Scale-up of the production of soy (Glycine max L.) protein films using tape casting: Formulation of film-forming suspension and drying conditions

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The processing variables used to produce biopolymer films may have great influence on the material’s properties and production time. The aim of this work was to determine i) a suitable formulation (concentration and rheological properties) for tape casting processing of soy protein suspensions ii) the effects of the drying method (heat conduction, C; and infrared radiation, IR) and temperature (40, 50 and 60 °C) on the drying kinetics and physical properties of resulting films. A soy protein isolate (SPI) concentration over 10.5% w/v was required for proper tape casting processing. Conduction drying at 60 °C (C60) and IR60 showed short drying times and similar drying kinetics. However, drying by IR60 yielded films having poor mechanical properties. Raising the drying temperature increased film’s tensile strength, Young’s modulus, glass transition temperature (Tg), and heat seal strength. The results of this study demonstrated that it is possible to produce soy protein films at large scale by tape casting. The influence of the processing method and drying conditions on soy protein film properties are discussed.

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1. Introduction

In the last decade, great efforts have been devoted to develop biodegradable materials derived from biopolymers of agricultural origin (Guilbert & Cuq, 2005) such as proteins. The properties of protein films have been shown to be strongly dependent on the conditions used throughout the production process (Denavi, Perez-Mateos, Anón, Montero, Mauri, & Gómez-Guillén, 2009; Denavi, Tapia-Blácido, Anón, Sobral, Mauri, & Menegalli, 2009; Foulk & Bunn, 2001; Lafargue, Lourdin, & Doublier, 2007; Tapia-Blácido, Sobral, & Menegalli, 2005). Small protein-based films (1–30 cm) are usually obtained in laboratory by casting. In this method a colloidal dispersion made up of the protein, a solvent and, frequently, a plasticizer is poured on an appropriate support. Afterwards, the suspension is commonly dried in convection ovens for relatively long times (e.g. 6–12 h). The film thickness is controlled by the mass of dispersion poured onto the support.

Tape casting is an upgrading of the casting method and has been widely used in paper, plastic, ceramics and paint industries (Cuq, Gontard, & Guilbert, 1998; Richard & Twiname, 2000, p. 293). In recent years it has been suggested as a suitable approach to scale-up biodegradable films, which can be negatively affected by some drying strategies (Moraes, Scheibe, Sereno, & Laurindo, 2013). This technique consists in spreading the film-forming suspension on a support with a doctor blade device, which allows controlling the suspension thickness. After that, the spread suspension is dried at a controlled temperature (Moraes, Scheibe, Augusto, Carciofi, & Laurindo, 2015; Moraes et al., 2013; Tanimoto, Hayakawa, & Nemoto, 2005). The heat supply during the drying step can be done by heat conduction, convection, infrared radiation, or by a combination of these mechanisms. Tape casting processing, requires film-formation suspensions with shear-thinning behavior and a low viscosity at the shear rates (to ensure appropriate flow conditions under the blade) and high viscosity at low-shear stresses.

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(to minimize undesired flow and sedimentation) (Moraes et al., 2013). Consequently, determining the concentration of each component used to obtain the film-forming suspension is fundamental to achieve the rheological-desired behavior.

Whereas several studies have determined the effects of film formulation on the properties of the resulting materials (Condés, Añón, Mauri, & Dufesne, 2015; Han, Shin, Park, & Kim, 2015; Ramos, Fernandes, Silva, Pintado, & Malcata, 2012) only few have focused on the influence of the drying process itself. Moreover, the influence of the drying temperature on the film properties depends on the characteristics of the film-forming suspension, such as preexisting gel phase or the tendency to thermal gelation during drying. Several phenomena may occur throughout drying such as the transition from a rubbery to a vitreous phase, phase separation or crystallization (Wittaya, 2012). Consequently, determining the concentration of each component used to obtain the film-forming suspension is fundamental to achieve the rheological-desired behavior.

