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Density and viscosity of sorbitol/maltitol in L-ascorbic acid aqueous solutions at $T = (293.15$ to $323.15)$ K

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ABSTRACT

Densities, ρ, and viscosities, η, of ternary solutions (sorbitol/maltitol + L-ascorbic acid + water) and binary solutions (sorbitol/maltitol + water) were measured at $T = (293.15$ to $323.15)$ K under atmospheric pressure. Guimarães equation and extended Jones–Dole equation were applied to correlate the densities and viscosities of the solutions, respectively. The apparent molar volume (V_φ), limiting partial molar volume (V_φ^0) and limiting partial molar volume of transfer ($\Delta_{tr}V_\varphi^0$) were calculated by densities. The free energy of activation per mole of solvent ($\Delta\mu_1^{0\neq}$) and free energy of activation per mole of solute ($\Delta\mu_2^{0\neq}$) were calculated through the obtained viscosity B-coefficients, respectively. These significant parameters are helpful to understand the interactions in sugar alcohols + L-ascorbic acid + water ternary solutions and also to study the substance conversion and transfer in living liquid system.

1. Introduction

Density and viscosity are the basic physical parameters frequently used in chemical engineering, and also the indispensable thermodynamic data for chemical researches and computations involving fluid flow, heat transfer and mass transfer [1–3]. The volumetric properties such as the partial molar volume and limiting partial molar volume of transfer, which can be calculated through density, are of crucial significance in studying phase equilibrium, equation of state and molecular interactions, as well as in designing reactor. Viscosity can be used to obtain the viscosity B-coefficients and the free energy of activation and so on which are important to analyze the transport properties and flowing feature of fluids [4,5]. Therefore density and viscosity play a very important role in further researches of solution thermodynamic and transport properties [6,7].

Along with the constantly improvement of people's living standards, there emerges much more requirements for the quality of functional or healthy food including confectionary, beverages and diet drinks. Increasing interests have been focusing on the non-sugar and low calorie products. Sugar alcohols, a new kind of functional food, owing to their desirable properties such as good taste, low calorie, no tooth decay, and suitability for people with diabetes, are applied in many aspects like food, pharmacy, cosmetics, and chemical engineering [8–10] and will occupy a wide market in the years to come. Since the solution thermodynamic properties can provide basic information for food, pharmacy, and chemical engineering, there have been extensive researches about the sugar alcohols solutions [11–13]. But for ternary solution of sugar alcohols and the bioactive compounds in water, it still needs further systematic investigations.

L-ascorbic acid, also called vitamin C (VC for short), is a ubiquitous and indispensable compound to biological system [14]. It is a kind of polyhydroxy compound with antioxidant property which could protect our body from radicals [15]. As is known, sugar alcohols themselves don't equip with the antioxidant property, however they could enhance the antioxidant ability of antioxidants to some extent. Since L-ascorbic acid and sugar alcohols cannot be synthesized in living system, there must exists molecular diffusion and interaction between them after entering the body through food or cosmetics. Therefore, it is essential to study the thermodynamic properties of sugar alcohols in L-ascorbic acid aqueous solutions. As the continuation to our thermodynamic study of sugar alcohols, we report here the densities and viscosities of sorbitol/maltitol in L-ascorbic acid aqueous solutions at $T = (293.15$ to $323.15)$ K. Through the experimental data, some volumetric and viscometric properties were calculated.

2. Experimental section

L-Ascorbic acid is biochemical reagent-grade. (2S,3R,4R,5R)-Hexane-1,2,3,4,5,6-hexol (sorbitol) and 2-(hydroxymet hyl)-6-[4,5,6-trihydroxy-2-(hydroxymethyl)oxan-3-yl] oxyoxane-3,4,5-triol (maltitol) belong to food additives. More details about the L-ascorbic acid, sorbitol and maltitol were given in Table 1. Prior to measurement, these chemicals were thoroughly dried over silica gel in a vacuum desiccator at 313.15 K for more than 24 h. Double-distilled water was used in the experiments. All the ternary and binary solutions were prepared by mass afresh before use at room temperature using an analytical balance (model FA2204B) with uncertainty of ±0.0001 g. The molality range of L-ascorbic acid was from

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Table 1
The specification of studied chemicals.

Components	CAS no.	$M/(g \cdot mol^{-1})$	Purity[a] (mass %)	Manufacturer
L-Ascorbic acid	50-81-7	176.12	≥0.997	Tianjin Jiangtian Chemical Reagent Co., Ltd.
Sorbitol	50-70-4	182.17	≥0.99	Zhengzhou Jianda Chemicals Inc.
Maltitol	585-88-6	344.31	≥0.99	Zhengzhou Jianda Chemicals Inc.

[a] Declared by the supplier.

0.0 to 0.4 $mol \cdot kg^{-1}$, while the molality ranges of erythritol/xylitol/mannitol aqueous solutions were from 0.0 to 1.0 $mol \cdot kg^{-1}$. All of the solutions were degassed with ultrasonic waves. The uncertainty of molality for all solutions is ± 0.0001 $mol \cdot kg^{-1}$.

The densities were measured through a sophisticated vibrating tube density meter DMA 4500 (Anton Paar, Austria) with an uncertainty of $\pm 5.0 \times 10^{-5}$ $g \cdot cm^{-3}$. Before each series of measurement, the apparatus was calibrated with doubly distilled water and dry air at atmospheric pressure. The temperature stability of the measuring cell was automatically controlled within ± 0.03 K with the help of two integrated Pt100 platinum thermometers together with built-in peltier elements. Triplicate measurements of each data were conducted to obtain the average value of density.

