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# Physicochemical and microstructural characterization of films prepared by thermal and cold gelatinization from non-conventional sources of starches

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## Abstract

Edible films were developed using different starch sources (banana, okenia and mango). Starch suspensions were cold gelatinized with NaOH or gelatinized by heat. Glycerol was used as plasticizer. Films were characterized by X-ray diffraction, Scanning Electron Microscopy (SEM), water vapor permeability and mechanical analyses. Films prepared with thermal gelatinization showed homogeneous surfaces without pores or cracks compared with starch films made with cold gelatinization, and additionally, thermal gelatinization produced films that during storage, recrystalized as a B-type compared with the A-type X-ray diffraction pattern of the films elaborated with cold gelatinization. These differences had an important role in the thickness, barrier and mechanical properties. In general, films prepared with the thermal gelatinization method, showed better barrier and mechanical properties, important characteristics when choosing the preparation method of the films. In some parameters studied, the starch source had an important role in the properties of the films. Non-conventional sources can be used for film preparation with certain barrier and mechanical characteristics.

*Keywords:* Starch films; Microstructure; Mechanical properties

## 1. Introduction

Consumers demand higher quality and longer shelf-life in foods, while reducing disposable packaging materials and increasing recyclability. Such concerns have caused increased interest in edible film research (Arvanitoyannis, 1999). Biodegradable films, by regulating water, oxygen, carbon dioxide, and lipid movement in food systems, provide potential solutions to such concerns (McHugh, Avena-Bustillos, & Krochta, 1993). The use of edible films and coatings is constantly increasing in the food industry. Edible films are used primarily to extend the shelf life and quality of foods by preventing changes in aroma, taste, texture, appearance, or handling characteristics (Arvanitoyannis, 1999; Tharanathan, 2003). Edible films often contain hydrophilic components, such as proteins or polysaccharides (Arvanitoyannis & Biliaderis, 1998), these films provide under certain conditions of relative humidity (RH) and temperature, a good barrier to gas transmission but a poor barrier to water vapor (Guilbert, 1986; Kester & Fennema, 1986; Petersson & Stading, 2005). Several types of composite films and coatings reported in the food literature over the last 10 years, contain a polysaccharide base (Averous & Boquillon, 2004). For these films, the advantages are more in the area of decreasing gas exchange rather than retardation of water loss due to their hydrophilic nature (Krochta & De Mulder-Johnston, 1997). With regard to biodegradable packaging, starch is the commonly used agricultural raw material, since it is a renewable source, inexpensive, widely available and relatively easy to handle (Lourdin, Della Valle, & Colonna, 1995). Starch is a reserve polysaccharide present in the endosperm of the grain of corn (*Zea mays L.*), banana pulp (*Musa paradisiaca*), yucca (*Manihot esculenta*) among others (Bello-Pérez, Agama-Acevedo, Sayago,

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& Figueroa, 2000); it is extracted and used in the food industry to impart functional properties, modify food texture and consistency and so on. Starch owes much of its functionality to two major high-molecular-weight carbohydrate components, amylose and amylopectin, as well as to the physical organization of these macromolecules into the granular structure (French, 1984). Amylose is responsible for the film-forming capacity of starches. However, the functional, organoleptic, nutritional and mechanical properties of an edible film can be modified by the addition of various chemicals in minor amounts. Plasticizers, such as glycerol, sorbitol and polyethylene glycol, are defined as an essentially high boiling, non-separating substance, which when added to another material, changes the physical and/or mechanical properties of that material (Arvanitoyannis & Biliaderis, 1998; Jansson & Thuvander, 2004; Mali, Grossmann, García, Martino, & Zaritzky, 2005). Water is a common plasticizer but is very difficult to control in biopolymers that are generally more or less hydrophilic. Plasticization of biopolymeric films depends on the RH of the packaging atmosphere and on the environment for unpacked products (Gontard et al., 1992). The plasticizers must be compatible with the polymer (starch), and these compounds decrease intermolecular attractions between adjacent polymeric chains, thus increasing film flexibility (Donhowe & Fennema, 1993). Incorporation of these additives may, however, cause significant changes in the barrier properties of the film (García, Martino, & Zaritzky, 2000a,b).

Native and modified starches (from traditional and alternative sources) have been used since ancient times as a raw material to prepare different products. They are employed in foodstuffs because of their good thickening and gelling properties. Actually, they are used for the formation of starch-based edible films and coatings (Jansson & Thuvander, 2004; Mali et al., 2005). However, the current tendency is to look for alternative sources for obtaining starch with better physicochemical and functional characteristics. In previous works, okenia (*okenia hypogaea*), banana (*musa paradisiaca*) and mango (*manguifera indica*) starches were isolated and characterized (Aparicio-Saguilan, 2001; Bello-Pérez Agama-Acevedo, Sánchez-Hernandez, & Paredes-López, 1999; Sánchez-Hernandez, Solorza-Feira, Méndez-Montealvo, Paredes-López, & Bello-Perez, 2002). In the present study, we propose to use these starches for film preparation and study some of their mechanical, physicochemical and microstructural characteristics.