The apparent viscosity (\(\eta_{app}\)) and flow behavior of film forming suspensions prepared with different SPI concentrations (5.0, 10.0, 10.5, 11.0 and 12.5 g SPI per 100 mL of suspension and 0.3 g glycerol/g SPI at pH 10.5. The film-forming suspensions were degassed under vacuum (2 min at 400 mm Hg) and immediately spread at a speed of 1.8 cm s\(^{-1}\) onto a 30 cm \(\times\) 84 cm Plexiglas\(^{®}\) plate, covered with a polyester film (Mylar\(^{®}\), DuPont, Brazil) to facilitate film removal after drying. A spreading system with a thickness control device (doctor blade BYK, Shanghai, China) (Fig. 1A) was used to spread the film-forming suspension on the tape casting support using a predefined thickness (2 mm). The Plexiglas\(^{®}\) plate was heated at 60 °C by a water circulating system through an internal coil.

### 2.2.2. Selection of protein concentration

Soy protein films were prepared by tape casting with film-forming suspensions containing 5.0, 10.0, 10.5, 11.0 and 12.5 g SPI per 100 mL of suspension and 0.3 g glycerol/g SPI at pH 10.5. The film-forming suspensions were degassed under vacuum (2 min at 400 mm Hg) and immediately spread at a speed of 1.8 cm s\(^{-1}\) onto a 30 cm \(\times\) 84 cm Plexiglas\(^{®}\) plate, covered with a polyester film (Mylar\(^{®}\), DuPont, Brazil) to facilitate film removal after drying.

The drying curves were obtained according to Moraes et al. (2015) by measuring the mass loss during drying. The weighing system consisted of an aluminum plate (15 cm \(\times\) 15 cm) with an internal electrical resistance controlled by a PID temperature controller. The plate had a Mylar\(^{®}\) film (thickness 250 μm) on its upper surface and placed on a semi-analytical scale (AS2000C, Brazil). The initial suspension thickness was adjusted to 2 mm. Drying rates were determined using Equation (2):

\[
R = \frac{\Delta X}{\Delta t} = \frac{X_t - X_t + \Delta t}{\Delta t}
\]

being \(R\) is the drying rate, \(X_t\) and \(X_t + \Delta t\) are the moisture contents at times \(t\) and \(t + \Delta t\), respectively. The experiments were done at least in duplicate.
Table 1
Nomenclature of the different drying process studied in this manuscript to obtain soy protein films by tape casting.

| Sample | Drying technology | Drying temperature (°C) |
|--------|-------------------|------------------------|
| CS0    | Conduction        | 40                     |
| CS50   | Conduction        | 50                     |
| C14    | Conduction        | 60                     |
| IR60   | Infrared light    | 60                     |

2.3. Films characterization

2.3.1. Thickness

Film thicknesses were measured with a digital coating thickness gauge (Check Line DCN-900, USA). Ten measurements were done for each drying condition evaluated.

2.3.2. Color

Color was determined with a Minolta Chromameter (CR 300, Minolta Chroma Co., Osaka, Japan) by placing the films on the surface of the white standard plate (color coordinates of \( L_{\text{standard}} = 97.3, a_{\text{standard}} = 0.14 \) and \( b_{\text{standard}} = 1.71 \)). The instrument was standardized by means of a set of three Minolta calibration plates. A CIE Lab color scale was used to measure the degree of lightness \( L \), redness \( +a \) or greenness \( -a \), and yellowness \( +b \) or blueness \( -b \) of the films. The total color difference \( \Delta E \) was calculated using Equation (3):

\[
\Delta E = \sqrt{(L_{\text{film}} - L_{\text{standard}})^2 + (a_{\text{film}} - a_{\text{standard}})^2 + (b_{\text{film}} - b_{\text{standard}})^2}
\]  

For each drying condition tested, nine measurements were done on different regions of the films and averaged. Three films were evaluated for each drying condition.

2.3.3. Opacity

Each film specimen was cut into a rectangular piece (0.5 cm \( \times \) 2 cm) and placed inside of the UV–vis spectrophotometer (Beckman DU650, Germany) at 500 nm. Air was used as reference for transparency measurements. The opacity of the films was calculated from the absorbance at 600 nm divided by the film’s thickness (mm) Measurements were performed in triplicate.

