Vol. 20, No. 1 (2021) 423-433
Revista Mexicana de Ingeniería Química

Application of modified vegetable oil for improvement of biodegradable materials based on thermoplastic starch and polylactic acid

Aplicación de aceite vegetal modificado para mejoramiento de materiales biodegradables a base de almidón termoplástico y ácido poliláctico

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Received: June 11, 2020; Accepted: December 1, 2020

Abstract

Currently, it is a challenge for academia and industry to develop materials capable of replacing conventional petroleum-derived polymers. The objective of the present study was to evaluate the physicochemical and morphological properties of biodegradable materials obtained from thermoplastic yam starch (TPS), and polylactic acid (PLA) improved with the addition of epoxidized sesame oil (ESO). The blends were made by extrusion, and the films were made by compression moulding. The interaction with water, mechanical properties and structural properties were studied. As a result, the addition the ESO on the TPS / PLA polymeric matrix caused a decrease in moisture content, surface wettability and lower permeability to water vapour. Furthermore, when adding ESO at 3%, elastic modulus and the tensile strength increased approximately double and the deformation capacity for the mixtures without ESO by more than 70%. Regarding structural properties, the addition of ESO promoted the formation of a TPS / PLA interface without marked separation and smoother surfaces. The materials obtained show promising properties for the development of food packaging with low moisture content.

Keywords: *Dioscorea rotundata*, Sesamum indicum, epoxidation, interface, physicochemical properties.

Resumen

Actualmente, es un desafío para la academia y la industria desarrollar materiales capaces de reemplazar los polímeros convencionales derivados del petróleo. El objetivo del presente estudio fue evaluar las propiedades fisicoquímicas y morfológicas de materiales biodegradables obtenidos a partir de almidón de ñame termoplástico (TPS) y ácido poliláctico (PLA) mejorado con la adición de aceite de sésamo epoxidado (ESO). Las mezclas se prepararon por extrusión y las películas se fabricaron mediante moldeo por compresión. Se estudió la interacción con el agua, las propiedades mecánicas y propiedades estructurales. Como resultado, la adición de ESO a la matriz polimérica de TPS/PLA provocó una disminución en el contenido de humedad, la humectabilidad de la superficie y una menor permeabilidad al vapor de agua. Además, al añadir ESO al 3%, el módulo de elasticidad y la resistencia a la tracción aumentaron aproximadamente al doble y la capacidad de deformación de las mezclas sin ESO en más de un 70%. En cuanto a las propiedades estructurales, la adición de ESO promovió la formación de una interfaz TPS/PLA sin separación marcada y superficies más lisas. Los materiales obtenidos muestran propiedades prometedoras para el desarrollo de envases alimentarios con bajo contenido de humedad.

Palabras clave: *Dioscorea rotundata*, Sesamum indicum, epoxidación, interfaz, propiedades fisicoquímicas.

1 Introduction

The main objective of food packaging is to maintain the quality and safety of food products during storage, transportation and to extend their shelf life, protecting it from external physical forces, chemical compounds, sunlight, permeable volatile compounds, oxygen, and moisture. (Castle *et al*., 2011). For this, the packaging materials must provide physical protection and establish appropriate physicochemical situations for the products that are essential to achieve good resistance, as well as defend food quality and safety problems (Dehnad *et al*., 2014).

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https://doi.org/10.24275/rmiq/Poly2164
ISSN:1665-2738, *issn-e*: 2395-8472

However, there is a substantial environmental problem generated by the waste of packaging made with conventional polymers, representing 39.7% of plastics produced worldwide (Plastics - the Facts, 2018). In Colombia, 56% of plastics production is directed to the packaging sector, and the materials most used for its production are polyethene (36%), propylene (20%), polyvinyl chloride (18%) and PET (13%) (Acoplásticos, 2017).