## 2. Materials and methods

### 2.1. Raw materials

Starches from mango (Aparicio-Saguilan et al., 2004), banana (Bello-Pérez Aparicio-Saguilan, Méndez-Montealvo, Solorza-Feria, & Flores-Huicochea, 2005) and okenia (Sánchez- Hernandez et al., 2002) were isolated as reported previously. Mango starch had an amylose content of 28.7% (Agustiniano-Osornio, 2004), banana starch of 35.0% (Bello-Pérez et al., 1999) and Okenia starch of 26.1% (González-Reyes, 2002).

### 2.2. Film preparation

Starch based films were prepared using two different methods: (a) cold gelatinization with NaOH and (b) thermal gelatinization

Aqueous solutions of 30 g/L starch were cold gelatinized with 10 g/L NaOH to obtain the films. These suspensions were then neutralized with 7 M $H_3PO_4$. Glycerol was added as plasticizer after neutralization, at a concentration of 15 g/L of formulation.

Thermal gelatinization was performed mixing starch (3 g) and glycerol (1.5 g) with distilled water (100 ml). The film forming dispersions were transferred quantitatively to the cup of a viscoamylograph (Brabender OHG, Duisburg, Germany), they were heated from 30 to 95 °C and maintained at 95 °C for 10 min, with regular shaking (125 rpm) and constant heating rate (3 °C/min).

For both gelatinization methods, the films were prepared by casting; gelatinized suspensions (20 ml) were immediately poured on each polyethylene Petri dish of 8 cm in diameter. The starch suspensions were dried at 65 °C until constant weight (approximately 5 h). An electrical oven model Imperial V (Lab line Instruments Inc., Illinois, USA) with leveled stages was used. The moisture content was determined gravimetrically ($130 \pm 2$ °C for 2 h) using 500 mg of sample. Three measurements were made on each film.

### 2.3. Thermal analysis

The thermal properties of starches, were studied using a differential scanning calorimeter (DSC, TA Instruments, model 2010, New Castle, USA) previously calibrated with indium. The gelatinization of the starches was evaluated by the method proposed by Paredes-López, Bello-Pérez & López (1994), 2 mg sample (dry basis) was weighed on an aluminum pan, 7 μL of deionized water was added. The pan was sealed tightly and then it was allowed to stand for 1 h before carrying out the analysis. An empty aluminum pan was used as reference. The sample was subjected to a heating program over a temperature range from 30 to 140 °C and a heating rate of 10 °C/min. The gelatinization or peak temperature ($T$p) and the transition enthalpy ($\Delta H$), were obtained directly using the software of TA Instruments OS/2 version 2.1.

### 2.4. Thickness determination

Thickness of films was determined using a digital coating thickness gauge Elcometer A 300 FNP 23 (England) for nonconductive materials on nonferrous substrates. Thickness of films was also observed on SEM micrographs of film cross sections.

### 2.5. Scanning electron microscopy (SEM)

For SEM study, the samples (starches from different sources) were fixed to a conductive tape of copper of double glue, which was covered with a layer of coal of 20 nm thickness. It was deposited at vacuum with an evaporator in a JEOL JSMP 100 (Japan) electron microscope. Later on, it was covered with a layer of gold of 50 nm of thickness in the ionizer of metals JEOL. This was observed in the microscope and registered photographically. Film pieces were mounted on aluminum stubs using a double-sided tape and then coated with a layer of gold (40–50 nm), allowing surface and cross-section visualization. All samples were examined using an accelerating voltage of 5 kV.

### 2.6. X-ray diffraction

Samples were analyzed between $2\theta=2°$ and $2\theta=60°$ with a step size $2\theta=0.02°$ in an X-ray diffractometer (Philips PW 1710, Netherlands) using Cu K$\alpha$ radiation ($\lambda=1.543$), 50 kV and 30 mA. The diffractometer was equipped with 1° divergence slit and a 0.1 mm receiving slit. Modifications of crystalline structure of films during storage of 60 days at 20 °C were evaluated.

### 2.7. Water vapor permeability

Water vapor permeability tests were conducted using ASTM (1987) method E96 with some modifications. Each film sample was sealed over a circular opening of 0.00181 $m^2$ in a permeation cell that was stored at 25 °C in a dessicator. To maintain a 75% RH gradient across the film, anhydrous calcium chloride (0% RH) was placed inside the cell and sodium chloride saturated solution (75%) was used in the dessicator. Water vapor transport was determinated from the weight gain of the permeation cell. After steady state conditions were reached (about 2 h), eight weight measurements were made over 24 h. Changes in the weight of the cell were recorded to the nearest 0.0001 g and plotted as a function of time. The slope of each line was calculated by lineal regression ($r^2>0.99$) and the water vapor transmission rate (WVTR) was calculated from the slope of the straight line (g/s) divided by the cell area ($m^2$). After the permeation tests, film thickness was measured and WVP (g $Pa^{-1}$ $s^{-1}$ $m^{-1}$) was calculated.

