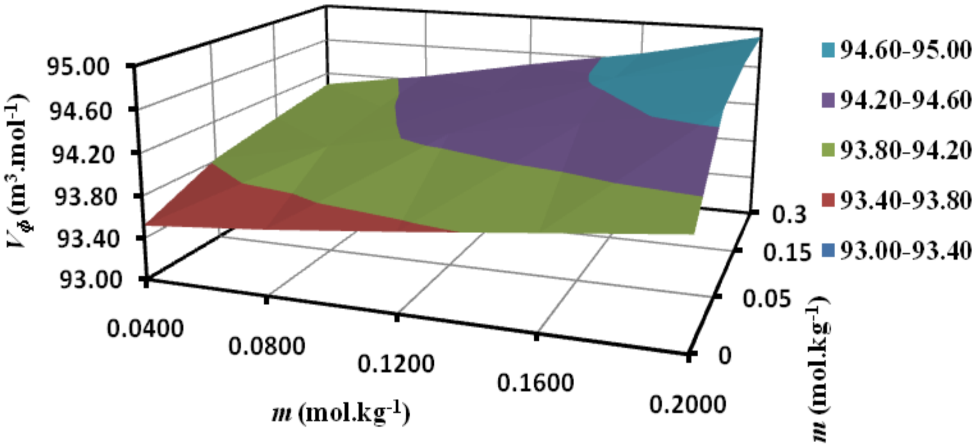

Fig. 2: Variation of V_ϕ of D(-)-Arabinose in water and in (0.05, 0.15, 0.3) sodium saccharin with molality, *m* ($mol.kg^{-1}$) at 298.15 K

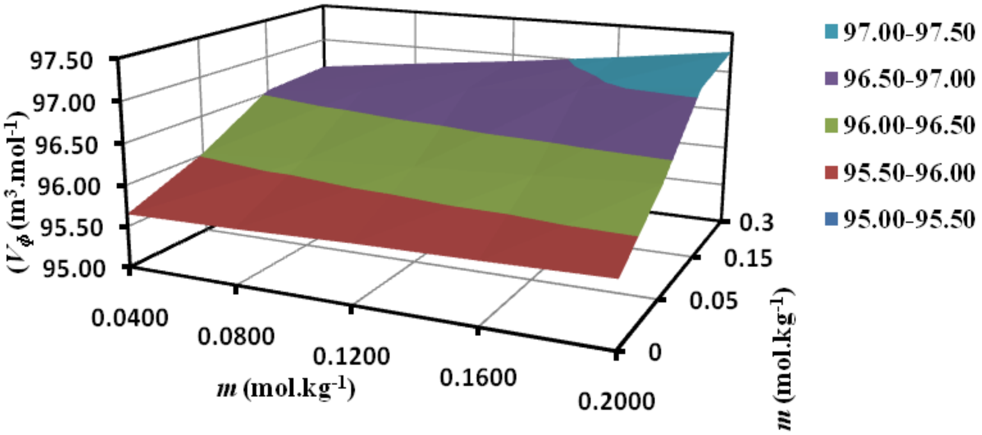

Fig. 3: Variation of V_ϕ of D(-)-Xylose in water and in (0.05, 0.15, 0.3) sodium saccharin with molality, m (mol.kg^{-1}) at 298.15 K

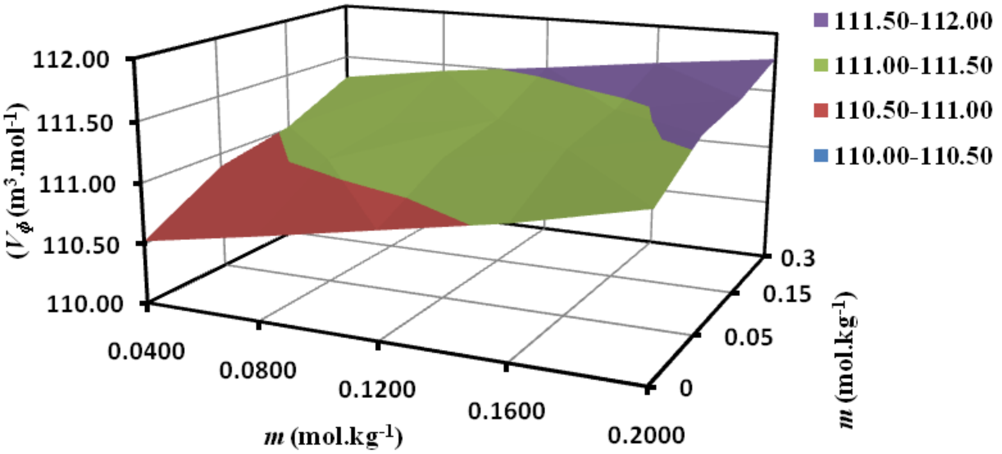

Fig. 4: Variation of V_ϕ of D(+)-Galactose in water and in (0.05, 0.15, 0.3) sodium saccharin with molality, m (mol.kg^{-1}) at 298.15 K