Today, a wide variety of polymers can be obtained from renewable resources. Some of these are based on natural polysaccharides; in the case of biopolymers originated from starch, they can be produced at low cost and on a large scale (Tabasum *et al*., 2019). This biopolymer can be obtained from different sources such as corn (Zea mays), potato (Solanum tuberosum), rice (Oryza sativa), yam (Dioscorea rotundata), among others. In the development of this work, yam was used, which in Colombia is grown around 500 thousand tons per year, with a yield of 13.8 tons per planted hectare, being a crop of small and medium farmers that constitutes in many regions of the country, the primary source of rural employment and food supply to its inhabitants (Villabona-Ortiz, Tejada-Tovar, & Ortega-Toro, 2020). Yam starch has a high amylose content compared to cassava starch, being less susceptible to enzymatic degradation. Besides, they have a higher gelatinization temperature requiring more energy for processing, but they are also less susceptible to thermal degradation (Villabona-Ortíz *et al*., 2020; Rodriguez-Lora *et al*., 2020). Starch-based materials have significant advantages such as biodegradability, compostability (Martins da Costa *et al*., 2020), high oxygen barrier. Also, it has good processability through conventional techniques such as extrusion, compression moulding, injection, among others (Ortega-Toro *et al*., 2014). On the other hand, it presents deficiencies in terms of its mechanical properties, water vapour barrier properties and changes in properties over time due to the retrogradation of starch (Ortega-Toro *et al*., 2016).

Plasticizers are incorporated into the starch matrix to improve flexibility and processability by increasing the space between molecular chains and increasing segmental mobility (Juansang *et al*., 2017). Starch has a unique characteristic that allows the transformation of native starch to a thermoplastic material (TPS) obtained by blending the biopolymer with plasticizer under conditions of pressure, temperature and cutting speed (Weerapoprasi & Prachayawarakorn, 2019). The deficiencies presented by TPS can be improved using different strategies, such as physical and chemical modifications or mixtures with other components, plasticizers or compatibilizers (García-Cruz *et al*., 2020). These blends are a simple, fast, and inexpensive method. Various authors have carried out mixtures with biodegradable polymers such as Poly (hydroxybutyrate) (PHB) (Florez *et al*., 2019), Poly (butylenes adipate-co-terephthalate) (PBAT) (Dammak *et al*., 2020), Acid Polylactic (PLA) (García-Cruz at al., 2020; Datta & Halder, 2019; Zhou *et al*., 2019), among others. Ortega-Toro *et al*. (2015) considered that the most promising mixtures are found in those made with hydrophobic polymers such as PLA because it allows to reduce the permeability to water vapour and in turn presents a moderate cost in the market. This aliphatic polyester can be synthesized from the ring-opening polymerization (ROP) of lactic acid obtained from the fermentation of starch. Among the characteristics that make PLA one of the most attractive materials is its excellent biodegradability, processability and biocompatibility (Ingrao *et al*., 2015). PLA exhibits balanced mechanical and barrier properties; it is transparent and shiny (Ferri *et al*., 2016). However, the use of PLA as a food packaging material is reduced because it shows brittleness and low barrier to oxygen (Râpă *et al*., 2016). Different strategies have been proposed to overcome their fragility. An attractive solution is to blend PLA with more flexible polymers like thermoplastic starch (TPS). Although physical blending is a cost-effective solution to overcome this drawback, the lack of miscibility between the components restricts the improvement in toughness. For this reason, some authors have proposed the use of coupling agents that promote their chemical affinity. Chemically modified vegetable oils represent an interesting alternative as coupling agents. In recent years, different modified vegetable oils (epoxidized, maleinized, hydroxylated, among others) have been used successfully as renewable plasticizers with a positive contribution to environmental efficiency. Some epoxidized vegetable oils are commercially available. In this way, epoxidized soybean oil (ESBO) (Xing & Matuana, 2016) and epoxidized linseed oil (ELO) (Alam *et al*., 2014) which is a low-cost epoxy compound derived from a renewable resource, biodegradable and readily available (Belhassen *et al*., 2014). The incorporation of epoxidized oils to the mixture improves the formation and properties of biodegradable films; these epoxidized oils have demonstrated to be useful in many applications. Balart *et al*. (2016) reported a possible reaction mechanism between epoxidized vegetable oil, PLA,

and cellulose (structure similar to starch). The epoxy groups of the modified oil can react with the terminal hydroxyls of both PLA and carbohydrate, generating significant improvements in polymer blends. This work aims to evaluate the physicochemical properties and morphology of biodegradable materials obtained from thermoplastic yam starch and polylactic acid improved with the addition of epoxidized sesame oil.