2.3.4. Moisture content (MC)

The films were weighed and dried in an oven at 105 °C until constant weight. MC was determined in triplicate for each drying condition and calculated as the percentage of weight loss relative to the initial weight (ASTM D644-94, 1994). Measurements were done in triplicate.

2.3.5. Solubility

Film solubility was determined in triplicate, according to Gontard, Guilbert, and Cuq (1992). Three film pieces (2 cm in diameter) were immersed in 50 mL distilled water at 22–25 °C and stirred for 24 h. After filtration, the insoluble material was taken and dried in a forced-air oven (105 °C for 24 h). Results were expressed in % and the measurements were done in triplicate.

2.3.6. Water-vapor permeability (WVP)

WVP tests were based on the ASTM method E96-80 (1989) with minor modifications (Gennadios, Weller, & Gooding, 1994). Films were put over a circular opening of 0.00185 m² in a permeation cell and stored at 20 °C in a desiccator. To maintain a 75% RH gradient across the film, anhydrous silica (0% RH) was placed inside the cell and a saturated NaCl solution (75%RH) in the desiccator. Water-vapor transport was determined from the weight gain of the permeation cell. After reaching steady-state conditions changes in the cell weight were recorded and plotted as a function of time. The results were represented by a straight line and calculated by linear regression (OriginPro8.5 software). The water vapor transmission rate was calculated from the slope of the straight line \((\text{g H}_2\text{O s}^{-1})\) divided by the cell area \((\text{m}^2)\). WVP \((\text{g H}_2\text{O/Pa s m})\) was calculated from Equation (4):

\[
\text{WVP} = \frac{\text{WVTR}}{P_{H_2O} (RH_d - RH_e) A} d
\]  

in which WVTR = the water-vapor-transmission rate, \( P_{H_2O} \) = vapor pressure of water (1753.35 Pa) at the test temperature \( (20 \degree C) \), \( RH_d \) = RH in the desiccator, \( RH_e \) = RH in the permeation cell, \( A \) = permeation area, and \( d \) = film thickness \((\text{m})\). Measurements were done in triplicate for each drying condition.

2.3.7. Glass transition temperature

The glass-transition temperature \((T_g)\) of the films was determined by differential scanning calorimetry using a DSC TA2010 calorimeter Model Q100 V9.8 Build296 (TA Instrument, New Castle, DE, USA) equipped with a TA5000 temperature controller and a quench-cooling accessory. Temperature and heat-flow calibrations were carried out according to ASTM indications, with lauric acid, stearic acid, and indium as standards. Hermetically sealed aluminum pans containing 10–15 mg of sample were scanned at 10 °C min⁻¹ over the range –100 to 220 °C. The Tg was considered to be the inflexion point of the base line caused by the discontinuity of the specific heat of the sample and was calculated by means of the Universal Analysis V4.2E software (TA Instruments, New Castle, DE, USA). All the assays were performed at least in duplicate.
2.3.8. Contact angle

Surface hydrophobicity was assessed by measuring the contact angle at room temperature, using a goniometer (Ramé-Hart Model 250, Succasunna, USA). A 5 μl drop of demineralized water was placed on the surface of the film using an automatic piston syringe and photographed. An image analyzer (DROPalmage Advanced v2.2) was used to measure the angle formed between the surface of the film and the drop of water. For each film, the hydrophobicity was obtained from the values of initial contact angle (average value of contact angles measured on both sides of the drop). Ten measurements were done on different areas of the film surface and averaged. Five film samples were measured for each drying condition tested.

2.3.9. Mechanical properties

Film mechanical properties were determined by performing tensile tests according to the ASTM D882-02 norm (ASTM, 2004), in a texture analyzer TA.XT2i (Stable Micro Systems, Surrey, England). The films were cut into strips, (6 mm x 80 mm) and mounted between two grips at opposite ends. Grip separation was 50 mm and the crosshead speed was 0.5 mm s⁻¹. The tensile strength and the elongation at break were determined directly from the stress-strain curves using Texture Expert V1.15 software (Stable Micro Systems, Surrey, England). The Young’s modulus (E) was calculated from the initial slope of the curve. At least six measurements were done for each drying condition tested.

