



Developing a green and edible film from Cassia gum: The effects of glycerol and sorbitol

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ABSTRACT

The feasibility of preparing a novel edible film based on cassia gum (CG) was studied. The mechanical, barrier, optical, and thermal properties as well as the contact angle were determined to evaluate the effect of glycerol (G), sorbitol (S) in various concentrations on the CG films. The rheology properties of the film-forming solutions were also analyzed. The Fourier transform infrared spectroscopy (FTIR) analysis showed that hydrogen bonds formed between the G or S and CG molecules. The viscosity tests revealed the film-forming solutions had a shear-thinning behavior and the dynamic measurements confirmed that the glycerol-added CG (CG-G) and sorbitol-added CG (CG-S) solutions exhibited classical gel properties. The CG-G films had higher elongation at break (EB) but lower water vapor permeability (WVP) and oxygen permeability (OP) than CG-S ones. Therefore, tensile strength (TS) increased with increasing G (except for 50%) and S concentrations. The TS of CG-G and CG-S films increased from 11.02 ± 0.47 to 18.53 ± 0.40 MPa, and from 12.69 ± 0.59 to 18.20 ± 0.76 MPa, respectively; the corresponding EB increased from 22.27 ± 0.47 to $38.00 \pm 0.59\%$ and from 7.90 ± 0.41 (CG-30S) to $25.33 \pm 0.90\%$, indicating that G or S played as not only a plasticizer but also a cross-linker. An increase in G and S concentrations also made the films more opaque and improved their thermal stability. CG is a promising material for the production of edible packaging films.

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1. Introduction

Packaging plays an important role in reducing food waste because it can improve food preservation and optimize space during handling, shipping, and storage (de Léis et al., 2017). In recent decades, synthetic polymers (plastic) have gradually taken over the market of food packaging for their lower cost, better durability and excellent waterproof performance. However, the degradation of plastic packaging products requires more than 100 years, which has raised issues regarding environmental pollution and food safety, increasing the interest towards the search for new packaging materials that should be biodegradable and environmentally friendly. Biopolymers could be a promising alternative to plastic on account of their biodegradability, renewability and abundance of resources. Polysaccharides (Prakash Maran et al., 2013), proteins (Oliveira et al., 2015) and lipids (Wang et al.,

2014) are the most commonly used biopolymeric materials. In recent years, there have been many reports on the preparation of edible films from biopolymers, such as gum ghatti (Zhang et al., 2016), starch (Wu et al., 2013), gelatin (Vanin et al., 2005), soy protein isolate (Garrido et al., 2014) and carboxymethyl cellulose (Dashipour et al., 2015).

Cassia gum (CG) is a hydrophilic colloid derived from the endosperm of the Cassia plant. The Cassia seed endosperm contains a large number of polysaccharides possessing the molecular structure of galactomannan. The high quality products were obtained by conventionally grinding the Cassia seed endosperm. CG is a long chain polymer composed of 1,4-β-D-mannopyranose units with 1,6 linked α-D-alactopyranose units (Fig. 1), and the galactose to mannose ratio is about 1:5. The molecular weight of CG has been observed to be approximately 100,000–300,000 Da. CG plays a key role as a coagulant in the industry of effluents treatment (Subramonian et al., 2015); moreover, it has been widely used in papermaking (Subramonian et al., 2014) and pharmaceutical industries (Kumar et al., 2016). Owing to its rheological properties (Kapoor et al., 1996) and composition, CG is a potential material for

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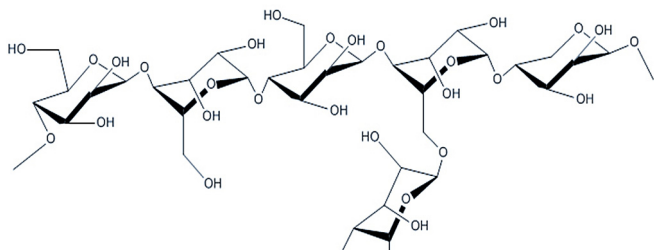


Fig. 1. Chemical structure of CG.

edible films. The production of edible films requires plasticizers to mix with the polysaccharides to compose the films' networks; however, to different degrees, the plasticizer changes the mechanical properties, solubility in water and barrier ability of the films (María A García and Noemí E. Zaritzky, 2000). Plasticizers could improve the flexibility of edible films via decreasing intermolecular forces and increasing the mobility of polymer chains (Zhang et al., 2016). Glycerol, sorbitol, propylene glycol and other polyols have been widely used to prepare edible films as plasticizers (Ghasemlou et al., 2011). Linseed oil is a vegetable-based plasticizer rich in essential oleic acid (Martins et al.), linoleic acid and α -linolenic acid (Salarbashi et al., 2013), which is thought to be beneficial to health, protecting against cardiovascular diseases. It has been used as plasticizer in soy protein isolate-based films (Hopkins et al., 2015). The TS of gum ghatti based edible films decreased but EB increased with an increase in glycerol or sorbitol dosage (Zhang et al., 2016). For the semi-refined kappa-carrageenan based edible films, the TS and EB both increased with glycerol and sorbitol concentration increasing from 20% to 30% (Farhan and Hani, 2017). It is found that the same plasticizer has different effects on the different film-forming materials. So far no specific systematic study has been carried out to discuss the effects of additive type and concentration on the properties of CG films. The objective of this work was to explore the effect of glycerol, sorbitol, and linseed oil in various concentrations on the mechanical, barrier, optical, thermal, micro-structural, rheological and hydrophilic behavior of the CG films and to prepare a novel edible CG-based film.

