

Full Paper

Volumetric Behavior of Sodium Saccharin in Water and (0.1, 0.3, and 0.5) *m* Fructose at (298.15, 303.15, 308.15, and 313.15) K

| Vol 9 | | No. 1 | | January-March 2017 |

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Article history: Received: 16 October 2016; revised: 14 January 2017; accepted: 27 February 2017. Available online: 30 March 2017. DOI: <u>http://dx.doi.org/10.17807/orbital.v9i1.921</u>

Abstract: In order to get the information regarding the sweetener-water and sweetener-sweetener interactions, densities of sodium saccharin in water and (0.1, 0.3, and 0.5) m fructose have been measured at (298.15, 303.15, 308.15, and 313.15) K by the use of bicapillary pycnometer. From density values, partial molar volumes, expansion coefficient, Hepler's constant, apparent specific volumes, partial molar volumes of transfer, doublet and triplet interaction coefficients have been calculated. From density study, it has been concluded that strong water-sodium saccharin interactions exist. Sodium saccharin is water structure maker. Strong interactions exist between sodium saccharin and fructose. In presence of fructose, the interactions exist between hydrophilic group (–OH, C=O, and –O–) of fructose and sodium ion of sodium saccharin in aqueous solutions of sodium saccharin. All investigated solutions exhibit sweet taste.

Keywords: density; fructose; sodium saccharin; sweetener

1. INTRODUCTION

The intense sweetener sodium saccharin is widely used in foods, beverages, and pharmaceuticals [1-4]. Furthermore, sugar solutions have great importance in bio-systems. Water is very important in sweet taste because no molecule can be tasted unless it is soluble and transportable to the receptors via oral fluid. Sweeteners establish their molecular interactions with receptor through the water molecule, which surround them. Therefore, understanding of the nature of sweetener-water (solute-solvent) and solute-solute interactions is important. Temperature and concentration dependence of density and ultrasonic velocity of aqueous solutions has been proved as one of the most appropriate methods for the study of solutesolvent and solute-solute interactions. Furthermore, thermodynamic properties have great importance in the study of taste behavior of sweeteners in mixed aqueous solutions. These properties of aqueous solutions of sweeteners are required for biological, pharmaceutical, and food processing studies. The objectives of the research work carried out are:

1.To generate data of thermodynamic properties of sodium saccharin solutions in presence or absence

of fructose.

2.To get the information regarding type of interactions in aqueous solutions of sodium saccharin in presence or absence of fructose.

3.To get the information regarding taste qualities of sodium saccharin solutions in presence of fructose.

This paper reports density study of sodium saccharin solutions in water and in (0.1, 0.3, and 0.5) m fructose at (298.15, 303.15, 308.15, and 313.15) K and at atmospheric pressure.

2. MATERIAL AND METHODS

Na-saccharin (Merck, purity ≥ 99.0 %) and fructose (Merck, purity ≥ 99.0 %) were used without further purification for this study. Aqueous solutions of sweeteners were prepared by using triply distilled water by weight by weight method in airtight stoppered glass bottle. Masses were recorded on Dhona balance accurate to ± 0.1 mg. Densities of solutions were measured by using 15 cc bi-capillary pycnometer [5-8]. Pycnometer was calibrated with triply distilled

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deionized water. Density measurements were undertaken in glass-walled bath. Uncertainty in density and temperature measurements were $5.8 \times 10^{-2} \text{ kg} \cdot \text{m}^{-3}$ and 0.006 K, respectively.

3. RESULTS AND DISCUSSION

In present investigation, aqueous solutions of sodium saccharin in presence of fructose have been studied at different temperatures. Density data of sodium saccharin in presence of (0.1, 0.3, and 0.5) *m* fructose have been measured at (298.15, 303.15, 308.15, and 313.15) K and at atmospheric pressure.

Table 1 displays the densities of aqueous solutions of sodium saccharin in absence and presence of (0.1, 0.3, and 0.5) *m* fructose at (298.15, 303.15, 308.15, and 313.15) K. It is confirmed that density of aqueous solutions of sodium saccharin varies linearly with molality of the solutions. As usual, density decreases with increase in the temperature of the solutions. Similar behavior of density of aqueous solutions of sodium saccharin in presence of (0.1, 0.3, and 0.5) *m* fructose has been observed. Moreover, it is observed that density of aqueous solutions of sodium saccharin in presence of (0.1, 0.3, and 0.5) *m* fructose increases with increase in the concentration of fructose.

