



## Volumetric properties of aqueous solutions of sodium cyclamate in presence of (0.1, 0.3, and 0.5) m fructose at different temperatures

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### ABSTRACT

Densities of sodium cyclamate in water and (0.1, 0.3, and 0.5) m solutions of fructose have been measured at (298.15, 303.15, 308.15, and 313.15) K. The molality range of sodium cyclamate studied was (0.02 to 0.1) mol.kg<sup>-1</sup>. From experimental values of densities, partial molar volumes, expansion coefficients and Hepler's constants have been calculated. The calculated parameters have been used to get the information regarding solute-solvent interactions and structure making or breaking behaviour of sodium cyclamate. Furthermore, the apparent specific molar volumes of sodium cyclamate in water and (0.1, 0.3, and 0.5) m fructose have been calculated. These values are used to get the information regarding taste quality of sodium cyclamate solutions.

**Keywords:** Density, partial molar volume, fructose, sodium cyclamate

### INTRODUCTION

Pharmaceutical and food industries need the intense sweeteners that have adequate characteristics for the use including sensory qualities (clean for use, no bitterness, odorless), solubility, low cost, and stability. In food and pharmaceutical industries, blends of the sweeteners are commonly used. By blending the sweetener, the user can take the advantages like lower use level, lowest cost, and improved taste. Water play significant role in the sweet responses. 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. Thermodynamic properties of aqueous solutions of sweeteners have an important role in many fields, for example biological, pharmaceutical and food processing studies. Sodium cyclamate is the intense sweetener, widely used in foods, beverages, and pharmaceuticals [1-4]. Susan Schiffman [5] demonstrated the blends of some sugars with sodium cyclamate.

Temperature and concentration dependence of density has been proved as one of the most appropriate methods for the study of solute-solvent and solute-solute interactions. From literature survey, it is revealed that there are no reports on density study of aqueous solutions of sodium cyclamate in presence fructose.

The paper reports the density study of the aqueous solutions of sodium cyclamate in presence of fructose at (298.15, 303.15, 308.15, and 313.15) K. The partial molar volumes, expansion coefficients and Hepler's constants have been calculated from experimental values of densities. Results are interpreted in terms of solute-solvent interactions, structure making/breaking behaviour of sodium cyclamate, and taste qualities of sodium cyclamate in presence of fructose.

### MATERIALS AND METHODS

Sodium cyclamate (Merck, Purity  $\geq 99.0$ ) and fructose (Merck, Purity  $\geq 99.0$ ) were used without further purification for this study. The aqueous solutions 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  $5.8 \times 10^{-2} \text{ kg m}^{-3}$  and 0.006 K, respectively.

### RESULTS AND DISCUSSION

The experimental values of densities of sodium cyclamate solution in water and in (0.1, 0.3, and 0.5) m fructose are presented in Table 1. Figure 1, Figure 2, Figure 3, and Figure 4 show the variation of density with molality. From Table 1, Figure 1, Figure 2, Figure 3, and Figure 4, it is understood that density varies linearly with molality of the solutions. Furthermore, it increases with increase in the concentration of fructose. Apparent molar volume provides the information regarding extent of interaction of the solute molecule. It measures packing efficiency of sweetener

molecules among the solvent molecules. Following equation [10-11] was used for calculation of apparent molar volume.

$$V_{\phi} = \frac{M}{\rho} - \frac{(\rho - \rho_0)}{(m\rho\rho_0)} \quad 1$$

where  $M$ ,  $m$ ,  $\rho_0$ , and  $\rho$  are the molar mass of the solute, molality of the solution, density of solvent (water), and the density of the aqueous solution, respectively. The density values of water have been taken from the literature [12] for the calculation of apparent molar volumes. The uncertainty in the apparent molar volume ranges from  $(0.01$  to  $0.005)10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$ . Figure 5, Figure 6, Figure 7, and Figure 8 present the variation of apparent molar volume with square root of molality at different temperatures. It is observed that the apparent molar volume varies linearly with square root of molality.