### 2.8. Film solubility in water

Pieces of film of 2×3 cm were cut from each film and were stored in a dessicator (o% HR) for 7 days. Samples were weighed to the nearest 0.0001 g and placed into test beakers with 80 ml of deonized water. The samples were maintained under constant agitation for 1 h at room temperature (approximately 25 °C) or at boiling temperature. The remained pieces of film after soaking were dried again in an oven at 60 °C to constant weight. The percentage of total soluble matter (% solubility) was calculated as follows:

$$\%\text{Solubility} = \frac{(\text{Initial dry weight} - \text{Final dry weight})}{\text{Initial dry weight}} \times 100$$

Samples were analyzed at least in duplicates.

### 2.9. Mechanical properties

Puncture tests were performed in a texturometer TA XT2i—(Stable Micro Systems, England). Results were calculated with the maximum force needed to cross the film. A cylindrical probe of 2 mm in diameter at a constant rate of 1 mm/s was used. Tensile tests were also performed with the same equipment using a tension grip system A/TG. Probes of 8 cm in length and 0.7 cm width were used; at least 4 probes were used for each film formulation. Distance between grips were of 6 cm, velocity of assay 10 mm/min.

Maximum breaking force (*N*) and deformation at break (extension at the moment of rupture, mm) were obtained from the force vs. deformation curves according to the ASTM D882-91 (1996). Tensile strength (MPa) was calculated by dividing maximum load by film cross-sectional area of the film. Percent elongation at break was expressed as percentage of change of the original length of a specimen between grips at break. Elastic modulus (***E***?) was determined from the slope of the linear regression performed on the initial points on the stress–strain curves.

## 3. Results and discussion

### 3.1. Raw materials

The non-conventional starches were studied previously before making the films. Scanning electron micrographs of okenia, mango and banana are shown in Fig. 1. Mango and okenia starch granules showed rounded shapes, mango starch with a diameter of 5–10 μm and Okenia starch with 1–3 μm, banana starch granules showed oval shapes with a diameter between 10–40 μm.

### 3.2. Thermal analysis

A thermal analysis of native starches was performed (Table 1), where banana starch showed the highest gelatinization temperature. This parameter is important in the gelatinization process for film preparation.

### 3.3. Thickness

Thickness of the film changes depending on starch source and gelatinization method (Table 2), and their density

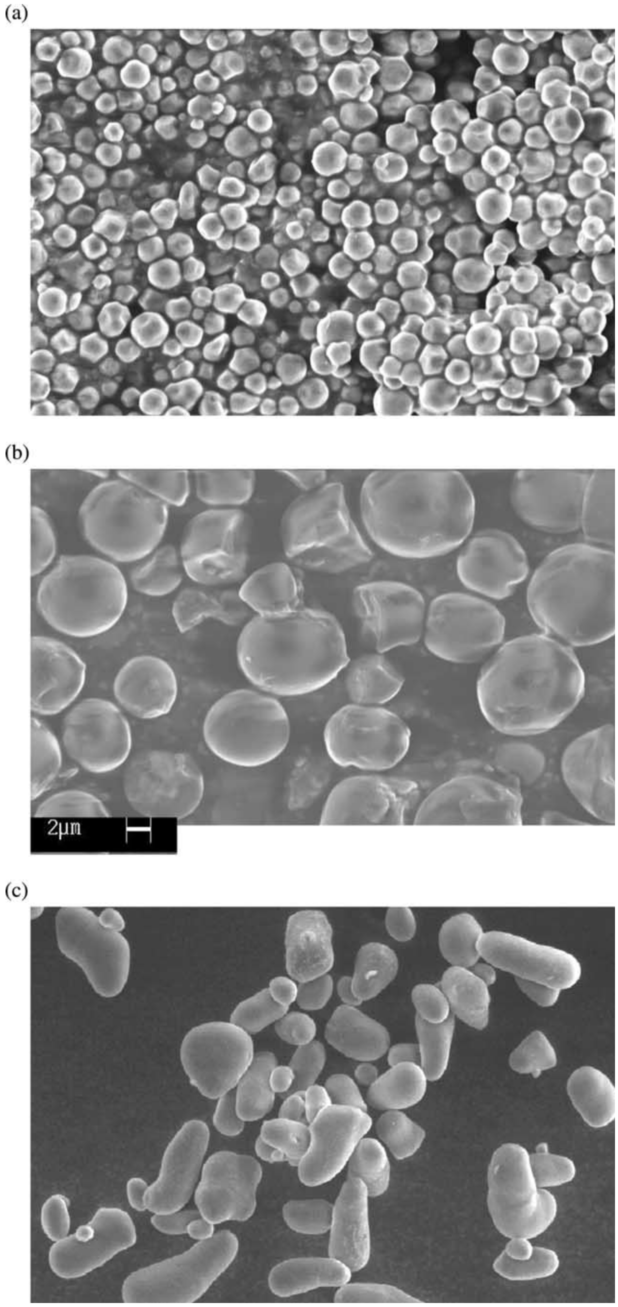

Fig. 1. SEM micrographs of banana, okenia and mango starch granules.

