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

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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 solute-solvent 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
deionized water. Density measurements were undertaken in glass-walled bath. Uncertainty in density and temperature measurements were $5.8 \times 10^{-2}$ kg·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 ($\rho$ / kg·m$^{-3}$) of sodium saccharin in water and (0.1, 0.3, and 0.5) $m$ fructose at different temperatures.

| $m$ (mol·kg$^{-1}$) | $\rho$ (kg·m$^{-3}$) |
|---------------------|----------------------|
|                     | 298.15 K             | 303.15 K             | 308.15 K             | 313.15 K             |
| Sodium saccharin + water |
| 0.000              | 997.07               | 995.67               | 994.06               | 992.24               |
| 0.020              | 998.92               | 997.51               | 995.89               | 994.05               |
| 0.040              | 1000.77              | 999.35               | 997.72               | 995.86               |
| 0.059              | 1002.60              | 1001.17              | 999.53               | 997.65               |
| 0.0799             | 1004.41              | 1002.97              | 1001.32              | 999.42               |
| 0.100              | 1006.22              | 1004.77              | 1003.11              | 1001.9               |
| Sodium saccharin + 0.1 $m$ fructose |
| 298.15 K           | 1000.23              | 1002.53              | 1004.41              | 1006.29              |
| 0.000              | 1003.98              | 1000.92              | 999.13               | 1017.73              |
| 0.020              | 1005.82              | 1002.72              | 1000.90              | 1019.55              |
| 0.040              | 1007.65              | 1004.51              | 1002.66              | 1021.36              |
| 0.060              | 1009.47              | 1006.96              | 1004.41              | 1023.16              |
| 0.080              | 1011.28              | 1009.75              | 1006.15              | 1024.95              |
| 0.100              | 1013.08              | 1011.53              | 1007.88              | 1026.73              |
| Sodium saccharin + 0.3 $m$ fructose |
| 298.15 K           | 1004.62              | 1007.53              | 1010.41              | 1013.29              |
| 0.000              | 1008.36              | 1005.23              | 1003.41              | 1016.22              |
| 0.020              | 1010.23              | 1006.72              | 1004.51              | 1018.02              |
| 0.040              | 1012.10              | 1008.65              | 1006.15              | 1019.81              |
| 0.060              | 1013.97              | 1010.96              | 1008.78              | 1021.59              |
| 0.080              | 1015.85              | 1013.37              | 1011.53              | 1023.16              |
| 0.100              | 1017.73              | 1015.73              | 1013.88              | 1025.12              |
| Sodium saccharin + 0.5 $m$ fructose |
| 298.15 K           | 1008.23              | 1011.53              | 1014.41              | 1017.21              |
| 0.000              | 1012.04              | 1014.92              | 1012.83              | 1015.24              |
| 0.020              | 1013.86              | 1016.72              | 1019.60              | 1022.73              |
| 0.040              | 1015.68              | 1018.65              | 1021.41              | 1025.12              |
| 0.060              | 1017.50              | 1020.37              | 1023.16              | 1027.21              |
| 0.080              | 1019.32              | 1022.10              | 1024.95              | 1029.21              |
| 0.100              | 1021.14              | 1023.88              | 1026.73              | 1031.69              |

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

$$V_A = \{(M / \rho) - [\rho / (\rho_0 / \mu)]\}$$

where $M$, $\rho$, and $\rho_0$ 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 $V_A$ at (298.15, 303.15, 308.15, and 313.15) K for (sodium saccharin + water) and (sodium saccharin + water + fructose) systems. The calculated $V_A$ are correlated with molality by the use of the Equation 2 [12].

$$V_A = V_A^0 + S_V m^{0.5}$$

where $V_A^0$ 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 $V_A$ of sodium saccharin in water and in (0.1, 0.3, and 0.5) $m$ fructose varies linearly with concentration of sodium saccharin. $V_A$ of sodium saccharin in water and (0.1, 0.3, and 0.5) $m$ fructose increases with increase in the temperature.
Vφ of sodium saccharin increases with increase in the concentration of fructose.

Table 2. Apparent molar volume (Vφ/m³·mol⁻¹) of sodium saccharin in water and (0.1, 0.3, and 0.5) m fructose at different temperatures.

| m          | 10⁴Vφ m³·mol⁻¹ | 298.15 K | 303.15 K | 308.15 K | 313.15 K |
|------------|----------------|----------|----------|----------|----------|
| Sodium saccharin + water |
| 0.020      | 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   |
| Sodium saccharin + 0.1 m fructose |
| 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   |
| Sodium saccharin + 0.3 m 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   |
| Sodium saccharin + 0.5 m 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 (Vφ) of sodium saccharin in water with m⁰⁵.

