Volumetric Properties of Sodium Cyclamate Solutions in Presence of Glucose and Sucrose

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Abstract. Densities of sodium cyclamate (Na-cycl) in water and (0.1, 0.3, and 0.5) m (glucose/sucrose) have been measured at (298.15, 303.15, 308.15, and 313.15) K. From density values, partial molar volumes \( (V^0) \), expansion coefficient \( (E^0) \), Hepler’s constant \( (\partial^2 V^0 / \partial T^2)_p \), apparent specific volumes (ASV), partial molar volumes of transfer \( (\Delta m_r V^0) \), doublet \( (V_{AB}) \) and triplet \( (V_{ABB}) \) interaction coefficients have been calculated. An increase in the values of \( V^0 \) and \( \Delta m_r V^0 \) was observed with increase in the concentration of glucose/sucrose. The positive values of \( E^0 \) and \( V^0 \) are due to the strong solute-solvent interactions. The positive values of \( (\partial^2 V^0 / \partial T^2)_p \) suggest structure making behaviour of sodium cyclamate in water and in presence of glucose and sucrose. The positive values of \( (\Delta m_r V^0) \) and \( V_{AB} \) may be due to the interactions between hydrophilic group (–OH, C=O, and –O–) of glucose/sucrose and sodium ion of sodium cyclamate. All solutions studied exhibit sweet taste because ASV of all solutions ranges from \( (0.569 \times 10^{-6}) \) m\(^3\)-kg\(^{-1}\) to \( (0.626 \times 10^{-6}) \) m\(^3\)-kg\(^{-1}\).

1. Introduction

Synthetic sugar substitutes, artificial sweeteners provide little or no calories or carbohydrates and do not increase blood sugar. Artificial sweeteners are known as intense sweeteners because they are highly sweeter than natural sugars. According to the United States Food and Drug Administration [1], a non-nutritive sweetener is a substance which has less than 2% of the caloric value of sucrose per equivalent unit of sweetening capacity. Artificial sweeteners are considered as non-nutritive and very minute quantity of their is required to achieve a desirable sweetness level in a food formulation. Due this reason the intense sweeteners are useful for the food manufacturer. A common practice in food and pharmaceutical industries is to blend the sweetener. By blending the sweetener, the user can take the advantages like lower use level, lowest cost, and improved taste. The intense sweetener sodium cyclamate and its blends with other sweeteners are commonly used in food industries and pharmaceuticals [2].

Water is very important in sweet taste because it is not possible to taste the molecule unless it is soluble and transportable to the receptors via oral fluid. An understanding of mechanism of the taste chemoreception might be achieved by studying water-solute (sweetener) interactions [3-5]. Density data can be used to study the water-sweetener and sweetener-sweetener interactions.

This paper reports densities of Na-Cycl in water and (0.1, 0.3 and 0.5) m (glucose/sucrose) at (298.15, 303.15, 308.15, and 313.15) K. The objectives of research work reported here are to generate the accurate data of densities of sodium cyclamate solutions in presence or absence of glucose and sucrose, to study the sweetener-water and sweetener-sweetener interactions and to get the information regarding taste qualities of solutions.
2. Materials and Methods

Na-cycl (Merck, purity ≥ 99.0), glucose (Merck, purity ≥ 99.0) and sucrose (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 0.1mg. Density measurements were undertaken by using 15 cc bi-capillary pycnometer [6-9]. Pycnometer was calibrated with triply distilled water. For measurements of density of aqueous solutions at different temperatures, glass-walled thermostat was used. Density measurements were made at (298.15, 303.15, 308.15, and 313.15) K. Uncertainties in the density and temperature measurements were 5.8 × 10^{-2} kg m^{-3} and 0.006 K, respectively.

