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Solution properties and taste behavior of lactose monohydrate in aqueous ascorbic acid solutions at different temperatures: Volumetric and rheological approach

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ABSTRACT

The densities and viscosities of lactose monohydrate in aqueous ascorbic acid solutions with several molal concentrations $m = (0.00\text{–}0.08)\ \text{mol kg}^{-1}$ of ascorbic acid were determined at $T = (298.15\text{–}318.15)$ K and pressure $p = 101$ kPa. Using experimental data apparent molar volume (ϕ_V), standard partial molar volume (ϕ_V^0), the slope (S_V^*), apparent specific volumes (ϕ_{Vsp}), standard isobaric partial molar expansibility (ϕ_E^0) and its temperature dependence $(\partial\phi_E^0/\partial T)_P$, the viscosity *B*-coefficient and solvation number (S_n) were determined. Viscosity *B*-coefficients were further employed to obtain the free energies of activation of viscous flow per mole of the solvents ($\Delta\mu_1^{0\neq}$) and of the solute ($\Delta\mu_2^{0\neq}$). Effects of molality, solute structure and temperature and taste behavior were analyzed in terms of solute-solute and solute-solvent interactions; results revealed that the solutions are characterized predominantly by solute-solvent interactions and lactose monohydrate behaves as a long-range structure maker.

1. Introduction

Lactose is a milk sugar composed of one galactose and one glucose molecule. It has significant role in the biological and food industrial processes (Banipal, Singh, Aggarwal, & Banipal, 2015). Food producers use lactose as filler in bread. As lactose does not have any flavor so using lactose in food does not change its flavor. In food industries it is used as an alternative sweetener. Additionally, as lactose can help to prevent food discoloration it can be used in canned and frozen vegetables. Most of the powdered food products such as soups, meal-replacement supplements and dehydrated potatoes generally contain lactose. Further some non-dairy foods like coffee creamers contain lactose in the state of dry milk solids or whey. In the pharmaceutical industry, lactose is used as a coating or filler in many prescription and over-the-counter drugs including birth control pills, antacids, vitamin pills and throat lozenges because of its excellent compressibility properties (Hwang & Peck, 2001; Zuurman, Riepma, & Bolhuis, 1994). However major problem with the diary product is their short shelf life, *i.e.*, they get expired over a short period of time. To get rid of this problem food additives are often used. *Substances* added for the preservation of food and to enhance its appearance and taste quality are generally known as food additives. Ascorbic acid commonly known as vitamin-C is used as antioxidant food additive (Jang, Zhu, & Ma, 2013). It is mainly applied to food and feed to increase product stability and quality. So ascorbic acid is added to foodstuffs to retain its characteristic aroma, nutrients and other properties during processing or before packing. It is also essential for growth, healthy teeth, gums, bones, skin and blood vessels and aiding the absorption of iron (Roy, Das, & Bhattacharjee, 2010).

Volumetric studies provide information regarding solute-solvent interactions and related structural changes affect the apparent molar volume, which in turn produces corresponding changes in the taste behavior of a solute (Banipal et al., 2015). Apparent specific volume is a parameter with great importance to foods as it gives a clear idea about the taste chemoreception mechanism (Birch, Parke, Siertsema, & Westwell, 1996). The determination of the viscosity data can be used to study the structural property and the solvation of the solute, which in turn determines the stability of the solute in the solution (Seuvre & Mathlouthi, 2010). Although a number of experiments have been made to explain the properties of lactose and ascorbic acid but such a study on the solution properties and taste behavior of lactose monohydrate in aqueous ascorbic acid solutions is not available in the literature (Banipal, Chahal, Hundal, & Banipal, 2010; Banipal, Singh, Banipal, & Singh, 2013; Jang et al., 2013; Pal & Chauhan, 2011; Verissimo, Ribeiro, Ribeiro, Rodrigo, & Esteso, 2014; Zhuo, Liu, Wang, Ren, & Wang, 2006). So the main object of the present work

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is to study the effect of ascorbic acid on the volumetric, rheological and taste properties of lactose monohydrate in aqueous solutions at temperatures from 298.15 to 318.15 K. The results have been interpreted in terms of solute-cosolute and solute-solvent interactions in terms of apparent molar volumes, standard partial molar volumes and viscosity *B*-coefficients and an attempt has been made to study the effect of ascorbic acid on the taste behavior of lactose monohydrate.

2. Experimental

2.1. Materials

Reagent grade lactose monohydrate (CAS: 64044-51-5; Sigma-Aldrich, mass fraction purity >0.980) and A. R grade ascorbic acid (CAS: 50-81-7, Sigma-Aldrich, mass fraction purity >0.990) were used for present study (Table 1). The chemicals were used as such but stored in *vacuo* over anhydrous $CaCl_2$ for several hours before use. Doubly distilled de-ionized water with a specific conductance $< 1 \cdot 10^{-6}$ S cm^{-1} at 298.15 K was used to prepare different aqueous solutions of ascorbic acid. Various mixed solvents were prepared by mass and necessary adjustments were done to achieve exact molal concentrations (m = 0.02, 0.04, 0.06 and 0.08) of ascorbic acid in the mixed solvents. The physical properties of these mixed solvents are given in Supplementary Table S1. Stock solutions of lactose monohydrate in different solvent mixtures were prepared by mass (measured by Mettler, AG 285, Switzerland) and all the experimental solutions were prepared afresh before use by mass dilution and uncertainty in mass measurements was $\pm 1 \cdot 10^{-4}$g. Molalities were converted into molarities by using experimental density data (Shoemaker & Garland, 1967). Uncertainty in molalities of lactose monohydrate in the studied solutions was estimated to be within $\pm 1 \cdot 10^{-4}$ mol kg^{-1}.