Table 1. Densities ($\rho / \text{kg·m}^{-3}$) of sodium saccharin in water and (0.1, 0.3, and 0.5) *m* fructose at different temperatures.

т	ρ									
(mol·kg ⁻¹)		(kg·m ⁻³)								
			298.15 K	303.15 K	308.15 K	313.15 K				
	Sodium saccharin + water									
0.0000			997.07	995.67	994.06	992.24				
0.0200			998.92	997.51	995.89	994.05				
0.0430			1000.77	999.35	997.72	995.86				
0.0599			1002.60	1001.17	999.53	997.65				
0.0798			1004.41	1002.97	1001.32	999.42				
0.1000			1006.22	1004.77	1003.11	1001.19				
	298.15 K	303.15 K	308.15 K	313.15 K	298.15 K	303.15 K	308.15 K	313.15 K		
	Sod	ium sacchari	n + 0.1 <i>m</i> fru	ctose	Sodium saccharin + 0.3 <i>m</i> fructose			ctose		
0.000	1003.98	1002.53	1000.92	999.13	1017.73	1016.22	1014.49	1012.56		
0.020	1005.82	1004.35	1002.72	1000.90	1019.55	1018.02	1016.27	1014.31		
0.040	1007.65	1006.16	1004.51	1002.66	1021.36	1019.81	1018.04	1016.05		
0.060	1009.47	1007.96	1006.29	1004.41	1023.16	1021.59	1019.80	1017.78		
0.080	1011.28	1009.75	1008.06	1006.15	1024.95	1023.36	1021.55	1019.50		
0.100	1013.08	1011.53	1009.82	1007.88	1026.73	1025.12	1023.29	1021.21		
	298.15 K 303.15 K 308.15 K 313.15 K									
	Sodium saccharin + 0.5 <i>m</i> fructose									
0.000			1030.56	1028.38	1026.83	1024.79				
0.020			1032.36	1030.17	1028.60	1026.53				
0.040			1034.15	1031.95	1030.36	1028.26				
0.060			1035.93	1033.72	1032.11	1029.98				
0.080			1037.70	1035.48	1033.85	1031.69				
0.100			1039.46	1037.23	1035.58	1033.39				

By the use of experimentally measured values of densities, the apparent molar volumes $(V_{\phi} / \text{m}^3 \cdot \text{mol}^{-1})$ of sodium saccharin in water and fructose solutions were calculated with the help of the Equation 1 [9-10].

$$V_{\phi} = \{ (M/\rho) - [(\rho - \rho_0)/(m\rho\rho_0)] \}$$
 1

where M, m, ρ and ρ_0 are the molar mass of the solute, molality of the sodium saccharin solution, density of solvent, and the density of the aqueous solution, respectively. For the calculation of apparent molar volumes, density values of water have been taken from the literature [11]. Table 2 reports the values of V_{ϕ} at (298.15, 303.15, 308.15, and 313.15) K for (sodium saccharin + water) and (sodium saccharin + water + fructose) systems. The calculated V_{ϕ} are correlated with molality by the use of the Equation 2 [12].

$$[V_{\phi} = V_{\phi}^{0} + S_{\rm V} m^{0.5}]$$

where V_{ϕ}^{0} and S_{V} are the partial molar volume and solute-solute interaction parameter, respectively. Table 2 and Figures 1, 2, 3, and 4 clarify that V_{ϕ} of sodium saccharin in water and in (0.1, 0.3, and 0.5) *m* fructose varies linearly with concentration of sodium saccharin. V_{ϕ} of sodium saccharin in water and (0.1, 0.3, and 0.5) *m* fructose increases with increase in the temperature. V_{ϕ} of sodium saccharin increases with increase in the concentration of fructose.

Table 2. Apparent molar volume ($V_{\phi}/\text{m}^3 \cdot \text{mol}^{-1}$) of sodium saccharin in water and (0.1, 0.3, and 0.5) *m* fructose at different temperatures.