3. Results and Discussion

Blends of sweetener with other sweeteners have great importance in pharmaceutical and food industries. Apparent specific volume and, isentropic compressibilities, and hydration numbers of binary mixtures of two bulk sweeteners (sucrose and maltitol) and three intense sweeteners (potassium acesulfame, aspartame, and sodium cyclamate) have been reported previously [10]. In present investigation, aqueous solutions of Na-cycl in presence of glucose and sucrose have been studied at different temperatures. Densities data of Na-cycl in presence of 0.1, 0.3, and 0.5 m (glucose/sucrose have been generated at (298.15, 303.15, 308.15, and 313.15) K and at atmospheric pressure. Table 1 reports the densities of aqueous solutions Na-cycl in absence and presence of (0.1, 0.3, and 0.5) m glucose/sucrose at (298.15, 303.15, 308.15, and 313.15) K.

The observed trend of density (ρ) of Na-cycl in water, glucose, and sucrose is as follows

ρ_{water} < ρ_{glucose} < ρ_{sucrose}

It is observed that density of aqueous solutions of Na-cycl varies linearly with molality of the solutions. As usual, density decreases with increase in the temperature of the solutions. Similar behaviour of density of aqueous solutions of Na-cycl in presence of (0.1, 0.3, and 0.5) m glucose/sucrose has been observed. Furthermore, it is observed that density of aqueous solutions of Na-cycl in presence of (0.1, 0.3, and 0.5) m glucose/sucrose increases with increase in the concentration of glucose/sucrose.

By the use of experimentally measured values of densities, the apparent molar volumes ($V_\phi$ / m³·mol⁻¹) of Na-cycl in water, glucose, and sucrose were calculated by using the Equation 1 [11-12].

$$V_\phi = \frac{M}{\rho} - \left(\frac{(\rho - \rho_0)}{(m \rho_0)}\right)$$

where $M$, $m$, $\rho_0$, and $\rho$ are the molar mass of the solute, molality of the Na-cycl solution, density of solvent, and the density of the aqueous solution, respectively. The density values of water have been taken from the literature [13] for the calculation of apparent molar volumes.

From Table 2, it is clear that:

a. $V_\phi$ of Na-cycl in water and in (0.1, 0.3 and 0.5) m glucose and sucrose increases linearly with concentration of sodium cyclamate.

b. $V_\phi$ of Na-cycl in water and (0.1, 0.3, and 0.5) m glucose and sucrose increases with increase in the temperature.

c. $V_\phi$ of Na-cycl increases with increase in the concentration of glucose and sucrose.
Table 1. Densities ($\rho$/kg·m$^{-3}$) of sodium cyclamate in water and (0.1, 0.3, and 0.5) $m$ glucose/sucrose at different temperatures.