2.2. Apparatus and procedure

Densities were measured by means of a vibrating-tube densimeter (DMA 4500 M, Anton Paar). The temperature was kept constant using an automatic built-in Peltier technique with an accuracy of ±0.01 K. Before measuring the solution densities test runs were performed with dry air and doubly distilled degassed water for calibration. The repeatability and accuracy of the densities were claimed to be $\pm 1 \cdot 10^{-5}$g cm^{-3} and $\pm 5 \cdot 10^{-5}$g cm^{-3}, respectively by the vendor (Anton Paar) and the uncertainty in the densities for most of the solutions was found to be ±0.01 kg m^{-3}.

Viscosities were measured with the help of an Ubbelohde type capillary viscometer thoroughly cleaned and dried. Calibration was done using purified methanol and triply distilled, degassed water. The viscometer was filled with experimental solution and it was placed in a thermostatic bath with a temperature stability ±0.01 K. After the attainment of thermal equilibrium the flow times were recorded in a digital stopwatch with a resolution of ±0.01 s. An average of five measurements was taken into account as the final efflux time (Sarkar, Pandit, & Sinha, 2016). The uncertainty in viscosity measurements was within 0.003 mPa s.

The absorption spectra of lactose monohydrate in aqueous ascorbic acid solutions were recorded on Jasco V-530 double beam UV–vis Spectrophotometer, coupled with thermostatic arrangement and maintained at 298.15 K. A quartz cell of 1 cm path length was used for holding the experimental solutions. Spectroscopic grade water was used as the reference solvent for all the absorption measurements. During spectrophotometric measurements, 2 mL of either lactose monohydrate or ascorbic acid solution ($1 \cdot 10^{-4}$mol L^{-1}) was placed in the quartz cell and the absorbance of the solution was measured. Then 40 μL of either ascorbic acid ($1 \cdot 10^{-4}$mol L^{-1}) or lactose monohydrate ($1 \cdot 10^{-4}$mol L^{-1}) solution was added in a stepwise fashion through a micropipette. The absorbance of the experimental solution was measured at each step. A Systronics digital pH meter, calibrated at pH = 4.00, was used to measure pH of the aqueous ascorbic acid solutions at 298.15 K. pH's were 3.12, 3.00, 2.92 and 2.86, respectively for 0.02, 0.04, 0.06 and 0.08 mol kg^{-1} aqueous solutions of ascorbic acid.

3. Results and discussion

The experimental molalities m, densities ρ, viscosities η, and apparent molar volumes ϕ_V of lactose monohydrate in various aqueous ascorbic acid solutions (used as solvents) at the experimental temperatures are reported in Supplementary Table S2.

3.1. Standard partial molar volumes

The experimental determination of partial molar volume involves the careful measurement of the densities of solutions of known concentrations. The calculation is simplified by the use of a related quantity called apparent molar volume (ϕ_V), which is the volume due to added solute per mole in the solution (Krakowiak, 2011). The apparent molar volumes ϕ_V of lactose monohydrate in various aqueous ascorbic acid solutions were obtained from the following relation:

$$\phi_V = \frac{M}{\rho} - \frac{1000(\rho - \rho_1)}{m\rho\rho_1} \quad (1)$$

where M is the molar mass of lactose monohydrate, m is its molality in the solution, ρ_1 and ρ are the densities of the solvent and solution, respectively. Uncertainties in ϕ_V values were within $\pm(0.11 - 0.62) \times 10^{-6}$m^3 mol^{-1}. Table S2 shows that apparent molar volumes ϕ_V increase with increasing temperature and ascorbic acid content in the aqueous solutions under study. Such trends in apparent molar volumes suggest that the interactions between solute and solvent as well as those between solute-solute or solute-cosolute change with solvent compositions and temperature. However, more clear information regarding solute-solute and solute-solvent interactions can be had from limiting apparent molar volumes at infinite dilution or rather in infinitesimal concentration, *i.e.*, standard partial molar volumes ϕ_V^0 of the solute. Due to the linear relationship between ϕ_V and square root of molar concentrations ($\sqrt{m}$) in the studied concentration range of lactose monohydrate at all experimental temperatures, Masson equation

Table 1
Provenance and purity of the chemicals used.

Chemical	Source	Purification method	Mass fraction purity	CAS No	Pub Chem. CID
Lactose monohydrate	Sigma-Aldrich, Germany	Non	0.980	64044-51-5	62223
Ascorbic acid	Sigma-Aldrich, Germany	Non	0.990	50-81-7	54670067