2.3.10. Heat seal strength

Heat seal strength was measured according to ASTM F88-00 (2004). Two film stripes (76 mm x 25 mm) were cut and thermo-sealed on a hot wire-sealing machine (Lipari CC400, Argentina). The thermo seal strength was evaluated in a texture analyzer TA.XT2i, (Stable Micro Systems, England). The samples were attached on both ends sides of the seal area with two grips and subjected to a tensile test. The force (N) required for thermo-seal failure was recorded. The stripes were visually inspected to determine the nature of the failure (adhesive, cohesive or delamination), according to ASTM F88-00 (2004). Five samples were evaluated for each drying condition tested.

2.4. Statistical analysis

Experimental results were expressed as mean ± standard deviation and subjected to analysis of variance (ANOVA). Means were compared on the Tukey’s least significant difference test, at a significance level of α = 0.05, using the SYSTAT v. 12 software (Systat Software, Inc., Chicago, USA).

3. Results and discussion

3.1. Characterization of the soy protein suspensions

This study was done with a commercial soy protein isolate, in which all proteins were denatured (Supplementary Fig. 1) as depicted from the lack of the typical denaturation DSC thermogram endotherms corresponding to the two main protein fractions: β-conglycinin (7S) and glicinin (11S) in native SPI, according to Denavi & Perez-Mateos et al., 2009; Denavi & Tapia-Blácido et al., 2009; Mauri, 2011; Salgado, Molina Ortiz, & Echeverría, 2006, 2008; Mauri & Añón, 2006, 2008; Salgado, Fernández, Dragó, & Mauri, 2011; Salgado, Molina Ortiz, Petruccelli, & Mauri, 2010). This level was not adequate for the tape casting process tested herein because the suspension flowed off before the spreading step. Only at higher protein concentration (>10.5% w/v of SPI) the film forming suspension showed an adequate flow properties for pouring and spreading on the tape casting support. A second set of experiments showed that was not possible to obtain homogeneous films at SPI concentrations higher than 10.5% due to incomplete degassing which, resulted in the presence of bubbles in the films. Thus, suspensions containing 10.5% SPI, exhibiting adequate viscosities and could be spread properly on the tape casting support and yielded films with no bubbles were selected to evaluate the influence of drying conditions on the physical properties of tape casting soy protein films. The blade gap was fixed at 2 mm and operated at a shear rate ca. 9 s⁻¹ (Table 2).

decreased as the strain rate gradient increased. This pseudoplastic behavior of the suspensions became progressively pronounced at higher protein content. Flow-behavior index (n) decreased and K parameter increased with increasing SPI concentration. As expected, higher protein concentration lead to higher apparent viscosity. As mentioned before, shear-thinning behavior is adequate for to the production of films by tape casting (Moraes et al., 2013).

Previous studies have usually generated soy protein films by casting by using suspensions containing 5% w/v protein SPI (Denavi & Perez-Mateos et al., 2009; Denavi & Tapia-Blácido et al., 2009; Echeverría, Eisenberg, & Mauri, 2014; Mauri & Añón, 2006, 2008; Salgado, Fernández, Dragó, & Mauri, 2011; Salgado, Molina Ortiz, Petruccelli, & Mauri, 2010). This level was not adequate for the tape casting process tested herein because the suspension flowed off before the spreading step. Only at higher protein concentration (>10.5% w/v of SPI) the film forming suspension showed an adequate flow properties for pouring and spreading on the tape casting support. A second set of experiments showed that was not possible to obtain homogeneous films at SPI concentrations higher than 10.5% due to incomplete degassing which, resulted in the presence of bubbles in the films. Thus, suspensions containing 10.5% SPI, exhibiting adequate viscosities and could be spread properly on the tape casting support and yielded films with no bubbles were selected to evaluate the influence of drying conditions on the physical properties of tape casting soy protein films. The blade gap was fixed at 2 mm and operated at a shear rate ca. 9 s⁻¹ (Table 2).
Table 2
Rheological properties of film forming suspensions containing different concentrations of soy protein isolates: (a) index of consistency (K), and flow-behavior (n) calculated by the Ostwald de Waele model; (b) Apparent viscosity values at 9 s⁻¹ shear rate (D).

