



## Effect of the concentration of starch and clove essential oil on the physicochemical properties of biodegradable films

### Efecto de la concentración de almidón y de aceite esencial de clavo de olor sobre las propiedades fisicoquímicas de películas biodegradables

O.I. Buso-Rios<sup>1</sup>, G. Velazquez<sup>2</sup>, L. Jarquín-Enríquez<sup>1</sup>, N.L. Flores-Martínez<sup>1\*</sup>

<sup>1</sup>Universidad Politécnica de Guanajuato. Avenida Universidad Sur 1001, Comunidad Juan Alonso, Cortazar, Gto., C.P. 38483. México.

<sup>2</sup>Instituto Politécnico Nacional. CICATA-IPN unidad Querétaro, Cerro Blanco No. 141, Col. Colinas del Cimatario, C.P. 76090, Santiago de Querétaro, Querétaro, México.

Received: November 4, 2019; Accepted: January 24, 2020

#### Abstract

Biodegradable films were obtained with the casting method using different concentrations of purple sweetpotato *Ipomoea batatas* (L.) starch and clove *Syzygium aromaticum* (L.) essential oil. Mechanical properties (tensile strength, percentage of elongation and Young's modulus), water vapor permeability and film adsorption isotherms were evaluated. In the formulations containing the intermediate concentrations of the components, a homogeneous surface, an improvement in fracture stress and a decrease in water vapor permeability were observed. The statistical analysis showed a significant difference in the mechanical properties of the composite films ( $p < 0.05$ ). The GAB model was used to adjust the experimental data of the adsorption isotherms and a higher moisture content of the films containing the highest concentration of starch and essential oil was observed. The F6 (starch 3.5%, clove essential oil 60 mg/L) formulation showed the lowest water vapor permeability values ( $1 \times 10^{-8}$  g/m s Pa) and the lowest moisture content in the monolayer, which is considered to be the material with the best characteristics for its possible application, avoiding the excessive use of raw material during its preparation.

**Keywords:** edible films, starch, essential oil, mechanical properties, GAB model, adsorption isotherms.

#### Resumen

Se elaboraron películas biodegradables por el método de vaciado en placa utilizando diferentes concentraciones de almidón de camote morado *Ipomoea batatas* (L.) y aceite de clavo de olor *Syzygium aromaticum* (L.). Evaluando propiedades mecánicas (tensión a la fractura, deformación y módulo de Young), permeabilidad al vapor de agua e isoterma de adsorción de las películas. En las formulaciones que contenían las concentraciones intermedias de los componentes, se observó una superficie homogénea, una mejora en la tensión a la fractura y una disminución de la permeabilidad al vapor de agua. El análisis estadístico mostró una diferencia significativa en las propiedades mecánicas de las películas compuestas ( $p < 0.05$ ). Se utilizó el modelo de GAB para ajustar los datos experimentales de las isoterma de adsorción y se observó un mayor contenido de humedad de las películas que contenían la mayor concentración de almidón y de aceite esencial. La formulación F6 (almidón 3.5%, aceite esencial de clavo 60 mg/L), mostró los valores menores de permeabilidad al vapor de agua ( $1 \times 10^{-8}$  g/m s Pa) y el menor contenido de humedad en la monocapa por lo que se considera como el material con mejores características para su aplicación evitando el uso excesivo de materia prima.

**Palabras clave:** película comestible, almidón, aceite esencial, propiedades mecánicas, modelo GAB, isoterma de adsorción.

## 1 Introduction

Currently, there are different techniques to extend the shelf-life of food products, such as vacuum packaging, cold storage and modified atmospheres or other techniques that help to provide a barrier to

water vapor and gases, as well as some mechanical protection. In recent years, packaging materials with desired mechanical properties and excellent durability have been developed, mostly non-biodegradable materials, which has caused negative impact on the environment. This has encouraged studies in the field of biodegradable or edible films and coatings

\* Corresponding author. E-mail: nlflores@upgto.edu.mx  
Tel. Tel. (461) 44 14 300, Ext. 4305  
<https://doi.org/10.24275/rmiq/Alim965>  
issn-e: 2395-8472

(Albertos *et al.*, 2019; Zheng *et al.*, 2019 and Sánchez *et al.*, 2015). On the other hand, as the raw material comes from renewable, abundant and economical sources, the industrial use is feasible. For this reason, several studies have been carried out using polysaccharides as a raw material for the formulation of edible films. In this sense, there is an increasing interest in manufacturing packaging materials using agro-industrial waste since they can help to delay the deterioration and decomposition of fresh products and, at the same time, do not cause environmental deterioration.

Over the years, these bio-packaging materials have been used to prolong the shelf-life of fresh foods, but it has been considered that those edible films based on a single component usually do not have good properties. These deficiencies could be overcome using additives or formulations that include hydrocolloid materials, plasticizers, emulsifiers or surfactants, resulting in a better structured polymer matrix with improved characteristics. The formulation of each edible film depends on the requirements of the intended application. The selection of biopolymers determines the mechanical and barrier properties of the films. There are different materials that can be used to produce biodegradable films including proteins, lipids and carbohydrates (gums, starch, cellulose derivatives and pectins) or the combination between them (Hassan *et al.*, 2018; Sánchez *et al.*, 2015; Domínguez and Jiménez, 2012). One of the most used biopolymers is starch, since it can be found in different sources such as cereals, legumes and in tubers including sweet potatoes, potatoes and cassava. Among the native tubers that contain a high percentage of starch is sweet potato having between 60 to 90% starch. The purple variety with white pulp *Ipomoea batatas* (L.), is an ancient traditional crop in Mexico and America. Various polymers have been used in the formulation of bio-packages including proteins (grenetin) and carbohydrates (starch). The use of these polysaccharides as raw materials for the formation of edible films results in moderate mechanical properties. However, applications are limited because the films are water soluble and permeable to water vapor. To improve these properties, it is possible to add hydrophobic compounds such as waxes or oils (clove oil) since lipids cannot form a film by themselves, and a plasticizer (glycerol) to give flexibility to the structure (Dash *et al.*, 2019; Dehghani *et al.*, 2018; Campos *et al.*, 2011; Vargas *et al.*, 2008). The present work aims to develop and characterize an edible film

prepared from different concentrations of sweet potato starch *Ipomoea batatas* (L.) and clove oil *Syzygium aromaticum* (L.).

