



Effect of protic ionic liquid on the volumetric properties and taste behaviour of sucrose



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ABSTRACT

The volumetric properties and taste behaviour of sucrose in aqueous solutions of a protic ionic liquid (3-hydroxypropylammonium acetate) have been studied at temperatures, $T = (293.15\text{--}318.15)$ K and at atmospheric pressure. Apparent molar volumes, $V_{2,\phi}$, apparent specific volumes, ASV, apparent molar isentropic compressibilities, $K_{s,2,\phi}$, and apparent specific isentropic compressibilities, ASIC, were calculated from measured density, ρ and speed of sound, u data. Partial molar volumes, V_2^* , and partial molar isentropic compressibilities, $K_{s,2}^*$ at infinite dilution, transfer parameters ($\Delta_T V_2^*$ and $\Delta_T K_{s,2}^*$), expansion coefficients, $[(\partial V_2^*/\partial T)_P]$ and $(\partial^2 V_2^*/\partial T^2)_P$, interaction coefficients, (Y_{AB} and Y_{ABB}) and hydration numbers, N_w , were also evaluated and discussed in terms of solute–cosolute interactions. Further, the effect of protic ionic liquid on the taste behaviour of sucrose has been discussed from ASV and ASIC parameters, as these parameters, which are sensitive to solvation behaviour of solute, are divided into four basic taste qualities occupying certain ranges.

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1. Introduction

Saccharides and their derivatives play an important role in various aspects of chemistry, biochemistry and biotechnology. They act as a source of building blocks for cofactors or biomolecules, and are used in the design of biocompatible and biodegradable materials (Ma, Sun, Chen, Zhang, & Zhu, 2014; Ramesh & Tharanathan, 2003). Saccharides are also used in pharmaceuticals, foods, and biomedical applications (Bordat, Lebrét, Demaret, Affouard, & Descamps, 2004; Ribeiro et al., 2011). Binary and ternary aqueous solutions containing saccharides (e.g., sucrose, glucose, and fructose) and additives (ethanol, glycerol, salts, etc.) have been widely used as a suitable immersion media for freezing fruits (Banipal, Singh, & Banipal, 2010; Pincu, Brauver, Gerber, & Buch, 2010). The thermochemical methods of energy production from sucrose biomass are gaining much interest and recently the feasibility of hydrogen production from the catalytic reforming of sucrose biomass has been reported (Tanksale, Wong, Beltramini, & Lu, 2007).

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Ionic liquids (ILs) are molten salts composed of cations and anions. They are considered as “green benign solvents” promising wide-spread industrial applications, possibly replacing conventional organic solvents (Elyasi, Khalilzadeh, & Karimi-Maleh, 2013; Freire et al., 2008; Singh, Chhotaray, & Gardas, 2014). ILs are also employed for chemical and enzymatic modifications of polysaccharides into useful chemicals and materials, having potential applications in the field of glycobiology, glycochemistry and glycototechnology. As a solvent, ILs are capable of dissolving various polar and non-polar compounds. Recently, imidazolium based ILs were used for cellulose dissolution (Murugesana & Linhardt, 2005; Swatloski, Spear, Holbrey, & Rogers, 2002; Zhang, Wu, Zhang, & He, 2005). Further, mixed solvent systems containing ILs are regarded as more efficient solvent for solubilising cellulose, as these solvents disrupt the hydrogen bonding network present in cellulose.

The application of ILs in carbohydrate chemistry is rapidly growing, both for their functionalization and dissolution, which can bring about attractive new methodologies and enhanced procedures for solution processing of lignocellulosic materials. Most of the published work (Conceicao, Bogel-Lukasik, & Bogel-Lukasik, 2012; Swatloski et al., 2002; Xu, Zhang, Zhao, & Wang, 2013) focus mainly on the dissolution and homogenous modification of cellulose,

starch, lignin, etc. or direct wood dissolution. The physicochemical properties of simple saccharides (mono-, di- or tri-saccharides) in the presence of ILs particularly, protic ionic liquids (PILs) are much less studied (Jin & Chen, 2011; Singh et al., 2014; Wu, Zhang, & Wang, 2009; Wu, Zhang, Wang, & Yang, 2008).