2 Materials and methods

2.1 Development of materials

Hawthorn yam (Dioscorea rotundata) (amylose 28.3% and amylopectin 71.7%) and sesame seeds (Sesamum indicum) were purchased from the local market in Cartagena de Indias (Colombia); PLA (LL700, with a density of 1.25 g/cm3) was supplied by Natureworks (U.S.A). Glycerol and other reagents were provided by Sigma-Aldrich and Panreac (Bogotá-Colombia).

Starch was obtained from previously dried yam (Dioscorea rotundata). Glycerol was added to manually perform mixing with a starch: glycerol ratio of 1: 0.25. The mixture was made by an extrusion process in a range of temperatures between 80 °C and 130 °C. The extruder had five sections programmed at 80 °C, 110 °C, 130 °C, 120 °C, and 100 °C. The screw speed was 10 rpm for a residence time of 2.5 min. During this first stage, thermoplastic starch (TPS) was obtained from yam.

Then a second extrusion process was carried out in which a mixture of TPS with PLA was passed, and epoxidized sesame oil (ESO) was added to improve the blend (the formulations are presented in Table 1). The extrusion conditions were the same used in the first stage. The sesame oil was epoxy as described in previous works (Zuleta *et al*., 2013; Boyacá & Beltrán, 2010) obtaining a 58.4% degree of substitution (density: 0.995 g / mL; iodine number: 1.715 gI2 / 100g; and Oxygen oxirane 3.64% p/p). Briefly, the reaction was carried out by adding the catalyst solution to a reactor; previously, the acetic acid was dissolved in the hydrogen peroxide used as the oxidant. The solution was fed during the first 10 minutes of reaction by using a peristaltic pump with a flow rate of approximately 10 mL/min.

The obtained blends were pelleted and conditioned at a relative humidity of 53% and 25 °C until pressing. Finally, the films were obtained by compression moulding at 130 °C for 10 minutes following the methodology reported in previous studies (Ortega-Toro *et al*., 2014; Ortega-Toro *et al*. 2015; Ortega-Toro *et al*., 2016). In general, the process for obtaining the films was divided into three stages, preheating of the material, pressing, and finally cooling the films obtained. The films were conditioned for two weeks at a relative humidity of 53% and 25 °C.

2.2 Material characterisation

2.2.1 Film thickness

The thickness of the films was measured using a micrometre. These measurements were taken in triplicate. They were taken in eight places in the film, later the arithmetic average and the respective standard deviation were reported.

2.2.2 Moisture content

The films were conditioned at 53% R.H and 25 °C for one week. Convection using an oven was used at 60 °C until a constant weight was obtained to determine its humidity. Moisture was calculated as the ratio of wet weight to dry weight. The test was carried out in triplicate, reporting the arithmetic mean and the respective standard deviation.

2.2.3 Water solubility

The films were immersed in containers with distilled water at a ratio of 1:10, film: water respectively, for 48 hours.

Table 1. Mass fraction of studied formulations.

Formulations	TPS	PLA	Glycerol	Epoxidized sesame oil
PLA	0	1	0	0
TPS	0.8	0	0.2	0
TPS_{50}/PLA_{50}	0.444	0.444	0.111	0
$TPS_{50}/PLA_{50}/ESO_{1.5}$	0.439	0.439	0.11	0.013
$TPS_{50}/PLA_{50}/ ESO_3$	0.433	0.433	0.108	0.026

The test was carried out in triplicate for each of the formulations. Next, samples were taken to a natural convection oven for a time of 24 hours with a temperature of 60 °C to remove the free water and then they were taken to a P2O5 desiccator at 25 °C for two weeks. This process was carried out in order to eliminate the water that is firmly bound. Finally, the solubility of the films will be estimated from the initial and final weights.

2.2.4 Contact angle

The contact angle of distilled water on the films studied was determined. It is measured by studying the shape of a drop of water (0.01 mL) after 10 seconds using a digital camera with a white background. The distance between the lens of the camera and the drop of water was constant (50 cm). Subsequently, the image analysis was carried out using Goniotrans v2.0 software to determine the contact angle formed between the drop of water and the surface of the film.