т	$10^6 V_{\phi}$								
(mol·kg ⁻¹)		m ³ ·mol ⁻¹							
			298.15 K	303.15 K	308.15 K	313.15 K			
	Sodium saccharin + water								
0.0200			112.10	112.63	113.17	114.23			
0.0430			112.33	112.87	113.40	114.46			
0.0599			112.44	112.97	113.51	114.57			
0.0798			112.54	113.07	113.61	114.67			
0.1000			112.61	113.14	113.68	114.74			
	298.15 K	303.15 K	308.15 K	313.15 K	298.15 K	303.15 K	308.15 K	313.15 K	
	Sod	Sodium saccharin + 0.1 <i>m</i> fructose				Sodium saccharin + 0.3 m			
0.020	112.88	113.90	114.94	116.49	113.54	114.54	115.56	117.08	
0.040	112.92	113.95	114.98	116.53	113.58	114.58	115.60	117.12	
0.060	112.96	113.99	115.03	116.58	113.62	114.62	115.64	117.17	
0.080	113.01	114.04	115.07	116.63	113.66	114.67	115.69	117.21	
0.100	113.05	114.08	115.12	116.67	113.70	114.71	115.73	117.26	
			298.15 K	303.15 K	308.15 K	313.15 K			
	Sodium saccharin $+$ 0.5 <i>m</i> fructose								
0.020			114.15	114.68	115.67	117.17			
0.040			114.18	114.72	115.71	117.21			
0.060			114.22	114.76	115.75	117.25			
0.080			114.26	114.80	115.79	117.29			
0.100			114.30	114.84	115.83	117.33			



Figure 1. Variation of (V_{ϕ}) of sodium saccharin in water with $m^{0.5}$.



Figure 2. Variation of (V_{ϕ}) of sodium saccharin in presence of 0.1 *m* fructose with $m^{0.5}$.



Figure 3. Variation of (V_{ϕ}) of sodium saccharin in presence of 0.3 *m* fructose $m^{0.5}$.



Figure 4. Variation of (V_{ϕ}) of sodium saccharin in presence of 0.5 *m* fructose with $m^{0.5}$.

Figures 1, 2, 3, and 4 show the variation of V_{ϕ} with $m^{0.5}$ at different temperatures. V_{ϕ}^{0} and S_{V} values were calculated using least square method. Table 3 reports V_{ϕ}^{0} values of sodium saccharin in water and in (0.1, 0.3, and 0.5) *m* fructose solutions. From Table 3, it is understood that:

- a. V_{ϕ}^{0} values of sodium saccharin in water and in (0.1, 0.3, and 0.5) *m* fructose solutions are positive.
- b. V^{0}_{ϕ} value increases with increase in the temperature.
- c. V^{0}_{ϕ} value increases with increase in the concentration of fructose

The experimentally observed V^{0}_{ϕ} values for sodium saccharin in water at (298.15, 303.15, 308.15, 313.15) K are (111.725, 112.256, 112.793, and 113.846) × 10⁻⁶ m³·mol⁻¹. The reported [13] values of V_{ϕ}^{0} for sodium saccharin (111.336, 111.85, 112.52, 113.90 × 10⁻⁶ m³·mol⁻¹ in water at (298.15, 303.15, and 313.15) K are very close to the literature values. Positive values of V^{0}_{ϕ} indicate strong sodium saccharin-water interactions. The S_V values for all systems studied are also positive but smaller than V_{ϕ}^{0} values, suggesting that solute-solute interactions are weaker than solute-solvent interactions. V_{ϕ}^{0} and S_{V} values increase with increase in the temperature. This suggests that at higher temperature the electrostriction effect of water reduces and water molecules in secondary solvation layer release into the bulk of the water. This result leads to the expansion of the solution [14]. V_{ϕ}^{0} varies with temperature according to the Equation 3.

$$[V^0_{\phi} = a_0 + a_1 T + a_2 T^2]$$
 3

where a_0 , a_1 , and a_2 are constants. Least square method was used for calculations of a_0 , a_1 , and a_2 .

