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TANNIN-MODIFIED SOYBEAN PROTEIN CONCENTRATE FOR WOOD ADHESIVE

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Abstract:

Soybean protein concentrate (SPC) modified with condensed mimosa tannin (CT) were employed as eco-friendly and formaldehyde-free adhesives for glued-wood joints. *Eucalyptus grandis* wood boards free of knots and cracks were used as substrate. Thermogravimetric analysis (TGA) showed that tannin provided higher thermal stability to the adhesive, which allowed expanding the temperature range for hot pressing. Apparent viscosity and dynamic contact angle were measured to evaluate the influence of tannin content on rheological behaviour and the wettability process. A classic shear-thinning behaviour was observed for all the adhesives. Apparent viscosity and equilibrium contact angle reached a maximum value for low CT content. This effect was attributed to the existence of associative interactions between CT and SPC. Bonding quality parameters (wood failure percentage and shear strength) of the glued-wood joints were measured according to EN 302-1:2004 standard. SPC adhesive modified with 1 % w/w CT showed the best performance in dry conditions. These adhesives were suitable for glued-wood joints for indoor environments.

Keywords: soybean protein concentrate; mimosa tannin; rheology properties; wettability; shear strength.

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1. Introduction

Over the last decades, significant efforts have been made for the development of environmentally sound adhesives as alternatives to the formaldehyde-based glues widely applied in the wood-based materials industry (Nordqvist et al., 2013; Khosravi et al., 2015). Unfortunately, synthetic adhesives come from petrochemical sources and most of them have formaldehyde in their formulations, which has been classified in 2004 as carcinogenic class I by the International Agency for Research of Cancer (IARC) and more recently, in 2016 the European Union has re-categorized formaldehyde as carcinogen class 1 B (Solt et al., 2019, Jang & Li, 2015).

Proteins have a long tradition as wood adhesives and have been subjected to a significant amount of research due to their low toxicity, in addition to its availability at large scale as by-products of other industries (Khosravi et al., 2015). Several protein-based adhesives have been reported, such as soy protein (Mo et al., 2003; Ciannamea et al.,2012; Chalapud et al., 2020, Nicolao et al., 2020), wheat protein (Nordqvist et al., 2013), cow blood (Lin & Gunasekaran, 2010), among others.

Soy proteins are being used as adhesives because they comply with the above-mentioned advantages of proteins: low cost, easy handling, low pressing temperature, and good ability to bond wood (Jang & Li, 2015). Accordingly, soybean proteins are good alternatives, particularly in Argentina, that is one of the largest producers of soybeans in the world (FAO, 2019). Soy proteins a commercially available under different grades depending on the protein content, including defatted soy flour (SF, ~50 % w/w protein), soy protein isolates (SPI ~90 % w/w protein) and soy protein concentrate (SPC ~65–70 % w/w proteins) (Ciannamea et al., 2010), which is economically more favourable than SPI (i.e., $2.05/kg for SPC vs $2.70/kg for SPI) (Hojilla-Evangelista, 2010; Song et al., 2011) and performs similarly (Ciannamea et al., 2010).

Although the great potential of SPC for adhesives, improvements in moisture resistance is mandatory to gain industrial uses. Grafting and crosslinking have been reported as some of the main strategies to improve protein stability in humid environments (Damodaran & Zhu, 2016; Ciannamea et al., 2012, Ghahri et al., 2021). Condensed tannins (CT), extracted from agroforestry sources as oligomers or polymers of flavonoids (Liu et al., 2017), have the ability of interact with a variety of proteins through covalent, hydrogen, ionic and hydrophobic bonding (Peña et al., 2010; de Freitas & Mateus, 2012). Several works report improvements in adhesion properties of protein-based adhesives by adding tannins (Ping et al.,

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2012; Ozdal et al., 2013; Ghahri et al., 2017; Liu et al., 2017; Ghahri et al., 2021). The control of tannin/protein ratio, which in turn determines tannin/protein interactions, is crucial for the formation of tannin/protein aggregates and gels, responsible for the rheological and mechanical properties of the adhesives (de Freitas et al., 2012).