| g SPI/100 mL | a. Parameters of Ostwald de Waele model | b. Apparent viscosity (mPa s) |
|-------------|---------------------------------------|-----------------------------|
|             | K (Pa sⁿ) | N | r² | D = 9 s⁻¹ |
| 5.0         | 0.068 ± 0.010 c | 0.792 ± 0.012 a | 0.999 | 53.7 ± 2.7 e |
| 10.0        | 0.979 ± 0.038 bc | 0.602 ± 0.008 b | 0.999 | 393.4 ± 6.1 d |
| 10.5        | 1.260 ± 0.196 bc | 0.588 ± 0.012 b | 0.999 | 510.4 ± 57.2 c |
| 11.0        | 2.062 ± 0.267 b | 0.530 ± 0.018 e | 0.999 | 740.6 ± 53.7 c |
| 12.5        | 6.568 ± 1.118 a | 0.396 ± 0.025 d | 0.997 | 2232.2 ± 117.7 a |

Values for each film are means ± standard deviations. Values with different superscript letters in a given column are significantly different (p < 0.05) according to the Tukey test.

Fig. 3. Drying curves of SPI films using different temperatures and methods (A); drying rates (B). All represented in duplicate.

3.2. Drying kinetics

The drying curves of soy protein films for C₄₀, C₅₀, C₆₀, and IR₆₀ are shown in Fig. 3. The thermographs showed in Fig. 3a pointed out that the temperature of the film was homogeneous until the late stages of drying. The drying processes showed a short initial period of temperature adaptation, a constant rate period and a falling rate period (Moraes et al., 2015). The constant drying rate was 0.10 g g⁻¹ min⁻¹ for IR₆₀ and 0.10 g g⁻¹ min⁻¹, 0.04 g g⁻¹ min⁻¹, and 0.014 g g⁻¹ min⁻¹ for C₆₀, C₅₀, and C₄₀, respectively. In all cases, the transition between constant rate and falling rate periods occurred in the range of 60–75% moisture content. As expected shorter drying times were found at higher drying temperatures (Daud & Armstrong, 1988; Moraes et al., 2014; Moreira et al., 2011; Reis et al., 2013). The drying time was 120 min for IR₆₀, drying method, and 580, 240 and 120 min for C₄₀, C₅₀, and C₆₀, respectively. This drying time is much shorter than the normally used for films prepared by casting (Denavi & Perez-Mateos et al., 2009; Denavi & Tapia-Blácido et al., 2009; Moraes et al., 2013).

3.3. Effect of drying conditions on film properties

Regardless of the temperature used, the films dried by conduction presented similar thickness (≈ 105 μm). In contrast the films dried by IR radiation were thicker (Table 3), implying a lower degree of compaction of the protein matrix probably due to a different degree of molecular unfolding or cross-linking within the protein network of the film. Both the drying mechanism and temperature modified protein conformation and determined fundamentally the ability of protein chains to interact with each other as well as with other components of the formulation under consideration during film formation. The type and number of interactions involved in the stabilization of a protein matrix (disulfide covalent bonds, hydrogen bonding, electrostatic attractions, and hydrophobic bonding) would determine the degree of cross-linking and the hydrophobic-hydrophilic character of the films and also correlate with the latter’s physicochemical, mechanical, and barrier properties (Denavi & Perez-Mateos et al., 2009; Denavi & Tapia-Blácido et al., 2009).