Table 1
Thermal analysis of starches isolated from non-conventional sources

| Starch source | Temperature of gelatinization (°C) | Enthalpy of gelatinization (J/g) |
|---|---|---|
| Banana | 77.6 | 23.4 |
| Mango | 73.8 | 21.2 |
| Okenia | 71.2 | 15.0 |

(tested at 25 °C) ranged between 1.02 and 1.04 g/cm$^3$. Films prepared after gelatinization in the viscoamylograph had a thicker thickness than those prepared using NaOH for cold gelatinization. This behavior can be explained because during NaOH treatment not only gelatinization was produced but also hydrolysis of amylose and external long chains of amylopectin did take place (García et al., 2000a, b). The extensive hydrolysis of starch components yields short chains that can align in higher proportion and produce films with thinner thickness. On the other hand, the starch source plays an important role in film thickness when cold gelatinization is carried out, but only in the case of okenia starch thinner films than those prepared with banana and mango starch were observed, no statistical differences were found ($\alpha = 0.05$) among these last ones. Thinner films produced with okenia starch was probably due because it has the smallest amylose content of the three starches. García, Martino, & Zaritzky (1999), prepared films using cold gelatinization with starch medium amylose content (MAS) and with high amylose content (HAS), adding glycerol and sorbitol as plasticizers. They found that HAS films were thicker than MAS films. Also, films with plasticizers were thinner than unplasticized films.

However, when thermal gelatinization was used, thickness of the films prepared with the three starch sources did not change ($\alpha = 0.05$). In the case of films prepared with normal corn and plasticizers, the thickness values ranged between 104–110 μm, which was higher than those determined in films prepared with the same gelatinization method but different starch source. Granule size can play an important role, because okenia starch has smaller granule size (Sánchez-Hernandez et al., 2002) than banana (Bello-Pérez et al., 2000) and mango (Aparicio-Saguilan, 2001) starches. However, further studies are needed in to make this point clearer.

The two gelatinization methods used to elaborate the films resulted in different moisture; those prepared with cold gelatinization have higher values than those obtained with thermal gelatinization when they were recently prepared (Table 2).

### 3.4. Microstructure observations

Starch films made with thermal gelatinization showed homogeneous surfaces without pores or cracks compared with starch films made with cold gelatinization. Cross-sections of thermal gelatinization starch films showed

Table 2
Effect of starch source and gelatinization method on thickness of starch-based films*

| Film | Humidity (%) | Thickness (μm) |
|---|---|---|
| *Cold gelatinization* | | |
| Banana starch | 15.96 ± 0.49 | 83.26 ± 7.78 |
| Okenia starch | 16.46 ± 0.31 | 54.55 ± 5.24 |
| Mango starch | 21.34 ± 1.72 | 86.65 ± 4.77 |
| *Thermal gelatinization* | | |
| Banana starch | 9.61 ± 0.68 | 113.05 ± 6.13 |
| Okenia starch | 8.15 ± 0.43 | 116.85 ± 8.77 |
| Mango starch | 5.86 ± 0.18 | 126.88 ± 8.56 |

*Average of three replicates ± standard error.

a dense structure while films made up with cold gelatinization showed an irregular structure, where the former are thicker than the latter. The homogeneous matrix of films is a good indicator of their structural integrity, and consequently, good mechanical properties would be expected (Mali, Grossmann, Garcia, Martino, & Zaritzky, 2002). A similar structure was obtained in films prepared with cold gelatinization as reported in unplasticized films (Garcia et al., 2000a,b; Mali et al., 2002), because amylose molecules without plasticizer produce brittle films (Donhowe & Fennema, 1993). There was no significant difference between the starch sources, when analysed only by the gelatinization method. This behavior was similar between the same manufacture methods.

### 3.5. X-ray diffraction

X-ray diffraction patterns of starch films made with thermal gelatinization are shown in Fig. 2. They are characterized by sharp peaks associated with the crystalline and amorphous zones. The three films had a B-type diffraction pattern with differences in the peak intensity, which is related with crystalline size, size distribution and by the particular imperfections of the crystalline lattice (Klug & Alexander, 1974). The same pattern was obtained for films prepared from potato starch and dried at 20 °C and 25% of relative humidity (Rindlav, Hulleman, & Gatenholm 1997). Films made with mango starch showed four sharp peaks at $2\theta=5$, 16, 19 and 21°, but in the case of banana films, peaks at $2\theta=19°$ and 21° were observed. Okenia films had low tendency to show a crystalline structure after storage, phenomenon that may be due to amylopectin structure of this starch with high amount of short chains (Sánchez-Hernández, 2001), that decreased the formation of crystalline structure as was reported by Yuan, Thompson, and Boyer (1993).