**Table 1. Densities of sodium cyclamate in water and (0.1, 0.3, and 0.5) m fructose at different temperatures**

$m$ (mol.kg <sup>-1</sup> )	$\rho$ (kg.m <sup>-3</sup> )			
	Na-Cyclamate + water			
	298.15 K	303.15 K	308.15 K	313.15 K
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
Na-Cyclamate + water + 0.1 m fructose				
0.00	1003.98	1002.53	1000.92	999.13
0.02	1005.53	1004.07	1002.44	1000.63
0.04	1007.07	1005.60	1003.95	1002.12
0.06	1008.60	1007.12	1005.45	1003.60
0.08	1010.12	1008.63	1006.94	1005.07
0.10	1011.63	1010.13	1008.42	1006.53
Na- Cyclamate + water + 0.3 m fructose				
0.00	1017.73	1016.22	1014.49	1012.56
0.02	1019.26	1017.74	1015.99	1014.03
0.04	1020.78	1019.25	1017.48	1015.49
0.06	1022.29	1020.75	1018.96	1016.94
0.08	1023.79	1022.24	1020.43	1018.38
0.10	1025.28	1023.72	1021.89	1019.81
Na- Cyclamate + water + 0.5 m fructose				
0.00	1030.56	1028.38	1026.83	1024.79
0.02	1032.07	1029.88	1028.31	1026.24
0.04	1033.57	1031.37	1029.78	1027.68
0.06	1035.06	1032.85	1031.24	1029.11
0.08	1036.54	1034.32	1032.69	1030.53
0.10	1038.01	1035.78	1034.13	1031.94

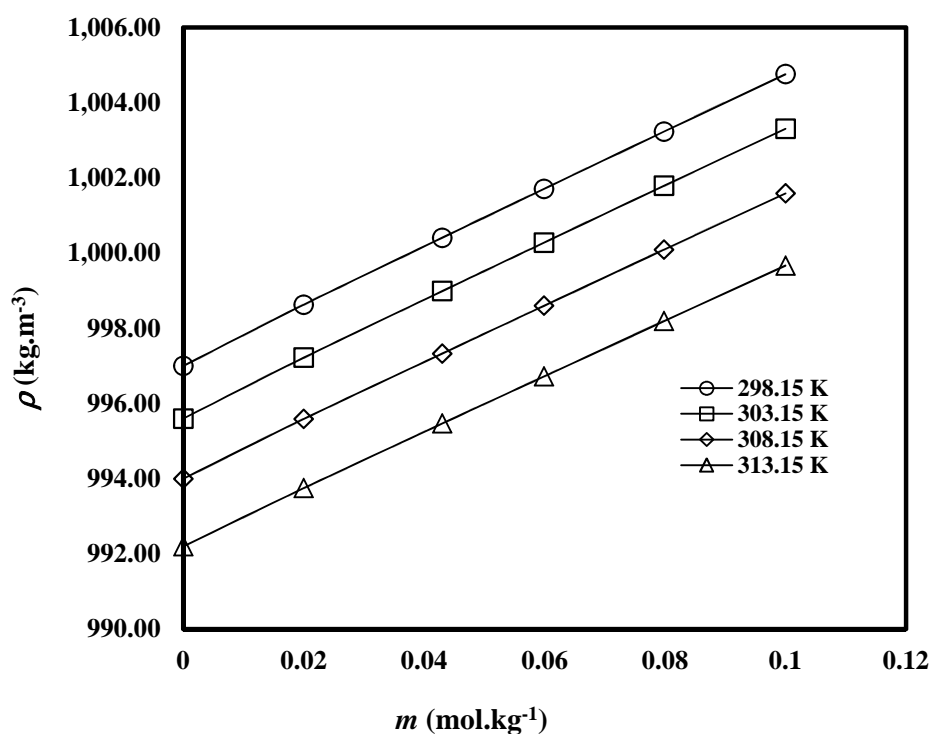


Figure 1. Plot of density ( $\rho$ ) vs molality ( $m$ ) of sodium cyclamate in water at different temperatures ( $T$ )

