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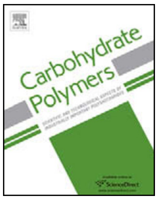

Influence of tragacanth gum in egg white based bioplastics: Thermomechanical and water uptake properties

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ABSTRACT

This study aims to extend the range of applications of tragacanth gum by studying its incorporation into bioplastics formulation, exploring the influence that different gum contents (0–20 wt.%) exert over the thermomechanical and water uptake properties of bioplastics based on egg white albumen protein (EW). The effect of plasticizer nature was also evaluated through the modification of the water/glycerol ratio within the plasticizer fraction (fixed at 40 wt.%). The addition of tragacanth gum generally yielded an enhancement of the water uptake capacity, being doubled at the highest content. Conversely, presence of tragacanth gum resulted in a considerable decrease in the bioplastic mechanical properties: both tensile strength and maximum elongation were reduced up to 75% approximately when compared to the gum-free system. Ageing of selected samples was also studied, revealing an important effect of storage time when tragacanth gum is present, possibly due to its hydrophilic character.

1. Introduction

Proteins and polysaccharides have been used as biopolymer sources for many years. A wide range of tailored functional properties may be achieved modulating the different interactions and intermolecular linkages that take place between and within heteropolymer molecules (Gómez-Martínez, Partal, Martínez, & Gallegos, 2009; Pommet, Redl, Morel, & Guilbert, 2003). Proteins like egg white or soy protein represent renewable materials that are produced massively. The suitability of some of these proteins for the manufacture of bioplastics has been shown by different studies (Kim, 2008; Mohanty et al., 2005; Tummala, Liu, Drzal, Mohanty, & Misra, 2006; Zheng, Tan, Ran Zhan, & Huang, 2003).

The processing of bioplastics generally requires the addition of plasticizers and, sometimes, disrupting agents in the formulation (Pommet, Redl, Guilbert, & Morel, 2005; Sothornvit, Krochta, & Han, 2005). A plasticizer is a component included to overcome brittleness and to avoid chipping and cracking of polymeric materials during subsequent handling and storage. Plasticizers, like glycerol or water, are molecules with low molecular weight and volatility, which reduce the intermolecular forces and increase the mobility of the polymeric chains, which results in a decrease in the material glass transition temperature, through modification of the three-dimensional structure of proteins (Matveev, Grinberg, & Tolstoguzov, 2000).

The barrier properties of biopolymeric films are important parameters in food packaging applications. Protein and polysaccharide films display generally good barrier properties against oxygen at low and intermediate relative humidity (RH) values, showing good mechanical properties, like elongation at break (Anker, Berntsen, Hermansson, & Stading, 2002). An adequate selection of composition and processing parameters may lead to materials with unique properties (Pommet et al., 2003). Recent works by Fernández-Espada, Bengoechea, Cordobés and Guerrero (2013) and González-Gutiérrez, Partal, Garcia-Morales and Gallegos (2010); González-Gutiérrez et al., 2011González-Gutiérrez, Partal, Garcia-Morales and Gallegos (2011), have revealed the feasibility of producing bioplastics from egg white protein (albumen). Those bioplastics obtained through thermo-mechanical methods showed suitable values for Young's modulus, stress at break or elongation for biodegradable food packaging applications. This would represent a novel alternative for egg white when compared to the traditional use given by the food industry due to its functional properties, such as gelling, foaming, heat setting and binding adhesion.

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Moreover, if modified atmosphere packaging (MAP) applications are pursued, a hydrophilic character may be desirable for the bioplastic material, as some processes leading to oxygen scavenging or CO_2 emitting have been proved to be moisture dependent. Thus, those processes would only take place after moisture has been absorbed from the food product or package atmosphere into the bioplastic material (Conte et al., 2013; Ščetar, Kurek, & Galić, 2010). Tragacanth gum is a hydrophilic and very complex heterogeneous anionic polysaccharide of high molecular weight, around 840 kg mol^{-1} (Weiping & Branwell, 2000), consisting of two main fractions: (a) a water-insoluble component called *bassorin*, which has the capacity to swell and form a gel; and (b) a water-soluble component called *tragacanthin* (Balaghi, Mohammadifar, Zargaraan, Gavlighi, & Mohammadi, 2011). Tragacanth gum has been reported, when mixed with glycerol and water, to form a useful excipient to bind pill masses in the pharmaceutical or cosmetic industries (Phillips & Williams, 2000). Tragacanth gum has been accepted since 1961 as generally recognized as safe (GRAS), according to FDA, and used for many years as a stabiliser, thickener, emulsifier and suspending agent in the food, pharmaceutical, cosmetic, textile and leather industries as well as in technical applications (Anderson & Bridgeman, 1985). It presents high viscosity at low concentration, good suspending action, unusually high stability to heat and acidity and effective emulsifying properties. It is also pourable and has creamy mouth feel and good flavour-release properties (Weiping & Branwell, 2000) and very long shelf life (Levy & Schwarz, 1958).