| $m$ (mol·kg$^{-1}$) | 298.15 K | 303.15 K | 308.15 K | 313.15 K |
|---------------------|----------|----------|----------|----------|
| Sodium cyclamate + water |          |          |          |          |
| 0.0000              | 997.07   | 995.67   | 994.06   | 992.24   |
| 0.0200              | 998.63   | 997.22   | 995.59   | 993.75   |
| 0.0430              | 1000.41  | 998.99   | 997.33   | 995.47   |
| 0.0599              | 1001.71  | 1000.28  | 998.6    | 996.72   |
| 0.0798              | 1003.23  | 1001.79  | 1000.09  | 998.19   |
| 0.1000              | 1004.76  | 1003.31  | 1001.59  | 999.67   |
| Sodium cyclamate + 0.1 $m$ glucose |          |          |          |          |
| 0.0000              | 1003.83  | 1002.39  | 1000.78  | 998.99   |
| 0.0200              | 1005.38  | 1003.93  | 1002.30  | 1000.48  |
| 0.0400              | 1006.92  | 1005.46  | 1003.81  | 1001.96  |
| 0.0600              | 1008.45  | 1006.98  | 1005.31  | 1003.43  |
| 0.0800              | 1009.97  | 1008.49  | 1006.80  | 1004.89  |
| 0.1000              | 1011.48  | 1009.99  | 1008.28  | 1006.34  |
| Sodium cyclamate + 0.3 $m$ glucose |          |          |          |          |
| 0.0000              | 1029.56  | 1027.76  | 1026.3   | 1024.21  |
| 0.0200              | 1031.07  | 1029.26  | 1027.77  | 1025.65  |
| 0.0400              | 1032.57  | 1030.75  | 1029.23  | 1027.08  |
| 0.0600              | 1034.06  | 1032.23  | 1030.68  | 1028.50  |
| 0.0800              | 1035.54  | 1033.70  | 1032.12  | 1029.91  |
| 0.1000              | 1037.01  | 1035.16  | 1033.55  | 1031.31  |
| Sodium cyclamate + 0.5 $m$ glucose |          |          |          |          |
| 0.0000              | 1040.02  | 1038.40  | 1036.51  | 1034.69  |
| 0.0200              | 1035.55  | 1033.96  | 1032.16  | 1030.43  |
| 0.0400              | 1037.05  | 1035.45  | 1033.62  | 1031.86  |
| 0.0600              | 1038.54  | 1036.93  | 1035.07  | 1033.28  |
| 0.0800              | 1040.02  | 1038.40  | 1036.51  | 1034.69  |
| 0.1000              | 1041.49  | 1039.86  | 1037.94  | 1036.09  |
| Sodium cyclamate + 0.1 $m$ sucrose |          |          |          |          |
| 0.0000              | 1029.56  | 1027.76  | 1026.3   | 1024.21  |
| 0.0200              | 1031.07  | 1029.26  | 1027.77  | 1025.65  |
| 0.0400              | 1032.57  | 1030.75  | 1029.23  | 1027.08  |
| 0.0600              | 1034.06  | 1032.23  | 1030.68  | 1028.50  |
| 0.0800              | 1035.54  | 1033.70  | 1032.12  | 1029.91  |
| 0.1000              | 1037.01  | 1035.16  | 1033.55  | 1031.31  |
| Sodium cyclamate + 0.3 $m$ sucrose |          |          |          |          |
| 0.0000              | 1034.04  | 1032.46  | 1030.69  | 1028.99  |
| 0.0200              | 1035.55  | 1033.96  | 1032.16  | 1030.43  |
| 0.0400              | 1037.05  | 1035.45  | 1033.62  | 1031.86  |
| 0.0600              | 1038.54  | 1036.93  | 1035.07  | 1033.28  |
| 0.0800              | 1040.02  | 1038.40  | 1036.51  | 1034.69  |
| 0.1000              | 1041.49  | 1039.86  | 1037.94  | 1036.09  |
| Sodium cyclamate + 0.5 $m$ sucrose |          |          |          |          |
| 0.0000              | 1040.02  | 1038.40  | 1036.51  | 1034.69  |
| 0.0200              | 1035.55  | 1033.96  | 1032.16  | 1030.43  |
| 0.0400              | 1037.05  | 1035.45  | 1033.62  | 1031.86  |
| 0.0600              | 1038.54  | 1036.93  | 1035.07  | 1033.28  |
| 0.0800              | 1040.02  | 1038.40  | 1036.51  | 1034.69  |
| 0.1000              | 1041.49  | 1039.86  | 1037.94  | 1036.09  |
Table 2. Apparent molar volume \( (V_\phi \text{ m}^3\text{mol}^{-1}) \) of sodium cyclamate in water and (0.1, 0.3, and 0.5) m glucose/sucrose at different temperatures.

