JOURNAL OF Applied Polymer SCIENCE

Special Issue: Bio-based Packaging

Guest Editors: José M. Lagarón, Amparo López-Rubio, and María José Fabra
Institute of Agrochemistry and Food Technology of the Spanish Council for Scientific Research

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Edible films based on chia flour: Development and characterization

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ABSTRACT: The aim of this study was to develop and evaluate the structural, physicochemical, barrier (water vapor), and mechanical properties of chia flour (CF)-based films. The films were prepared with the casting method. Three ratios of CF to maize starch (MS), namely, 1:0, 1:1, and 1:2 w/w, were evaluated. A fixed total solid content of 6% w/v and a glycerol concentration of 1% w/v were used for all of the tested proportions. The transparency of the films increased as a function of the addition of MS. Moreover, CF-based films presented a low solubility and a satisfactory water vapor barrier and showed UV-radiation-shielding properties. The presence of MS in the composite film elevated the molecular interactions and led to a more compact structure; this resulted in improved strength, although the elongation still remained low. CF-based films are a promising material for the formulation of edible films with adequate physicochemical and mechanical properties and high nutritional quality. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 42455.

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INTRODUCTION

The development of new edible and biodegradable films from agricultural biopolymers could be an alternative to substitute the nonbiodegradable petrochemical-based plastics that have caused serious environmental problems through their waste accumulation. Biopolymers, such as polysaccharides, proteins, lipids, and their blends, are considered promising candidates for promoting a new source of packaging materials because of their biodegradability and abundance.[1,2] An alternative to create new opportunities for materials in the area of edible films is the use of flours prepared from agricultural crops, which are naturally occurring complex blends of polysaccharides, protein, lipids, and fibers.[3,4] The combination of natural mixtures directly obtained from agricultural sources takes advantage of each component in their original system and could be an alternative for increasing their applications in the market.[5]

A few studies have investigated the use of different flours as a raw materials suitable for the preparation of edible films in the last decade. Some authors have reported on the potential application of flours obtained from whole materials, such as banana, soy, amaranth, achira, rice, or wheat, for film production.[2–4,6–11] These films feature excellent characteristics because of the natural and intrinsic molecular interactions taking place between their polysaccharide, protein, lipid, and fiber components.[9] Chia seed (*Salvia hispanica* L.), which contains a natural mixture of natural compounds, is an attractive, renewable, raw material for the preparation of edible and biodegradable films. Chia's native environment is southern Mexico and northern Guatemala. The seeds are small in size (1–2 mm) with an oval, flattened shape; they range in color from dark coffee to beige with small darker spots.[12] Chia was revived by a group of scientists and farmers because of its nutritional and functional characteristics.[13] Foods, such as cereal bars, biscuits, pasta, bread, snacks, yogurt, and cakes, among others, can be produced with chia seeds as an ingredient.[14,15] Chia seeds are an excellent source of omega-3 fatty acids, protein, antioxidants, and dietary fiber plus vitamins and minerals.[14] The protein content of chia seed is higher

Additional Supporting Information may be found in the online version of this article.

(19–23 g/100 g) than those of other traditional crops, such as wheat, corn, rice, oat, barley, and amaranth.[16,17] It has a significant quantity of oil (ca. 25–38 g/100 g). This lipid fraction contains polyunsaturated fatty acids: the omega-3 α-linolenic acid (which has been said to be the highest known percentage of α-linolenic fatty acid of any known vegetable source, comprising ~60%) and the omega-6 linoleic acid (comprising ~20%).[18] Both essential fatty acids are required by the human body for good health, and they cannot be artificially synthesized. The total chia dietary fiber (34–40 g/100 g) has been researched for its outstanding soluble and insoluble fiber ratio, and it has greater values than quinoa, flaxseed, and amaranth. Chia soluble dietary fiber is partially expelled from the seed when it comes in contact with water; this results in a clear mucilaginous gel (ca. 5–6% seed weight). The dietary fiber portion includes lignin, which contains antioxidant compounds and has some hypocholesterolemic effects. The major phenolic compounds found in chia seeds are chlorogenic and caffeic acids, followed by myricetin, quercetin, and kaempferol.[14,19]

Edible plasticized films are thin, flexible materials made from a biopolymer base material, a solvent, and a plasticizer. Plasticizers must be added at a certain amount to increase the film flexibility and workability. Glycerol is one of the most preferred and used plasticizers; it has a relatively small hydrophilic molecule, which can be incorporated between adjacent polymeric chains to increase the molecular mobility.[20,21]

Chia flour (CF) mainly consists of fiber, protein, and lipids, and these components might provide properties to form edible films; nevertheless, the absence of starch in this natural flour could result in a poor resistance to break. The use of maize starch (MS) for producing edible films has a long history. Such films have the advantage of low cost compared to other alternative high-tensile-strength (TS) materials. Starch films present good mechanical properties, but their sensitivity to moisture is a major drawback.[22] In this study, to investigate its synergistic effect with CF, MS was added to increase the mechanical strength and to improve films stiffness. CF-based films have commercial potential because they can protect food products and, at the same time, if ingested, increase its nutraceutical characteristics. Furthermore, the possibility to form films from chia seed flour either alone or in combination with MS has not yet been explored. In this context, the objectives of this study were to investigate the use of CF for the elaboration of edible films (with or without MS) and to compare different formulations of CF blends in relation to their edible film structural, physicochemical, water-vapor-barrier, and mechanical properties.