Fig. 5 shows the variation of transfer volume at infinite dilution ($\Delta_{trs}V_\phi^0$) is plotted against the molalities, m of sodium saccharin. In aqueous medium of sodium saccharin, the $\Delta_{trs}V_\phi^0$ values for all examined saccharide systems are positive, and their values increase with increasing sodium saccharin concentration. The findings for saccharides are arranged as follows:

$(\Delta_{trs}V_\phi^0)_{\text{D-arabinose}} < (\Delta_{trs}V_\phi^0)_{\text{D-xylose}} < (\Delta_{trs}V_\phi^0)_{\text{D-galactose}}$

The observed pattern shows that sodium saccharin concentration, stereochemical features of saccharides, and temperature influence solute-cosolute interactions. The increase in transfer volume ($\Delta_{trs}V_\phi^0$) is due to increased molecular complexity from arabinose to xylose to galactose. It shows that molar mass increases molecular complexity. Dhondge et al. [44] found similar outcomes. The two possible types of interactions in ternary systems are:

I. Hydrophilic-ionic interactions between saccharide hydrophilic groups (-C=O,- OH and -O-) and the Na$^+$ ion of sodium saccharin.
II. Hydrophobic-ionic interactions between saccharide and cosolute ion hydrophobic groups.

According to the "co-sphere overlap model" [45], positive contribution to $\Delta_{trs}V_\phi^0$ values with type I, whereas type II interactions contribute negative values. Positive values of $\Delta_{trs}V_\phi^0$ indicate that type I interactions dominate over type II interactions for D-arabinose, D-galactose and D-xylose. Similarly, increase in $\Delta_{trs}V_\phi^0$ values for all saccharides examined indicates that type I interactions are reinforced across the whole concentration range (0.05, 0.15, and 0.3) *m*. With increasing molecular complexity, the overall trend in values of V_ϕ^0 and corresponding $\Delta_{trs}V_\phi^0$ values of saccharides increase. The same tendency was observed in KCl and NaCl aqueous solutions reported by Banipal et al. [46, 47]. The hydrophilic-ionic interactions between saccharide and sodium saccharin molecules add positive values to the transfer volume.

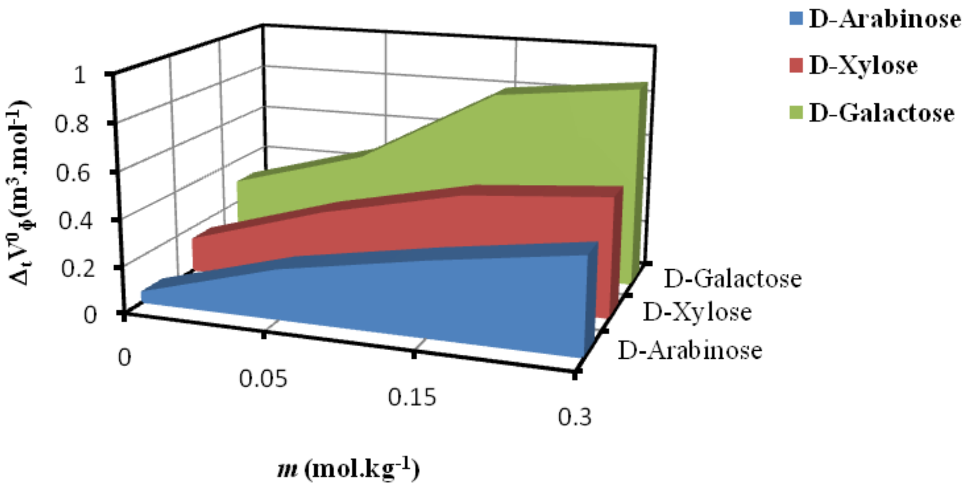

Fig. 5: Variation of Partial molar volume of transfer, ($\Delta_{trs}V_\phi^0$) with molalities (0.05, 0.15, 0.3) *m* of sodium saccharin at 298.15 K

To illustrate the solute-cosolute interactions, Ferrell et al. [35] used equation 7, in which the V_ϕ^0 of a non-electrolyte at infinite dilution is related as,

$$V_\phi^0 = V_{\text{v.w}} + V_{\text{void}} - V_{\text{shrinkage}} \qquad 7$$

The empty void is denoted by V_{void}, while Van der Waal's volume is denoted by $V_{\text{v.w}}$. $V_{\text{shrinkage}}$ is the volume shrinking produced by hydrogen bonding group interactions with water. When $V_{\text{v.w}}$ and V_{void} have the same value in both water and sodium saccharin, positive transfer values for saccharides are obtained. This could be due to volume contraction mediated by interactions between sodium saccharin and the saccharides hydrophilic -OH groups. Saccharin's interactions with saccharide inhibit saccharin's further structure-breaking effect on water. Zhuo et al.[48], and Kharat [49] also reported solute-cosolute interactions using the above equation.