2.2.5 Water vapour permeability

To determine the properties of the water vapour barrier, the protocols reported in previous studies were used (Ortega-Toro *et al*., 2014), following the standard method ASTM E96-95 implementing some modifications. A gradient humidity of 53% RH at 100% RH was used, at 25 °C. Distilled water was placed in Payne permeability cups, exposing the samples to 100% RH on one side. After the films were secured, each cup was placed a controlled relative humidity cabinet at 25 °C. The RH of the cabinets (53%) was constant using oversaturated solutions of magnesium nitrate-6-hydrate. Once a stable state was reached, the cups were periodically weighed (0.0001 g), and the water vapour transmission (WVTR) was determined from the slope obtained from the regression analysis of the weight loss data versus time. The thickness of the films was considering for this determination.

2.2.6 Mechanical properties

The procedure was carried out following the provisions of the ASTM D 882 standard. Specific properties such as tensile strength, Young's modulus and percentage of elongation were determined. The equipment used for the study was a universal testing machine with a 500 N cell and a head speed of 50 mm/min. The film samples measured 25 mm x 100mm; the gap between the jaws was 50 mm. The thickness measurement of the samples was determinate using a micrometre.

2.2.7 Structural properties

The surface and cross-sections of the biodegradable films analysed were studied using an optical microscope (Primo Star HD binocular microscope) integrated with a high definition camera. The photomicrographs captured at 40X were processed using the Image Pro-Plus version 5.1 software. Also, cross-sectional micrographs of the formulations were acquired by an SEM microscope (Hitachi model SU8010). Films previously dried under vacuum were cryo-fractured using liquid N2 and laminated with a layer of Au-Pd. The samples were fixed on copper slides, and an acceleration voltage of 10 kV was used.

2.2.8 Statistic analysis

All results were analysed using Statgraphics Plus for Windows 5.1 software (Manugistics Corp., Rockville, MD). Carrying out an analysis of variance (ANOVA) and the means were evaluated using the Fisher's Least Significant Differences (LSD) test with 95% confidence.

3 Results and discussion

3.1 Interaction with water

Table 2 presents the results for thickness, moisture content, water-solubility, contact angle, and water vapour permeability. The thickness of the films showed a significant change in those films where ESO was added compared to those of pure TPS. The presence of epoxidized oil acts as a plasticizing agent, being able to present a tendency to decrease the thickness of the films as a result of the fluidity provided to the material. PLA exhibits thermic softening that allow it to flow in the compression moulding process; this material has a lower softening temperature than TPS. To evaluate how the addition of ESO could affect the hydrophilicity of the films measurements of contact angles with water (θw) were carried out for each of the formulations. In the TPS_{50}/PLA_{50} formulation where there is no presence of epoxy groups, the contact angle with water had values of 43°, which is indicative of a more hydrophilic surface compared to those with 1.5% and 3% ESO.

Table 2. Mean values and standard deviation of the thickness (μm), moisture content (g of water/100 g dry film), solubility in water (g soluble film/100g dry film), contact angle (°) and water vapour permeability (g·mm/kPa·h·m^2) of the studied formulations.

Formulations	Thickness	Moisture	Solubility in water	Contact angle	Water vapor permeability
PLA	175 ± 26^{a}	0.36 ± 0.05^{a}	0.30 ± 0.09^{a}	63.3 ± 1.5^{d}	0.36 ± 0.11^{a}
TPS	439 ± 34^{c}	11.0 ± 0.8^{d}	28.4 ± 0.8^{c}	52.3 ± 0.6^{b}	9.43 ± 0.76^{d}
TPS_{50}/PLA_{50}	231 ± 24^{b}	5.3 ± 0.5^{c}	13.4 ± 0.3^{b}	43.3 ± 0.6^{a}	3.68 ± 0.81^{c}
$TPS_{50}/PLA_{50}/ESO_{1.5}$	177 ± 25^{a}	$4.8 \pm 0.4b^{c}$	13.0 ± 0.6^{b}	46.0 ± 2.8^{a}	$2.63 \pm 0.5b^{c}$
$TPS_{50}/PLA_{50}/ESO_{3}$	157 ± 25^{a}	4.2 ± 0.5^{b}	13.7 ± 0.7^{b}	57.0 ± 1.4^{c}	2.17 ± 0.42^{b}

Hydrophilicity is expected due to the presence of high density of hydroxyl groups within the main chain of starch, but also due to the incorporation of glycerol (Belhassen *et al*., 2014).