2. Materials and methods

2.1. Materials

Cassia gum (CG, food grade) was supplied by Anli Fine Chemical Co., Ltd. (Henan, China). Sorbitol was supplied by Huishi Biochemical reagent Co., Ltd. (Shanghai, China). Linseed oil (viscosity = 7.4; average molecular weight of fatty acid = 279–307; saponification value (mg KOH/g oil = 189–196)) was obtained from Fufei Chemical Co., Ltd. (Guangzhou, China). Glycerol and ethyl alcohol was analytical reagent grade purchased from Yongda Chemical Reagent Co., Ltd. (Tianjin, China). Calcium chloride anhydrous was supplied by Tianli Chemical Reagent CO., Ltd. (Tianjin, China).

2.2. Preparation of CG films

Preliminary experiments showed that films prepared without plasticizers were too brittle to be peeled off from the plate. Consequently, glycerol (G), sorbitol (S) and linseed oil (L) were used to improve the flexibility and treatability of CG films. The films containing L were discontinuous, opaque and fragile, therefore only G and S were selected for further study and the results showed that

the films containing G or S exceeding 50% were too sticky to further process.

To prepare the films, firstly, CG powder was dispersed with 10 mL ethyl alcohol. Subsequently, distilled water was added and the mixture was stirred at 45 °C for 30 min to obtain a solution of 0.6 wt%. After dissolution, G or S was added into the CG solution at different concentrations (30, 35, 40, 45, and 50%, w/w based on dry weight of CG) and the mixtures continue to stir for 15 min. Finally, the CG film-forming solution was cast into a Plexiglas plate (28 cm × 29 cm × 5 cm) and dried at 65 °C for 30 h.

2.3. Characterization

A Nicolette 6700 spectrometer (Thermo Fisher Science Co., Ltd., MA, USA) was used to detect the FTIR spectra of samples. The FTIR spectra were conducted in attenuated total reflection (Shak and Wu, 2017) mode between 500 and 4000 cm^{-1} at a resolution of 4 cm^{-1} .

Micrographs of the CG films were observed by using a Quanta 200 scanning electron microscope (Philips-FEI Co., AMS., The Netherlands) with an accelerating voltage of 5 kV. The cross-section of samples was obtained by liquid nitrogen embrittlement. And then, the samples were pasted on the sample stage and sputtered with a thin layer of gold before observation.

2.4. Rheological analysis

The rheological behavior of CG film-forming solutions without and with different concentrations G or S was conducted by a TA AR2000EX (Newcastle, DE, USA) rotational rheometer with a 40 mm parallel-plate geometry. The steady shear tests were carried out at a shear rate range of 0.1–100 s^{-1} at 25 °C with a selected gap of 1000 μm . The rheological properties of all film-forming solutions were simulated using the Ostwald-de Wale (Eq. (1)) and cross models (Eq. (2)):

$$\tau = \kappa \gamma^n \quad (1)$$

where τ is the shear stress (Pa), κ indicates the consistency coefficient ($\text{Pa}\cdot\text{s}^n$), γ represents the shear rate (s^{-1}), and n is the flow index.

$$\eta = \eta_\infty + (\eta_0 - \eta_\infty) / (1 + \kappa \gamma^m) \quad (2)$$

where η denotes the viscosity (Pa.s), η_0 indicates the zero shear viscosity (Pa.s), η_∞ represents the infinite shear viscosity (Pa.s), γ is the shear rate (s^{-1}), and m denotes a dimensionless constant.

The viscoelasticity of the CG film-forming solutions was studied via dynamic oscillation measurements within the linear viscoelastic region. The chosen strain of 1% during the frequency range of 0.1–100 rad/s in the linear viscoelastic range. Elasticity modulus (G') and viscosity modulus (G'') were recorded as functions of frequency to investigate the dynamic viscoelasticity.

2.5. Property measurements

2.5.1. Light transmission measurement

An ultraviolet–visible (UV–vis) spectrophotometer (UV-2600, Shimadzu, Kyoto, Japan) was employed to analyze the light transmittance of CG films in the range of 200–800 nm. CG films were uniformly cut into 4 cm × 2 cm.

2.5.2. Water vapor permeability and oxygen permeability

The water vapor permeability of the samples at 75% RH (25 °C) was measured by the gravimetric method (Pérez-Gago and Krochta,

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