

Tannin-Based Extracts of Mimosa Tenuiflora Bark: Features and Prospecting as Wood Adhesives

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1 **Tannin-based extracts of *Mimosa tenuiflora* bark: features and prospecting as wood**
2 **adhesives**

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21

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Abstract

Mimosa tenuiflora (Willd.) (MT) Poir. is an underutilized plant specie since its wood is mostly used for energy production. Nonetheless, the bark from this forest plant has a high amount of tannins and other valuable compounds. Tannins have high worldwide importance and, because of that, there is an increasing number of researches on biorefinery systems aiming at maximizing their exploitation. The present study evaluated tannins extracted from the MT bark and their use for producing wood adhesives. Three types of powdered tannins were extracted using different solvents: a pure aqueous solution, a 5% sodium hydroxide (NaOH) aqueous solution, and a 5% sodium bisulfite (NaHSO₃) aqueous solution. Also, distilled water, wheat flour, and formaldehyde were used as a solvent, a glue extender, and a catalyst, respectively. These adhesives were applied for bonding pine wood joints and their shear strengths were determined. All the MT-based adhesives showed high viscosities and, if compared to each other, yielded glue lines with similar shear strengths and similar shear deformations. That tannin-based glue incorporated with the tannin extracted using NaOH/NaHSO₃ stood out and yielded a similar bonding performance compared to a commercial adhesive applied as a positive control.

Keywords: Jurema-preta, Valuable raw material, Powder tannins, NaOH/NaHSO₃, Tannin adhesives

Introduction

Located at the semi-arid Brazilian region, the caatinga vegetation has a restricted timber production for several commercial purposes, such as furniture, lumbers, and civil engineering. Most of these available trees have high tortuosities and are overturned before developing high diameters. Therefore, sustainable management of non-timber forest products (NTFPs) emerges as an important complementary alternative for some communities, which depend on this biome to survive.

In the last decades, the concept of sustainability has gained prominence, especially after the Rio-92 Conference. This term can be briefly defined as the ability of humans to use available resources and meet their current needs, preserving the ability of future generations in also meeting their needs. According to the Brazilian Institute of Geography and Statistics – IBGE [1], NTFPs have been progressively exploited in Brazil and reached their highest production value in 2018. For instance, açai berry (R\$ 592.0 million), yerba mate (R\$ 468.4 million), Brazil nuts (R\$ 130.9 million), carnauba wax (R\$ 189.2 million), and babassu almonds (R\$ 92.1 million) also reached outstanding production values in their respective markets.

Vegetable tannins (natural polyphenolic compounds) also stand out for their high market values (0.7-1.5

59 € kg⁻¹) [2]. Among these resources, condensed tannins are responsible for the largest global production (about
60 90%) with about 200 thousand tons per year [3-4]. For this reason, numerous forest species have been used as
61 matrices for extracting tannins, including *Acacia mearnsii*, *Acer* sp., *Quercus* sp., *Pinus* spp., and *Schinopsis* spp.
62 In this context, MT (commonly called as jurema-preta) is a native plant from the *caatinga* vegetation and its bark
63 has a high amount of tannins [5-6].

64 In general, vegetable tannins have high reactivity [7-8], ability to unite proteins [9] and metals [10], as
65 well as antifungal [11] and antioxidant [12] activities. These substances have a highly branched chemical structure
66 [2, 13], which hinders their isolation, purification, and even their characterization. For this reason, they are usually
67 sold as liquid or powdered extracts. Missio et al. [12] studied a powdered industrial tannin (IT) and reported that
68 it is rich in condensed tannins (70-80%) and also have a minor portion of non-tannic materials, such as hydrocolloid
69 gums, sugars, and small molecules (20-30%). Chemical modifications can be performed in condensed tannins
70 through the incorporation of reactants, like hydrolysis and sulfonation reactions [2]. These reactions may increase
71 the extraction yield and the overall quality of the tannin extract (including its solubility) in order to adjust these
72 features of tannin-based extracts to each required use, like adhesives for wood products.

73 Currently, several bio-based adhesives have been developed in order to gradually replace synthetic ones
74 designed for the timber industry. The use of renewable raw materials from plant sources for developing new
75 materials or improving existing ones has increased significantly in recent years. These ecological solutions intend
76 to replace conventional materials related to the oil production chain and other toxic materials with technical,
77 environmental and economic advantages. Although suitable bonding qualities were already attributed to these
78 green compounds for glueing wood surfaces, there are numerous physical and chemical influential factors related
79 to pressing conditions, adhesive, wood, and their interfaces [14-16].