The formulations $TPS_{50}/PLA_{50}/ESO_{1.5}$ and $TPS_{50}/PLA_{50}/ESO_3$ were added with ESO, and they exhibit an increase in the contact angle. The values reported for these formulations were 46° and 57° respectively, indicating that as the percentage of ESO in the TPS/PLA matrix increases, the greater will be the contact angle of the material. For said material to be considered with hydrophobic behaviour, it must present an angle greater than 65° (Vogler, 1998). However, the values obtained are not above this limit; it is evidenced that the addition of epoxidized sesame oil leads to a tendency towards higher hydrophobicity. Therefore, although the amount of ESO added is small, a significant improvement in the hydrophobicity of TPS was observed. This slight increase in the hydrophobicity of the plasticized film can be attributed to the hydrophobic nature of the fatty acid segments of the oil (Requena *et al*., 2016; Carbonell-Verdu *et al*., 2017).

Regarding moisture content, films prepared with pure TPS have the highest moisture content, all due to the massive amounts of hydrophilic hydroxyl groups that starch has (Zhou *et al*., 2019). Unlike PLA, which has the lowest values due to its hydrophobic nature. In the case of TPS_{50}/PLA_{50}, when combining TPS with PLA, the moisture content was reduced by approximately 56% for pure TPS. By adding ESO to the polymeric matrix, the decrease in moisture content continued progressively as the concentration of ESO increased. This fact may be due to the incorporation of hydrophobic oils that can affect the ability of the film to retain water (Nordi *et al*., 2020). About these findings, they were similar to those reported by (Ghasemlou *et al*., 2013) where they made use of essential oils from Zataria multiflora Boiss and Mentha pulegium to improve the physical and mechanical properties of corn starch films.

The water solubility of materials is an essential factor when choosing a film for specific applications. As expected, water solubility exhibits a similar trend to moisture content. The water solubility values of the films are relatively high, consistent with the hydrophilic nature of the polymers. However, the blend films exhibited significantly less solubility than pure TPS. This fact suggested that there was a decrease in the hydrophilic nature of the matrix in the blend, probably due to the establishment of polymeric interactions, which led to a lower affinity for water (Olewnik *et al*., 2019).

The presence of the epoxidized oil decreased the water solubility of the film. The added ESO can become trapped within the TPS/PLA matrix, creating a strong interaction with the polymeric network through the epoxide groups of ESO and the hydroxyl groups of TPS and PLA, which causes a decrease in the affinity of the films towards the water and reduces the solubility of the films (Nordi *et al*., 2020). Therefore, the addition of PLA and ESO in TPS-based materials can reduce solubility, improving the water-resistance of packaging materials to help keep food fresh.

One of the main functions of food packaging is to prevent or minimize the transfer of moisture between the food and the surrounding atmosphere. Therefore, the water vapour permeability (WVP) should be as low as possible to optimize the food package environment and potentially increase the shelf life of the food product. The values reported in Table 2 follow the same trend as the results obtained in the evaluation of the properties of moisture content and solubility in water. The addition of ESO to the TPS/PLA matrix shows a significant decrease in the permeability to water vapour, which in the case of pure TPS presented the highest WVP value, this may be associated with

the polarity of the starch and the forces of attraction between the film and the water (Haq *et al*., 2016). When mixing PLA and TPS, the values of this parameter decrease by 61% compared to pure TPS.