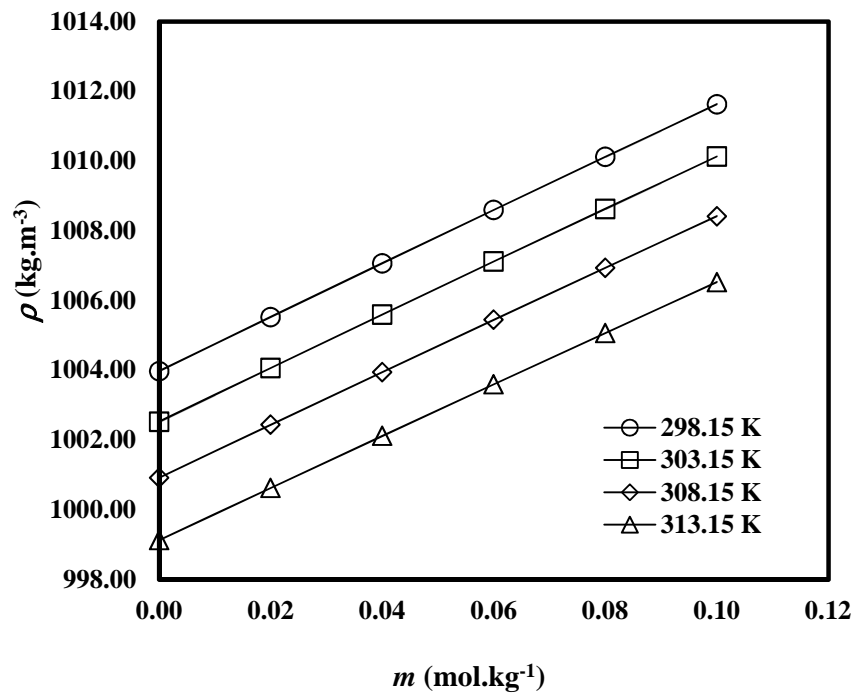


Figure 2. Plot of density ( $\rho$ ) vs molality ( $m$ ) of sodium cyclamate in 0.1  $m$  fructose at different temperatures ( $T$ )

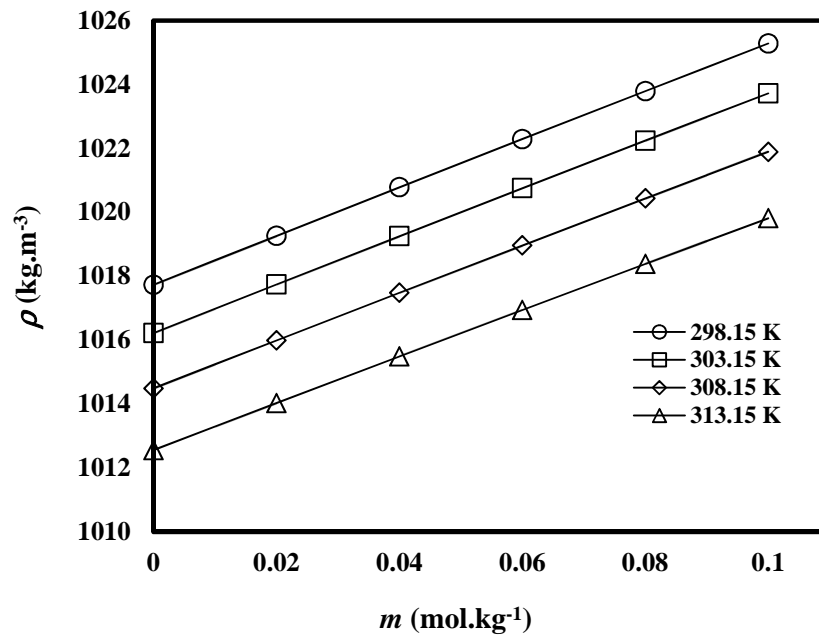


Figure 3. Plot of density ( $\rho$ ) vs molality ( $m$ ) of sodium cyclamate in 0.3  $m$  fructose at different temperatures ( $T$ )