| \( M \) (mol.kg\(^{-1}\)) | \( 10^6 V_\phi \) (m\(^3\)mol\(^{-1}\)) | 298.15 K | 303.15 K | 308.15 K | 313.15 K |
|-----------------------------|-------------------------------------|----------|----------|----------|----------|
| Sodium cyclamate + water     |                                     |          |          |          |          |
| 0.0200                      |                                     | 123.16   | 123.73   | 124.81   | 125.92   |
| 0.0400                      |                                     | 123.27   | 123.80   | 125.05   | 126.09   |
| 0.0599                      |                                     | 123.32   | 123.89   | 125.15   | 126.26   |
| 0.0798                      |                                     | 123.40   | 123.97   | 125.19   | 126.30   |
| 0.1000                      |                                     | 123.51   | 124.08   | 125.27   | 126.38   |
| Sodium cyclamate + 0.1 m glucose |                                 |          |          |          |          |
| 0.0200                      |                                     | 123.35   | 123.92   | 124.99   | 126.58   |
| 0.0400                      |                                     | 123.41   | 123.98   | 125.05   | 126.65   |
| 0.0600                      |                                     | 123.47   | 124.04   | 125.11   | 126.71   |
| 0.0800                      |                                     | 123.53   | 124.10   | 125.18   | 126.78   |
| 0.1000                      |                                     | 123.59   | 124.16   | 125.24   | 126.84   |
| Sodium cyclamate + 0.3 m glucose |                                 |          |          |          |          |
| 0.0200                      |                                     | 124.03   | 124.60   | 126.10   | 127.65   |
| 0.0400                      |                                     | 124.09   | 124.66   | 126.16   | 127.71   |
| 0.0600                      |                                     | 124.14   | 124.71   | 126.22   | 127.77   |
| 0.0800                      |                                     | 124.20   | 124.77   | 126.28   | 127.83   |
| 0.1000                      |                                     | 124.26   | 124.83   | 126.34   | 127.89   |
| Sodium cyclamate + 0.5 m glucose |                                 |          |          |          |          |
| 0.0200                      |                                     | 123.80   | 124.35   | 125.86   | 127.37   |
| 0.0400                      |                                     | 123.86   | 124.41   | 125.92   | 127.43   |
| 0.0600                      |                                     | 123.91   | 124.47   | 125.98   | 127.49   |
| 0.0800                      |                                     | 123.97   | 124.52   | 126.03   | 127.55   |
| 0.1000                      |                                     | 124.03   | 124.58   | 126.09   | 127.61   |

The calculated \( V_\phi \) are correlated with molality by the use of the Equation 2 [11-12].

\[
V_\phi = V_\phi^0 + S_V m^{0.5},
\]

where \( V_\phi^0 \) and \( S_V \) are the partial molar volume and solute-solute interaction parameter, respectively.

The least square method was used for calculations of \( V_\phi^0 \) and \( S_V \). Table 3 reports the values of \( V_\phi^0 \) of Na-cycl in water and in (0.1, 0.3, and 0.5) m glucose/sucrose.

From Table 3 it is understood that:

a. \( V_\phi^0 \) values of Na-cycl in water and in (0.1, 0.3 and 0.5) m glucose/sucrose are positive.

b. \( V_\phi^0 \) value increases with increase in the temperature and also increase in the concentration of glucose/sucrose.
The experimentally observed value of $V_0$ for Na-cycl in water at (298.15, 303.15, 308.15, 313.15) K are (122.87, 124.49, 125.54) $\times 10^{-6}$ m$^3$-mol$^{-1}$. The reported [14] values of $V_0$ for Na-cycl at (298.15, 303.15, and 313.15) K are (122.05, 123.31, and 124.55) $\times 10^{-6}$ m$^3$-mol$^{-1}$. Observed values of $V_0$ for Na-cycl in water at (298.15, 303.15, and 313.15) K are very close to the literature values. The observed values of $S_V$ for all systems studied are also positive but smaller than $V_0$ values. The $S_V$ values give the information related to solute-solute interactions. The $V_0$ 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]. The experimentally observed values of $S_V$ are positive but smaller than $V_0$ values, suggesting that solute-solute interactions are weaker than solute-solvent interactions.

$V_0$ varies with temperature according to the Equation 3.

$$V_0 = a_0 + a_1 T + a_2 T^2,$$

where $a_0$, $a_1$, and $a_2$ are constants.

Expansion coefficient $E^\infty$ can be calculated with help of Equation 4

$$E^\infty = (\partial V_0 / \partial T) = a_1 + 2a_2 T.$$

Table 3 compiles $E^\infty$ values for Na-cycl-water, Na-cycl-glucose, and Na-cycl-sucrose. $E^\infty$ values are positive and decrease with increase in the concentration of glucose/sucrose. The positive values of $E^\infty$ indicate strong solute-solvent [15] interactions in all investigated solutions. Furthermore, $E^\infty$ value increases with increase in temperature at all composition of glucose/sucrose. In ternary mixtures, same effect of temperature on $E^\infty$ has been reported previously by some researchers [15-16].