In the results obtained for $TPS_{50}/PLA_{50}/ESO_{1.5}$ and $TPS_{50}/PLA_{50}/ESO_3$ (2.63 g.mm/kPa.h.m^2 and 2.17 g.mm/kPa.h.m^2, respectively) it is evidenced as the use of ESO reduced the WVP values of the films due to the reduction of the water transmission caused by its hydrophobic nature and higher molecular packing promoted by its coupling effect. Ge *et al*. (2019) reported similar results with the use of acrylate epoxidized soybean oil (AESO) as a plasticizer in the starch films. They observed that the moisture permeability of the films decreased with increasing AESO concentration, the WVP value decreased notably from 531.81 to 46.79 g/m^2*224 h with increasing AESO concentration from 0% to 30%. For their part, Ghasemlou *et al*. (2013) reported WVP values for the film samples and the results to suggest that there were significant differences in the WVP values observed between the starch films with the addition of essential oil Zataria multiflora Boiss (ZEO) or Mentha pulegium (MEO) and the control film. The addition of ZEO or MEO improved the barrier properties of the starch films, decreasing the WVP by up to 50% relative to the control sample.

3.2 Tensile properties

Table 3 shows the mechanical behaviour (EM: Elastic Modulus; TS: tensile strength; E: maximum deformation at the breaking point) of the studied formulations. The blend of a more flexible and ductile polymer, such as TPS, with a more rigid one such as PLA (TPS50/ PLA50), decreased the elastic modulus of the latter by approximately 81%, in the same way, it presented a decrease in tensile strength by 85% and a greater than 50% increase in maximum deformation at the fracture point. The epoxidized sesame oil (ESO) added at 1.5% and 3% to the TPS/PLA matrix, increased the elastic modulus and tensile force value approximately double and increased the deformation capacity by more than 70%. These changes in properties reveal that ESO can act as an interface agent in the TPS/PLA composite. Also, the addition of ESO provides a plasticizing effect on the TPS/PLA matrix with a notable increase in ductile mechanical properties, in turn reducing the high intrinsic brittleness of pure PLA (Carbonell-Verdu *et al*., 2017). It should be noted that the increase of ESO, as observed in $TPS_{50}/PLA_{50}/ESO_3$ formulation regarding $TPS_{50}/PLA_{50}/ESO_{1.5}$, there is a non-significant increase in the rigidity of the material and a brief decrease in tensile force and deformability. Carbonell-Verdu *et al*. (2017) consider that, by increasing the ESO content, the system could be saturated and the properties would not tend to improve. These researchers reported system saturations at concentrations greater than 10% epoxidized oil addition. According to the above, the epoxidized oil has a double effect, both plasticizer and coupling agent. The modified oil possibly acts as a plasticizer in the PLA-rich phase, favouring the increase in the material's deformation capacity. On the other hand, there are interactions between TPS and ESO in the system and at the same time interactions between ESO and PLA, generating the effect of a coupling agent that exhibits increases in EM and TS.

3.3 Structural properties

Interfacial adhesion and dispersion of the system in the polymer blend play an essential role in determining the overall properties that the final product will possess. Figure 1 shows the surface morphology of each of the formulations studied, showing the effect of epoxidized sesame oil on the TPS/PLA matrix. In the case of PLA, a compact and very uniform surface can be seen, compared to TPS, which presents topography unevenness very pronounced.

Table 3. Mean values and standard deviation of the elastic modulus (EM), tensile force (TS) and deformation of the studied films (E).

Formulations	EM (MPa)	TS (MPa)	E(%)
PLA	1276 ± 84^{d}	48 ± 3^{c}	4.5 ± 0.3^{a}
TPS	95 ± 12^{a}	8.1 ± 1.1^{a}	35 ± 5^{c}
TPS_{50}/PLA_{50}	242 ± 5^{b}	7.2 ± 0.8^{a}	6.8 ± 0.8^{a}
$TPS_{50}/PLA_{50}/ESO_{1.5}$	485 ± 9^{c}	15.2 ± 0.9^{b}	12.1 ± 0.5^{b}
$TPS_{50}/PLA_{50}/ESO_3$	501 ± 7^{c}	14.1 ± 1.1^{b}	11.8 ± 1.1^{b}

Different superscript letters mean significant differences ($p < 0.05$) between formulations.