## 2 Materials and methods

---

### 2.1 Materials

The starch was extracted from the sweet potato variety with white pulp *Ipomoea batatas* (L.). Gelatin Knox<sup>TM</sup> was provided by Con Alimentos S.A de C.V (Mexico City, Mexico), Glycerol ACS (catalog number 06441) was supplied by Productos Químicos de Monterrey, S.A de C.V. (Monterrey, N.L., Mexico) and the clove essential oil was extracted by hydrodistillation of *Syzygium aromaticum* (L.).

### 2.2 Methods

#### 2.2.1 Film preparation

The film-forming solution (Table 1) was prepared following the same procedure for each formulation, in the case of starch concentration, it was determined by bibliographic analysis, taking as reference the work of Müller *et al.* (2011) using starch concentrations in order to ensure a high amylose content to facilitate the formation of resistant and firm films. Sweet potato starch was dispersed in 90 mL of distilled water and the dispersion was taken to a boiling bath with constant stirring. Grenetin was added gradually (2%), simultaneously, water (10 mL) was added to incorporate with the starch, mixed with glycerol 0.5% w/w and finally the clove oil was added. The temperature was monitored at 80 °C for 20 minutes, maintaining constant agitation. To obtain the films, 100 mL of the solution were poured into Petri dishes and dried in a convection oven for 1 h at 55 °C, then, to finish drying, the films were placed in an area delimited by a mesh for 48 h at room temperature. After drying, the films were peeled off and stored in plastic sheet protectors.

#### 2.2.2 Films thickness

The film thickness (mm) was measured using a handheld digital micrometer (F1000/30-3, Käfer Messuhrenfabrik GmbH & Co., Villingen-Schwenningen, Germany) with a resolution of 0.001 mm.

Table 1. Code and components of biodegradable film based on starch and clove essential oil.

Code of edible films	Starch (g)	Grenetin (g)	Clove essential oil (mg/ L)	Glycerol (g)
C1	0	2	0	0.5
F1	2	2	30	0.5
F2	2	2	45	0.5
F3	2	2	60	0.5
F4	3.5	2	30	0.5
F5	3.5	2	45	0.5
F6	3.5	2	60	0.5
F7	5	2	30	0.5
F8	5	2	45	0.5
F9	5	2	60	0.5

Measurements were performed at ten random locations in the film and the mean thickness value was used in the calculation of the permeability and mechanical properties.

### 2.2.3 Solubility

The solubility was determined following the methodology reported by Palma *et al.*, (2017). Samples of 2 x 3 cm were cut from each film and stored in a desiccator with silica gel at 0% RH, for 7 days. The weight of the films was recorded and subsequently placed in a glass bottle with distilled water (80 mL) on a heating plate (60 °C) and stirred at approximately 125 rpm for 5 minutes. The solution was filtered and dried in a convection oven at 60 °C for 2 h. The dried film was weighed and finally the percentage of soluble matter (% solubility) was determined. The test was carried out for each formulation with 3 replicates.

### 2.2.4 Water adsorption isotherms

The water adsorption isotherms were obtained according to with the ASTM E104-02 method. Samples were cut with dimensions of 3 x 3 cm (in triplicate), placed in sealed acrylic containers, each one containing a supersaturated solution from 11 to 90% of relative humidity of equilibrium at 30 °C (Table 2). The film samples were weighed using an analytical balance (Model VE-303). After 5 days that the samples showed a stable weight indicating an equilibrium state, they were dried until obtaining a constant weight to calculate the equilibrium moisture content of the films in each microclimate. The moisture content in the monolayer ( $X_m$ ) was calculated using GAB model.

### 2.2.5 Water vapor permeability

The water vapor permeability (WVP) was determined in accordance with the ASTM E96-10 method. In a glass cell containing distilled water (100% RH) with an internal diameter of 30.3 mm, the cut-out films with a diameter of 54 mm were placed between two silicon gaskets sealing the permeability cell. The cell and the film were placed on the balance plate inside of a permeability chamber containing silica gel (0% HR) with temperature controlled at 30 °C. The weight change was recorded automatically every minute for 8 h. The slope of the weight loss ( $m$ ), the transfer area ( $A$ ), the pressure difference in both sides of the film ( $\Delta P$ ) = 3183.8 Pa corresponding to 30 °C and the thickness ( $l$ ) were used to calculate the water vapor permeability (Eq. 1).

$$\text{Water vapor Permeability} = \frac{m \cdot l}{A \cdot \Delta P} \left( \frac{g}{msPa} \right) \quad (1)$$

### 2.2.6 Mechanical properties

The tensile strength, percentage of deformation and Young's modulus were evaluated to measure the mechanical properties of the films. The samples were cut into rectangles of 1 x 8 cm with a precision cutter in accordance with the specifications of the ASTM D882-02 method and preconditioned in a microclimate containing an oversaturated NaBr solution (57% RH) for 5 days. Previously, the thickness of each film was determined at 10 random points. The mechanical properties were analyzed in a texturometer (Texture Analyzer TA plus) using mechanical clamps with an initial separation of 60 mm and operating at a crosshead speed of 2 mm/s. Three replicates were evaluated for each formulation.

Table 2. Preparation of salts to obtain the relative humidity of equilibrium.