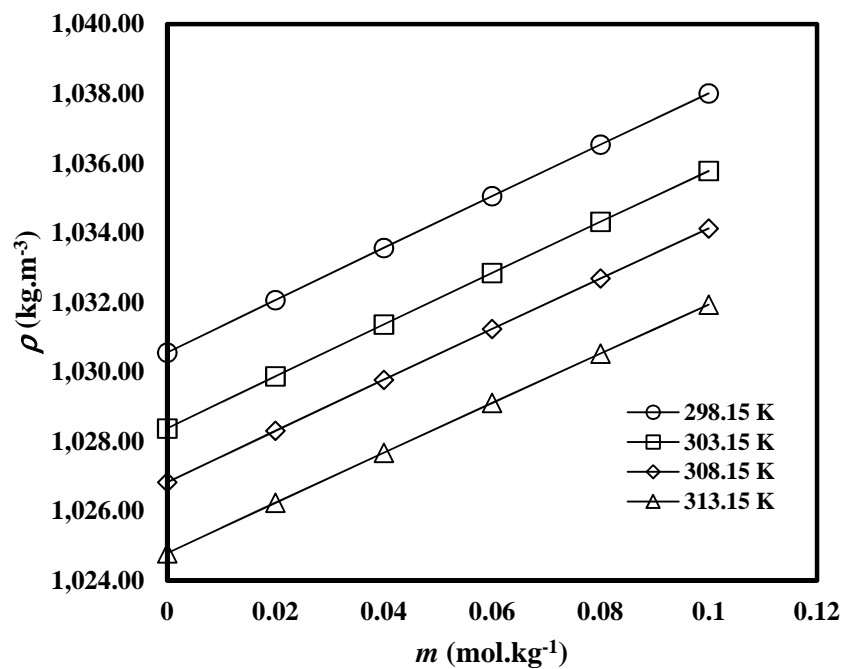


Figure 4. Plot of density ( $\rho$ ) vs molality ( $m$ ) of sodium cyclamate in 0.5  $m$  fructose at different temperatures ( $T$ )

The calculated apparent molar volumes are correlated with molality by the use of the equation [13].

$$V_{\phi} = V_{\phi}^0 + V_s m^{0.5}$$

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where  $V_{\phi}^0$ ,  $V_s$ , and  $m$  are the partial molar volume, solute-solute interaction parameter, and molality. The least square method was used for calculations of  $V_{\phi}^0$  and  $V_s$ . The  $V_{\phi}^0$  value provides valuable information regarding the strength of the solute-solvent interactions [14-16].

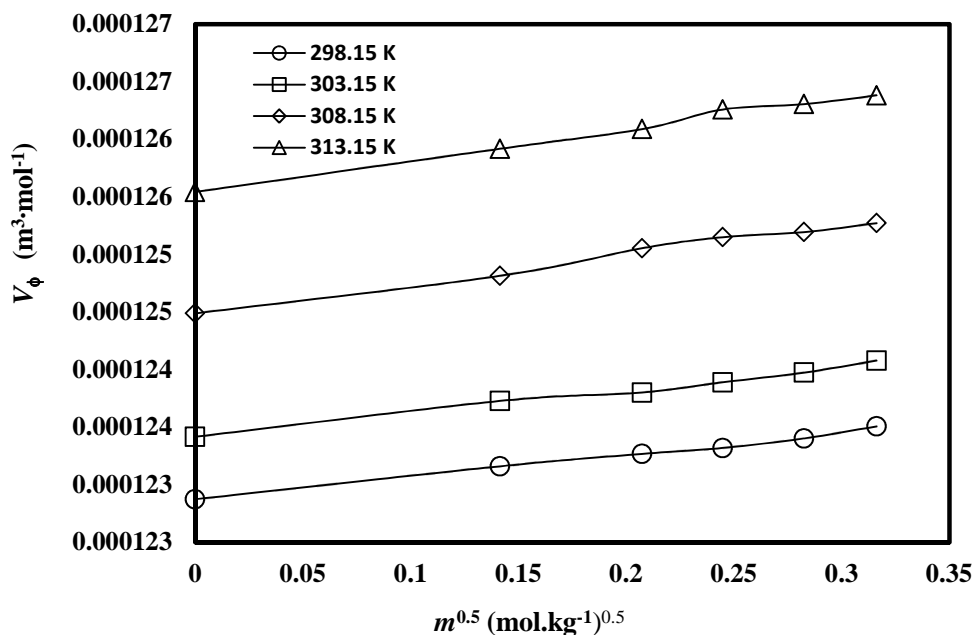


Figure 5. Plot of apparent molar volume ( $V_{\phi}$ ) vs  $m^{0.5}$  of sodium cyclamate in water at different temperatures ( $T$ )