80 The use of condensable tannins (extracted from the MT bark) as raw materials to obtain novel products
81 and/or by-products may follow some concepts, like bioeconomy and integrated biorefinery systems. This means
82 that the valuable chemical compounds belonging to biomasses must be extracted based on profitable pathways and
83 the involved environmental issues must be taken into account from an economic perspective. In this paper, tannin-
84 based extracts were obtained from MT bark using different solvents and their potential to produce wood adhesives
85 were ascertained. Additionally, an industrial tannin was used as a positive control.

86

87 **Material and methods**

88 **Raw materials**

89

90 The vegetal sample came from *Fazenda Experimental do Núcleo de Pesquisas do Semiárido*
91 (NUPEÁRIDO), which is belonging to Universidade Federal de Campina Grande (UFCG) and has the geographic
92 coordinates of 07°05'10" S and 37°15'43" W. A random sampling of ten (shrubs and trees) MT were performed
93 during the summer and these individuals were overturned. Subsequently, their barks were removed, mixed, and
94 air-dried in the shade. Then, they were fragmented in a hammer mill (forage) coupled to a 150 mesh screen
95 (opening of 1.00 mm). The homogenized material was then hermetically sealed using nylon bags.

96 The IT (Phenotan M[®]) consisted of a powder extraIT from *Acacia mearnsii* De Wild. Bark and was
97 donated by TANAC S/A, which is located at Montenegro/Brazil. Detailed information on the constitution and the
98 performed industrial extraction was reported in previous manuscripts [2, 12, 17]. Pine wood samples were obtained
99 from a local market located at Patos/Brazil. Sodium hydroxide (NaOH) and sodium bisulfite (NaHSO₃), both of
100 them in micro-pearls and analytical grade, as well as formaldehyde (CH₂O, 37 wt.% aqueous solution) and
101 hydrochloric acid (HCl, 10 M aqueous solution) were purchased from Neon Comercial (Suzano/Brazil). All these
102 chemicals were used as supplied.

103

104 **Production of the tannin-based extracts**

105

106 For obtaining both tannin-based liquid (LE) and powdered (PTE) extracts, the moisture content (MC) of
107 the bark (sample with 10 g) was determined according to the ASTM D6403 [18]. Three solvents were used, namely
108 distilled water (5000 mL), 0.5% NaOH aqueous solution, and 0.5% NaHSO₃ aqueous solution. The aqueous
109 solvent and the MT bark (air-dried) sample were mixed at a 10:1 weight ratio in all extractions. The extraction
110 process was carried out into a vertical autoclave (a stainless steel cylindrical reservoir) at a constant temperature
111 of 120 °C and a pressure of 0.9 MPa for 150 min. Four replicates (extractions) were performed for each solvent.

112 After the extractions, the LE was purified by sieving on a quantitative filter paper (grade 40: 8 µm)
113 coupled to a Millipore system and a vacuum pump. The pH and temperature of the solutions and purified LE in
114 were determined (triplicates) along with both extraction and purification steps. Soon after the filtration step, the
115 purified LE (each repetition) was immediately reduced to a dry powder in an oven with forced air circulation
116 adjusted at 70 ± 2 °C for 48 h. The mass was registered for determining the crude PTE content (Equation 1).

$$EY_c = \left(\frac{M_c}{M_p} \right) \times 100 \quad (1)$$

117 Where: EY_c is the crude powder extract content (in %), M_c is the dry mass of crude powder extract (in g), and M_p
118 is the dry mass of raw material (in g).

119

120 The crude PTE was processed in a Wiley mill coupled to a 60 mesh screen (< 0.25 mm). The refined
121 extracts were packed in opaque plastic bottles and hermetically sealed. The refined PTE content was calculated
122 according to Equation 2.

$$EY_r = \left(\frac{M_r}{M_p} \right) \times 100 \quad (2)$$

123 Where: EY_r is the refined powder extract content (in %), M_r is the dry mass of refined powder extract (in g), and
124 M_p is the dry mass of raw material (in g).