All the films were homogeneous and had the yellow hue typically observed in soy protein biopolymers (González, Strumia, & Alvarez-Igarzabal, 2011; Song, Tang, Wang, & Wang, 2011). No differences among color parameters (L*, a*, b*, and ΔE) were observed among films (Table 3). Denavi and Perez-Mateos et al. (2009); Denavi and Tapia-Blácido et al. (2009) also reported that the drying

Table 3
Thickness, color (L*, a*, b*, ΔE), and opacity of SPI films processed by tape casting and dried at 40, 50 and 60 °C by conduction or at 60 °C by IR radiation.

| Sample | Thickness (μm) | Hunter-lab color parameters | Opacity (μA/mm) |
|--------|---------------|----------------------------|----------------|
|        |               | L* | a* | b* | ΔE |                        |
| C₄₀    | 105.8 ± 17.3 b | 84.5 ± 1.4 a | -1.8 ± 0.3 a | 31.2 ± 3.1 a | 32.2 ± 3.3 a | 1.5 ± 0.05 a |
| C₅₀    | 107.8 ± 12.5 b | 86.1 ± 0.5 a | -2.2 ± 0.1 a | 29.0 ± 0.9 a | 30.1 ± 1.0 a | 1.0 ± 0.05 b |
| C₆₀    | 104.2 ± 14.6 b | 86.1 ± 2.0 a | -2.1 ± 0.4 a | 28.0 ± 2.6 a | 28.7 ± 3.0 a | 1.3 ± 0.01 b |
| IR₆₀   | 185.3 ± 31.4 a | 85.4 ± 3.3 a | -1.9 ± 0.7 a | 30.7 ± 5.4 a | 31.4 ± 6.1 a | 0.6 ± 0.02 c |

Values for each film are means ± standard deviations. Values with different superscript letters in a given column are significantly different (p < 0.05) according to the Tukey test.
conditions of soy protein films prepared by casting did not affect color. However, the opacity of the films decreased at higher drying temperatures (Table 3), which could be due to extensive protein cross-linking. The mechanisms determining the differences in opacity and thickness between C60 and IR60 (Table 3) is unclear. The differences in the initial drying rate may be critical for the properties of the films (Denavi & Perez-Mateos et al., 2009; Denavi & Tapia-Blázido et al., 2009; Reis et al., 2013; Salgado et al., 2016). The location of the heating medium in the lower film surface in conduction drying could have also favored the formation of microbubbles, which may in turn affect the film transparency. Further work is needed to address that.

Table 4 shows the moisture content, solubility, water vapor permeability (WVP), contact angle (°) and glass transition temperature of the films processed by tape casting and dried at 40, 50 and 60 °C by IR radiation. The WVP values obtained are in the range of those published for soy protein films prepared by casting (Denavi & Perez-Mateos et al., 2009; Denavi & Tapia-Blázido et al., 2009; Salgado et al., 2011) without the dependence on film thickness that has been reported for hydrophobic films (Ghorpade, Li, Gennadios, & Hanna, 1995; McHugh, Avena-Bustillos, & Krochta, 1993).

The contact angles values suggest that the drying method influenced film's surface energy. Increasing the drying temperature by conduction resulted in lower surface hydrophobicity (lower contact angles) (Table 4). Moreover, the films dried by IR radiation presented higher surface hydrophobicity than those obtained by conduction drying at the same temperature (60 °C). Even though the kinetics of drying by IR60 and by C60 did not show differences, in the former the heat was delivered from the top to the bottom of the film, while in the latter the heat was delivered in the opposite direction. These facts could explain differences in the films surface. During drying, water is progressively eliminated, changing proteins conformation and the interactions that can be established among protein chains (Denavi & Perez-Mateos et al., 2009; Denavi & Tapia-Blázido et al., 2009). It is worth noting that the conformational changes that may contribute to explain the modification in film’s surface hydrophobicity did not affect water content and WVP.