The Hepler’s constant [17] provides the qualitative information regarding hydration of a solute from the thermal expansion of aqueous solutions. Following equation was used for calculations of Hepler’s Constant.

$$\text{Hepler’s constant} = ((\partial^2 V_0 / \partial T^2)_p) = 2a_2.$$

The values of the Hepler’s constants for all the systems studied are reported in Table 3. For structure making solute, the Hepler’s constant should be positive [18]. The positive values of Hepler’s constants suggest structure making behaviour of Na-cycl in water and in (0.1, 0.3, and 0.5) m glucose/sucrose.

The partial molar volume of transfer at infinite dilution ($\Delta_{trs}V_0$) of Na-cycl from water to aqueous glucose/sucrose solutions were calculated by the using the equation 6 [19].

$$\Delta_{trs}V_0 = V_0(\text{in aqueous glucose solutions}) - V_0(\text{in water}) \quad (6)$$
The calculated values of $\Delta_{trs}V_{i}^{0}$ for all the systems studied are reported in the Table 3. The values of $\Delta_{trs}V_{i}^{0}$ for Na-cycl in aqueous glucose/sucrose solutions are positive and increase with increase in the concentration of glucose/sucrose. Similar results have been obtained in (NaCl + water + Glucose) [20], (NaI + water + sucrose) [23], and (1-histidine + water + sucrose) [24] systems. The concentration effect of glucose/sucrose on $\Delta_{trs}V_{i}^{0}$ can be interpreted in terms of the hydration model.

The standard partial molar volume of ion ($V_{i}^{0}$) can be expressed by using the Equation 7 [25-27]:

$$V_{i}^{0} = V_{i} + V_{c} + V_{r} + V_{e}$$  \hspace{1cm} (7)$$

where $V_{i}$, $V_{c}$, $V_{r}$, and $V_{e}$ are the intrinsic volume, the cavity volume, the solvent-structure reaction volume, and the electrostriction volume, respectively.
But,
$$\Delta V_e = V_c + V_r + V_e$$
(8)
Therefore,
$$V_i^o = V_i + \Delta V_e$$
(9)
and
$$\Delta V_e = V_i^o - V_e$$
(10)

The $\Delta V_e$ has large negative value [28]. With increase in the molality of glucose/sucrose, the partial dehydration of sodium and cyclamate ion will take place compared with that in water. The glucose/sucrose molecules have less polarity and larger volume than water, their electrostriction effect by the ions is weaker than that of water. Therefore, $\Delta V_e$ value will become less negative with increase in molality of glucose/sucrose. This is the possible reason for the fact that $\Delta_{trs} V_\phi^0$ increases with increase in the molality of glucose/sucrose.

Theory based on McMillan-Mayer [29] theory of solutions was proposed by Kozak et al. [30]. 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. In order to include solute-cosolute interactions in the solvation spheres, the approach was further discussed by Friedmann and Krishnan [31] and Franks [32]. Same approach was used by many researchers to study the solute-cosolute interactions in aqueous solutions [33-36].

Equation 11 [29-30] can be used for calculation of volumetric interaction parameters doublet $V_{AB}$ (m$^3$·mol$^{-2}$·kg) and triplet $V_{ABB}$ (m$^3$·mol$^{-3}$·kg$^2$)
$$\Delta_{trs} V_\phi^0 = 2 V_{AB} m_B + 3 V_{ABB} m_B^2 + \ldots$$
(11)
where $A$ denotes Na-cycl (solute) and $B$ denotes glucose/sucrose/ (co-solute). For calculations of $V_{AB}$ and $V_{ABB}$, least square method was used. The volumetric interaction parameters doublet ($V_{AB}$) and triplet ($V_{ABB}$) are listed in Table 4.