125

126 **Characterization of the powdered tannin-based extracts (PTE) and CT**

127 Attenuated total reflectance with Fourier transform infrared spectroscopy (ATR/FTIR) was used to
128 analyse the refined PTE and CT. For that, a Nexus 470 (Thermo Nicolet brand) infrared spectrophotometer was
129 adjusted to total absorbance. For each sample (1.0 mg), 32 scans were performed at the 4000 to 600 cm^{-1} spectral
130 range with an applied resolution of 4.0 cm^{-1} . The obtained spectra were normalized and their baseline were
131 corrected.

132 For preparing analytical solutions, 6.0 g of each tannin sample was gradually mixed with 1000 mL of
133 distilled water at a constant temperature of 95 °C. Then, the material was naturally cooled and stored according to
134 the D3790, D4904, and D4905 standards [19-21]. Total solids content (TS) was determined according to D4902
135 and D4903 [22-23]. The MC was calculated by the difference between the total mass (100%) and the TS. The MC
136 influences the other parameters (soluble and insoluble solids, condensed tannins, and non-tannins) and, because
137 of that, these chemical parameters of the PTE were adjusted to its MC using Equation 3.

$$M_{0\%} = M_s \times \left[1 - \left(\frac{MC}{100} \right) \right] \quad (3)$$

138 Where: $M_{0\%}$ is the dry mass (in g), M_s is the air-dry mass (in g), and MC is the moisture content (in %).

139

140 A 100 mL aliquot of the analytical solution was filtered in a quantitative filter paper (grade 40: 8 µm with
141 medium speed and retention). Thus, the soluble solids content (SS) was determined by the gravimetric method,
142 based on the dry mass (filtrate liquid) measured after a drying process performed in an oven with forced air
143 circulation (100 ± 4 °C). The insoluble solids content (IS) was the difference between the TS and the SS according
144 to the NBR 14362 and D6402 [24-25]. Condensed tannins content (CT) was measured based on the reactivity of
145 the extract (aliquot with 0.4% pure condensed tannins) with formaldehyde using Equation 4, as reported in the
146 literature [26-28]. For this, an amount of 100 mL of filtered aliquot was subjected to a Stiasny reaction by adding
147 2 mL of HCl and 8 mL of CH₂O. This mixture was kept under reflux for 30 min and stirring every 5 min. The
148 generated precipitate was filtered on quantitative filter paper (40: 8 µm grade with medium speed and retention)
149 and was then washed using distilled water and dried at 100 ± 4 °C in an oven with forced air circulation adjusted.
150 The non-tannins content (NT) was obtained by the difference between the SS and the IT (Equation 5). The
151 condensed tannins content in the aliquot of the analytical solution was determined using Equation 6. When any
152 repetition did not meet a concentration of 0.4 ± 0.025% (using 6.0 g), an adequate amount of air-dried powder
153 extract was used as a replacement (Equation 7).

$$CT = \left(\frac{M_{pt}}{M_{0\%}} \right) \times 100 \quad (4)$$

$$NT = SS - CT \quad (5)$$

$$Ct = \left[\frac{(Ma_t \times 100)}{A} \right] \quad (6)$$

$$Me_c = \left[\frac{(Ma \times 0.4)}{Ct} \right] \quad (7)$$

154 Where: CT is the condensed tannins content (in %), Mpt is the dry mass of the precipitate extrapolated to the total
155 volume of the beneficiary's extract solution (in g), M_{0%} is the dry mass of the powder extract used in the preparation
156 of the analytical solution (in g), NT is the non-tannins content (in %), SS is the soluble solids content (in %), Ct is
157 the concentration of pure condensed tannins in the aliquot (in %), Ma_t is the dry mass of the precipitated condensed
158 tannins (in g), A is the aliquot volume (in mL), Me_c is the appropriate mass of the powder extract to a concentration
159 (in g), and Ma is the air-dried mass of the powder extract used in the preparation of the analytical solution (in g).

160

161 **Characterization of the tannin-based adhesives**

162

163 Tannin-based adhesives were prepared using both the refined PTE and IT according to the formulations
164 displayed in Table 1. The experiment was carried out in triplicates for each adhesive formulation. All the adhesives
165 were prepared as indicated by each supplier, except the MT + NaOH (refined PTE) since its concentration hinders
166 adequate hydration of the extract. The powdered extract was mixed with distilled water at a constant 60 °C and
167 was then kept at room temperature for 24 h until reaching its equilibrium moisture content. Wheat flour and CH₂O
168 were applied as a glue extender and a catalyst, respectively. The flour had a particle size smaller than 0.25 mm (60
169 mesh).

