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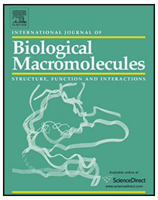

Influence of temperature, mono- and divalent cations on dilute solution properties of sage seed gum

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ABSTRACT

The functional properties of food hydrocolloids are remarkably affected by the quality of solvent/cosolutes and temperature in a food system. In this paper, dilute solution properties of sage seed gum (SSG) as a function of salt type (NaCl, KCl, $MgCl_2$ and $CaCl_2$), salt concentration (10, 50, 100 and 200 mM) and temperature (25, 45 and 65 °C) were investigated. Among various models, Higiro model showed a higher performance to determine intrinsic viscosity of SSG at all temperatures and cosolutes. From 25 to 65 °C for every 20 °C rise in temperature, intrinsic viscosity decreased about 18.99 and 63.86%, respectively. The divalent cations had more reduction effect on intrinsic viscosity than monovalet cations. More flexibility of SSG in monovalent salts solutions compared with divalent ones was observed. A high value for activation energy (2.53×10^7 J/kg mol) and chain flexibility (3046.45) of SSG was obtained, which was higher than many hydrocolloids. The shape factor of SSG macromolecules at 25–65 °C was an oblate or prolate and for all used cosolutes, the shape was roughly found to be ellipsoidal.

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1. Introduction

Hydrocolloids are widely used in food systems to enhance their quality by affecting the physical and organoleptic attributes as thickening and gelling agents, stabilizers and texture modifiers [1]. Nowadays, the demand for hydrocolloids from plants (e.g. plant cell walls, tree exudates, seeds, seaweeds) is greater than those from animals (hyaluronan, chitin, chondroitin sulphate) because of more benefits and friendly image toward consumers [2].

There are some Iranian endemic plants that their seeds can be used as a new hydrocolloid sources for food and pharmaceutical systems [3]. *Salvia* contains about 700–900 species of herbaceous and woody plants of the mint family, Lamiaceae. About 200 species of this genus grow in some provinces of Iran [4]. Wild sage (*Salvia verbenaca*) is one these endemic plants and its seed (*Salvia macrosiphon*) mucilage has a potential alternative to some commercial gums [5]. The extraction conditions for sage seed mucilage was optimized using response surface methodology [6]. The steady shear flow behavior of sage seed gum (SSG) demonstrated that it has strong shear thinning characteristics at different temperature and concentrations which is comparable to xanthan [3]. Structural characteristics of SSG revealed that mannose (61.50%) and galactose (33.15%) are the main carbohydrate fractions, but glucose (2.78%), arabinose (1.41%) and rhamnose (1.17%) are insignificant ones [4]. It was concluded that SSG polysaccharide is a galactomannan with a 1.78–1.93:1 mannose/galactose ratio. It was recently found that SSG enhanced the stability of oil-in-water emulsion containing whey protein concentrate (5–15% w/v) by modification of the flow behavior and induction of intermolecular interactions [7]. The dynamic rheological properties of SSG at all concentration within the range of 0.5–2% (w/w) have shown weak gel behavior like xanthan and psyllium gums that was more elastic compared with other galactomannans [2].

The viscosity behavior of macromolecular substances in a solution is one of the most frequently used approaches to determine its specification. In dilute solution, it is assumed that macromolecule chains are separated without intermolecular interactions [8,9]. Investigation of molecular properties such as macromolecule-solvent interaction, molecular weight, molecular shape and conformation seems to be useful to understand and control the behavior of a hydrocolloid in dilute solution at different conditions.

Intrinsic viscosity $[\eta]$ is a measure of the capability of a polymer in solution to increase the viscosity of the solution. Much information for fundamental properties of a solute and its interaction with

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a specific solvent can be obtained by determination of intrinsic viscosity [10,11]. There are some linear (Huggins and Kraemer) and non-linear (Tanglertpaibul & Rao and Higiro) equations to determine intrinsic viscosity [12–15]. Razavi et al. [9] found that the Tanglertpaibul and Rao and Higiro models were the best ones for intrinsic viscosity determination of SSG at different temperatures (20, 30 and 40 °C) and salts concentrations (0.5, 20 and 50 mM), respectively. Behrouzian et al. [16] also showed that the Tanglertpaibul and Rao model was the best one to determine intrinsic viscosity of cress seed gum dilute solutions. Since intrinsic viscosity affect by changes in solvent properties like ionic strength, then study of its change could be a good instrument to track the changes in molecular attributes [1,9,11,16]. Literature shows that higher ionic strength and temperature lead to decrease in solution viscosity [9,11,15,17–20]. For instance, Behrouzian et al. [16] found that increase in concentration of NaCl (25–100 mM), $CaCl_2$ (5–15 mM), sucrose (up to 30%) and lactose (up to 5%) caused a reduction of intrinsic viscosity. Razavi et al. [9] also showed that $CaCl_2$ had a more pronounced effect on intrinsic viscosity of SSG than NaCl. The similar results were reported by Mohammad Amini and Razavi [11] for Balangu (*Lallemantia royleana*) seed gum. Furthermore, they reported that the effect of temperature on intrinsic viscosity of Balangu seed gun was significant, so that each 10 °C temperature increase from 20 °C to 50 °C caused a decrease in intrinsic viscosity approximately 15.12%, 24.10%, and 30.84%, respectively. In salt-free situation or low salt concentration, the expansion of polymer chain occurs because of interachain electrostatic repulsion. At higher salt concentration, screening of the charge occurs and the electrostatic interactions diminish and the chain conformation becomes more compact.