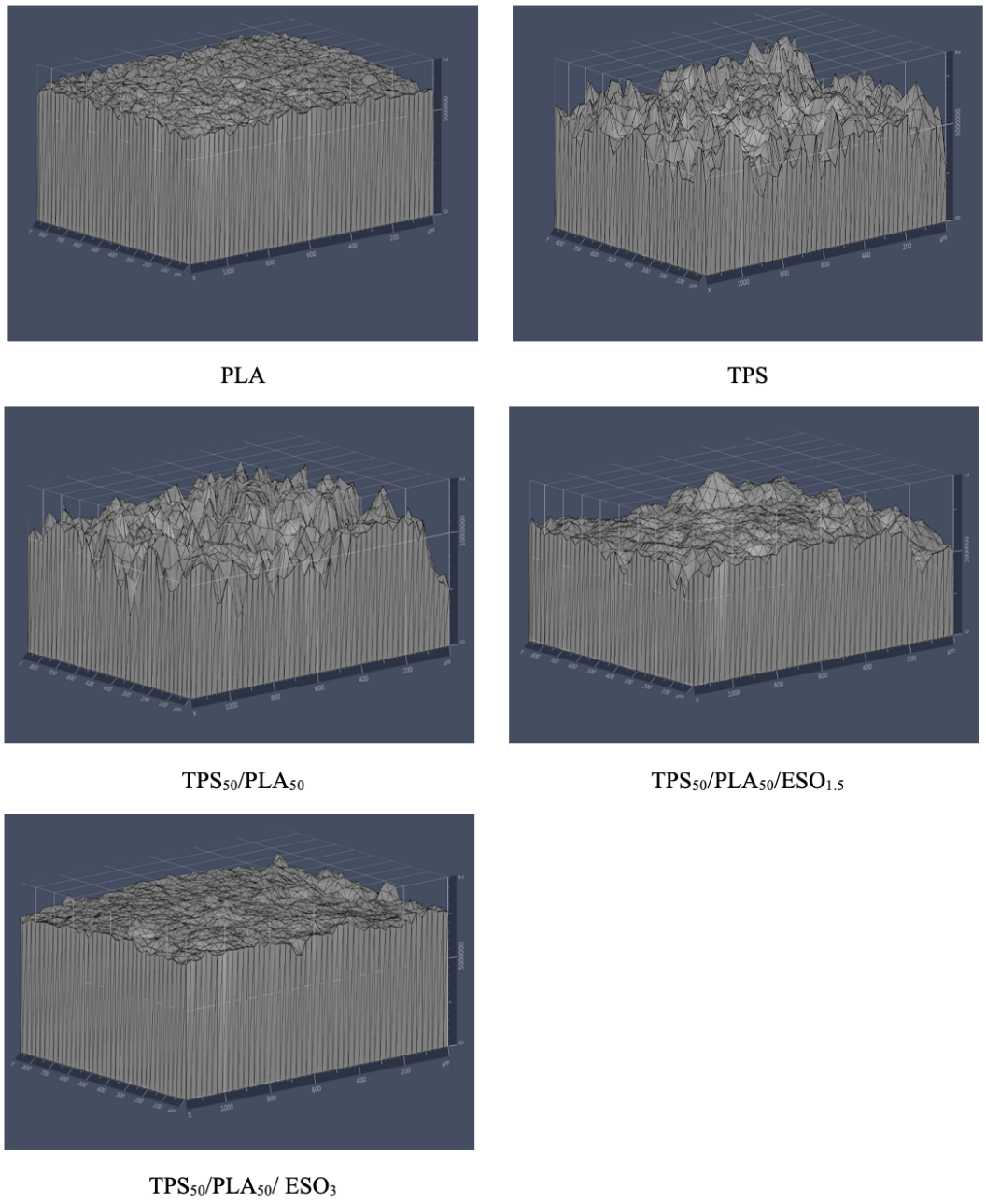
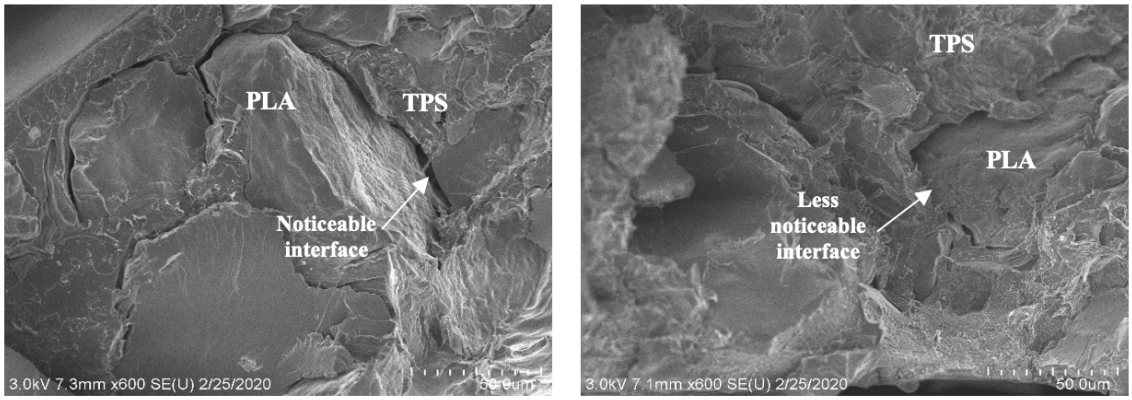