X-ray diffraction patterns of films made with cold gelatinization are shown in Fig. 3. There is a difference in the patterns of films depending on the gelatinization method. In mango films there are still 4 peaks but are located in different places $2\theta=10$, 20, 26 and 36°. Okenia has one sharp peak in $2\theta=10°$ and three little ones at $2\theta=26$, 27 and 36°. These patterns have a similar shape to those found by Garcia et al. (2000a,b), working with the same gelatinization method. Bader and Göritz (1994), reported X-ray patterns of amylomaize films obtained by autoclaving, they attributed their findings to an A-type starch pattern, similar to those found in this work.

### 3.6. Water vapor permeability

One of the major problems found in the applications of edible films is the high permeability due to the hydrophilic character of the starch molecule. There were differences between the two methods of elaboration to obtain the films (Fig. 4). Films made by cold gelatinization had less permeability than those made by thermal gelatinization. Differences in permeability can be attributed to alterations in film structure. The permeability of films made with the cold gelatinization agreed with those of Arvanitoyannis, Psomiadou, Nayayama, Aiba, and Yamamoto (1997), who found similar results for gelatin and soluble starch using the

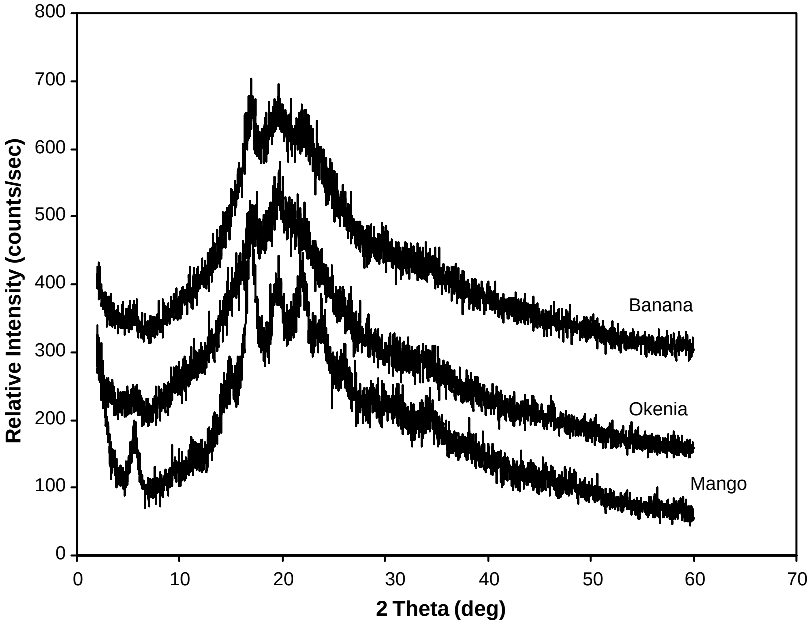

Fig. 2. X-ray diffraction pattern of starch films made by thermal gelatinization at 60 days of storage at 25 °C.

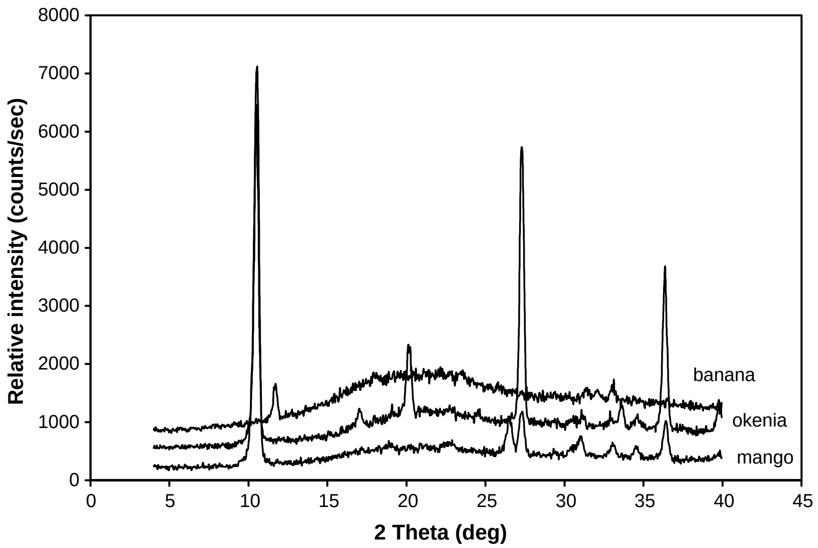

Fig. 3. X-ray diffraction pattern of starch films made by cold gelatinization at 90 days of storage at 25 °C.

same plasticizers (Table 3). In the case of cold gelatinization, starch the source had an effect on the permeability, because films prepared with okenia had a lower value than mango and banana films; however, the films obtained with thermal gelatinization did not show differences ($\alpha = 0.05$). These results are similar to those found for films prepared with mixtures of potato and corn starch (García et al., 1999), corn starches with different amylose content (Garcia et al., 2000a,b), yam starch and glycerol (Mali et al., 2002), the mixtures gelatin/soluble starch/glycerol (Arvanitoyannis et al., 1997) and sodium caseinate/soluble starch/glycerol (Arvanitoyannis & Biliaderis, 1998). However, a lower value was determined in films prepared with native potato starch and acetylated monoglyceride (Petersson & Stading, 2005).