The aim of the present study was to develop our previous works about SSG. For this purpose, the influence of different mono- and divalent ions (NaCl, KCl, $MgCl_2$ and $CaCl_2$) and temperatures (25, 45 and 65 °C) on intrinsic viscosity of SSG was investigated. In addition, some molecular parameters of SSG including conformation, relative stiffness, persistence length, chain flexibility, voluminosity, shape factor, coil radius and volume were determined and their relation with the intrinsic viscosity of SSG were discussed.

2. Materials and methods

2.1. Preparation of materials

The sage seeds were purchased from a local market in Mashhad, Iran. Several cleaning steps manually were used to ensure removing all of undesirable stuffs. Extraction of the sage seed gum was performed using the method described by Bostan et al. [6]. Finally, the SSG was subjected to a force convection oven (Model 4567, Kimya Pars Com., Iran) overnight at 70 °C prior to be milled and sieved using a mesh 18 sifter. The SSG powder contained, on averagely, 6.72% moisture, 0.85% lipid, 8.17% ash, 2.84% protein, 1.67% crude fiber, and 79.75% carbohydrate.

2.2. Preparation of solutions

Stock SSG solutions were prepared at concentration of 0.25% by mixing 0.1 g (d.b.%) of SSG powder in 40 ml of de-ionized water and a range of NaCl, KCl, $MgCl_2$ and $CaCl_2$ concentrations (10, 50, 100 and 200 mM) at room temperature. For this reason, the prepared gum powder was gradually added into the vortex formed due to whirl of magnetic stirrer. After that, the attained deionized water, NaCl, KCl, $MgCl_2$ and $CaCl_2$-SSG suspensions were mixed using a roller mixer (Hematology Cell Mixer; Pars, Iran) for 15 min without heating and was retained 24 h for complete hydration. In order to discard any insoluble residues, all of the prepared SSG solutions were centrifuged at 10,000 × g. Eventually, the supernatant parts of the centrifuged samples were filtered via a methyl-cellulose membrane with a pore size of 0.45 μm. It should be noted that the dilute solutions of SSG were obtained by diluting the filtrated part (0.25%, d.b.).

2.3. Density measurements

Density of solvent (ρ_0) and solution (ρ) were obtained by means of a standard 25 ml pycnometer. The temperature for density measurements was similar to that for viscosity measurements. The partial specific volume ($\bar{v}$) of SSG solution in deionized water was determined through density increment ($\Delta\rho$) vs. concentration curve at 25 °C.

2.4. Viscosity measurement

Dilute SSG solutions were made by adding distinct extent of the solvents (de-ionized water, NaCl, KCl, $MgCl_2$ and $CaCl_2$ solutions) to the stock solutions. The viscosity of SSG solutions was measured using a Cannon-Ubbelohde viscometer (Cannon Instruments, USA; viscometer constant, $k = 0.007690\ \mathrm{mm^2/s^2}$) immersed in a paraffin bath to maintain at 25 °C for 15 min. The kinematic viscosity was measured by allowing the solutions to flow due to their gravity through the capillary part of the viscometer. All the measurements were done as triplicates and the average values are reported. Intrinsic viscosity $[\eta]$ can be determined by measuring the viscosity of very low concentration solutions through the calculation of the following viscosities:

$$\eta_{rel} = \frac{\eta}{\eta_s} \tag{1}$$

$$\eta_{sp} = \eta_{rel} - 1 \tag{2}$$

$$[\eta] = \lim_{C\to 0} \frac{\eta_{sp}}{C} \tag{3}$$

where η is the solution viscosity, η_s is the solvent viscosity, η_{rel} is relative viscosity and η_{sp} is the specific viscosity.

There are several developed equations to determine the intrinsic viscosity. According to Huggins model [12] (Eq. (4)), the intrinsic viscosity $[\eta]$ obtained by extrapolating η_{sp}/C data to zero concentration simply through a linear regression:

$$\frac{\eta_{sp}}{C} = [\eta] + k'[\eta]^2 C \tag{4}$$

where k' is the Huggins constant. Kraemer [13] reported that the intrinsic viscosity $[\eta]$ could be obtained by extrapolation of $\ln \eta_{rel}/C$ values to zero concentration (Eq. (5)):

$$\frac{\ln \ \eta_{rel}}{C} = [\eta] + k''[\eta]^2 C \tag{5}$$

where k'' is the Kraemer constant. It is demonstrated that the methods in which the intrinsic viscosity is calculated based on the slopes of plots had higher correlation coefficient and lower standard errors in comparison with those are calculated through intercepts of plots [9,21]. Based on this finding, three equations are shown as follow to determine the intrinsic viscosity of the solutions based on the slope of plots:

Tanglertpaibul and Rao [14]:

$$\eta_{rel} = 1 + [\eta]C \tag{6}$$

Higiro et al. [15]:

$$\eta_{rel} = e^{[\eta]C} \tag{7}$$

$$\eta_{rel} = \frac{1}{1 - [\eta]C} \tag{8}$$

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