Salt	Salt amount(g)	Water volume (mL)	Relative humidity (%RH)
LiCl	200	70	11.15
MgCl <sub>2</sub> 6H <sub>2</sub> O	200	25	32.73
K <sub>2</sub> CO <sub>3</sub> 2H <sub>2</sub> O	200	90	43.30
NaBr	200	70	57.70
NaCl	200	50	75.32
BaCl <sub>2</sub> 2H <sub>2</sub> O	200	60	90.26

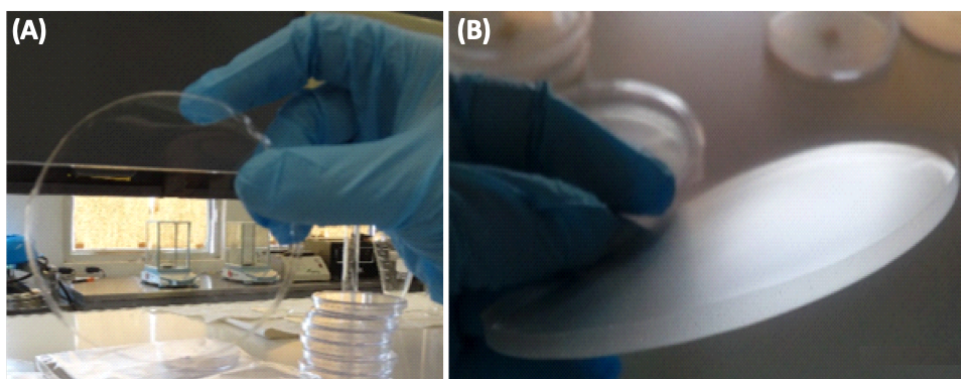


Figure 1. Appearance of biodegradable films. (A) based on grenetin and glycerol. (B) All formulations added with starch and clove essential oil.

### 2.2.7 Color properties

Color values of the films were determined using a colorimeter (CR-400, Minolta, Konica Ramsey, NJ, USA) by the method described by Thakur *et al.* (2017) and Zuo *et al.* (2019), with some modifications. The color scale was used for measuring color parameters: L\*, the lightness variable; a\*, from green to blue; and b\*, from yellow to red. The total color difference ( $\Delta E$ ) was calculated using Eq. (2):

$$\Delta E = \sqrt{\Delta a^2 + \Delta b^2 + \Delta L^2} \quad (2)$$

where  $\Delta E = L^*_{standard} - L^*_{sample}$ ,  $\Delta a = a^*_{standard} - a^*_{sample}$  and  $\Delta b = b^*_{standard} - b^*_{sample}$ . The standard plate (calibration plate L\* = 94.90 a\* = -0.45 b\* = 3.93) was used as a standard. The film was cut with a diameter of 54 mm and covered. Five measurements were collected on each film, one at the center and four around the perimeter.

### 2.2.8 Statistical analysis

The studied factors were sweet potato starch and clove oil concentration and a factorial design 3<sup>2</sup> was used to analyze the individual behavior of the factors and the interaction between them. The response variables

were tensile strength (TS), percentage of deformation (% E), Young's modulus, water vapor permeability (WVP) and solubility (S). The analysis of variance was carried out using the Statistica Software version 7. The adsorption data were adjusted by the GAB model (Eq. 3), using the solver function in Excel, obtaining the GAB parameters ( $Xm$ ,  $C$  and  $k$ ) and the value of R<sup>2</sup>.

$$m = \frac{XmCk a_w}{[(1 - k a_w)(1 - k a_w + C k a_w)]} \quad (3)$$

## 3 Results and discussion

### 3.1 Characterization of the films

#### 3.1.1 Films preparation

The formulations, coded from F1 to F9, visually have a similar appearance. The film C1, with no sweet potato starch and clove oil, became thinner and translucent (Figure 1). The results are consistent with those reported by Rodríguez *et al.* (2007) who mentioned that, due to the retrogradation of the starch granule, as the temperature increases, the hydrocolloid film will become opaque.

Table 3. Total color difference of biodegradable film as a function of starch and clove essential oil.

Code	Starch (g)	Clove essential oil (mg/L)	Total color difference ( $\Delta E$ )
C1	0	0	$86.10 \pm 1.63^{abc}$
F1	2	30	$87.12 \pm 0.11^{ab}$
F2	2	45	$86.69 \pm 0.22^{abc}$
F3	2	60	$86.55 \pm 0.20^{abc}$
F4	3.5	30	$86.11 \pm 0.15^{abc}$
F5	3.5	45	$87.70 \pm 0.08^a$
F6	3.5	60	$86.86 \pm 0.30^{abc}$
F7	5	30	$85.31 \pm 0.04^c$
F8	5	45	$85.93 \pm 0.47^{bc}$
F9	5	60	$86.16 \pm 0.54^{abc}$

Results are expressed as the mean  $\pm$  SD of triplicates.

Means with a common letter are not significantly different ( $p > 0.05$ )

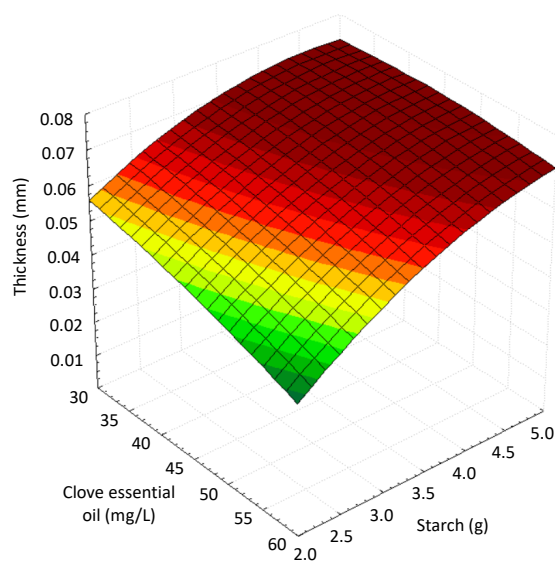


Figure 2. Thickness of biodegradable film as a function of starch and clove essential oil.