In this context, the aim of this research was to evaluate the rheological behaviour, the wettability and wood bonding performance of aqueous adhesive based on soy protein concentrate (SPC) and condensed mimosa tannin (CT).

2. Materials and Methods

2.1. Materials

Soybean protein concentrate (SPC, Solcom S 110; Isoelectric point (Ip) = 4.5), containing 7 % moisture, 69 % protein, 1 % fat, 3 % fibres, 5 % ash and about 15 % non-starch polysaccharides (mainly cellulose, non cellulose polymers and pectin polysaccharides) as mean composition and has an average particle size that could pass through a 100 mesh, was supplied by Cordis SA (Buenos Aires, Argentina). Condensed mimosa tannin extract (CT) was purchased from SETA (Brasil). Sodium hydroxide (NaOH) and all chemical reagents (p.a. grade), were provided by Anedra (San Fernando, Argentina). 26-year-old *Eucalyptus grandis* trees grown in Concordia, Entre Ríos (Argentina), were supplied by Aserradero Ubajay. Boards with a cross section of 5 mm x 150 mm were obtained from wooden log according to the scheme shown in Fig. 1. Test samples were carefully inspected and selected avoiding major defects such as knots or cracks.

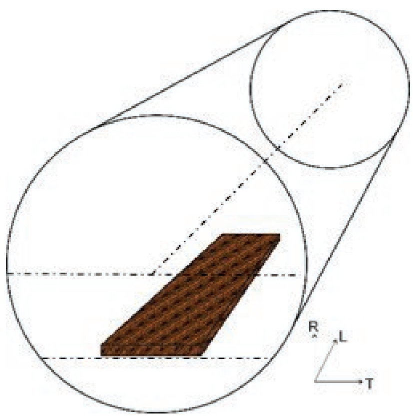

Figure 1: Boards obtained from wooden log. L, R and T indicate the longitudinal, radial and tangential directions, respectively.

2.2. Adhesive preparation

All adhesive dispersions were prepared as described in our previous works (Leiva et al., 2007; Ciannamea et al., 2010; Ciannamea et al., 2012). For it, alkali-modified soybean protein concentrate adhesive was prepared by dispersing SPC (10 g) in 0.2 % w/w NaOH solution (100 mL) under constant stirring at 500 rpm for 2 h at 25 ± 2 °C (Cole-Parmer IL, USA). Condensed tannin-modified SPC adhesives (CT-SPC), were obtained similarly by adding different CT amount (0, 1, 5, and 10 % w/w on SPC dry basis) into the SPC slurry. Adhesives were named as XCT-SPC, where X correspond to CT concentration (w/w SPC).

2.3. Adhesive characterization

Thermogravimetric analysis (TGA) was performed using a Shimadzu 50 (Shimadzu Corp., Japan) thermal analyser. Temperature was raised from 25 °C to 600 °C, at a heating rate of 10 °C/min and under nitrogen atmosphere (20 mL/min), to avoid thermo-oxidative degradation. Thermal parameters such as the onset of the degradation process at 5 % of conversion of the main weight loss step (T_5), the temperature of the maximum degradation rate (T_{max}) and the residue 600 °C (R_{600}) were taken from the experimental curves. Viscosity measurements were performed using a rheometer (Anton Paar MCR 301, Graz, Austria), with a plate and cone geometry at 20 ± 0.2 °C, over a shear rate range of 0-100 s^{-1}.

2.4. SPC adhesive bonding performance on wood

2.4.1. Determinations on wood

Wood board (150×150×5 mm^3) were sanded on one face along the fibre direction with a 600-grit sandpaper in order to get a uniform roughness. Roughness (Ra) was measured with a Taylor Hobson (Surtronic 3+ Model) roughness tester (Japan). Wood board surface were divided in eight areas in order to measure the roughness. The tracing length in each area was 4 mm. Finally, wood boards were stored in an environmental chamber at 20 ± 2 °C and 65 ± 5 % relative humidity for 7 days before testing.

