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Physicochemical and mechanical properties of extruded laminates from native and oxidized banana starch during storage

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ABSTRACT

Changes in some mechanical and physicochemical properties of extruded Laminates, made from native (NBS) and oxidized banana starch (OBS) with 3 g/100 g of sodium hypochlorite, as affected by storage time (0–45 days) were evaluated. Micrographs showed cracks and pores with some continuity on the surface of both OBS and native banana starch (NBS) laminates. Extruded Laminates of OBS were more transparent, soluble and homogeneous than those from NBS. Little differences were observed in water vapor permeability between NBS and OBS Laminates. Laminates solubility decreased with storage time. The X-ray diffraction from NBS and OBS Laminates, showed similar type B patterns and percent of crystallinity. Throughout storage time, an increase in temperature and enthalpy of melting was observed for all Laminates, however, the enthalpy values for OBS Laminates were lower than those of NBS. The tensile strength, percent of elongation and elasticity modulus values of OBS Laminates, were higher than those of NBS. Overall, OBS might be a suitable raw material to produce extruded Laminates with adequate functional properties.

1. Introduction

Nowadays, the indiscriminate use of plastic bags has become such an increasing environmental pollution problem, that local governments have passed laws in order to limit their use. Over the last years, the interest in biodegradable films made from renewable and natural polymers has increased. Water-soluble polysaccharides such as starch, cellulose derivatives, alginate and pectin can form biodegradable and edible films (Garcia, Ponotti & Zaritzky, 2006). Thus, for example, the substitution of plastic films by short term degradation ones, mainly those from natural sources, has been strongly encouraged. The production of films or laminates (thickness > 1 mm), based on biological materials uses film-forming agents (e.g., macromolecules as polysaccharides, starch and proteins), a solvent (usually water) and a plasticizer (e.g., polyols as glycerol, sorbitol, ethylene glycol).

Starch consists of two chemically distinct polysaccharides, namely; amylose and amylopectin, conforming characteristic structures called granules with both amorphous and crystalline structures. The amylose is an essentially linear macromolecule of glucose units linked mainly by α-D-(1-4) bond, although there may be some α-D-(1-6) bonds. It is distributed mostly in the starch granule amorphous domains (lamella), with small amounts in the semicrystalline granule ring. The amylopectin is a branched glucose polymer, linked mainly by α-D-(1-4) bonds (about 96 g/100 g) and the remaining 4–6 g/100 g by α-D-(1-6) bonds, conforming the crystalline lamella (Hermansson & Svegmark, 1996). The proportion of these two polymers and their physical organization inside the granules, confer singular physicochemical characteristics and functional properties to starch (Thomas & Atwell, 1999). Starch granules are semi-crystalline polymeric systems, where the degree of crystallinity (15–45 g/100 g), is due to short-chain fraction of the amylopectin arranged as double helices and packed in small crystallites organized in a tri-dimensional structure (Du, MacNaughtan & Mitchell, 2011).

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Banana is a general term involving a number of species or hybrids in the genus *Musa* of the family *Musacea*. Plantains are generally the larger, more angular starchy fruits of hybrid cultivars in the banana family, edible raw when fully ripe, but also suitable for cooking. They are mainly produced in developing countries, being of major importance to people in the growing areas, where a lot of the produce is lost because of poor post-harvest handling methods. Nonetheless, banana starch has the potential to be a commodity starch because of its specific properties and its potential production from low-cost, cull bananas (Zhang, Whistler, BeMiller &Hamaker, 2005). Various authors have attempted to produce films from starch isolated from various sources including maize, potato, cassava, tapioca, flaxseed and recently banana (Zhang et al., 2005). The most used techniques for film processing include casting and extrusion, being the latter a convenient technique for practical purposes. Starch film extrusion is more complex than that of conventional polymers due to multi-phase transitions involved during processing. There are many works done on films produced by casting, but further investigation is needed on the physical properties of those obtained by extrusion.

Starch granules during extrusion are subjected to high shear stress and temperature, these factors produce the destruction of granules, elimination of the crystalline structure, at times the molecular weight reduction, and also, the formation of an amorphous molten mass (thermoplastic starch polymer, TSP), which is then pressurized and shaped by the die head (Su et al., 2009).