EXPERIMENTAL

Materials

The chia seeds (*Salvia hispanica* L.) used in this study were grown in the region of the state of Jalisco in Mexico. The seeds were cleaned (we manually separated the dust, broken seeds, and straw from the threshed seeds) and stored at −18°C in vacuum-sealed bags until they were tested. MS (Unilever Alimentos, Guaranhuns, Brazil) was obtained from the local market in Porto Alegre, Brazil. Glycerol PA was purchased from Nuclear (CAQ-Casa da Quimica Indústria e Comércio Ltda, Diadema, São Paulo, Brazil).

Proximate Analysis of the CF

Chia seeds were ground with the aid of a kitchen blender (Arno, model wwB3 400W, Brazil) and screened through a 60-mesh sieve to obtain CF with a standard granulometry of 250 μm or less and stored in sealed containers until they were used. Chemical analyses were performed to determine the proximate composition of the CF (mesh 60) used to elaborate the films according to the standard Association of Official Analytical Chemist methods.[23] The total protein content was determined by the Kjeldahl method with a nitrogen-to-protein conversion factor of 6.25. The lipid content was determined with a Soxhlet extractor (Foss Soxtec, model 2055, Denmark). The ash content was measured in a muffle furnace (ElektroTherm Linn, 312.6 SO LM 1729, Germany) set to 550°C. The moisture content (MC) was determined by desiccation of the sample in an oven at 105°C (DeLeo, model TLK 48, Porto Alegre, Brazil). The total dietary fiber, soluble and insoluble, was determined by the enzymatic–gravimetric method. The carbohydrate content was determined by the difference from the other components. All analyses were performed in triplicate. The results are expressed as grams per 100 g of dry base.

Preparation of the Films Containing CF

The films were prepared according to the casting technique. The total solid content, glycerol concentration, pH value, drying conditions, and chia film technique were established according to preliminary tests. Film-forming solutions were prepared by the dispersion of CF and MS in 6% w/v (6 g of total solids/100 mL of water) in distilled water with different ratios of CF to MS (1:0, 1:1, and 1:2 w/w, respectively). Dispersions were mechanical stirred (Fisatom, 713-D, São Paulo, Brazil) for at least for 1 h at room temperature (25°C), and their pH values were adjusted to 7.5 with 0.1*M* NaOH to dissolve the protein. The solutions were then heated in a water bath at 70°C under stirring for 15 min. After heating, glycerol (1% w/v) was added as a plasticizer, and the solution was stirred for more 30 min. Then, solutions were poured into a series of acrylic plates (40 g of each solution on a plate 14 cm in diameter; 0.26 g/cm^2) and were dried for 12–14 h at 35°C in an oven with air-flow circulation (DeLeo oven, model TLK 48, Porto Alegre, Brazil). After this time, the dried films solutions were peeled off the casting surface, cut into adequate samples, and conditioned at 25°C and 52% relative humidity (RH) with saturated solutions of $Mg(NO_3)_2$ for 48 h before characterization in terms of MC, tensile and puncture properties, and water vapor permeability (WVP). All of the experiments were done in triplicate unless otherwise indicated.

Film Characterization

Film Thickness. Film thickness (in millimeters) was determined with a digital micrometer (Digimess, model IP40, São Paulo, Brazil) with an accuracy of 0.001 mm. The measurements were determined from an average measurement of five films for each CF/MS ratio at five different positions for each film specimen.

Film Morphology. To study the microstructural changes of the films, the surface and cross-sectional (fractured under liquid

nitrogen before visualization) morphologies of the dried films were examined with a scanning electron microscope (JEOL, model JSM-5800, Tokyo, Japan) with accelerating voltages from 5 to 8 kV. Before the analyses, all of the samples were mounted on aluminum stubs with double-sided adhesive tape and coated with a thin layer of platinum.

MC and Solubility in Water. MC was determined in triplicate by the weight loss of the film samples. The initial weight of the dry matter (W_i) was calculated by the difference between the weight of the original sample and the MC. First, samples were equilibrated at 52% RH. Then, three replicates of each film sample (2 cm in diameter) were weighed and dried with a laboratory oven (Weiss-Gallenkamp, BS model OV 160, Leicestershire, United Kingdom) at 105°C for 24 h. The dried pieces of the samples were directly used to measure the solubility.