The films dried at 60 °C by conduction presented higher tensile strength and Young modulus and lower elongation at break than films dried 40 and 50 °C or dried by IR radiation at 60 °C (Fig. 4). Previous studies have shown that disulfide bridges are favored at high pHs and temperatures, presenting a major role in the stabilization of soy proteins, which in turn influence the mechanical properties (Mauri & Anón, 2006; Salgado et al., 2010). Denavi and Perez-Mateos et al. (2009); Denavi and Tapia-Blázido et al. (2009) reported that higher drying temperatures increased the tensile strength of soy protein films prepared by casting. However, in this case higher elongation values were also found. This suggests that the processing method may affect the level protein interactions within the film matrix. Further studies may be conducted to

### Table 4

| Sample | Moisture content (%) | Solubility (%) | WVP \times 10^{-10} (g/m Pa s) | Contact angle (°) | Tg (°C) DSC |
|--------|----------------------|---------------|-------------------------------|------------------|-------------|
| C0     | 12.04 ± 0.51 a       | 33.70 ± 0.01 a| 1.32 ± 0.19 a                 | 75.13 ± 4.40 a   | −20.8 ± 0.8 b|
| C50    | 12.79 ± 0.19 a       | 33.62 ± 2.42 a| 1.47 ± 0.33 a                 | 67.66 ± 7.41 ab  | −21.1 ± 0.5 b|
| C60    | 12.15 ± 0.12 a       | 31.23 ± 1.26 a| 0.95 ± 0.21 a                 | 56.07 ± 5.33 b   | −16.5 ± 0.7 a|
| IR60   | 12.25 ± 0.22 a       | 32.69 ± 1.95 a| 1.61 ± 0.68 a                 | 66.03 ± 5.80 ab  | −19.8 ± 0.3 b|

Values for each film are means ± standard deviations. Values with different superscript letters in a given column are significantly different (p < 0.05) according to the Tukey test.

**Fig. 4.** Tensile strength (A), Young’s modulus (B) and elongation at break (C) of films processed by tape casting, dried at 40, 50 and 60 °C (C40, C50, C60) by conduction and at 60 °C by IR radiation (IR60). Error bars are shown. Different letters indicated significant differences based on Tukey test at a level of significance of P ≤ 0.05.
determine the degree of protein interactions in the different films, as affected by the drying conditions and methods. The potential increase of protein crosslinking in C60 films, which may have contributed to improve the mechanical properties, was in line with the higher glass transition temperature presented by this sample (Table 4).

All the films were able to produce continuous thermo-seals, which may be appealing for certain technological uses (Scheibe, De Moraes, & Laurindo, 2014). Higher drying temperatures resulted in films with improved thermo-seal resistance. Conduction drying yielded films with higher thermo-seal resistance than IR radiation drying. Noteworthy, there was a negative correlation between the film contact angle and thermo-seal resistance (Fig. 5). The facts that in all cases the seal failure was adhesive suggest a major role of film’s surface on the thermo-seal resistance. However, thermo-sealing processes are affected by several factors that may exceed the film surface.

4. Conclusions

Results from this study demonstrate that soy proteins-based films can be produced by tape casting in large scale. The film-forming suspension should have the minimal protein concentration of 10.5% w/v to allow its correct application on the support. The drying conditions (heat transfer method and temperature) have great impact on the physical properties of the resulting films. Drying at 60 °C not only reduced the drying time, but also yielded in films with superior mechanical properties and heat seal strength than films dried at lower temperatures. Although a similar drying time was observed for IR60, these films had poorer mechanical properties than those dried by conduction.

In the tape taping process, a solution or suspension is spread onto a flat plate, heated with hot water circulating in a reservoir, which supplies the heat required by the drying process. One advantage of this drying method is the possibility of producing larger films in short times, when compared to the casting procedure, which result in films with no more than 30 cm. Tape casting uses relatively low drying temperature and short drying times, resulting in films with good mechanical properties. The use of infrared heating is an alternative to improve drying, when associated with heat conduction from the hot plate. Therefore, tape casting is a suitable process to produce films at industrial scale. Though further work is needed to determine the changes in protein structure and interaction in response to different drying treatments, the information presented herein may be useful for projects aiming to scale up the production of custom-designed protein films.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.foodhyd.2016.12.029.

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