Viscosities of sorbitol/maltitol in L-ascorbic acid aqueous solutions were measured using an iVisc capillary viscometer (LAUDA, Germany). A thoroughly cleaned and dried viscometer filled with experimental solutions was placed exactly vertical in a glass sided water thermostat controlled to ± 0.05 K. After the thermal equilibrium, computer software connected to the viscometer recorded automatically the flow time of liquids with an uncertainty of ± 0.01 s. The flow time for each sample at a specified temperature was taken an average of at least four readings within the deviation of ± 0.2 s. Since all flow times were greater than 100 s and the capillary diameter (0.5 mm) was far less than its length (120 mm), the kinetic energy and the end corrections were found to be negligible. The viscosity η of the solutions could be calculated from the following equation [16]:

$$\eta/\eta_w = \rho t/\rho_w t_w \tag{1}$$

where η, ρ, t and η_w, ρ_w, t_w are viscosities, densities, and flow times of the solutions and pure water, respectively. The viscosity of pure water was obtained from Lange's Handbook of Chemistry [17]. The accuracy of experimental viscosities was ± 0.0003 $mPa \cdot s$.

3. Results and discussion

3.1. Volumetric properties

The experimental densities of sorbitol/maltitol + L-ascorbic acid + water ternary solutions and sorbitol/maltitol + water binary solutions at $T = (293.15, 303.15, 313.15$ and $323.15)$ K were listed in Table 2. The measured densities of binary solutions showed a great agreement with the data in the literature [18].

From Table 2, the densities of ternary and binary solutions decrease with the increase of temperature whereas increase with the increase of the molality of sorbitol/maltitol. Furthermore, from Fig. 1, the densities of solutions show a very good linear relationship with the molar concentration of sorbitol/maltitol. It could be observed that for the same solute, the densities increase monotonously with the molality of L-ascorbic acid. When the molality of L-ascorbic acid increases, the molecule distance gets shortened, that leads to the increase of the densities of both sorbitol and maltitol solutions. However, for the same molality of L-ascorbic acid, the densities of maltitol + L-ascorbic acid + water solutions are larger compared to sorbitol + L-ascorbic acid + water solutions (see Fig. 1). It's known that the larger the molar weight of solute is, the larger the density is. That's why maltitol solutions show a larger density than sorbitol solutions.

Fig. 1 vividly presented the relationship of density of sorbitol/maltitol versus temperature and solute molality. Thus the Guimarães equation was applied to correlate the experimental density data [19].

$$\rho = A_1 + A_2 T + A_3 C \tag{2}$$

where A_1, A_2, and A_3 are the empirical constants, T is the temperature, and C is the molar concentration of sorbitol/maltitol in L-ascorbic acid aqueous solutions at 293.15 K. The fitting parameters, A_1, A_2, and A_3 were shown in Table 3 alongside with the values of standard deviation (SD) and the average deviation (AD).

The standard deviation (SD) and the average deviation (AD) were calculated as follows:

$$\mathrm{SD} = \left[\sum_{i=1}^{n}\left(y_{\mathrm{exp},i} - y_{\mathrm{cal},i}\right)^2/(n-m)\right]^{1/2} \tag{3}$$

$$\mathrm{AD} = \frac{1}{n}\sum_{i=1}^{n}\left|\left(y_{\mathrm{exp},i} - y_{\mathrm{cal},i}\right)/y_{\mathrm{exp},i}\right| \tag{4}$$

where n is the total number of experimental data and m is the number of parameters. $y_{\mathrm{exp},i}$ and $y_{\mathrm{cal},i}$ refer to the experimental values and the calculated values, respectively. From Table 3, the maximum values of AD and SD were 0.088% and 0.0012 $g \cdot cm^{-3}$, respectively.

The apparent molar volume (V_φ) could be calculated by the experimental densities according to the following equation [20,21]:

$$V_\varphi = M/\rho - 1000(\rho - \rho_0)/m\rho\rho_0 \tag{5}$$

where M is the molar weight of the solute, m is the molality of sorbitol/maltitol in solvent, ρ and ρ_0 are the densities of the solution and solvent, respectively. The calculated apparent molar volumes, V_φ, were also included in Table 2.

The apparent molar volumes, V_φ, was found to have a good linear relationship with the molar concentration of the solute. Consequently, the limiting partial molar volume, V_φ^0 was obtained through the following equation:

$$V_\varphi = V_\varphi^0 + S_v C \tag{6}$$

where S_v is the slope of the straight line of V_φ to C. The values of V_φ^0 were obtained by the least-square regression analysis and displayed in Table 4. To further study the transfer properties of the ternary system, limiting partial molar volumes of transfer, $\Delta_{tr}V_\varphi^0$ at infinite dilution of sorbitol/maltitol from pure water to L-ascorbic acid aqueous solutions were calculated as follows:

$$\Delta_{tr}V_\varphi^0 = V_\varphi^0[\text{L-ascorbic acid + water}] - V_\varphi^0[\text{water}] \tag{7}$$

The calculated partial molar volumes of transfer, $\Delta_{tr}V_\varphi^0$ were also summarized in Table 4. And the $\Delta_{tr}V_\varphi^0$ of sorbitol/maltitol in L-ascorbic acid aqueous solutions versus the molality of L-ascorbic acid was plotted in Fig. 2. The uncertainty of V_φ^0 is ± 0.03 $cm^3 \cdot mol^{-1}$ for