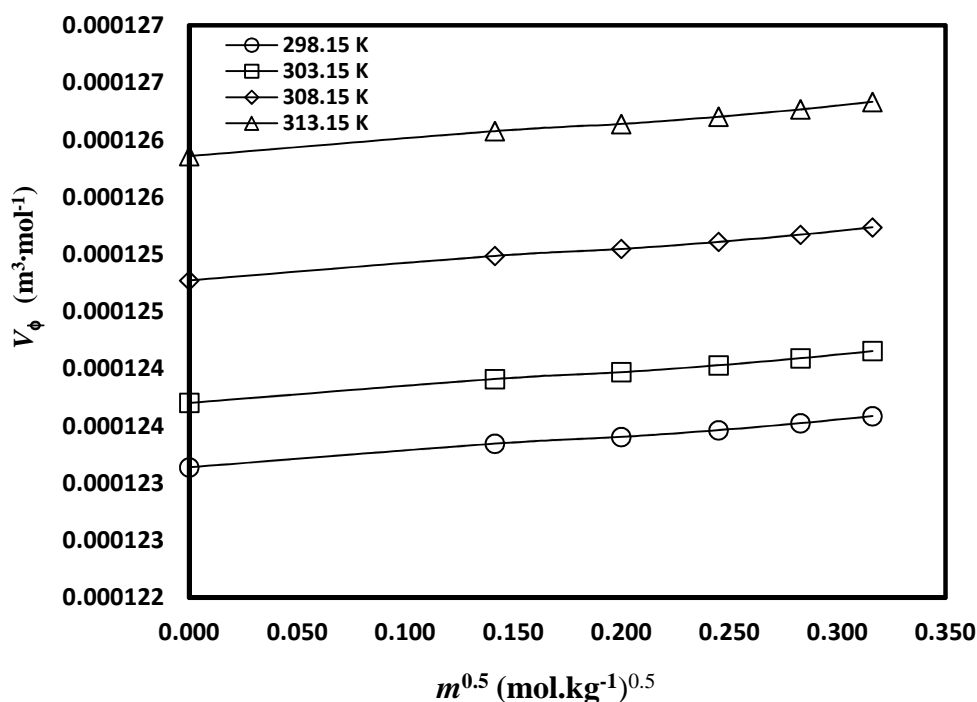


Figure 6. Plot of apparent molar volume ( $V_{\phi}$ ) vs  $m^{0.5}$  of sodium cyclamate in 0.1  $m$  fructose at different temperatures ( $T$ )

Table 2 reports the values of  $V_{\phi}^0$  at different temperatures. The positive values of  $V_{\phi}^0$  indicate strong solute-solvent interactions.  $V_{\phi}^0$  increases with increase in concentration of fructose and also with increase in the temperature.

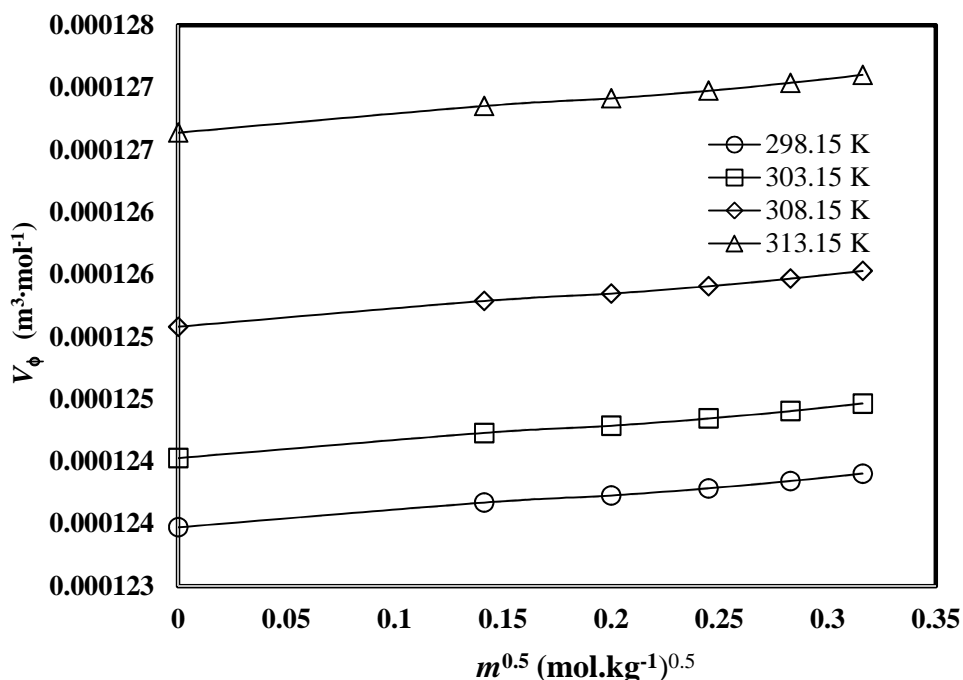


Figure 7. Plot of apparent molar volume ( $V_\phi$ ) vs  $m^{0.5}$  of sodium cyclamate in 0.3  $m$  fructose at different temperatures ( $T$ )