### 3.7. Water solubility

Solubility of edible film indicates their integrity in an aqueous environment, and higher solubility would indicate lower water resistance (Gnanasambandam, Hettiarachy, & Coleman, 1997; Handa, Gennadios, Froning Kuronda, & Hanna, 1999). The gelatinization method had a remarkable effect on water solubility of the films. In the case of thermal gelatinization (Fig. 5a), addition of plasticizer increased water solubility, with statistical differences among the films prepared with the different starches. Okenia film showed the highest solubility (around 32%) and banana films the lowest one (approximately 23%). For control films (without plasticizer), banana and mango films did not present differences ($\alpha = 0.05$), and mango films had the lowest water solubility value. For films prepared with cold gelatinization (Fig. 5b), the water solubility were higher than those obtained with thermal gelatinization. Banana films (with plasticizer and control) were not different ($\alpha = 0.05$) and they had the highest values (approximately 60%). Okenia and mango films with plasticizer were not different (around 57%) and the same pattern was obtained for their control films (around 44%). Water solubility values of highly carboxymethylated starch films (Kim, Ko, & Park, 2002) was similar to the value obtained in this work for banana film with cold gelatinization. For these films with high water solubility, their application may be as edible film and candy wrap edible film, which should be dissolved quickly while they melt softly in the mouth (Perez-Gago, Nadaud, & Krochta, 1999; Sothornvit & Krochta, 2000).

### 3.8. Mechanical properties

The tensile strength (TS) of the films prepared from diverse starch sources is presented in Fig. 6a. The gelatinization method is important in this mechanical property of the films, because cold gelatinization decreased

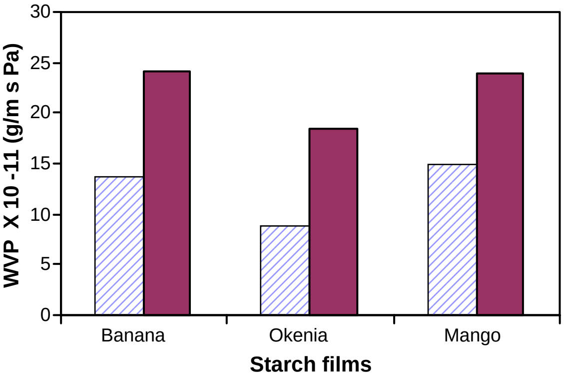

Fig. 4. Water vapor permeability of starch-based films prepared with two methods. ■, cold gelatinization; ▨, thermal gelatinization.

Table 3
Mechanical properties and water solubility of hydrophilic films

| Polymer | Tensile strength (MPa) | Elongation (%) | Water vapor permeability (g/m s Pa)×10 $^{-11}$ | Reference |
|---|---|---|---|---|
| Medium amylose starch from potato, cold gelatinization | Nd | Nd | 36.6 | García et al., 1999. |
| High amylose corn starch, cold gelatinization | Nd | Nd | 26.3 | García et al., 1999. |
| Yam starch, glycerol 50% | Nd | Nd | 18.10 | Mali et al., 2002. |
| Corn starch, glycerol (20 g/L) cold gelatinization | Nd | Nd | 25.7 | García et al., 2000a,b |
| High amylose corn starch, glycerol (20 g/L), cold gelatinization | Nd | Nd | 21.4 | García et al., 2000a,b. |
| Gelatin/ soluble starch (1:1), glycerol 25% | 35.8 | 38.0 | 19.3 | Arvanitoyannis et al., 1997. |
| Native potato starch and acetylated monoglyceride | 29.9 | 2.3 | 0.24 | Petersson and Stading, 2005. |
| Soluble starch / sodium caseinate (1:1) glycerol 30% | 22.5 | 26.8 | 15.1 | Arvanitoyannis and Biliaderis, 1998. |
| Thermoplastic starch and 10% vol. cellulose fibres | 2.2 | 0.22 | Nd | Averous and Boquillon, 2004. |
| Yam starch/ glycerol 3.3:1.3 | 7.84 | 10.3 | Nd | Mali et al., 2005 |
| Highly carboxymetylated starch glycerol | 9.7 | 7.7 | Nd | Kim et al., 2002 |

Nd, Not determined.