170

171 **Table 1 Formulation of the different tannin-based adhesives**

| Sample code | Base agent ^a (g) | Solvent (g) | Glue extender ^b (w/w) | Catalyst ^c (%) |
|-------------------------|--------------------------------|----------------|-------------------------------------|------------------------------|
| MT | 50 | 50 | 10 | 20 |
| MT + NaOH | 30 | 70 | 10 | 20 |
| MT + NaHSO ₃ | 50 | 50 | 10 | 20 |
| CT | 50 | 50 | 10 | 20 |

172 Where: ^aMoisture content ~ 6-8%; ^bBased on the mass of the resin mix; ^cPercentage in relation to the mass of base
173 agent (powdered tannin).

174

175 The solid content (SC) of the tannin-based adhesives was determined by evaporation of the moisture in a
176 1.0 g sample oven-dried with forced air circulation adjusted at 100 ± 4 °C. The SC was calculated according to
177 Equation 8. Viscosity was determined using an EEQ-9031 analog rotational viscometer (Brookfield-LV type) [29],
178 with a rotor spindle no. 3 and a rotation speed suitable to each adhesive. The pH at room temperature (25 ± 2 °C)
179 was determined before and after the addition of CH₂O using a LUCA-210 digital benchtop pH meter [30]. The gel
180 time was measured based on that method reported by [31-32] after the incorporation of the CH₂O (in a proportion
181 of 20% of the mass of SC).

$$SC = \left(\frac{M_d}{M_a} \right) \times 100 \quad (8)$$

182 Where: SC is the solid content of adhesive (in %), Md is the dry mass of adhesive sample (in g), and Ma is the
183 mass of adhesive sample (in g).

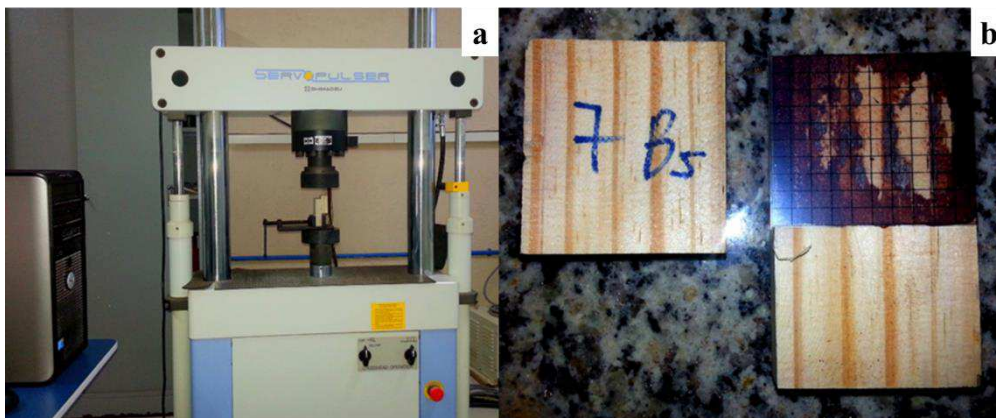
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185 **Bonding of the wood joints and their evaluation**

186 The used pine wood boards presented basic density, apparent density, and moisture content of 0.37 g cm^{-3}
187 3 , 0.43 g cm^{-3} , and 9.53%, respectively. This raw material was transformed into prismatic specimens with the
188 dimensions of $1.90 \times 5.10 \times 5.40 \text{ cm}^3$ (tangential \times radial \times longitudinal). To prepare the wood surfaces for
189 bonding, they were sanded in the longitudinal direction using an electric circular sander (sandpaper no. 120). A
190 weight of $300 \text{ g}\cdot\text{m}^{-2}$ of each tannin-based adhesive was applied on tangential plane and spread with a glass trowel.
191 Then, a manual assembly was carried out for 15 min, which was followed by cold pre-pressing for 30 min, and
192 then a hot pressing at $120 \text{ }^\circ\text{C}$ and 0.5 MPa for 60 min. Finally, the samples were post-cured at room temperature
193 for 15 days.