### 3.1.2 Films thickness

According to ASTM-D882-02, the thicknesses should be less than one mm for flexible films. Figure 2 shows the thicknesses as a function of the starch and clove essential oil concentrations. The formulation with the highest concentration of starch and clove essential oil showed a significant difference (5% - 60 mg/L). Thickness depends on the film composition. According to Wu *et al.* (2013), Wittaya (2012) and Pajak *et al.* (2019) the structure of amylose network is very stable with strong molecular orientation and forms denser films than amylopectin.

### 3.1.3 Total color difference of films

Table 3 shows the total color difference for each of the formulations. The values indicate is that the concentration of starch has a slightly significant effect to produce opacity in the film, while clove essential oil has no effect on the opacity of the film.

### 3.1.4 Solubility

Results of solubility behavior show that as the concentration of solutes increases the solubility decreases. There are natural factors of the solute and the solvent that promote the interaction between the atoms of the solute and the solvent separately affecting their percentage of solubility. Bertuzzi *et al.* (2006) mentioned that an increase in temperature causes a slight decrease in solubility and an increase in the diffusion of water vapor through the film. This increase in diffusivity is due to a greater movement of the polymers and the increase in the energy levels of the molecules.

## 3.2 Characteristics of the films

### 3.2.1 Water adsorption isotherms

The performance of the films can be affected when they are subjected to different conditions of relative humidity as in the adsorption isotherms. Figures 3, 4 and 5 show the adsorption isotherms at 30 °C. The experimental data is shown in symbols and the solid line shows the fitted isotherm using the GAB model. As expected, when the  $a_w$  increases, the moisture content in the equilibrium also increases. However, the increase in moisture is greater as the starch content in the film increases and decreases when the clove oil in the formulation increases.

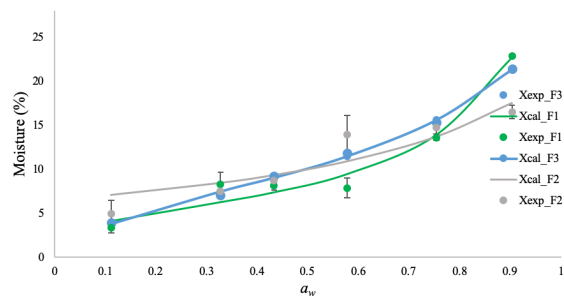


Figure 3. Adsorption isotherms for formulations F1, F2 and F3 (0.5 g of glycerol, 2 g of grenetin, 2 g of starch and 30, 45, and 60 mg / L of clove oil).

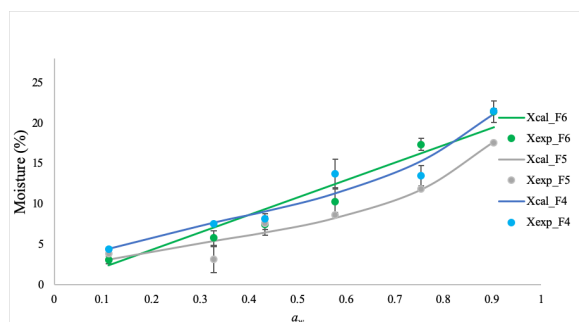


Figure 4. Adsorption isotherms for formulations F4, F5 and F6 (0.5 g of glycerol, 2 g of grenetin, 3.5 g of starch, and 30, 45 and 60 mg / L of clove oil).

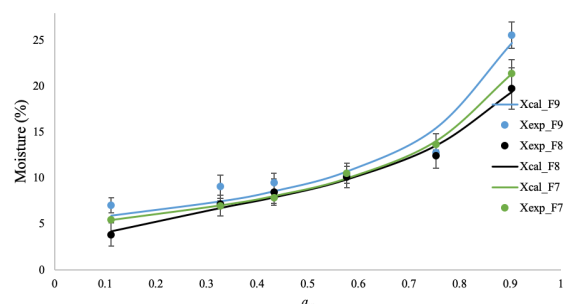


Figure 5. Adsorption isotherms for formulations F7, F8, and F9 (0.5 g of glycerol, 2 g of grenetin, 5 g of starch and 30, 45 and 60 mg / L of clove oil).

The type III adsorption isotherm is distinguished, which is representative of systems where the adsorbate-adsorbent interactions are weak. This type of isotherms can be found in the adsorption of a solvent or plasticizer such as glycerol. At low moisture values, the water is strongly adsorbed to the binding sites of the film surface. As the moisture content increases, due to the swelling of the network, new binding sites are available, causing an increase in

the moisture content in the equilibrium (Chen *et al.*, 2009). The moisture content in the equilibrium of the starch films (2 and 3.5 g) with the different concentrations of clove oil, decreases indicating that the incorporation of the essential oil improves the resistance to water, especially at low content of starch. However, as the starch (5 g) increases, the moisture content in the equilibrium does not decrease with the increase of clove oil.

This behavior could be explained because increasing the starch content promotes the formation of more hydrogen bridges with the hydroxyl groups of the oil making a stronger structure and forming a more water-resistant matrix. This behavior is in agreement with the results of other researchers (Šešlija, 2018). Sorption isotherms are important to predict the stability and quality changes that could occur during packaging and storage of the food product (Srinivasa *et al.*, 2003).

The parameters of the GAB model ( $X_m$ ,  $C$  and  $k$ ) and the  $R^2$  values are shown in Table 4. The adsorption isotherms of food products give important information about their behavior in a wide array of environments from dry to high moisture. GAB model is widely used to describe the sorption isotherms in a wide range of  $a_w$  values. The estimated parameters of  $X_m$  for corn starch films reported by Šešlija *et al.* (2018) show a similar behavior suggesting that the addition of glycerol and clove oil reduces the number of binding sites where the water molecules can interact with the functional groups in the starch chains, reducing the moisture adsorption in starch films (Slavutzky *et al.*, 2012). On the other hand, Hernández-Carrillo *et al.* (2017) mentioned that the incorporation of additives decreased the equilibrium moisture content.