The film solubility was determined in triplicate according to Gontard *et al.*[24] and expressed as the percentage of dry matter of the film solubilized after 24 h of immersion in water. Dried pieces of the film samples were immersed in 30 mL of distilled water, and the system was gently shaken for 24 h at room temperature. The samples were then filtered with desiccated, preweighed filter paper. The filter paper, containing an unsolubilized fraction of the film, was dried for 24 h at 105°C (Weiss-Gallenkamp, BS model OV 160, Leicestershire, United Kingdom), and the resulting material was weighed to determine the final weight of the dry matter (W_f). Thus, the solubility in water (S; %) of the films was calculated according to eq. (1):

$$S = \frac{W_i - W_f}{W_i} \times 100 \quad (1)$$

WVP. WVP tests were conducted according to ASTM E 96-95[25] with some modifications.[26] The samples were placed in permeation cells (*i.d.* = 63.5 mm, height = 25 mm) over a circular opening of 0.0032 m^2 filled with granular anhydrous calcium chloride and hermetically sealed with six screws around the permeation cell circumference. The permeation cells were placed in a glass chamber with a saturated sodium chloride solution to obtain an RH gradient of 0–75% at 25°C. We determined the mass gain by weighing the cell permeation on an analytical balance (Shimadzu, AY 220, Kyoto, Japan) at intervals of 2 h during the first 24 h. The WVP of the samples was determined in triplicate with eq. (2):

$$\mathrm{WVP} = \frac{wL}{At\Delta p} \quad (2)$$

where w is the weight of water permeated through the film (g), L is the thickness of the film (mm), A is the permeation area (m^2), t is the time of permeation (h), and Δp is the water vapor pressure difference between the two sides of the film (kPa).

Color Measurement. The color values of the films were measured with a colorimeter (Minolta, CR-400, Osaka, Japan). Film specimens were placed on the surface of a white standard plate, and the Hunter L, a, and b color values were measured. The three color coordinates ranges were 0 (black) to 100 (white) for L, − (greenness) to + (redness) for a, and − (blueness) to + (yellowness) for b. The total color difference (ΔE) was calculated with eq. (3):

$$\Delta E = \sqrt{(L_{\mathrm{film}} - L_{\mathrm{standard}})^2 + (a_{\mathrm{film}} - a_{\mathrm{standard}})^2 + (b_{\mathrm{film}} - b_{\mathrm{standard}})^2} \quad (3)$$

The standard values refer to the white calibration plate ($L = 94.23$, $a = -0.55$, and $b = 9.68$). Three replicates of each film were evaluated. Five readings were made for each replicate through changes in the position of the colorimeter over the film.

Light Transmittance and Transparency Value. The light transmittance of the films was measured as described by Shiku *et al.*[27] in the UV and visible range from 200 to 800 nm with an ultraviolet–visible spectrophotometer (Shimadzu Corp., UV-1800, Kyoto, Japan). Film specimens were cut into rectangles and directly placed in a spectrophotometer test cell. Spectral transmittances were determined against air as a reference. The transparency value of the films was calculated by the equation of Han and Floros[28] as the ratio between the absorbance at 600 nm (A_{600}) and the film thickness (in millimeters) and was expressed as A_{600}/mm. A greater transparency value represented a lower transparency of the film.

Mechanical Properties. The tensile mechanical properties were determined on a dynamic mechanical thermal analysis machine (TA Instruments, model RSA-3, New Castle, DE) controlled by Orchestrator version 7.2.0.4 software according to ASTM D 882-12.[29] The dimensions of the film samples used in the tests were 70 × 20 mm^2, as cut with sharp scissors. Before the mechanical tests, the samples were conditioned at 52% RH (25°C for 48 h) for dry tensile testing and at 90% RH (25°C for 48 h) for wet tensile testing (to measure the wet strength). The values of TS, elongation at break (EB), and Young's modulus (YM) were determined from five replicates for each film formulation. The samples were clamped between grips, and then, the force and deformation were recorded during extension at 20 mm/min with an initial separation between the grips of 60 mm.

The puncture properties were determined with a texture analyzer (TA.XT2 Plus, Texture Technologies Corp., Hamilton, MA) to determine the puncture force (PF) and puncture deformation (PD). A stainless cylinder probe 4 mm in diameter was used to puncture the uncut films (which were preconditioned at 52% RH and 25°C for 48 h). The circular film samples (50 mm in diameter) were fixed in an acrylic plate with a hole at the center 20 mm in diameter and perforated by the probe, which was moving at 60 mm/min and had a trigger force of 0.01 N. PF was determined directly from the force–displacement curves. PD was calculated with eq. (4), as described by Sobral *et al.*:[30]

$$\mathrm{PD} = \frac{\Delta l}{l_o} = \frac{\sqrt{D^2 + l_o^2} - l_o}{l_o} \quad (4)$$

where l_0 is the initial length of the film corresponding to the disc radius (10 mm) and D is the displacement of the probe until the break point. Five measurements were done for each film.