To calculate Expansion Coefficient E^{∞} , Equation 4 was used.

$$[E^{\infty} = (\partial V^{0}_{\phi} / \partial \mathbf{T}) = (\mathbf{a}_{1} + 2\mathbf{a}_{2}\mathbf{T})]$$

$$4$$

Table 3 includes E^{∞} values for sodium saccharin-water and sodium saccharin-fructose systems. The values of E^{∞} are positive and decrease with increase in the concentration of fructose. The positive values of E^{∞} indicate strong solute-solvent [14] interactions in all investigated solutions. Furthermore, E^{∞} value increases with increase in temperature at all composition of fructose. In ternary mixtures, same effect of temperature on E^{∞} has been reported previously by some researchers [14-15]. To get qualitative information regarding hydration of a solute, Hepler's constant [16] $(\partial^2 V^0_{\phi}/\partial T^2)_p$ was calculated by using the Equation 5.

$$\left[\left(\partial^2 V^0_{\phi}/\partial T^2\right)_{\rm p} = 2a_2\right]$$

 $(\partial^2 V_{\phi}^0 / \partial T^2)_p$ values are positive for all studied systems. Therefore, sodium saccharin behaves as structure maker in water and in (0.1, 0.3, and 0.5) *m* fructose solutions.

The thermodynamic property, partial molar volume of transfer at infinite dilution $(\Delta_{\rm trs}V^0_{\phi})$ of sodium saccharin from water to aqueous fructose solutions was calculated by the use of the following equation [17].

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

From Table 3 and Figure 5, it is clear that $(\Delta_{trs}V^0_{\phi})$ values of sodium saccharin in aqueous fructose solutions are positive and increase with increase in the concentration of fructose. Similar results have been obtained in (NaCl + water + Glucose) [17], (NaI + water + glucose) [18], (NaBr + water + glucose) [19], (diglycine + water + fructose) [20], and (1-histidine + water + glucose) [21] systems. Variation of $(\Delta_{trs}V^0_{\phi})$ with molality is depicted in Figure 5.

Kozak et al. [22] proposed theory which was based on McMillan-Mayer [23] theory of solutions. The proposed theory allows the formal separation of the effects due to interactions between pairs of solute molecules and those due to the interactions involving three or more than three molecules. To include solutecosolute interactions in the solvation spheres, the approach was further discussed by Friedmann and Krishnan [24] and Franks [25]. Same approach was used by many researchers to study the solute-cosolute interactions in aqueous solutions [26-29]. Equation 7 [21-23] can be used for calculation of volumetric interaction parameters doublet V_{AB} (m³·mol⁻²·kg) and triplet V_{ABB} (m³·mol³·kg²).

$$[(\Delta_{\rm trs} V^0_{\phi}) = 2 \, V_{\rm AB} \, m_{\rm B} + 3 \, V_{\rm ABB} \, m^2_{\rm B}]$$
 7

where A denotes sodium saccharin (solute) and B denotes fructose (co-solute).

Least squares method was used for estimation of V_{AB} and V_{ABB} values. The calculated values of V_{AB} for sodium saccharin + fructose at (298.15, 303.15, 308.15, and 313.15) K are 0.849, 1.317, 1.191, and 2.065 and those of V_{ABB} are -0.803, -1.747, -2.3237, and -2.899. For all solutions studied, the values of V_{AB} are positive and the values of V_{ABB} are negative. Positive values of V_{AB} and negative values of V_{ABB} suggest the strong interactions between sodium saccharin and fructose. Increase in the V_{AB} value with

concentration of fructose is mainly due to the increase in the sodium saccharin-fructose interactions.

Table 3. (V_{ϕ}^0) , S_V , (E^{∞}) , $(\partial^2 V_{\phi}^0/\partial T^2)_p$, (ASV), and $(\Delta_{trs}V_{\phi}^0)$ of sodium saccharin in (0.1, 0.3, and 0.5) *m* fructose at different temperatures.