Modified starch has been used to replace native starch in various applications, because of its appropriate functional and physicochemical properties as a result of its granule modification (Kuakpetoon & Wang, 2008). A commonly used method of starch modification is its oxidation with chemical agents differing in their oxidation power. Oxidized starch is widely used in many industries, particularly in applications where film formation and adhesion properties are desired. The major application of oxidized starches is as surface sizing agents and coating binders in the paper industry (Sangseethong, Termvejsayanon & Sriroth, 2010). Oxidized starch is commonly produced by reaction of starch with an oxidizing agent under controlled temperature and pH. Previous studies on corn starches have found that, when sodium hypochlorite (NaOCL) has been used, NaOCL concentrations of 2 g/100 g solution or higher, are enough to produce real changes on such starches physicochemical characteristics (Kuakpetoon Wang, 2008). During the oxidation process, hydroxyl groups on starch molecules are oxidized to carbonyl and carboxyl groups, contributing to improved stability of starch paste. The reaction also causes degradation of starch molecules resulting in a modified starch with low viscosity. This allows the use of oxidized starch in applications where high solid concentration is needed (Wurzburg, 1986) The addition of plasticizers is necessary to obtain a TSP by reduction of intermolecular forces, and increase in the mobility of polar polymer chains. In addition, plasticizers help to overcome the brittleness of starch films and improve their flexibility and extensibility. Glycerol has been one of the most used plasticizers, since it improves film extensibility (Gurgel, Vieira, da Silva, dos Santos & Beppu, 2011). To define possible uses and shelf life of these types of materials, studies on the effect of storage on the physicochemical properties are needed, but those studies are not available in the literature.

The purpose of this work was to evaluate the changes in some physical and physicochemical properties of extruded laminates, made from native and oxidized banana starches, as affected by storage time.

2. Material and methods

2.1. Materials

Unripe banana (*Musa paradisiaca* L.) selected with no yellow color on its peel (about 18 weeks from blossom), was obtained from the province of Veracruz, Mexico.

2.2. Starch isolation

Native banana starch was obtained as described by González (2003). This consisted in peeling, cutting and immersing the banana slices obtained in 0.25 g/100 g citric acid solution. The slices were blended three times with distilled water (water:banana ratio 1:1) using a Waring blender. The blended fruit pulp was sieved using an electric sieving machine (Testing Equipment, Model RNU). Sieves with mesh sizes 40, 100, 200 and 325 (0.425, 0.15, 0.075 and 0.045 mm, respectively), were used one at a time. Each paste formed in every sieve was washed with water at least three times (10 parts of water: 1 part of banana pulp) until the wash water was clean. The pulp cake was eliminated. The filtrate contained starch and smaller particles suspended. The starch was allowed to precipitate during 24 h at room temperature (about 25 °C). The excess of water was decanted until getting a solid concentration within the range of 30–35 g/100 g. The slurry obtained was dried with a Niro Atomizer spray-dryer (Model 230 EA 11 No. 21, Denmark). The processing parameters were: inlet temperature of about 140 °C; outlet temperature of about 68 °C, with a feeding flow of 14 L/h. The dehydrated native starch was standardized with a 100 mesh sieve (0.15 mm) and stored at room temperature in a glass container, till use for further experiments.

2.3. Starch chemical analysis

For the sake of comparison, the total starch and also the apparent amylose content (Gilbert & Spragg, 1964), was obtained with potato starch as standard. For this last component, about 1.5 mg of starch was weighed into an aluminium pan and transferred to a 50 ml volumetric flask. Then, about 0.5 ml of a solution NaOH 1 mmol equi/L was added and the mixture simmered during 3 min in a boiling water bath. It was then cooled down and neutralized with 0.5 ml of 1 mmol equi/L HCl, and 0.07 g of potassium bitartrate was added and diluted with distilled water until obtaining an approximate volume of 45 ml. Once the bitartrate was dissolved, 0.5 ml of a solution of iodine (2 mg/ml of Iodine and 20 mg/ml of KI) was added and the volume completed to 50 ml with distilled water. The solution was mixed and allowed to rest during 20 min at room temperature, and the absorbance was measured at 680 nm in a Genesys 5 spectrophotometer (Spectronic Instruments, USA). The amylose content in the starches was quantified by interpolation of the absorbance values in a standard curve. The amylopectin content was obtained by difference.

For total starch content (Goñi, García & Saura-Calixto, 1997), a 50 mg sample was dispersed in 2 mol/equi/L KOH (30 min) and incubated in a controlled-temperature water bath (60 °C, 45 min, pH 4.75) with amyloglucosidase (Roché No. 102 857, Roche Diagnostics, Indianapolis, IN, USA); the glucose hydrolyzate obtained was measured with a GOD/POD reagent (SERA-PAK$^{®}$ Plus, Bayer de México, SA de CV). Total starch content was calculated as glucose (mg) × 0.9; potato starch was used as standard.

2.4. Starch oxidation

To obtain oxidized banana starch (OBS), native banana starch (NBS) and sodium hypochlorite solution (NaOCl) with 3 g/100 g