Statistical Analysis

The experimental results were recorded as the means plus or minus the standard deviations. Unless otherwise specified, all analyses were done in triplicate. Statistica 8.0 from Statsoft, Inc., was used for statistical analysis. Analysis of variance and

Table I. Thickness, MC, Solubility, and WVP Values of the CF-Based Films

CF/MS (w/w)	Thickness (mm)	MC (%)	*S* (%)	WVP (g mm kPa^{-1} h^{-1} m^{-2})
1:0	0.247 ± 0.014^a	12.23 ± 0.95^a	25.57 ± 0.54^a	0.567 ± 0.01^a
1:1	0.203 ± 0.008^b	8.29 ± 0.41^b	23.39 ± 0.27^b	0.326 ± 0.02^b
1:2	0.194 ± 0.001^b	7.27 ± 0.29^b	25.34 ± 0.94^a	0.293 ± 0.00^b

S, solubility. The data are presented as the means plus or minus the standard deviations. Means in the same column followed by different letters were significantly different ($p < 0.05$) according to Tukey's test.

Tukey's test were used to test to significance of differences between the means at a p level of 0.05.

RESULTS AND DISCUSSION

Proximate Analysis of CF

CF (mesh 60) was composed on a dry basis of $4.77 \pm 0.16\%$ MC, $23.08 \pm 0.67\%$ protein, $32.15 \pm 0.98\%$ lipids, $4.71 \pm 0.27\%$ ash, $33.26 \pm 1.58\%$ total fiber, $32.59 \pm 0.02\%$ insoluble fiber, $0.67 \pm 0.80\%$ soluble fiber, and $6.80 \pm 0.88\%$ carbohydrate. These results were in accordance with those of Ayerza and Coates.[13]

The results of proximate analyses showed that CF had a low carbohydrate content. Moreover, it is well known that CF does not possess starch. Therefore, it was likely that the films produced with this raw material would have poor resistance to break. As such, MS was added to improve the mechanical strength and stiffness.

Film Characterization

Film Thickness. The thickness values for the CF-based films ranged from 0.194 ± 0.001 to 0.247 ± 0.014 mm (Table I). The films prepared with only CF and without any MS (1:0 ratio) exhibited a significantly higher thickness ($p < 0.05$) compared to the other films prepared with CF/MS ratios of 1:1 and 1:2. Similar results were found by Wu *et al.*[31] in their study of maize- and potato starch-based films made with the incorporation of flaxseed meal, where the thickness ranged from 0.278 to 0.242 mm for MS-based films prepared with 0 and 15% flaxseed meal, respectively. The thickness ranged from 0.279 to 0.354 mm for potato starch-based films prepared with 0 and 5% flaxseed meal, respectively.

Film Morphology. The scanning electron microscopy of the surfaces and cross sections of the CF-based films is presented in Figure 1.

The surface microstructure with a ratio of 1:0 [Figure 1(a)] revealed a nonhomogeneous structure and a less smooth surface with some imperfections, whereas those with ratios of 1:1 [Figure 1(c)] and 1:2 [Figure 1(e),] displayed a more uniform surface. The larger amount of components of different sizes present in CF, as compared to MS, may have afforded the rougher surface of the 1:0 films. It is also noteworthy that the CF-based films with no MS added exuded some oil after peeling; this did not occur with the films containing MS (1:1 and 1:2).

The irregularities on the cross sections of the CF-based films [Figure 1(b,d,f)] may have been related to the presence of more than one macromolecule in the polymer matrix (starch, protein, lipids, and fiber) and to the interactions between these components such as starch–protein and starch–cellulose (fiber) interactions, which play an important role in the final film structure. Moreover, some microsegregation of the phases might have occurred. Even though with some irregularities, the films with 1 : 1 and 1:2 ratios showed a more compact and dense structure than the 1:0 film, and this was favored with the increasing addition of MS. Furthermore, these observations correlated with the other properties of the films.

MC and Solubility in Water. Table I shows the results for the MC and solubility in water of the CF-based films. MC for the ratio 1:0 was significantly higher ($p < 0.05$) than those of the 1 : 1 and 1:2 ratios (Table I); therefore, MC of the films increased with the amount of MS added.

The decrease in MC in the CF/MS films with the addition of MS could be explained by the interactions between CF and MS. These interactions lowered the availability of the hydrophilic components (protein and fiber) in the flour, which, in turn, limited the CF interactions through hydrogen bonding (interactions with water molecules). These results for MC were similar to those reported by Tapia-Blácido *et al.*[2] for films elaborated from amaranth flour (13.78%) and those reported by Pelissari *et al.*[5] for films elaborated from banana starch (12.2%) and slightly lower than those for films of banana flour (15.1%). However, the results were considerably lower than those reported by Wang *et al.*[32] for active films based on chitosan incorporated with tea polyphenols in the range from 25.95 to 42.71%.