Dynamic contact angle determinations were carried out using a Standard Goniometer Model 250 (Succasunna, NJ, USA). A 5 µL drop of adhesive was placed over the surface of a wood board (roughness between 6.10 and 6.20 µm). The image of the drop was captured by a video camera from 0 to 120 s every 5 s and the angle was determined by using an image analysis software. All tests were carried out at 25 ± 2 °C. Reported values are the average of five replicates for each formulation.

2.4.2. Glued-wood joints preparation and characterization

SPC-based adhesives were evaluated in terms of their ability to bond two wood surfaces. Preconditioned wood samples with surface roughness in the range of 6.10-6.20 were selected. Adhesives were applied to one side of each board at a spreading rate of 2.5 mg/cm^2 (dry adhesive basis). The two glued-wood board were stacked together and hot-pressed (EMS, Buenos Aires, Argentina) at 140 ± 2 °C for 10 min at 1.2 MPa. After that, all samples were conditioned in an environmental chamber at 20 ± 2 °C and 65 ± 5 % relative humidity for 7 days.

Glued quality was evaluated measuring the shear strength and wood failure percentage according to EN 302-1:2004 standard with a crosshead speed of 0.5 mm/min (Instron Testing Machine 4467, England). Samples were subjected to three different environments treatments before testing: A1, preconditioned 7 days in an environmental chamber at 20 ± 2 °C and 65 ± 5 % RH; A2, similar to A1 followed by soaking in distillate water at 20 ± 2 °C for 4 days; A3, similar to A2 followed by storing at 20 ± 2 °C and 65 ± 5 % RH for 7 days. Reported values were the average of ten measurements. Experimental mechanical data were statistically analysed using the one-way analysis of variance (ANOVA) along with Tukey's tests at 95 % confidence interval (a = 0.05)

3. Results and discussion

3.1. Thermal stability

The thermal stability of CT-SPC adhesives was analysed from normalized weight loss curves and their derivatives (Fig. 2). Calculated thermal parameters (T_5, T_{max}, R_{600}) are listed in Table 1. The thermal decomposition of control SPC occurred in two stages; the first one in the range of 25-150 °C corresponding to the evaporation of residual moisture and the second one, from 150–600 °C was related to the random cleavage of peptide bonds in the protein backbone, resulting in smaller peptides (Ghahri et al., 2017, 2018). The final residue was about 32%. The inclusion of increasing CT shifted T_5 and T_{max} toward higher values (Table 1) because protein-CT interactions as well as the chemical ring-fused structure of CT provides higher thermal resistance (Ghahri et al., 2018). The carbonaceous residue also increased with CT (Table 1) confirming the stabilizing effect of CT.

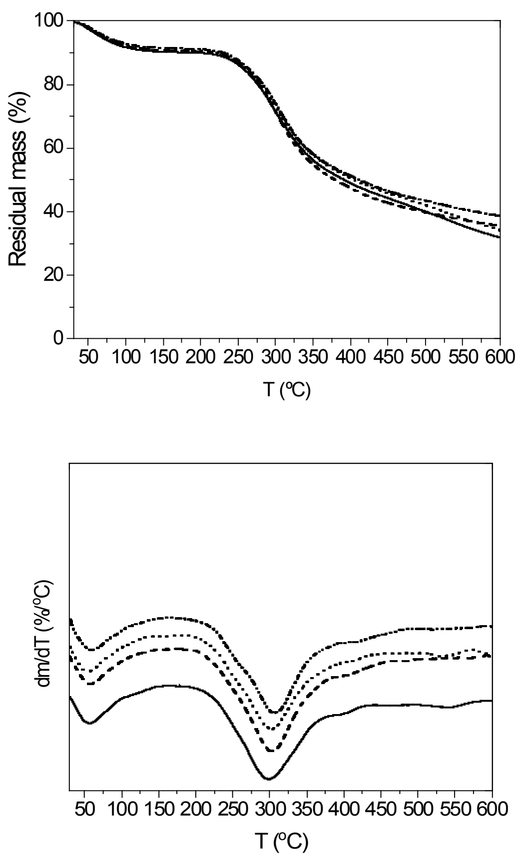

Figure 2: TGA/DTG curves of CT- SPC adhesives. 0CT-SPC (solid line), 1CT-SPC (dashed line), 5CT-SPC (dot line), 10CT-SPC (dash-dot line).