Figure 2. Variation of (Vφ) of sodium saccharin in presence of 0.1 m fructose with m⁰⁵.

Figure 3. Variation of (Vφ) of sodium saccharin in presence of 0.3 m fructose m⁰⁵.

Figure 4. Variation of (Vφ) of sodium saccharin in presence of 0.5 m fructose with m⁰⁵.
Figures 1, 2, 3, and 4 show the variation of $V^0_θ$ with $m^{0.5}$ at different temperatures. $V^0_θ$ and $S_θ$ values were calculated using least square method. Table 3 reports $V^0_θ$ 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. $V^0_θ$ values of sodium saccharin in water and in (0.1, 0.3, and 0.5) $m$ fructose solutions are positive.

b. $V^0_θ$ value increases with increase in the temperature.

c. $V^0_θ$ value increases with increase in the concentration of fructose

The experimentally observed $V^0_θ$ 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) $× 10^6$ m$^3$·mol$^{-1}$. The reported [13] values of $V^0_θ$ for sodium saccharin (111.336, 111.85, 112.52, 113.90) $× 10^6$ m$^3$·mol$^{-1}$ in water at (298.15, 303.15, and 313.15) K are very close to the literature values. Positive values of $V^0_θ$ indicate strong sodium saccharin-water interactions. The $S_θ$ values for all systems studied are also positive but smaller than $V^0_θ$ values, suggesting that solute-solute interactions are weaker than solute-solvent interactions. $V^0_θ$ and $S_θ$ 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]. $V^0_θ$ varies with temperature according to the Equation 3.

\[ V^0_θ = a_0 + a_1 T + a_2 T^2 \] 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 $E^ο$, Equation 4 was used.

\[ E^ο = (\frac{∂V^θ_θ}{∂T}) = (a_1 + 2a_2 T) \] 4

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

\[ (\frac{∂^2V^θ_θ}{∂T^2}) = 2a_2 \] 5

($\frac{∂^2V^θ_θ}{∂T^2})$ 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 ($Δ_n/V^θ_n$) of sodium saccharin from water to aqueous fructose solutions was calculated by the use of the following equation [17],

\[ (Δ_n/V^θ_n) = [V^θ_n in (aqueous fructose solutions) - V^θ_n (water)] \] 6

From Table 3 and Figure 5, it is clear that ($Δ_n/V^θ_n$) 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 ($Δ_n/V^θ_n$) 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 solute-cosolute 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 $V_{AB}$ (m$^3$·mol$^{-1}$·kg) and triplet $V_{ABB}$ (m$^3$·mol$^{-1}$·kg$^2$).

\[ [(Δ_n/V^θ_n) = 2 V_{AB} m_B + 3 V_{ABB} m_B^2] \] 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 method 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. $(V_0^\phi, S_V, (E^\infty), (\partial^2 V_0^\phi / \partial T^2)_p, (ASV),$ and $(\Delta_{trs} V_0^\phi)$ 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 |          |          |          |          |
| $10^6 V_0^\phi$     | 111.725  | 112.256  | 112.793  | 113.846  |
| $10^6 V_0$          | 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    |
| $10^8 (\partial^2 V_0^\phi / \partial T^2)_p$ | 0.1046   |          |          |          |

| Sodium saccharin + 0.1 m fructose | 298.15 K | 303.15 K | 308.15 K | 313.15 K |
| Sodium saccharin + 0.3 m fructose | 298.15 K | 303.15 K | 308.15 K | 313.15 K |
| Sodium saccharin + 0.5 m fructose | 298.15 K | 303.15 K | 308.15 K | 313.15 K |

Negative values of $V_{ABB}$ for all systems studied suggest the absence of sodium saccharin-fructose-fructose 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.

$$ [ASV = V_0/M]$$

where $V_0$ 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 $(0.51 \times 10^{-6})$ m$^3$kg$^{-1}$ to $(0.71 \times 10^{-6})$ m$^3$kg$^{-1}$. The ASV for ideal sweet taste lies at center of the range [35] $(0.618 \times 10^{-6})$ m$^3$kg$^{-1}$. From Table 3 it is observed that ASV of all studied solutions ranges from $(0.544 \times 10^{-6})$ m$^3$kg$^{-1}$ to $(0.570 \times 10^{-6})$ m$^3$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.

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