The results presented in Table I show that the films with ratios of 1:0 and 1:2 showed no significant differences ($p > 0.05$) between their solubility in water and showed values significantly greater than the film with a ratio of 1:1 ($p < 0.05$). The presence of lipid in CF increased the hydrophobicity of the films and decreased the water affinity. On the other hand, other components in the flour matrix, such as protein and fiber, were responsible for the increase in MC because of the increase in water retention in the film. The presence of fibers in the flour restrained the water sensitivity and favored an almost constant equilibrium water content in the films. Therefore, the amount of protein and fiber in the flour were not enough to cause a greater solubility in the water in all of the tested formulations. In addition, the protein and the lipids were homogeneously distributed throughout the film, as depicted in Figure 1. The distributed protein and lipid interacted with the MS and decreased the solubility of the films.[3–5] In general, the solubility of the

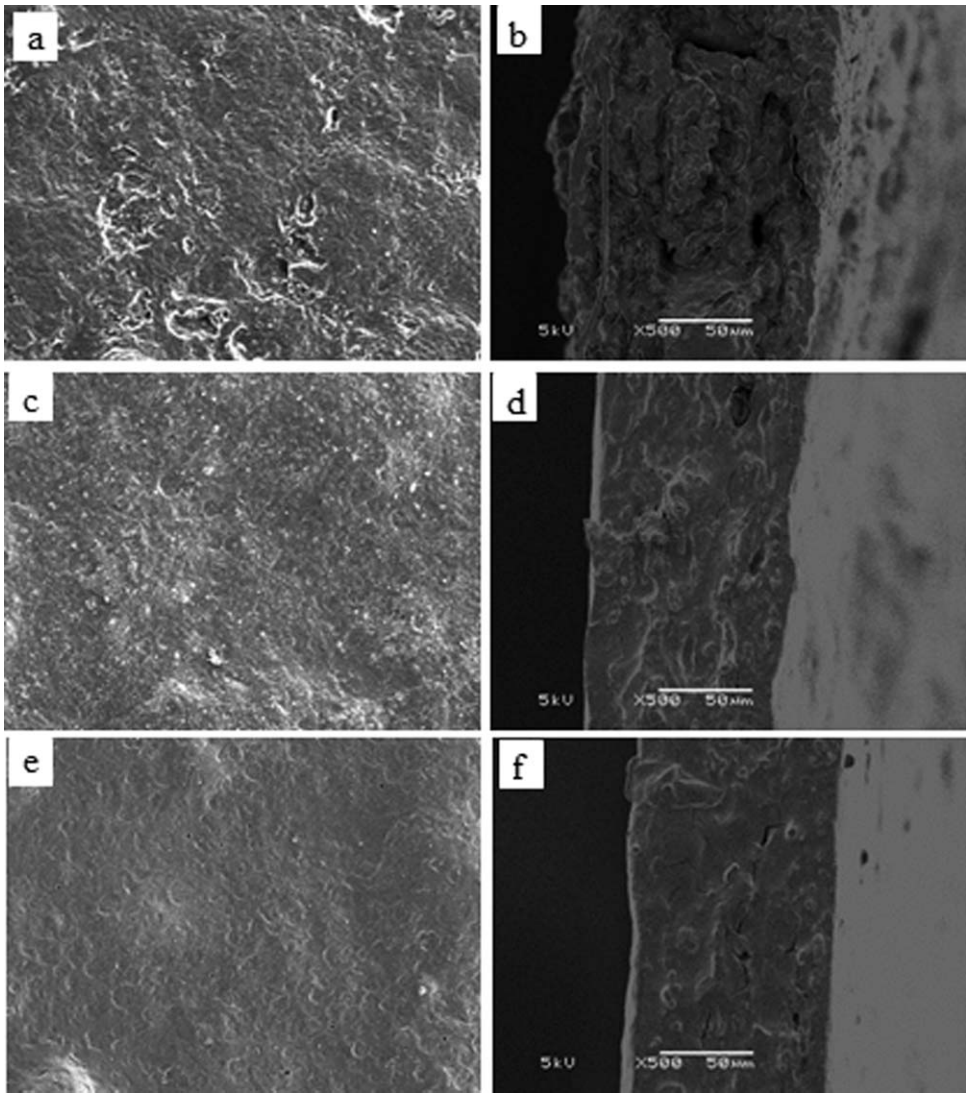

Figure 1. SEM microscopy of the surface (left column, magnification of 200×) and cross section (right column, magnification of 500×) of the CF-based films: (a,b) 1:0, (c,d) 1:1, and (e,f) 1:2 w/w CF/MS ratios, respectively.

tested films were around 25%. Independent of their solubility values, all of the tested films did not loose their integrity when they were immersed in water. According to Pelissari *et al.*,[5] the comparison of the solubility of a flour or a starch from different botanical sources is a challenge because the solubility is related to many factors, including the type of material used for the formation of the polymer matrix, the type of interaction that occurs in the matrix, the plasticizers used, and the process conditions. Moreover, the desired value for the solubility of a film will depend on its application or intended use.[5]

The solubility values for the CF-based films were lower than those found in films elaborated from amaranth flour (41.9%),[2] achira flour (38.3%),[3] and potato peel (41.2%)[33] but within the range of films elaborated from banana flour (27.9%) and banana starch (21.3%).[5] Hence, the CF-based film tended to be less soluble than other films; this suggested a good degree of fiber–matrix interactions because these fibers were spontaneously distributed in CF.