Table 1: Thermal properties of CT-SPC adhesives.

Adhesive	T_5 (°C)	T_{max} (°C)	R_{600} (% w/w)
0CT-SPC	67	300	32
1CT-SPC	69	302	35
5CT-SPC	68	303	34
10CT-SPC	73	307	39

3.2. Rheological behavior

The ability of SPC–based adhesives to wet, flow over and penetrate into the substrate without losing the adhesiveness is a key requisite to achieve a proper bond result (Ciannamea et al., 2012). These factors are directly dependent on the viscosity of adhesive and *Eucalyptus* microstructure. Lumen size, pit frequency, vessels size, occlusions by extractives or tyloses, and physical properties such as density and moisture content affect the glued quality (Hunt et al., 2018). Low viscosity–adhesives can excessively penetrate into substrate and reduce the amount of adhesive in the glue line. Conversely, a very high viscosity generates a poor penetration rate into substrate and therefore its ability to generate mechanical interlocking is less favoured (Nordqvist et al., 2013, Ciannamea et al., 2012, 2017). Representative viscosity curves of CT-SPC based adhesives are depicted in Fig. 3. A classic shear-thinning behaviour was record for all adhesives (viscosity starts to decrease immediately as shear rate increase), confirming the observations reported by other authors for different soybean protein-based adhesives (Wang et al., 2007; Ciannamea et al., 2012; de Freitas et al., 2012).

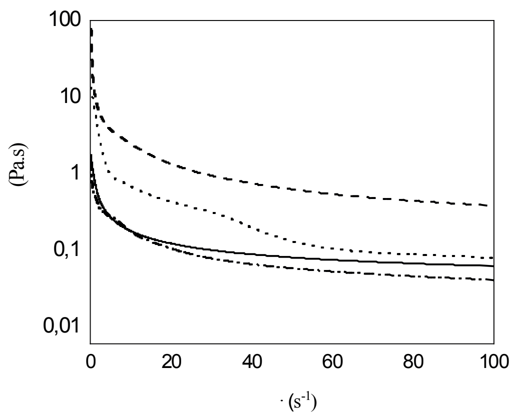

Figure 3: Viscosity as function of shear rate of soy bean adhesives. 0CT-SPC (solid line), 1CT-SPC (dash line), 5CT-SPC (dot line), 10CT-SPC (dash-dot line)

This rheological behaviour can be adequately expressed by the Herscher-Bulkley model (Wang et al., 2007):

$$\tau=\tau_0+K\cdot(\dot{\gamma}^n) \quad (1)$$

where τ is the shear stress (Pa); $\dot{\gamma}$ the shear rate (s^{-1}); *n* is the flow behaviour index and *K* is the consistency index (Pa s^n). The values of *n* and *K* were obtained from plots of log (τ-τ_o) vs log $\dot{\gamma}$. The τ_o value was obtained using the Casson equation:

$$\sqrt{\tau} = \sqrt{\tau_0} + \sqrt{(\mu_C . \dot{\gamma})} \quad (2)$$

where μ_c is the Casson viscosity (Hagenimana et al., 2007).

The values of τ_o, *n*, and *K* obtained applying Eq. 1 and 2 are summarized in Table 2. The flow behavior index varied from 0.64 to 0.69 indicating a pseudoplastic behaviour ($n < 1$). K τ_o, and apparent viscosity varied with CT content showing the highest values for 1CT-SPC formulation.

Table 2: Rheological properties of CT-SPC adhesives.