Egg white based bioplastic materials have been previously studied, displaying water absorption capacities under 100% (Fernández-Espada et al., 2013; Jerez, Partal, Martinez, Gallegos, & Guerrero, 2007a,b). A potential way to improve the water uptake of egg white based bioplastic materials would be to include a hydrocolloid in its formulation. Tragacanth gum has proved to swell and form gels (Weiping & Branwell, 2000), having been used in food products due to its attractive functional properties (e.g. high ability to bind water or effective emulsifying properties). More recently, potential applications of tragacanth gum in the field of packaging or biomaterials have also been studied (Mostafavi, Kadkhodaee, Emadzadeh, & Koocheki, 2016; Ranjbar-Mohammadi, Prabhakaran, Bahrami, & Ramakrishna, 2016). However, just few studies have investigated the properties of tragacanth gum as part of moulded bioplastic materials (López-Castejón, Bengoechea, García-Morales, & Martínez, 2015).

The main objective of this work was to evaluate the effect of tragacanth gum on the water uptake capacity, linear viscoelasticity and tensile properties of egg white protein matrices when they were plasticized with mixtures of varying water/glycerol ratios. Bioplastics were obtained through a conventional thermo-mechanical process, keeping the biopolymer/plasticizer ratio constant but modifying the composition within each fraction.

2. Experimental

2.1. Materials and sample preparation

2.1.1. Materials

Spray-dried egg white albumen (designated EW; with 73 wt.% protein (dry basis), 6 wt.% ashes and 8 wt.% moisture) provided by OVOSEC S.A. (Spain) was used as base material for bioplastics manufacture. EW was produced by the automatic shelling of eggs, which prior to their homogenisation and pasteurisation are desiccated by atomisation (Navarro, 2003). After this, the product is subjected to strict physico-chemical and bacteriological tests. On the other hand, tragacanth gum (designated T) (39–42% carbon content) was supplied by Sigma-Aldrich (USA). In relation with the plasticizers, glycerol, from Panreac Química, S.A. (Spain), and distilled water were designated G and W, respectively.

2.1.2. Sample preparation

Different egg white/tragacanth gum/glycerol/water (EW/T/G/W) compositions, always keeping a constant biopolymer (EW + T)/plasticizer (G + W) ratio of 60/40 have been studied, following an established protocol (López-Castejón et al., 2015). Three different plasticizer contents have been used: W40, with only water; G20W20, using both glycerol and water at a ratio equal to 1; and G40, containing only glycerol. Moreover, three different biopolymer contents have also been studied: EW60, with no tragacanth gum; EW50T10, with a tragacanth gum content of 10% (w/w); and EW40T20, with a gum content of 20% (w/w). A summary of the 9 compositions studied is shown in Table 1.

With regard to the bioplastics manufacture, this was accomplished by a thermo-mechanical process, which includes two stages:

a) Mixing of ingredients: it was carried out for 10 min in the kneading tool (Rheomix 600p) of a torque-rheometer (Polylab, Thermo Haake GmbH, Germany) equipped with two counter-rotating rollers turning at 50 rpm (Jerez, Partal, Martínez, Gallegos, & Guerrero, 2005). Temperature, starting at 25 °C, was allowed to naturally evolve over this period (no heating/cooling). The Polylab mixer used allowed the record of the torque and temperature along the mixing time. In every case, 100 g of blend was obtained.
b) Compression-moulding: the resulting dough-like material was subjected to pressure of 10 MPa and temperature of 120 °C for 10 min in a hot-plate press, as described by Jerez et al. (2007a,b). Two types of moulds were used: one to obtain rectangular 3-mm-thick specimens for both DMTA and water uptake capacity measurements; and a second one to obtain type IV-dumbbell specimens (2003) for tensile tests.

After preparation and before testing, these samples were placed in desiccators at relative humidity of 53% with a saturated solution of $Mg(NO_3)_2{\cdot}6H_2O$ at room temperature (Nyqvist, 1983). Samples were always stored at least for 24 h prior any test was conducted.

2.2. Testing

2.2.1. Water uptake capacity

Water uptake tests, according to ASTM D570 (2005), were carried out on rectangular probes ($50 \times 10 \times 3\,mm^3$) immersed into distilled water for 24 h. Three replicates were done for each sample, and the water uptake percentage calculated as:

$$Wateruptake\,(wt.\%) = \frac{wetwt. - reconditionedwt.}{conditionedwt.} \times 100 \qquad \text{(Eq. 1)}$$

where: conditioned weight, is the initial weight of the probe; wet weight, refers to the weight of the probe just after 24 h of water immersion; and reconditioned weight, is the final weight of the wet sample after 24 h of drying in an oven at 50 °C.

2.2.2. Tensile properties

Tensile tests were performed with a MTS Insight 10 kN (USA), according to ASTM D638 (2003), with an extension rate of 5 mm/min, at room temperature. An extensometer was used in order to accurately register the sample elongation. At least, five tests were carried out for each dumb-bell shaped type IV sample (thickness: 3.2 mm).

2.2.3. Dynamic mechanical thermal analysis (DMTA)

DMTA experiments were performed with a RSA3 rheometer (TA Instruments, USA) in dual cantilever bending mode on rectangular