WVP. The food deterioration in packages depends on water transfer between the internal products and the surroundings. As such, water-barrier properties of packages are required to limit that deterioration.[34,35]

The film with a CF/MS ratio of 1:0 showed a higher WVP (0.567 ± 0.01 g mm kPa^{-1} h^{-1} m^{-2}) than the films with CF/MS ratios of 1:1 and 1:2 (0.326 ± 0.02 and 0.293 ± 0.00 g mm kPa^{-1} h^{-1} m^{-2}, respectively) as shown in Table I. WVPs of the films produced in this study significantly decreased ($p < 0.05$) with increasing MS proportion. The higher WVPs found for the films with a CF/MS ratio of 1:0 could be explained by their more open, more porous structure and lower density (Figure 1) than the structures containing MS; this facilitated the migration of water vapor molecules through the film.[5,7,36] The lower WVPs for films with CF/MS ratios of 1:1 and 1:2 were attributed to structural changes in the film morphology with the addition of the starch, which decreased the WVP values. Regardless of this fact, the protein–protein and protein–lipid

Table II. Color Measurements of the CF-Based Films

CF/MS (w/w)	Color			
	L	*a*	*b*	Δ*E*
1:0	52.15 ± 1.03^{c}	8.80 ± 0.27^{a}	30.48 ± 0.34^{a}	47.87 ± 0.82^{a}
1:1	70.14 ± 0.55^{b}	4.45 ± 0.14^{b}	27.84 ± 0.25^{b}	30.58 ± 0.60^{b}
1:2	77.54 ± 0.75^{a}	2.71 ± 0.16^{c}	21.67 ± 0.77^{c}	20.81 ± 1.07^{c}

The data are presented as the means plus or minus the standard deviations. Means in the same column followed by different letters (a–c) were significantly different ($p < 0.05$) according to Tukey's test.

interactions forming the film matrix, together with the starch–starch interactions and starch–protein interactions (for the films containing MS), allowed the CF-based films to present satisfactory water-vapor-barrier properties.[4]

The rate of migration of water molecules through a biopolymer film decreases with decreasing film biopolymer size. This low migration increases the moisture-barrier properties of the film because of the increase in the tortuous path of water molecules to travel through the film.[37] MS had a smaller particle size than CF, and this may explain the reduced WVP for the films containing MS.

The moisture-barrier properties of the CF-based films were lower than those of many other biopolymers films, including apple-peel-based films (4.20–7.56 g mm kPa^{-1} h^{-1} m^{-2})[21], potato-peel-based films (2.99–5.30 g mm kPa^{-1} h^{-1} m^{-2}),[33] defatted-mustard-seed-meal-based films (3.40–4.96 g mm kPa^{-1} h^{-1} m^{-2}),[36] a calcium caseinate film (7.91 g mm kPa^{-1} h^{-1} m^{-2}),[38] a wheat gluten film (4.52 g mm kPa^{-1} h^{-1} m^{-2}),[39] and a whey protein isolate film (whey protein isolate/glycerol = 1:1, 5.16 g mm kPa^{-1} h^{-1} m^{-2}).[26] However, the moisture-barrier properties of the CF-based films were higher than those obtained from an amaranth flour-based film (0.0093 g mm kPa^{-1} h^{-1} m^{-2})[11] and synthetic films, such as a high-density polyethylene film (0.0012 g mm kPa^{-1} h^{-1} m^{-2}) and a polyester film (0.0091 g mm kPa^{-1} h^{-1} m^{-2}).[26]

Color Measurement. The color parameters values (*a, b, L,* and Δ*E*) for the CF-based films are shown in Table II. All of these attributes showed significant differences ($p < 0.05$) between the samples, and this indicated that the color was affected by the different CF/MS proportions in the films.

The color parameter *L* provides a measure of lightness. Color values range from 0 to 100, with 0 designating a perfect black and 100 designating a pure light. With regard to the Δ*E* parameter, greater values of Δ*E* indicate films with a higher color intensity.[40] The differences in color (Δ*E*) were significantly different ($p < 0.05$) between the films with all of the tested ratios. The highest value obtained from films with a 1:0 ratio, followed by the films with 1:1 and 1:2 ratios. These differences could be interpreted because of the chia seed dark coloration. This same behavior was also observed for the color parameters *a* and *b*. As shown in Table II, the films with a ratio of 1:0 were more yellowish (greater value for *b*) and more reddish (greater value for *a*) than the other films. The lower value for Δ*E* and the higher value for *L* (degree of lightness) of the 1:2 films was explained by the higher amount of starch in these films compared to the values at the others ratios, so when more MS was added to the formulation, the film was clearer.