Fig. 1. Optical micrographs of the studied films at 40X.

In the case of the combination of TPS_{50}/PLA_{50} polymers, it shows similar behaviour to TPS, with a slight improvement. The morphology of TPS_{50}/PLA_{50} can be better appreciated by cross-sectional micrographs obtained using a matrix scanning electron microscope (Figure 2). The TPS/PLA blend without the addition of ESO presented cracks that show a separation of phases and voids in the interfaces between the two compounds, which suggests a mixture with immiscible and poorly compatible characteristics. These images show the typical brittle fracture of PLA with a smooth fracture surface due to no plastic deformation and a more ductile fracture corresponding to TPS (Jullanun & Yoksan, 2020).



TPS_{50}/PLA_{50} at 600X **$TPS_{50}/PLA_{50}/ ESO_3$ at 600X**

Fig. 2. Cross-section micrographs obtained by matrix scanning electron microscope without epoxidized oil (TPS50/PLA50) and with epoxidized oil (TPS50/PLA50/ ESO3).

PLA is a hydrophobic polymer, while TPS is a highly hydrophilic polymer, this different polarity is responsible for the low affinity between these two polymers, and this leads to phase separation (Ferri *et al*., 2016). For this reason, strategies must be applied to improve TPS/PLA mixtures and thus generate materials with more convenient properties.

The addition of ESO to the TPS/PLA matrix improved the surface morphology of the material, as can be seen in Figure 1. The epoxidized oil shows the ability to plasticize the material allowing the formation of smoother and soft surfaces, appearing to be a material more compact. In Figure 2, it is observed that, when ESO is added to the mixture, the interface presents fewer cracks, being the material more homogeneous and compact. This fact would indicate a coupling effect of the epoxidized oil. As previously reported, the epoxy groups of ESO could form hydrogen bonds with the terminal hydroxyl groups of TPS and PLA (Balart *et al*., 2016).

Conclusions

Materials were developed based on yam thermoplastic starch (TPS) and polylactic acid and incorporating epoxidized sesame oil (ESO), utilizing extrusion mixing and compression moulding. Evidence of the plasticizing and coupling effect of epoxidized sesame oil on TPS/PLA blend was found. The addition of ESO promoted the decrease in moisture content and permeability to water vapour, while the solubility in water was not significantly affected, and the contact angle with water increased, indicating greater surface hydrophobicity. On the other hand, the mechanical properties exhibited a drastic increase in the modulus of elasticity, tensile strength, and deformation capacity when the epoxidized oil was added to the TPS/PLA blends. This result indicates an improvement in the interfacial adhesion and the plasticization of the material. Finally, the optical micrographs reveal smoother and soft surfaces, and more compact structures when ESO is added, and the cross-sectional micrographs obtained by SEM show more compact polymeric interfaces with the addition of epoxidized oil, showing a coupling agent effect. The addition of epoxidized oil to TPS/PLA mixtures is a suitable strategy to improve the functional properties of materials for the design of food packaging.

Acknowledgements

The researchers thank the Universidad de Cartagena (Colombia) for financing this work through Project 079-2018.

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