Adhesive	τ_o (Pa)	*n*	*K* (Pa·s^n)
0CT-SPC	0.71	0.64	0.18
1CT-SPC	17.29	0.53	1.85
5CT-SPC	3.16	0.58	0.91
10CT-SPC	0.88	0.69	0.14

This formulation displayed rheological parameters (τ_o and *K*) one order of magnitude higher than the control (see Table 2). In addition, the hight τ_o value for 1CT-SPC adhesive have also technological implications because it avoids the excessive losses by blasting during the colocation on the wood substrate. As the tannin content increased, τ_o, *K* decreased showing for the 10CT-SPC adhesive, similar rheological properties to the control adhesive. This behaviour would be associate with the different interactions that can occur between CT and SPC in solution. In the formulated adhesive under alkaline conditions, SPC and CT can interact by means of ionic interaction and hydrophobic effects (de Freitas et al., 2012). Recently, Ghahri et al. (2021) verified the occurrence of ionic and covalent bonding between soybean protein and condensed tannin even at ambient temperature (Ghahri et al., 2021). Also, hydrogen bonding can be established between the hydroxyl groups of phenolic compounds and carbonyl and amide groups of proteins while hydrophobic interactions can occur between the benzenic ring of phenolic compounds and the apolar side-chains of amino acids in proteins (Ozdal et al., 2013). The rheological behavior depends on these interactions, which are in turn a function of the relative concentration of CT-SPC and the number of accessible groups of both the protein chains and the carbohydrates of SPC (Ozdal et al., 2013, Santos-Buelga and de Freitas, 2009).

3.3. Wettability

Dynamic contact angle (θ_D) of CT-SPC adhesives was determined in order to analyse the wettability on wood substrate. Since wood is a porous material, the wetting process includes all the information on the contact angle formation, spreading, and penetrating ability of adhesive. Smaller angle contact represents better wettability and adhesion because of an increment in the interaction between adhesive and substrate (Aydin et al., 2007). The variation of θ_D for different CT-SPC adhesives on wooden surface is shown in Fig. 4. 1CT-ASPC adhesive formulation was impossible to evaluate due to their very high τ_o. This condition did not allow the drop to self-form on the wood surface; however, this did not prevent the adhesive from spreading on the surface by mechanical action. For 0CT-SPC, 5CTSPC and 10CTSPC adhesives, θ_D decreased abruptly during the first 20 s to reaching a stable value up to the end of the test (~60 s) (Fig. 4).

S-D model (Xu et al., 2012) was used to predict the penetration and spreading rate at a particular moment in time.

$$\theta_D = \frac{\theta_i \theta_e}{\theta_i + (\theta_e - \theta_i) exp\left[S\left(\frac{\theta_e}{\theta_e - \theta_i}\right)t\right]} \quad (3)$$

where θ_i is the initial contact angle, θ_e equilibrium contact angle and *S* the spreading/penetration parameter.

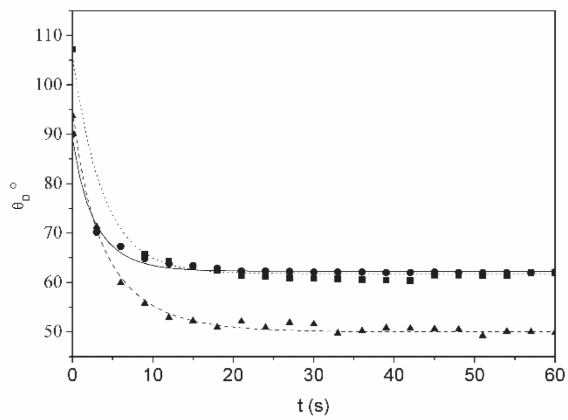

Figure 4: Representative curves of θ_D versus time (▲) 0CT-SPC, (■) 5CT-SPC, (●) 10CT-SPC, (lines) S-D model.

Table 3: Experimental θ_i and θ_e values and *S* parameter calculated from S-D model. Values followed by different letters are significantly different ($p < 0.05$).