TS compared with thermal gelatinization. This behavior is related with the higher thickness of the films obtained with thermal starch gelatinization. Banana films with thermal gelatinization had the highest TS value (approximately 25 MPa), however, mango and okenia films did not differ significantly ($\alpha = 0.05$).

This behavior is similar to those found by Myllärinen, Partanen, Seppälä, & Forsell, 2002, they worked with amylose and amylopectin films with glycerol.

For cold gelatinization, okenia films had the lowest TS value, but banana and mango were similar ($\alpha = 0.05$). Kim et al. (2002) reported TS values for highly carboxymethylated starch films and glycerol at different concentrations that ranged between 9.7–15.3 MPa, and this mechanical property was higher when the plasticizer concentration decreased; values that were lower than those found in this work (Table 3). Higher TS values were obtained in films prepared with mixtures Gelatin/soluble starch (Arvanitoyannis et al., 1997) and native potato starch/acetylated monoglyceride (Petersson & Stading, 2005). Using a thermoplastic starch and cellulose fibres, TS was lower than those determined in this study (Averous & Boquillon, 2004). Also, a TS value of 7.84 MPa was obtained in films prepared with a starch isolated from a non-conventional source (Mali et al., 2005).

The elastic modulus of starch films (Fig. 6b) prepared with cold gelatinization were smaller than those made with thermal gelatinization; the film of okenia starch had the smallest value.

Puncture values (Fig. 6c) were higher in films obtained by thermal gelatinization, without statistical differences ($\alpha = 0.05$) among the three starches. The maximum force for penetrating these films was in average 4.5 N. Films prepared with cold gelatinization, did not show differences ($\alpha = 0.05$) in puncture values, but they were lower than those of thermal gelatinization (average 2 N). Very few studies with starch films have reported this parameter that is important to determine the force needed to penetrate the film.

The elastic modulus of the puncture strength (Fig. 6d), was higher when starch films were prepared using the thermal gelatinization method, with no effect of starch source, but when using the cold gelatinization method, it could be seen that okenia has the smallest value.

The percent of elongation is shown in Fig. 6e. In general, films elaborated by the thermal gelatinization were greater than those of cold gelatinization but in the case of okenia

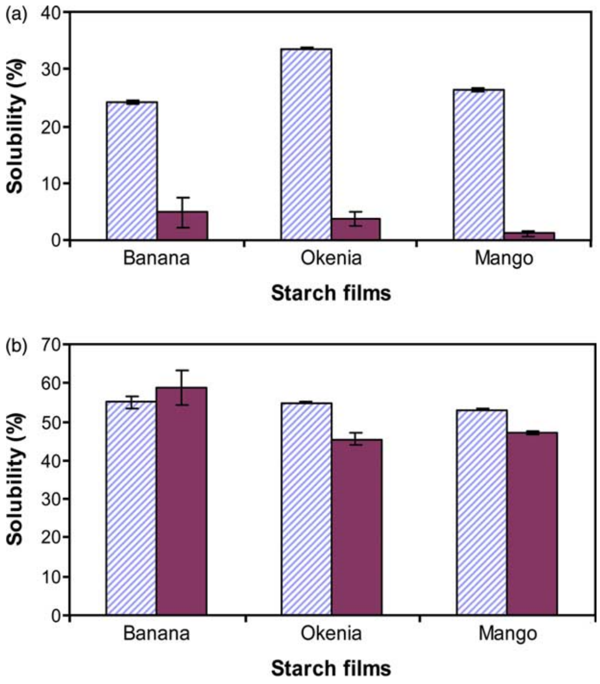

Fig. 5. Water solubility of films prepared with starches from non-conventional sources: (a) thermal gelatinization (b) cold gelatinization. ▨, control; ■, glycerol 50%.