			298.15 K	303.15 K	308.15 K	313.15 K			
	Sodium saccharin + water								
$10^6 V_{\phi}^0$			111.725	112.256	112.793	113.846			
$10^{6} V_{s}$			2.866	2.877	2.890	2.903			
$10^7 E^{\infty}$			0.595	1.118	1.641	2.165			
10 ⁶ ASV			0.544	0.547	0.550	0.555			
$10^8 \left(\frac{\partial^2 V_{\phi}^0}{\partial T^2} \right)_p$				0.1	046				
Ţ	298.15 K	303.15 K	308.15 K	313.15 K	298.15 K	303.15 K	308.15 K	313.15 K	
	Sodiu	ım sacchariı	n + 0.1 <i>m</i> fru	ctose	Sodium saccharin + 0.3 <i>m</i> fructose				
$10^6 V_{\phi}^0$	112.729	113.752	114.785	116.329	113.396	114.400	115.415	116.929	
$10^{6} V_{s}$	0.993	1.012	1.033	1.062	0.929	0.949	0.971	1.003	
$10^7 E^{\infty}$	1.586	2.107	2.627	3.146	1.557	2.067	2.578	3.088	
10 ⁶ ASV	0.549	0.554	0.559	0.560	0.553	0.557	0.562	0.569	
$\Delta_{trs} V_{\phi}^0 \times 10^6$	1.003	1.496	1.992	2.482	1.671	2.144	2.622	3.083	
$10^8 \left(\frac{\partial^2 V_{\phi}^0}{\partial T^2} \right)_p$	0.104				0.102				
Ψ r			298.15 K	303.15 K	308.15 K	313.15 K			
	Sodium saccharin + 0.5 <i>m</i> fructose								
$10^{6} V_{\phi}^{0}$			114.013	114.546	115.536	117.023			
$10^{6} V_{s}$			0.876	0.894	0.916	0.949			
$10^7 E^{\infty}$			5.720	1.526	2.481	3.436			
10 ⁶ ASV			0.556	0.558	0.563	0.570			
$\Delta_{trs} V_{\phi}^0 \times 10^6$			2.288	2.289	2.744	3.177			
$10^8 \left(\frac{\partial^2 V_{\phi}^0}{\partial T^2} \right)_p$	0.191								



Figure 5. Variation of partial molar volume transfer $\Delta_{trs} V_{\phi}^0 / m^3 \cdot mol^{-1}$ at infinite dilution of sodium saccharin from water to fructose solutions with concentration (*m*) of fructose at different temperatures.

Negative values of V_{ABB} for all systems studied suggest the absence of sodium saccharin-fructosefructose interactions. Wang et al. [17] reported the positive values of V_{AB} and negative values V_{ABB} for NaCl-glucose-water, NaCl-arabinose-water, and NaCl-galactose-water systems at 298.15 K.

According to group additivity model [30] four types of interactions present between electrolyte and saccharides. These are cation –R (Alkyl group), anion R, cation –O (–OH, C=O, and –O–) and anion O. A

strong electrolyte, sodium saccharin dissociates into ions in aqueous solution. According to structural interaction models [30-31], only cation interactions give positive contribution to V_{AB} whereas other three types of interactions have negative contribution to V_{AB} . Therefore, the positive value of V_{AB} may be due to the interactions between hydrophilic group (–OH, C=O, and –O–) of fructose and sodium ion of sodium saccharin. Due to the strong solute-cosolute interactions V_{AB} values of all studied systems increase with increase in the temperature.

To calculate Apparent Specific Volume (ASV) of sodium saccharin in water and in aqueous solutions of fructose following equation [32-33] was used.

$$[ASV = V^{0}_{\phi}/M]$$

where V_{ϕ}^{0} and *M* are the partial molar volume and molar mass of the solute, respectively. ASV values of sodium saccharin in water and fructose solutions are reported in Table 3. On the basis of taste quality, parameter ASV can be used to distinguish sweeteners as salty, sweet, bitter, and sour [34]. For sweet molecules ASV ranges from (0.51×10^{-6}) m³·kg⁻¹ to (0.71×10^{-6}) m³·kg⁻¹. The ASV for ideal sweet taste lies at center of the range [35] (0.618×10^{-6}) m³·kg⁻¹. From Table 3 it is observed that ASV of all studied solutions ranges from (0.544×10^{-6}) m³·kg⁻¹ to (0.570×10^{-6}) m³·kg⁻¹. Therefore, all studied solutions exhibit sweet taste.

4. CONCLUSION

From volumetric study of aqueous solutions of intense sweetener sodium saccharin in presence of sugar fructose it is revealed that sodium saccharin is water structure maker. In presence of fructose, the interactions exist between hydrophilic group (-OH, C=O, and -O-) of fructose and sodium ion of sodium saccharin in aqueous solutions of sodium saccharin. The interactions between sodium saccharin and fructose increase with increase in concentration of fructose. All studied solutions exhibit sweet taste.

5. ACKNOWLEDMENTS

This work has been carried out at Chemistry Department, HPT Arts and RYK Science College, Nashik. SMM thanks Principal of HPT Arts and RYK Science College, Nashik for providing the facilities to perform this research work.

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