The color values in Table II suggest that the CF-based films were darker and more red and yellow than biofilms made from amaranth flour,[4,11] banana flour,[5] and quinoa starch.[20] On the other hand, CF-based films were less yellowish than defatted-mustard-seed-meal-based film[36] and less yellowish and reddish than a carrot-puree-based film.[22]

Light Transmittance and Transparency Value. Transmission of UV and visible light in the range of 200–800 nm of the CF-based films are presented in Table III and in Figure S1 in the Supporting Information. The films had excellent barrier

Table III. Light Transmittance and Transparency Values of the CF-Based Films

Sample CF/MS (w/w)	Light transmittance (%) at different wavelengths (nm)								
	200	280	350	400	500	600	700	800	A600 (mm)
1:0	0.01	0.01	0.01	0.32	1.71	2.93	3.83	4.55	7.38 ± 0.27^{a}
1:1	0.01	0.01	0.24	3.42	7.63	10.24	12.20	13.85	5.03 ± 0.30^{b}
1:2	0.01	0.05	1.81	9.72	15.73	18.86	21.10	23.05	4.04 ± 0.32^{c}
Synthetic films[a]									
OPP	4.22	71.79	81.09	83.33	86.10	87.51	88.25	88.71	1.57
LDPE	0.45	27.64	35.76	39.97	45.53	49.81	53.24	56.33	4.26

The data are presented as the mean plus or minus the standard deviation. Means in the same column followed by different letters (a–c) were significantly different ($p < 0.05$) according to Tukey's test.
[a] Data obtained from Guerreiro *et al.*[41]

Table IV. Tensile Properties of the CF-Based Films under Dry and Wet Conditions

	TS (MPa)		EB (%)		YM (MPa)	
CF/CS (w/w)	Dry	Wet	Dry	Wet	Dry	Wet
1:0	0.77 ± 0.13^{c}	0.58 ± 0.11^{c}	5.16 ± 0.82^{a}	9.73 ± 1.50^{b}	25.64 ± 0.07^{c}	10.40 ± 1.99^{c}
1:1	4.78 ± 0.07^{b}	1.53 ± 0.14^{b}	1.52 ± 0.09^{b}	10.19 ± 1.61^{b}	374.62 ± 28.85^{b}	27.48 ± 7.05^{b}
1:2	6.26 ± 0.48^{a}	2.18 ± 0.09^{a}	1.05 ± 0.09^{b}	13.24 ± 1.76^{a}	681.39 ± 94.02^{a}	39.92 ± 5.08^{a}

The data are presented as the means plus or minus the standard deviations. Means in the same column followed by different letters (a–c) were significantly different ($p < 0.05$) according to Tukey's test. Dry conditions: 52% RH, 25°C, and 48 h. Wet conditions: 90% RH, 25°C, and 48 h.

properties to light in the UV range (200–280 nm). These results indicate that the CF-based films had a protective ability against UV radiation because of their UV-barrier capabilities; this suggests their potential preventive effect on product oxidation induced by UV light. In contrast, some synthetic polymers films, such as oriented polypropylene (OPP) and low-density polyethylene (LDPE), did not prevent the passage of UV light above 280 nm, as reported by Guerreiro *et al.*[41] and as shown in Table III.

The evaluation of the transparency values (Table III) revealed that the films with a 1:2 ratio had a significantly higher transparency (4.04 A_{600}/mm) than the films prepared at a 1:1 ratio (5.03 A_{600}/mm); in turn, the films prepared at a 1:0 ratio (7.38 A_{600}/mm) had the lowest transparency. The change in transparency was related to the composition of the raw materials in the films. The higher levels of protein, lipids, and fiber present in CF in addition to the presence of phenolic compounds might have contributed to the lower transparency (and greater opacity) of the 1:0 films. Moreover, in comparison with commercial films used for packaging purposes, the transparency of the CF-based films was lower than that of OPP but closer to LDPE in the films with ratios of 1:1 and 1:2.[41]

Mechanical Properties. TS is a property that is commonly used to evaluate the resistance of films.[3] In this study, the evaluation for the tensile properties (TS, EB, and YM under dry and wet tensile conditions) is summarized in Table IV. The stress–strain curves of the dried CF-based films are presented in Figure S2 in the Supporting Information. The mechanical data were influenced by the conditions before testing.

With regard to the dried films, the films with a ratio of 1:0 had significantly ($p < 0.05$) lower values of PF, TS, and YM compared to those with other ratios (containing MS). The results indicate that the 1:0 films were more flexible than the 1:1 and 1:2 films. However, the 1:1 and 1:2 films were more resistant and rigid than the 1:0 films. The CF/MS films plasticized with glycerol presented low to intermediate TS values, which ranged from 0.77 to 6.26 MPa. These results confirm that the protein and lipids present in the films could collaborate with the plasticizing effect and that the protein did not contribute to the formation of a stronger network in this case.[5,30] Meanwhile, in their attempt to clarify the lower TS values for rice-flour-based films compared to rice-starch-based film, Dias *et al.*[7] found that these results could be explained by the existence of irregularities at the microstructural level and the presence of lipids in the flour, as lipids are unable to form a cohesive and continuous matrix. Thus, these findings may explain the lower TS value for the 1:0 films found in this study. Even more, the rice flour used to produce the previously cited film contained a high amount of starch in its structure, which did not occur in the 1:0 films. The rice-flour-based films developed by Dias *et al.*[7] presented TSs of 10.3 and 1.3 MPa for 20 and 30% glycerol based on total solids, respectively. These TS values were within the results obtained from the 1:1 and 1:2 CF-based films in this study, with the consideration that only 17% of glycerol based on total solids was used. Furthermore, compared to other biodegradable films, the 1:0 ratio film showed the lowest value for TS. Films with 1:1 and 1:2 ratios had higher TS values than those obtained from biofilms based on amaranth flour (1.5 MPa; 22.5% glycerol).[11] However, the TS values of these ratios were in agreement with those reported in the literature for biofilms based on quinoa flour (4.1 MPa, 21% glycerol)[44] and achira flour (7.0 MPa, 17% glycerol)[3] and were lower than what is reported for biofilms based on banana flour (9.2 MPa, 19% glycerol).[5]