Šešlija *et al.* (2018) also mentioned that the addition order of the components to the filmogenic solution can change the behavior since the first compound incorporated will have the most interaction with the water and this interaction could increase the value of  $X_m$ .

### 3.2.2 Water vapor permeability

Figure 6 shows the behavior of water vapor permeability ( $\text{g/m s Pa}$ ) for the different biodegradable films. A low water vapor permeability value is desirable for a packaging film as it indicates a good performance as a barrier material. As shown in Figure 6, the film with the lowest permeability (30 mg/L, 2 g and 60 mg/L, 3.5 g) had a WVP value of  $1 \times 10^{-8} \text{ g/m s Pa}$ . Although permeability is independent of thickness,

Table 4. GAB parameters estimated for biodegradable film as a function of starch and clove essential oil.

Code	Starch (g)	Clove essential oil (mg/ L)	Xm	C	K	R <sup>2</sup>
F1	2	30	4.82	30.82	0.87	0.96
F2	2	45	6.51	8.54	0.69	0.96
F3	2	60	7.5	9.7	0.73	0.96
F4	3.5	30	6.89	15.57	0.75	0.99
F5	3.5	45	4.66	15.62	0.82	0.91
F6	3.5	60	3.31	4.34	0.001	0.99
F7	5	30	5.17	20.37	0.83	0.99
F8	5	45	5.66	21.96	0.79	0.93
F9	5	60	5.38	17.43	0.86	0.99

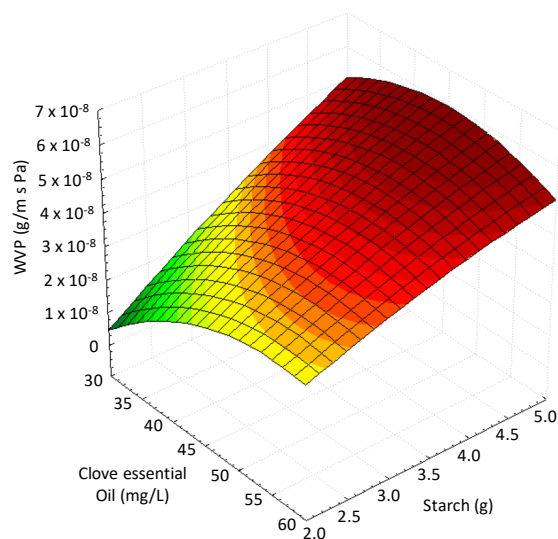


Figure 6. Water vapor permeability (WVP) of biodegradable film as a function of starch and clove essential oil.

the permeance, related with the performance of the film when used as packaging materials, is dependant of thickness. Salama *et al.* (2019) reported that the water vapor permeability was reduced upon increasing the frankincense essential oil content. Hashemi and Mousavi-Khaneghah (2017) and Abdel-Aziz *et al.* (2018) reported the WVP was decreased significantly by incorporation of oregano essential oil and ricinus essential oil respectively. Thakur *et al.* (2017) reported that increasing the starch in the formulation, the films were thicker; on the other hand, Moradi *et al.* (2016) the thickness of the films was not changed, contrary, increasing OEO contents from 1 to 2% significantly increased the thickness of quince seed mucilage films. In general, including essential oils into the formulations of biodegradable films decreases the water vapor permeability. The essential oil increases the hydrophobicity interrupting the continuity of the

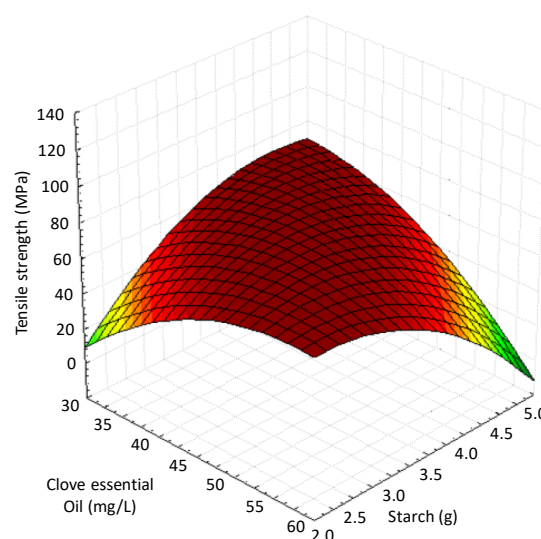


Figure 7. Tensile strength of biodegradable film as a function of starch and clove essential oil.

structure for mass transfer through the polymer matrix. Zuo *et al.* (2019) mentions a very important point "high permeability is not recommended for packaging applications".

### 3.2.3 Mechanical properties

The tensile strength refers to the maximum resistance to stress that a film can withstand while being stretched before its rupture, and depends on the elaboration conditions of the film (López-Hernández *et al.*, 2018), for this reason in the figure 7 shows the behavior of the tensile strength of the films with different starch and clove oil content. The concentration of clove oil (30, 45 and 60 mg / L) is shown on the X axis; the concentration of sweet potato starch (2, 3.5 and 5 g) is observed in the Z axis and in the axis Y the tensile strength is expressed in MPa. The 3D surface plot shows a different behavior with the

combination of these two compounds. The red area of the surface reflects the highest point of tensile strength. This behavior is given by the formulations with high content of clove oil and low concentration of starch, resulting in high tensile strength values. When a material is subjected to a certain force and deformation in the elastic region, the response of the material is proportional to the applied force. At this point, if the applied force is removed, the material will return to its original form.