**Table 4.** Solute-solute interaction parameters doublet ($V_{AB}$) and triplet ($V_{ABB}$), in aqueous solutions of sodium cyclamate in the presence of glucose/sucrose.

| Interaction parameters | 298.15 K | 303.15 K | 308.15 K | 313.15 K |
|------------------------|----------|----------|----------|----------|
| $V_{AB}$ $\times 10^6$ (m$^3$·mol$^{-2}$·kg) | 2.417 | 2.534 | 4.207 | 8.171 |
| $V_{ABB}$ $\times 10^6$ (m$^3$·mol$^{-3}$·kg$^2$) | -0.999 | -1.167 | -2.688 | -8.835 |
| $V_{AB}$ $\times 10^6$ (m$^3$·mol$^{-2}$·kg) | 3.136 | 3.225 | 5.826 | 9.589 |
| $V_{ABB}$ $\times 10^6$ (m$^3$·mol$^{-3}$·kg$^2$) | -0.918 | -1.109 | -3.189 | -9.332 |

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 Na-cycl and glucose/sucrose. Increase in the $V_{AB}$ value with concentration of glucose/sucrose is mainly due to the increase in the Na-cycl-glucose/sucrose interactions. Negative values of $V_{ABB}$ for all systems studied suggest the absence of Na-cycl-glucose-glucose, Na-cycl-sucrose-sucrose interactions. Wang et al [20] 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.

Group additivity model [37] describes four types of interactions between electrolyte and saccharides. Cation – R (Alkyl group), anion R, cation –O (–OH, C=O, and –O–) and anion O. Na-cycl is an electrolyte it dissociates into ions. According to structural interaction models [38-39], 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 glucose/sucrose and sodium ion of Na-cycl. In all systems studied, $V_{AB}$ value increases with increase in the temperature due to the increase in the solute-cosolute interactions.

Apparent specific volume (ASV) of Na-cycl in water and in aqueous solutions of glucose/sucrose can be calculated by the use of the Equation 12 [10, 40].

$$\text{ASV} = \frac{V_0^\phi}{M},$$  \hspace{1cm} (12)

where $V_0^\phi$ and $M$ are the partial molar volume and molar mass of the solute, respectively. Table 3 includes values of ASV for Na-cycl-glucose/sucrose systems.

On the basis of taste quality, parameter apparent specific volume (ASV) can be used to distinguish sweeteners as salty, sweet, bitter, and sour [41]. For sweet molecules ASV ranges from $(0.51 \times 10^{-6})$ $m^3\cdot kg^{-1}$ to $(0.71 \times 10^{-6})$ $m^3\cdot kg^{-1}$. The ASV for ideal sweet taste lies at centre of the range [42] $(0.618 \times 10^{-6})$ $m^3\cdot kg^{-1}$. From Table 3, it is observed that ASV of solutions studied ranges from $(0.569 \times 10^{-6})$ $m^3\cdot kg^{-1}$ to $(0.626 \times 10^{-6})$ $m^3\cdot kg^{-1}$. Therefore, the solutions studied exhibit sweet taste.

4. Conclusions

Density of Na-cycl in water and in (0.1, 0.3 and 0.5) $m$ glucose and (0.1, 0.3, and 0.5) $m$ sucrose were measured at (298.15, 303.15, 308.15, and 313.15) K. Apparent specific volume (ASV), partial molar volume of transfer ($\Delta_{\text{trs}}V_0^\phi$) at infinite dilution, and the volumetric interaction parameters doublet ($V_{AB}$) and triplet ($V_{ABB}$) were calculated. Positive values of $V_{AB}$ and $\Delta_{\text{trs}}V_0^\phi$ suggest hydrophilic-ionic interactions in Na-cycl aqueous solutions in presence of glucose and sucrose. Solutions studied exhibit sweet taste because their ASV ranges from $(0.569 \times 10^{-6})$ $m^3\cdot kg^{-1}$ to $(0.626 \times 10^{-6})$ $m^3\cdot kg^{-1}$.

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