3.3. Apparent Specific Volume

Apparent Specific Volume (ASV) is a taste quality metric that would be used to classify aqueous solutions as salt, sweet, salty, or tart [50]. The ASV of sweet molecules is between 0.51 and 0.71 X 10^{-6} m^3.kg^{-1}, with the optimal value [51] being in the middle of the range (0.618 X 10^{-6} m^3.kg^{-1}). The ASV of sugars in water and sodium saccharin can be calculated using the partial molar volume V_ϕ^0, and the solute's molar mass, M.

$$\text{ASV} = V_\phi^0/M \qquad 8$$

Table 3 contains the ASV values for monosaccharide in water at 298.15 K. At the examined temperature, the ASV values for arabinose, xylose, and galactose ranged from (0.612 to 0.642) X 10^{-6} m^3.kg^{-1}. All saccharides examined in this research kept their sweetness when dissolved in sodium saccharin solutions.

3.4. Interaction Coefficients

To examine volumetric interaction coefficients, Kozak et al. suggested the "McMillan-Mayer theory" [52, 53] of solutions. Friedmann, Krishnan, and Franks et al. [54] examined solute-cosolute interactions in solvation spheres based on this. Numerous researchers extensively apply the theory to the study of interactions in aqueous solutions. (55-57)

The transfer volume is also related as:

$$\Delta_{trs}V_\phi^0 = 2V_{AB}m_B + 3V_{ABB}m_B^2 + ---- \qquad 9$$

A and B represent saccharides (solute) and sodium saccharin (co-solute), respectively. These parameters were determined by analyzing experimental data and using the least-squares approach to equation 9. At 298.15 K, the calculated values of V_{AB} for arabinose, xylose, and galactose are (1.674, 2.209, and 2.633) $10^6(m^3.mol^{-2}.kg)$ respectively, while those for are (-2.005, -1.993, and -2.713).10^6 $(m^3.mol^3.kg^2)$. Positive values are contributed by the doublet interaction parameter (V_{AB}), whereas negative values are contributed by the triplet interaction parameter (V_{ABB}). Positive V_{AB} values show that saccharides and sodium saccharin have strong interactions. Negative values of V_{ABB} on the other hand, indicate the absence of saccharide-saccharin-saccharin interactions. At 298.15 K Jiang and coworkers [58] obtained positive$V_{AB}$and negativeV_{ABB}values for (CsCl-fructose-water) and (CsCl-glucose-water) systems.

In order to explore the interactions between sodium saccharin (electrolyte) and saccharide (non-electrolyte) the "Group Additivity Model" [59] developed four basic types of pair interactions.

a) Na^+ - R (-R is an alkyl group) - It makes a minor negative contribution to V_{AB}.

b) Anion - R, it also contributes negatively to V_{AB} pair interaction.

c) Na^+- O (-O indicates hydrophilic groups in saccharide). it is more dominant and contributes positively to V_{AB} value.

d) Because both groups have a negative charge, the repulsive effect is stronger in anion - O, and this leads to the negative value of V_{AB} but less than (a).

In aqueous solutions, the electrolyte sodium saccharin dissociates completely into ions. The pair interaction coefficient is positive due to the interactions of Na^+ with hydrophilic groups of saccharides (-OH, C=O, and -O-). Both theories proposed that interactions between solutes (saccharides) and cosolutes (sodium saccharin) are taking place.

4. CONCLUSION

The apparent molar volume and partial molar volume of monosaccharides were calculated from experimental densities at 298.15 K in water and in (0.05, 0.15, and 0.3) *m* sodium saccharin. The apparent specific volume (ASV), standard partial molar volumes of transfer $(\Delta_{trs} V_{\phi}^0)$and interaction coefficients (V_{AB}, and V_{ABB}) have been determined. The transfer volume of arabinose, xylose, and galactose transferred from water to aqueous sodium saccharin is positive, and the magnitude increase as concentration increased. Because the ASV varies from (0.612 to 0.642) $10^{-6}.m^3.kg^{-1}$, all of the systems evaluated had a sweet flavor. Positive V_{AB} values suggested that the saccharide (solute) and sodium saccharin cation had strong interactions (co-solute).

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Conflicts of Interest

The authors declare no conflict of interest.

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