Cao *et al.* (2018) they showed that the initiation of glycerol or sorbitol played not only as a plasticizer but also as a crosslinker. An increase in glycerol and sorbitol variations also made the films more opaque and improved their thermal stability and Mali *et al.* (2005) conducted a study on the behavior of mechanical properties as a function of plasticizers (sorbitol and glycerol) added to cassava starch-based films. The authors mention that the stress decreases when the two plasticizers are incorporated but with glycerol, due to its composition, shows a greater decrease in stress. This is consistent with a study conducted by Jongjareonrak *et al.* (2006) where the properties of edible films, formulated with different concentrations of glycerol, polyethylene glycol, ethylene glycol and sorbitol, were measured. In general, films formulated with glycerol showed greater tensile strength. In all cases, increasing the concentration of plasticizer increased the elongation percentage and the tensile strength was reduced. Because the plasticizer reduces internal hydrogen bonds while increasing intermolecular spacing of polymers, plasticizers modify mechanical properties and produce more flexible films thus decreasing their tensile capacity (Cao *et al.*, 2018; Campos *et al.*, 2011; Liberman and Gilbert, 1973).

The percentage of deformation of the different formulations is shown in Figure 8. The behavior of the 3D surface shows that the formulations with a high concentration of oil and starch (60 mg / L with 3.5 and 5 g) had the highest %E values; however, the percentage obtained from deformation is too low, same behavior reported Flores-Martínez *et al.* (2017).

Šešlija *et al.* (2018) reported that in corn starch films incorporated with an additive, strong interactions by hydrogen bonds with starch are created, improving tensile strength and Young's modulus, but in turn, affecting the flexibility of starch chains by decreasing the deformation value. This behavior has been observed by other researchers in starch films (Huang *et al.*, 2006; Chung *et al.*, 2010; Müller *et al.*, 2011).

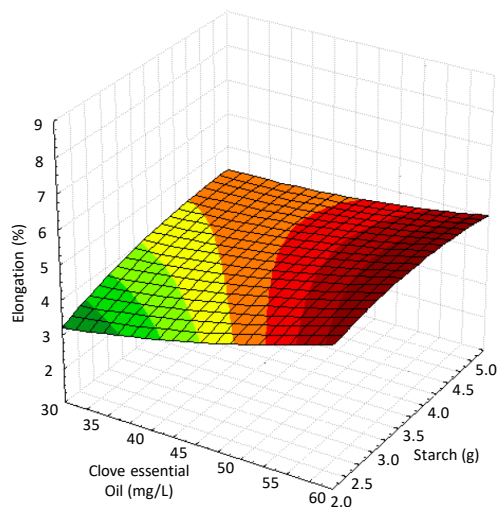


Figure 8. Percentage of elongation of biodegradable film as a function of starch and clove essential oil.

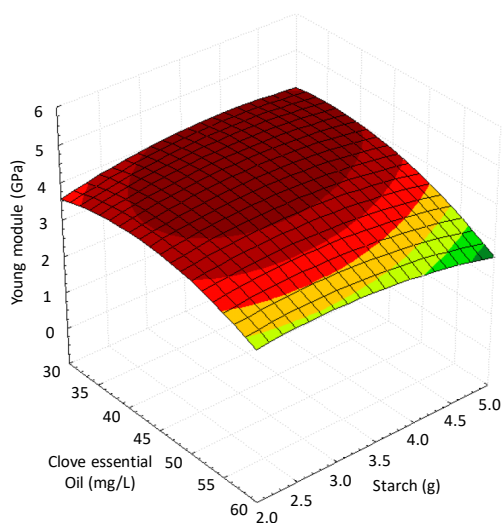


Figure 9. Young module of biodegradable film as a function of starch and clove essential oil.

Figure 9 shows the behavior of Young's module, which represents the stiffness of the film. This value has a similar behavior than that of tensile strength because the more rigid a material is, the more force it will take to break it. Olivas and Barbosa (2005) mention that a plasticizer can be added to avoid brittle films when biopolymers are used as a raw material. Young's modulus is independent of the effort as long as it does not exceed a maximum value called the elastic limit.



## Conclusions

Edible films based on sweet potato starch *Ipomoea batatas* (L.) and clove essential oil *Syzygium aromaticum* (L.) were able to form an organized structure that resulted in edible films.

The physicochemical characterization demonstrated the stability of the polymeric chains that formed the edible film contributing to the behavior of solubility, transparency, mechanical properties, adsorption isotherms and water vapor permeability. The final properties suggest that the films could be used as a food packaging material.

The formulation containing F6 (starch 3.5%, clove essential oil 60 mg/L), showed the lowest water vapor permeability value ( $1 \times 10^{-8}$  g /m s Pa) and a low moisture content in the monolayer, making this material the best option in applications intended to reduce the moisture migration.

## Acknowledgements

The first author thanks the support of the Universidad Politécnica de Guanajuato (UPG) for the scholarship to study the Maestría en Ciencias en Biotecnología Agroalimentaria in the postgraduate program of the UPG.

## Nomenclature

l	Thickness, mm
t	Time, h
S	Solubility, %
TS	Tensile strength, MPa
%E	Percentage of elongation, %
MY	Young's modulus, MPa
GAB	GAB model
WVP	Water vapor permeability
Xm, C, K	GAB parameters
Xm	Moisture content
RH	Relative humidity
A	transfer area, m <sup>2</sup>

## Greek symbols

$\Delta P$	Water vapor pressure differential (3167 Pa at 25 °C)
$\Delta m$	Weight differential, g
$\Delta L^*$	lightness
$\Delta a^*$	Change from green to blue
$\Delta b^*$	Change from yellow to red
$\Delta E$	Color differential