Parameter	0CT-SPC	5CT-SPC	10CT-SPC
θ_i	85.7±6.0[a]	97.6±3.6[b]	94.4±4.2[b]
θ_e	45.1±4.2[a]	61.3±7.4[b]	58.1±5.6[b]
S (s^{-1})	0.32±0.11[a]	0.13±0.03[b]	0.16±0.05[b]
R^2	0.98	0.95	0.97

As seen in Fig. 4, experimental dynamic contact angle values of CT-SPC adhesives were fitted according to Eq. (3). The main parameters derived from contact angle are summarized in Table 3. The greater the *S* value of adhesive the faster its penetration and spreading ability on the wood surface. No significant differences were observed in *S* and θ_e in formulations containing 5 and 10 % w/w CT, evidencing the influence of CT in spreading/penetration ability of the adhesives

CT-SPC adhesives showed excellent dry (A1) and soak-dry (A3) shear strength (Figure 4). All tested samples in A1 and A3 condition presented a high percentage of wood cohesive failure similar to those reported by Piter et al, (2007), using the same wood specie and urea-melamine-formaldehyde adhesive. In wet conditions, the failure occurred completely in the glue line (Fig. 5) regardless the CT content. After wet treatment (A2 in Fig. 4) hydrogen bond interactions between SPC/wood could be broken down by water molecules, thus decreasing the adhesion performance (Mo et al., 2011; Wang & Wu, 2012). For A3 where water was evaporated before testing, interactions between SPC and wood surface seems to be recovered and also improved, since better adhesion strength was obtained as compared with A2 condition (Wang & Wu, 2012).

Dry, wet and soak shear strength showed the same tendency with CT content (Fig. 4). Regardless the type of treatment (A1, A2 or A3), shear strength displayed a maximum for 1CT-SPC adhesive in terms of mean values. For this formulation, dry, wet and soak shear strength increased 25, 87 and 16 %, respectively as compared to control SPC adhesive. The same trend was observed in the wood failure percentage except for wet conditions (A2), where the failure occurred fully in the adhesive irrespective of CT content.

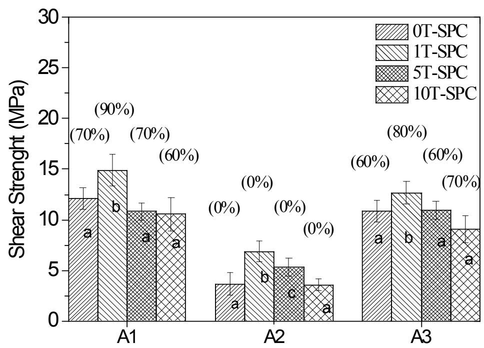

Figure 5: Shear strength of CT-SPC adhesives with different soaking treatments. Bars followed by different letters are significantly different ($p < 0.05$) (each type of treatment A1, A2, or A3, was independently evaluated). Data between parentheses indicate the average wood failure percentage.

As it was discussed above, 1CT-SPC adhesive showed the highest initial viscosity, which defines the impregnation degree (spreading and penetration into the porous structure of the wood interface), achieving a better bonding strength. Based on our results, 1CT-SPC formulation combined the best properties (viscosity and penetration/spreading rate) to obtain a good bonding strength. An optimum penetration is needed to enhance adhesion strength by developing an interactive zone at the interface. Less penetration would limit the formation of the three-dimensional zone at the interface. Too deep or too much penetration would result in 'dryout' at the interface (Cheng & Sun, 2006), resulting in reduced adhesion strength as observed for adhesive formulations with CT>1 % (w/w) and control (with lower viscosities).

4. Conclusions

The effect of tannin concentration on the performance of *Eucalyptus* wood boards bonded with CT-SPC adhesives was evaluated. The 1CT-SPC formulation showed the more adequate values of viscosity and spreading/penetration rate to reach the highest dry, wet and soak shear strengths and percentages of wood failure. The differences in shear strength found for CT-SPC adhesives could be attributed to the influence of tannins on/into soy protein chemical structures as well as physicochemical characteristics such as rheological properties (viscosity), and wettability. Regarding the adhesive performance, the formulation with a 1 % w/w of tannin showed the better result. Interesting, the best result for the 1CT-SPC adhesive was obtained under wet conditions where the shear strength increased 87% respect to control adhesive. Higher tannin level and lower viscosities than that of 1CT-SPC might fail due to the "dryout". Wood joints prepared with the SPC adhesives modified with low CT concentration were found to be an environmentally option for applications under indoor environments. Ongoing work is being undertaken to improve adhesive/substrate interactions by combining CT with a biobased aldehyde crosslinking agent.

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