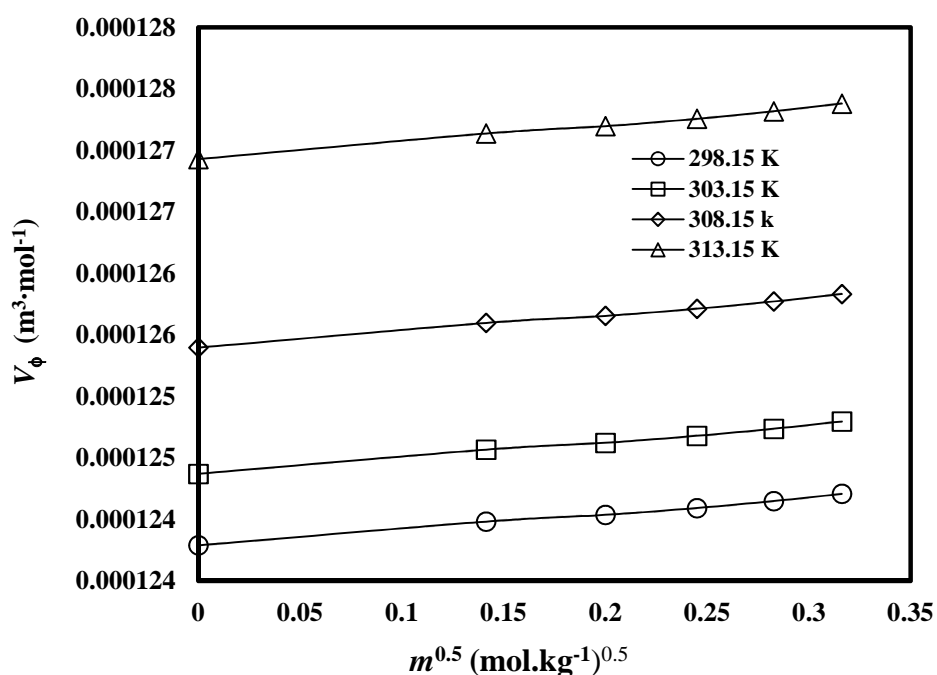


Figure 8. Plot of apparent molar volume ( $V_\phi$ ) vs  $m^{0.5}$  of sodium cyclamate in 0.5  $m$  fructose at different temperatures ( $T$ )

The increase in the  $V_\phi^0$  with increase in the concentration of fructose is due to the strong solute-co-solute interactions between sodium cyclamate and fructose.

Following expression can be used to describe the temperature dependence of  $V_\phi^0$ .

$$V_\phi^0 = a_0 + a_1 T + a_2 T^2 \quad 3$$

where  $T$  is the absolute temperature in Kelvin. For the calculations of  $a_0$ ,  $a_1$ , and  $a_2$  values, least squares method was used. Differentiation of expression 3 with respect to temperature gives the partial molar expansion ( $E^\infty$ ).

$$(E^\infty) = (\partial V_\phi^0 / \partial T)_p = a_1 + 2a_2 T \quad 4$$

The values of ( $E^\infty$ ) are included in Table 2. Positive values of ( $E^\infty$ ) suggest the strong solute-solvent interactions.

To get the information regarding structure making/breaking behaviour of the solute the Hepler's constant can be calculated by the use of following expression.

$$(\partial C_p^\infty / \partial P)_T = -T (\partial^2 V_\phi^0 / \partial T^2)_p \quad 5$$

Table 2 complies the values of Heplers constants. The positive values of Hepler's constants suggest structure making behaviour of solute in solution. Sodium cyclamate shows the structure making effect in fructose solutions. From Table 2, it is understood that the structure making effect of sodium cyclamate increases with increase in the temperature.