## References

- Abdel Aziz, M. S., Salama, H. E., and Sabaa, M. W. (2018). Biobased alginate/castor oil edible films for active food packaging. *Food Science and Technology* 96, 455–460. <https://doi.org/10.1016/j.lwt.2018.05.049>
- Albertos, I., Martin-Diana, A., Burón, M. and Rico, D. (2019). Development of functional bio-based seaweed (*Himanthalia elongata* and *Palmaria palmata*) edible films for extending the shelflife of fresh fish burgers. *Food Packaging and Shelf Life* 22, 1-9. <https://doi.org/10.1016/j.fpsl.2019.100382>
- ASTM D882. (2002). Tensile properties of thin plastic sheets. American Society for Testing and Materials. U.S.
- ASTM E96. (2010). Transmission of water vapor materials. American Society for Testing and Materials. U.S.
- ASTM E104. (2002). Maintenance of the relative humidity by mean of aqueous solutions. American Society for Testing and Materials. U.S.
- Bertuzzi A, Castro F, Armada M., Gottifredi C. (2006). *Water Vapor Permeability of Edible Starch Based Films*. Elsevier Ltd. All rights reserved. <http://doi:10.1016/j.jfoodeng.2006.07.016>.
- Campos, C. Gerschenson, L., Flores, S. (2011). Development of edible films and coatings with antimicrobial activity. *Food Bioprocess Technology* 4, 849-875. <https://doi.org/10.1007/s11947-010-0434-1>
- Cao, L., Liu, W., and Wang, L. (2018). Developing a green and edible film from *Cassia* gum: The effects of glycerol and sorbitol. *Journal of Cleaner Production* 175, 276–282. <https://doi.org/10.1016/j.jclepro.2017.12.064>
- Chen, Y., Liu, C., Chang, R., and Cao, I. and Anderson, D. (2009). Bio nanocomposites based on pea starch and cellulose Nano whiskers hydrolyzed from pea hull fibre: Effect of hydrolysis time. *Carbohydrate Polymers*

- 76, 607-615. <https://doi.org/10.1016/j.carbpol.2008.11.030>
- Chung, Y., Ansari, S., Esteves, L., Hayrapetyan, S., Giannelis, E. and Lai, H. (2010). Preparation and properties of biodegradable starch-clay nanocomposites. *Carbohydrate polymers* 79, 391-396. <https://doi.org/10.1016/j.carbpol.2009.08.021>
- Dash, K. K., Ali, N. A., Das, D., Mohanta, D. (2019). Thorough evaluation of sweet potato starch and lemon-waste pectin based-edible films with nano-titania inclusions for food packaging applications. *International Journal of Biological Macromolecules* 139, 449-458. <https://doi.org/10.1016/j.ijbiomac.2019.07.193>
- Dehghani, S., Hosseini, S. V., and Regenstein, J. M. (2018). Edible films and coatings in seafood preservation: A review. *Food Chemistry* 240, 505-513. <https://doi.org/10.1016/j.foodchem.2017.07.034>
- Domínguez, C. and Jiménez, M. (2012). Películas comestibles con polisacáridos: propiedades y aplicaciones. *Temas Selectos de Ingeniería de Alimentos* 6-2, 110-121.
- Flores-Martínez, N. L., Valdez-Fragoso, A., Jiménez-Islas, H. and Pérez-Pérez, M. C. (2017). Physical, barrier, mechanical and microstructural properties of Aloe vera-gelatin-glycerol edible films incorporated with *Pimenta dioica* L. Merril essential oil. *Revista Mexicana de Ingeniería Química* 16(1), 109-119. <http://www.rmiq.org/iqfvp/Pdfs/Vol.%2016,%20No.%201/Alim5/eAlim5.html>
- Hassan, B., Chatha, S. A. S., Hussain, A. I., Zia, K. M., and Akhtar, N. (2018). Recent advances on polysaccharides, lipids and protein based edible films and coatings: A review. *International Journal of Biological Macromolecules* 109, 1095-1107. <https://doi.org/10.1016/j.ijbiomac.2017.11.097>
- Hashemi, S. M. B., and Mousavi Khaneghah, A. (2017). Characterization of novel basil-seed gum active edible films and coatings containing oregano essential oil. *Progress in Organic Coatings* 110, 35-41. <https://doi.org/10.1016/j.porgcoat.2017.04.041>
- Hernández-Carrillo, J. G., Mújica-Paz, H., Welti-Chanes, J., Spataffora-Salazar, A. S. and Valdez-Fragoso, A. (2019). Sorption behavior of citric pectin films with glycerol and olive oil. *Revista Mexicana de Ingeniería Química* 18(2), 487-500. <https://doi.org/10.24275/uam/izt/dcbi/revmexingquim/2019v18n2/Hernandez>
- Huang, M., Yu, J. and Ma, X. (2006). High mechanical performance MMT-urea and formamide plasticized thermoplastic cornstarch biodegradable nanocomposites. *Carbohydrate* 63, 393-399. <https://doi.org/10.1016/j.carbpol.2005.09.006>
- Jongjareonrak, A., Benjakul, S., Visessanguan, W., Tanaka, M. (2006). Effects of plasticizers on the properties of edible films from skin gelatin of bigeye snapper and brownstripe red snapper. *European Food Research and Technology* 22, 229-235. <https://doi.org/10.1007/s00217-005-0004-3>
- Liberman, E and Gilbert, S. (1973). Gas permeation of collagen films as affected by cross linkage, moisture, and plasticizer content. *Journal of Polymer Science* 41, 33-43. <https://doi.org/10.1002/polc.5070410106>
- López-Hernández, L. H., Calderón-Oliver, M., Soriano-Santos, J., Severiano-Pérez, P., Escalona-Buendía, E. and Ponce-Alquicira, E. (2018). Development and antioxidant stability of edible films supplemented with a tamarind seed extract. *Revista Mexicana de Ingeniería Química* 3, 975-987. <https://doi.org/10.24275/uam/izt/dcbi/revmexingquim/2018v17n3/Lopez>
- Mali, S. Sakanaka, S. Yamashita, F. and Grossmann, E. (2005). Water sorption and mechanical properties of cassava starch films and their relation to plasticizing effect. *Carbohydrate Polymers* 60,283-289. <https://doi.org/10.1016/j.carbpol.2005.01.003>
- Moradi, M., Tajik, H., Rohani, S. M. R. and Mahmoudian, A. (2016) Antioxidant and antimicrobial effects of zein edible film impregnated with *Zataria multiflora* Boiss. essential oil and monolaurin. *LWT Food Science Technology* 72, 37-43. <https://doi.org/10.1016/j.lwt.2016.04.026>