Furthermore, the thermodynamic properties (apparent specific volumes and apparent specific isentropic compressibilities) of different sapid substances (including saccharides) have been studied (Aroulmoji, Hutteau, Mathlouthi, & Rutledge, 2001; Aroulmoji, Mathlouthi, Feruglio, Murano, & Grassi, 2012; Jamal, Khosa, Rashad, Bukhari, & Naz, 2014; Parke, Birch, & Dijk, 1999) in aqueous and mixed aqueous solutions (Aroulmoji, Mathlouthi, & Birch, 2000; Banipal, Singh, Banipal, & Singh, 2013; Seuvre & Mathlouthi, 2010), in order to understand the role of water–solute interactions and the influence of additives on the taste quality of these substances (saccharides). Moreover, the role of water in sweet taste chemoreception has also been reported (Birch, 2002) because the changes in the hydration layer and centre of hydration of solute in the solvent affects the transport of the solute to the taste epithelium (Parke et al., 1999).

The solvation and taste behaviour of sucrose has been reported in the presence of various additives e.g., gluconate salts of sodium, potassium and magnesium (Aroulmoji et al., 2000) and sodium acetate (Banipal et al., 2013). Furthermore, the taste quality of aqueous solutions of salts varies as the anionic part of salt is changed (“anion effects”) (Delwiche, Halpern, & Desimone, 1999). So, in the present work, 3-hydroxypropylammonium acetate (3-HPAAc) was used as an additive to understand the effect of its cationic and anionic part on the volumetric properties of sucrose and its plausible taste effects. The choice of solute (sucrose) was based on the fact that it has been used as a reference standard for sweet substances (Aroulmoji et al., 2000, 2001). The volumetric properties of sucrose in water and in m_B (molality of 3-HPAAc) = (0.10, 0.20, 0.30, and 0.40) mol kg⁻¹ aqueous 3-HPAAc solutions have been studied at temperatures, $T = (293.15–318.15)$ K, with 5 K interval and at atmospheric pressure. Various parameters such as partial molar expansion coefficients $[(\partial V_2/\partial T)_P]$ and second derivatives $(\partial^2 V_2/\partial T^2)_P$, interaction coefficients (Y_{AB} and Y_{ABB}), and hydration numbers, N_w were also evaluated and analysed in terms of solute–solute and solute–solvent interactions occurring between IL and sucrose. The taste behaviour of sucrose in presence of IL has been studied on the basis of range covered by the apparent specific volume, ASV and apparent specific isentropic compressibility, ASIC parameters.

2. Materials and methods

2.1. Materials

Sucrose (CAS No. 50-99-7, 98%) was purchased from Finar Chemical Ltd., India. Acetic acid (CAS No. 64-18-6, ≥95%) and 3-amino-1-propanol (CAS No. 156-87-6, 99%) were purchased from Sigma Aldrich Chemical Co. Sucrose was used as such without further purification after drying in a vacuum desiccator at room temperature for 48 h. The 3-amino-1-propanol and acetic acid were also used without further purification.

2.2. Synthesis of 3-hydroxypropylammonium acetate

3-Hydroxypropylammonium acetate was synthesised (Chhotaray & Gardas, 2014) by neutralization of equimolar 3-amino-1-propanol with acetic acid. The reaction was carried out with constant stirring, in a two necked round bottom flask fitted with a dropping funnel (containing acetic acid) and condenser. Acetic acid was added drop wise into 3-amino-1-propanol at a temperature below 283.15 K. On complete addition of acid, the

resultant mixture was continuously stirred for next 24 h at room temperature. The resulting viscous liquid was dried under high vacuum for 2 days at room temperature, so as to remove excess of reactant (if any) and moisture. The newly synthesised protic ionic liquid was then stored under nitrogen atmosphere.