On the basis of taste quality, parameter apparent specific volume (ASV) can be used to distinguish sweeteners as salty, sweet, bitter, and sour [17]. For sweet molecules ASV ranges from  $0.51 \times 10^{-6} \text{ m}^3 \cdot \text{kg}^{-1}$  to  $0.71 \times 10^{-6} \text{ m}^3 \cdot \text{kg}^{-1}$ . The ASV [18] for ideal sweet taste lies at centre of the range  $0.618 \times 10^{-6} \text{ m}^3 \cdot \text{kg}^{-1}$ . It can be calculated by the use of following equation

Apparent specific volume (ASV) = Partial molar volume / molar mass

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Table 2 reports ASV values of sodium cyclamate in water and in (0.1, 0.3, and 0.5) *m* fructose at (298.15, 303.15, 308.15, and 313.15) K. The experimentally observed ASV values for Sodium cyclamate in water at (298.15, 303.15, 308.15, and 313.15) K. are  $0.610 \text{ m}^3 \cdot \text{kg}^{-1}$  and  $0.619 \text{ m}^3 \cdot \text{kg}^{-1}$ . The reported value [17] of ASV for sodium cyclamate in water at 293.15 K is  $0.605 \text{ m}^3 \cdot \text{kg}^{-1}$ . From Table 2, it is observed that ASV of sodium cyclamate increases with increase in the concentration of fructose. From Table 2, it is observed that ASV of all solutions studied ranges from  $0.569 \times 10^{-6} \text{ m}^3 \cdot \text{kg}^{-1}$  to  $0.626 \times 10^{-6} \text{ m}^3 \cdot \text{kg}^{-1}$ . Therefore, all the solutions studied exhibit sweet taste.

**Table 2. Partial molar volume ( $V_\phi^0$ ), solute-solute interaction parameter ( $V_s$ ), expansion coefficient ( $E^\infty$ ), Hepler's constant  $(\partial^2 V_\phi^0 / \partial T^2)_p$ , and apparent specific volume (ASV)**

Parameter	298.15 K	303.15 K	308.15 K	313.15
<b>Na- cyclamate + water</b>				
$(V_\phi^0) \times 10^6$	122.87	123.42	124.49	125.54
$(V_s) \times 10^6$	1.917	1.999	2.552	2.716
$E^\infty \times 10^7$	0.557	1.614	2.670	3.726
$(\partial^2 V_\phi^0 / \partial T^2)_p$	2.113 $\times 10^8$			
$(ASV) \times 10^6$	0.611	0.613	0.619	0.624
<b>Na- cyclamate + 0.1 m fructose</b>				
$(V_\phi^0) \times 10^6$	123.14	123.70	124.77	125.86
$(V_s) \times 10^6$	1.37	1.39	1.42	1.46
$E^\infty \times 10^7$	0.616	1.632	2.649	3.665
$(\partial^2 V_\phi^0 / \partial T^2)_p$	2.033 $\times 10^8$			
$(ASV) \times 10^6$	0.612	0.615	0.620	0.625
<b>Na- cyclamate + 0.3 m fructose</b>				
$(V_\phi^0) \times 10^6$	123.47	124.03	125.08	126.64
$(V_s) \times 10^6$	1.324	1.343	1.375	1.422
$E^\infty \times 10^7$	0.612	1.609	2.606	3.602
$(\partial^2 V_\phi^0 / \partial T^2)_p$	1.993 $\times 10^8$			
$(ASV) \times 10^6$	0.614	0.616	0.622	0.629
<b>Na- cyclamate + 0.5 m fructose</b>				
$(V_\phi^0) \times 10^6$	123.79	124.37	125.40	126.93
$(V_s) \times 10^6$	1.284	1.307	1.339	1.387
$E^\infty \times 10^7$	0.719	1.607	2.495	3.382
$(\partial^2 V_\phi^0 / \partial T^2)_p$	1.775 $\times 10^8$			
$(ASV) \times 10^6$	0.615	0.618	0.623	0.631

## CONCLUSION

Positive values of  $V_\phi^0$  indicate strong solute-solvent interactions. Positive values of ( $E^\infty$ ) suggest the strong solute-solvent interactions. The positive values of Hepler's constants suggest structure making behaviour sodium cyclamate in solution. All solutions studied exhibit sweet taste because their ASV ranges from  $0.569 \times 10^{-6} \text{ m}^3 \cdot \text{kg}^{-1}$  to  $0.626 \times 10^{-6} \text{ m}^3 \cdot \text{kg}^{-1}$ .

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