- Müller, C. M. O., Laurindo, J. B. and Yamashita, F. (2011). Effect of nanoclay incorporation method on mechanical and water vapor barrier properties of starch-based films. *Industrial Crops and Products* 33(3), 605-610. <https://doi.org/10.1016/j.indcrop.2010.12.021>
- Olivas, G. I. and Barbosa-Cánovas, G. V. (2005). Edible coatings for fresh-cut fruits. *Critical Reviews in Food Science and Nutrition* 45(7), 657-670. <https://doi.org/10.1080/10408690490911837>
- Pajak, P., Przetaczek-Roznowska, I., and Juszczak, L. (2019). Development and physicochemical, thermal and mechanical properties of edible films based on pumpkin, lentil and quinoa starches. *International Journal of Biological Macromolecules* 138, 441-449. <http://doi.org/10.1016/j.ijbiomac.2019.07.074>
- Palma H, Salgado R, Páramo D, Vargas A, Meza M. (2017). Caracterización parcial de películas biodegradables elaboradas con almidón de plátano y proteínas séricas de la leche. *Acta Universitaria* 27(1), 26-33. doi: [10.15174/au.2017.1215](https://doi.org/10.15174/au.2017.1215).
- Rodríguez, E. Sandoval, A. and Fernández, A. (2007). Evaluación de la retrogradación del almidón de harina de Yuca precocida. *Revista Colombiana de Química* 36(1), 13-30. [10.15446/rev.colomb.quim](https://doi.org/10.15446/rev.colomb.quim)
- Salama, H. E., Abdel-Aziz, M. S., and Sabaa, M. W. (2019). Development of antibacterial carboxymethyl cellulose/chitosan biguanidine hydrochloride edible films activated with frankincense essential oil. *International Journal of Biological Macromolecules* 139, 1162-1167. doi: [10.1016/j.ijbiomac.2019.08.104](https://doi.org/10.1016/j.ijbiomac.2019.08.104)
- Sánchez, D. Aldana, J. Contreras, C. Nevárez, G. and Aguilar, C. (2015). Caracterización de películas comestibles a base de extractos pépticos y aceite esencial de limón mexicano. *CyTA. Journal of Food* 13, 17-25. Doi: [10.1080/19476337.2014.904929](https://doi.org/10.1080/19476337.2014.904929).
- Šešljija, S., Nešić, A., Rušić, J., Kalagasidis Krušić, M., Veličković, S., Avolio, R. and Malinconico, M. (2018). Edible blend films of pectin and poly(ethylene glycol): Preparation and physico-chemical evaluation. *Food Hydrocolloids* 77, 494-501. <https://doi.org/10.1016/j.foodhyd.2017.10.027>
- Slavutzky, A. M., Bertuzzi, M. A. and Armada, M. (2012). Water barrier properties of starch clay nanocomposite film. *Brazilian Journal of Food Technology* 15(3), 208-218. <http://dx.doi.org/10.1590/S1981-67232012005000014>
- Srinivasa, C., Ramesh, N., Kumar, R., and Tharanathan, N. (2003). Properties and sorption studies of chitosan-polyvinyl alcohol blend films. *Carbohydrate Polymers* 53(4), 431-438. [https://doi.org/10.1016/S0144-8617\(03\)00105-X](https://doi.org/10.1016/S0144-8617(03)00105-X)
- Thakur, R., Saberi, B., Pristijono, P., Golding, J., Stathopoulos, C., Scarlett, C., Bowyer-Zou, G. Song, X., Chen, F. and Shen, Z. (2017). Physical and structural characterization of edible films made with zein and corn-wheat starch. Production and hosting by Elsevier B.V. on behalf of King Saud University. <https://doi.org/10.1016/j.jssas.2017.09.005>
- Vargas, M. Pastor, C. Chiralt, A. McClements, D. and Gonzalez, C. (2008). Recent advances in edible coatings for fresh and minimally processed fruits. *Critical Reviews in Food Science & Nutrition* 48(6), 496-511. <https://doi.org/10.1080/10408390701537344>.
- Wittaya, T. (2012). Rice starch-based biodegradable films: Properties enhancement, in: A.A. Eissa (Ed.), *Structure and Function of Food Engineering*, Intech Open, United Kingdom, 103-134. <https://doi.org/10.5772/47751>
- Wu, M., Wang, L., Li, D., Mao, Z. and Adhikari, B. (2013). Effect of flaxseed meal on the dynamic mechanical properties of starch-based films. *Journal of Food Engineering* 118, 365-370. <https://doi.org/10.1016/j.jfoodeng.2013.04.017>.
- Zheng, K., Xiao, S., Li, W., Wang, W., Chen, H., Yang, F., and Qin, C. (2019). Chitosan-acorn starch-eugenol edible film: Physico-chemical, barrier, antimicrobial, antioxidant and structural properties. *International Journal of Biological Macromolecules* 135, 344-352. <https://doi.org/10.1016/j.ijbiomac.2019.05.151>
- Zuo, G., Song, X., Chen, F. and Shen, Z. (2019). Physical and structural characterization of

edible bilayer films made with zein and corn-wheat starch. *Journal of the Saudi Society of Agricultural Sciences* 18(3), 324-331. [https:](https://doi.org/10.1016/j.jssas.2017.09.005)

[//doi.org/10.1016/j.jssas.2017.09.005](https://doi.org/10.1016/j.jssas.2017.09.005)