2.3. Apparatus and procedure

The simultaneous measurement of density, ρ , and speeds of sound, u , of sucrose in water and in $m_B = (0.10, 0.20, 0.30, \text{ and } 0.40)$ mol kg⁻¹ aqueous solutions of 3-HPAAc at temperatures, $T = (293.15, 298.15, 303.15, 308.15, 313.15 \text{ and } 318.15)$ K was carried out by using vibrating-tube digital density, and a sound velocity meter (Anton Paar, DSA 5000 M). The density and sound velocity cells were temperature controlled by a built-in Peltier thermostat (PT-100) having an accuracy of ±0.01 K. Calibration of the instrument was done by millipore quality, freshly degassed water and dry air at atmospheric pressure. The uncertainty in the measurement of density is $\pm 7 \times 10^{-3}$ kg m⁻³ and for speed of sound is ± 0.5 m s⁻¹.

Fresh solutions were made on mass basis in air tight glass vials by using a Sartorius balance (Model CPA225D), having a precision of ±0.01 mg. Millipore quality freshly degassed water was used for making solutions.

3. Results and discussion

3.1. Characterisation of 3-HPAAc

The proton NMR of 3-HPAAc was recorded on Bruker Avance 500 MHz spectrometer using deuterated DMSO as solvent. ¹H NMR: $\delta = 4.1$ ppm (broad, 4H, OH and N_{H3}N), $\delta = 3.45$ ppm (t, 2H, CH₂-N), $\delta = 2.72$ ppm (t, 2H, CH₂-O), $\delta = 1.71$ ppm (s, 3H, CH₃-C), $\delta = 1.60$ ppm (qn, 2H, CH₂-C). IR spectra were recorded on JASCO FT/IR-4100 spectrometer. The characteristic ammonium peak, ν (N-H) and ν (O-H) stretching vibration appeared in the range of (3600–2600) cm⁻¹. The characteristic carbonyl, ν (C=O) stretching and δ (N-H) plane bending vibrations broad band appeared around 1600 cm⁻¹. An Analab Karl Fischer Titrator (Micro AquaCal 100) was used for determining water content of synthesised 3-HPAAc and was found to be ≈5538 ppm.

3.2. Volumetric properties and taste behaviour

The apparent molar volumes, $V_{2,\phi}$ and apparent molar isentropic compressibilities, $K_{s,2,\phi}$ of sucrose in water and in (0.10, 0.20, 0.30, and 0.40) mol kg⁻¹ aqueous solutions of 3-hydroxypropylammonium acetate (cosolute) at different temperatures were calculated from the experimentally determined density, ρ , and speed of sound, u , data by using the following Eqs. (1) and (2):

$$V_{2,\phi} = [M/\rho] - [(\rho - \rho_0)/(m \cdot \rho \cdot \rho_0)], \quad (1)$$

$$K_{s,2,\phi} = (\kappa_s \cdot M/\rho) - [(\kappa_s^\circ \cdot \rho - \kappa_s \cdot \rho_0)/(m \cdot \rho \cdot \rho_0)] \quad (2)$$

where ρ and ρ_0 are the densities of solution and solvent (water or water + 3-HPAAc), M and m are the molar mass and molality of the solute (sucrose), κ_s and κ_s° are the isentropic compressibilities of solution and solvent, respectively. The isentropic compressibilities, κ_s were evaluated by using the Eq. (3):

$$\kappa_s = 1/u^2 \cdot \rho. \quad (3)$$

The ρ , $V_{2,\phi}$, u and $K_{s,2,\phi}$ values in water and in aqueous solutions of PIL at different temperatures are given in Supporting Information (Table S1). The standard uncertainty in $V_{2,\phi}$ values resulting from various experimentally measured quantities [$u(m) = 5.2 \cdot 10^{-6}$ mol kg⁻¹, $u(\rho) = 7.0 \cdot 10^{-3}$ kg m⁻³, $u(T) = 0.01$ K] ranges from (0.108

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