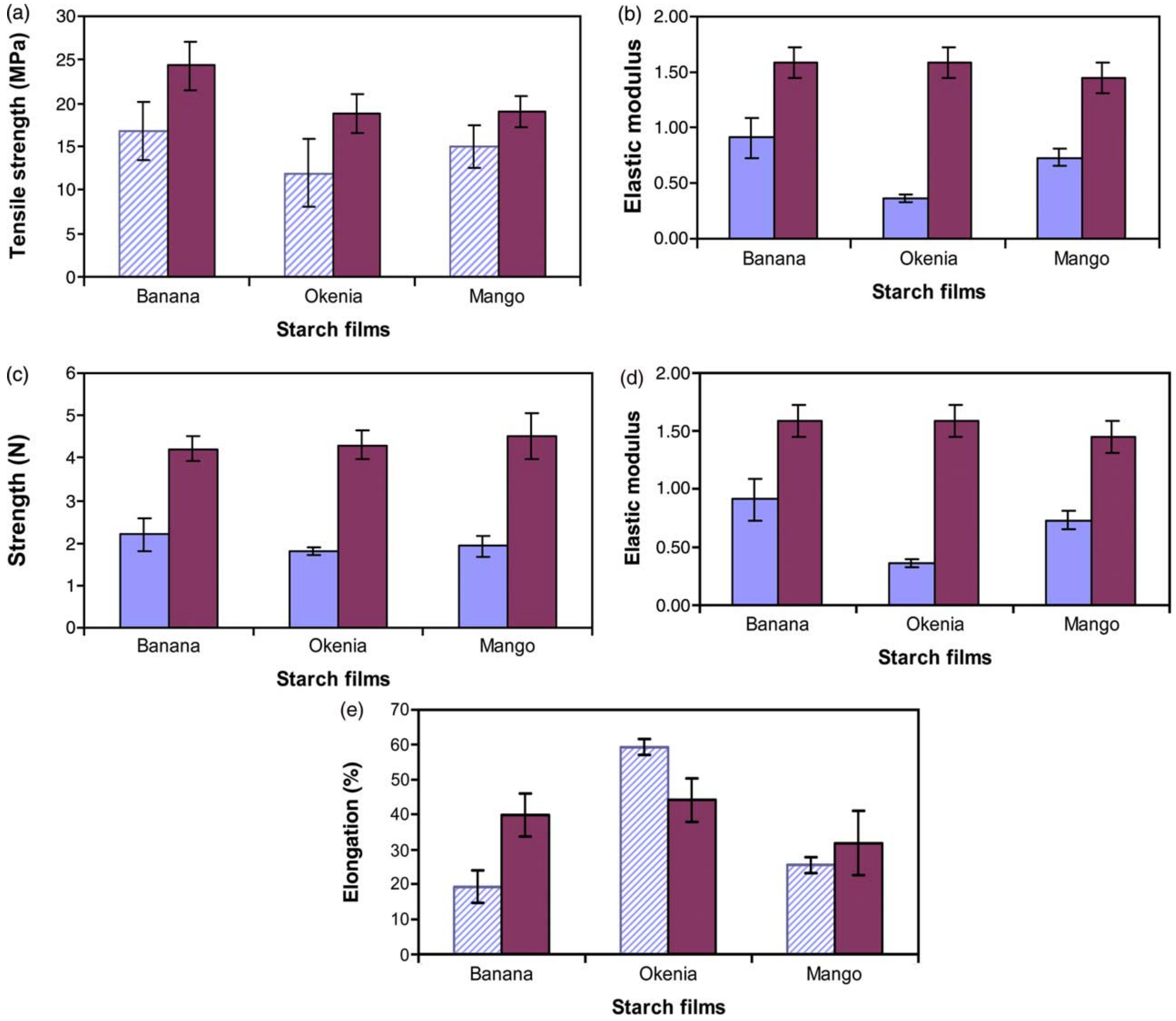

Fig. 6. Mechanical properties of starch films made with two gelatinization methods: (a) tensile strength of banana, okenia and mango starch. ▨, cold gelatinization; ■, thermal gelatinization., (b) elastic modulus of tensile strength in starch films, (c) puncture strength of starch, (d) elastic modulus of puncture strength, (e) elongation at break of banana, okenia and starch films.

film, the greatest value was found by the cold gelatinization method. The starch source plays an important role on this mechanical property, perhaps the small granule size of okenia starch plays an important role in this parameter. It has been reported that glycerol concentration increased *E* values, an inverse pattern to that found for TS (Kim et al., 2002). Higher elongation percentages were measured in films prepared with mixtures gelatin/soluble starch (Arvanitoyannis et al., 1997) and sodium caseinate/soluble starch (Arvanitoyannis & Biliaderis, 1998) (Table 3). However, lower elongation percentages were determined in films elaborated with different starch sources (Table 3). Plasticizers are added to polymers to reduce brittleness, since they work as spacers between polymer chains, decreasing intermolecular forces and thus increasing flexibility and extensibility of polymers (Lai & Padua, 1998).

## 4. Conclusions

Native starches isolated from non-conventional sources showed different shapes and sizes; okenia starch had the smallest size and the lowest gelatinization temperature, important parameters in films preparation. Starch-based films prepared with the thermal gelatinization method, had higher thickness than those prepared with cold gelatinization one; starch source playing an important role in these values. Starch films prepared with thermal gelatinization showed homogeneous surfaces without pores or cracks compared with starch films made with cold gelatinization, this integrity is important in the mechanical properties of the films. After storage, the films studied presented a B-type (thermal gelatinization) and A-type (cold gelatinization) X-ray diffraction pattern. The differences found in the physicochemical parameters of the films prepared with the two gelatinization methods, had influence in water vapor permeability and water solubility. Banana films made with thermal gelatinization method had the highest TS value; however, mango and okenia films did not differ significantly. Elastic modulus, puncture and elongation values were higher in films obtained by thermal gelatinization. Starch source plays an important role on these mechanical properties. Starches isolated from non-conventional sources can be used for films preparation with adequate physicochemical, mechanical and barrier properties.

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