The dried CF-based films presented a low flexibility, as shown by EB values ranging from 1.05% (1:2 ratio) to 5.16% (1:0 ratio), with a significant difference ($p < 0.05$) between these ratios. There was no significant difference ($p > 0.05$) between the 1:1 and 1:2 ratios. The MC and lipids could account for these different values. In fact, the larger moisture and lipids content in the 1:0 films exerted an important and well-known plasticizer effect; this reduces the mechanical resistance and increases the flexibility (EB) of biopolymer films.[5,11] Dias *et al.*[7] found an EB of 2.7% for rice flour films (plasticized with 20% glycerol), and Kang and Min[33] found an EB of 5.3% for potato-peel-based films (plasticized with 30% glycerol).

YM is an indicator of film rigidity. The YM values for the dried CF-based films were between 25.64 ± 0.07 MPa (ratio = 1:0) and 681.39 ± 94.02 MPa (ratio = 1:2). The existence of higher protein, lipid, and fiber contents in the 1:0 ratio significantly influenced the film rigidity ($p < 0.05$, Table IV) and decreased the value for such films. In addition, the increased rigidity of the films with 1:1 and 1:2 ratios took into account the formation of a network between MS and CF. Such a network enabled the formation of a denser polymer matrix with a greater TS and, consequently, a greater YM. Dias *et al.*[7] found a YM value of 560.7 MPa for rice flour films plasticized with 20% glycerol.

The wetted films (Table IV) showed quite different mechanical properties than the dried films. The wetted CF-based films exhibited lower TS and YM and higher EB than the corresponding dried ones. The reason might have been the

Table V. Puncture Properties of the CF-Based Films

CF/CS (w/w)	PF (N)	PD (%)
1:0	1.38 ± 0.07^{b}	1.03 ± 0.23^{a}
1:1	6.73 ± 0.39^{a}	1.09 ± 0.93^{a}
1:2	7.02 ± 0.28^{a}	1.42 ± 0.11^{a}

The data are presented as the means plus or minus the standard deviations. Means in the same column followed by different letters were significantly different ($p < 0.05$) according to Tukey's test.

hydroplastization induced by water (higher humidity conditions). Similar behavior was found by Liu *et al.*[42] and Wen *et al.*[43]

With regard to the mechanical properties derived from the dried conditions of the puncture tests (Table V), similar results to those of the tensile tests (dried conditions) were obtained. PF followed the same trend as TS. However, PD did not present significant differences ($p > 0.05$) for any of the tested formulations; this was a different behavior than EB presented. Andrade-Mahecha *et al.*[3] studied achira-flour-based films and reported values of 5.8 N for PF and 7.8% for PD, whereas Araujo-Farro[44] reported a PF value of 7.0 N and a PD value of 2.2% from quinoa-flour-based films.

Overall, the results from SEM, WVP, and mechanical analysis show that the more ordered and homogeneous microstructure for the 1:1 and 1:2 films led to films with lower WVP and higher TS values compared to those for the film without MS (1:0).

CONCLUSIONS

We conducted this work to investigate the feasibility of the use of chia seed flour to form edible films. CF had the ability to form films without the addition of a starch; however, the resistance to break and elongation of the film was impaired, so the incorporation of a starch (MS in this study) into the film formulation showed the ability to increase these properties because of the formation of a more compact structure. The results from studies of WVP and water solubility indicate that the CF-based films had a relatively better water resistance compared to other edible films made from different sources in previous studies. Such properties could be explained by the presence of native lipids, proteins, and fiber in the biofilms. Thus, this evidence shows the potential of such films to be applied in food products over a large range of MCs. Furthermore, the developed films possessed dark coloration (red to yellow), low to intermediate transparencies, and protection against UV radiation. The color parameters changed as more MS was added, with the films becoming clearer with more MS.

With the actual trend toward an increasing consumption of functional foods and with the high nutritional value of chia seeds and, consequently, its films taken into account, it will be interesting to explore the development of edible films made from this raw material and to use it as a packaging or a coating material that might be eaten together with the food product. Nevertheless, the properties of CF-based films could be further improved to obtain films with greater mechanical properties.

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