194 The glue line of the pine wood joints was analysed by scanning electron microscopy (SEM) using a
195 Tescan (VEGA-3) equipment. The radial plane was coated with gold particles to ensure a suitable electrical
196 conductivity and the SEM images were acquired under an accelerating voltage of 10 kV. The glue line was
197 evaluated for shear strength following the ASTM D905 [33]. For that, a compressive load was applied on the
198 transverse plane (i.e. perpendicularly to the glued plane, which became a shear plane). All mechanical tests were
199 carried out using a Shimadzu universal testing machine with 100 kN capacity (Figure 1a). Afterwards, the glued
200 plane was visually evaluated and the percentage of failed wood was determined according to ASTM D5266 [34].
201 Based on the total shear plane, this indicates the percentage of cohesive failure in the wood. For that, it was used
202 a transparent checkered grid with one hundred equal parts and the dimensions of $4.40 \times 5.10 \text{ cm}^2$ (length \times width)
203 (Figure 1b). Also, the MC of the samples was determined by gravimetric analyses soon after the mechanical tests.

204



205 **Fig. 1** Photographs of the shear test setup (a) and the glued plane of a failed pine wood sample (b).

206

207 **Statistical analyses**

208 Data normality and homoscedasticity of variances were verified using Shapiro-Wilk tests and Bartlett
209 tests, respectively. A completely randomized design was applied. The data were subjected to analysis of variance
210 (ANOVA) followed by Fisher-Snedecor tests. When the null hypothesis was rejected, a comparison of means was
211 performed using Fisher tests. A significance level of 5% was adjusted for all tests.

212

213 **Results and discussion**

214 **Evaluation of the extraction process**

215

216 The water (used in all treatments) presented a slightly acidic pH, as shown in Table 2. The addition of the
217 MT bark induced strong acidification in the aqueous solution due to the leaching of some organic extractives, like
218 condensed tannins (for instance, proanthocyanidins and saponins), sugars (c.a. glucose, starch, terpenoids, and
219 alkaloids) [35-38], and minerals (a minor amount). This occurs due to the high polarity of the water (used as a
220 solvent), which causes progressive solubilisation of some hydrophilic compounds from the MT bark. Among the
221 extractives belonging to tree barks, some water-soluble tannins (polyphenolic molecules) and weak acids release
222 H⁺ ions in the aqueous solution, leading to the deprotonation of hydroxyls. Similar results were obtained with the
223 addition of bark and NaHSO₃, which induced a slight increase in the pH of the purified LE. The low pH of the
224 sulphated LE can be attributed to the acid character of NaHSO₃, which generally ranges from 4.0 to 4.7.

225

226 **Table 2 Temperature and pH of the different solutions of *M. tenuiflora* bark**

| Extraction Condition | Solution Sample | pH | Temperature (°C) |
|----------------------|--------------------------------|--------------|------------------|
| MT | Water | 6.30 ± 0.37 | 26.4 ± 0.3 |
| | Bark Mix ^a | 4.76 ± 0.19 | 26.6 ± 0.3 |
| | Purified LE | 4.21 ± 0.16 | 26.4 ± 0.1 |
| MT + NaOH | Water | 6.30 ± 0.33 | 26.9 ± 0.1 |
| | Bark and NaOH Mix ^a | 12.48 ± 0.17 | 26.6 ± 0.4 |
| | Purified LE | 8.55 ± 0.14 | 26.0 ± 0.6 |

| | | | |
|-------------------------|--|-------------|------------|
| | Water | 6.55 ± 0.43 | 27.0 ± 0.6 |
| MT + NaHSO ₃ | Bark and NaHSO ₃ Mix ^a | 4.66 ± 0.15 | 27.0 ± 0.7 |
| | Purified LE | 4.87 ± 0.06 | 27.7 ± 2.5 |

227 ^aAfter 15 min. Where: numerals refer to averages ± standard deviations.

228

229 The incorporation of bark and NaOH led to increases in the pH of the solution (normally strongly alkaline)
 230 due to the dissociation of hydroxyl ions (OH⁻). The solution generated strong odour gases when heated and the
 231 purified LE demonstrated a decrease in pH after the extraction process. The NaOH is a strong base prone to react
 232 in an aqueous medium through an exothermic chemical process [39], which promotes the attack of free radicals
 233 present in the vegetable raw material (c.a. bark) and leads to the removal and conversion of soluble substances
 234 belonging to the extract.

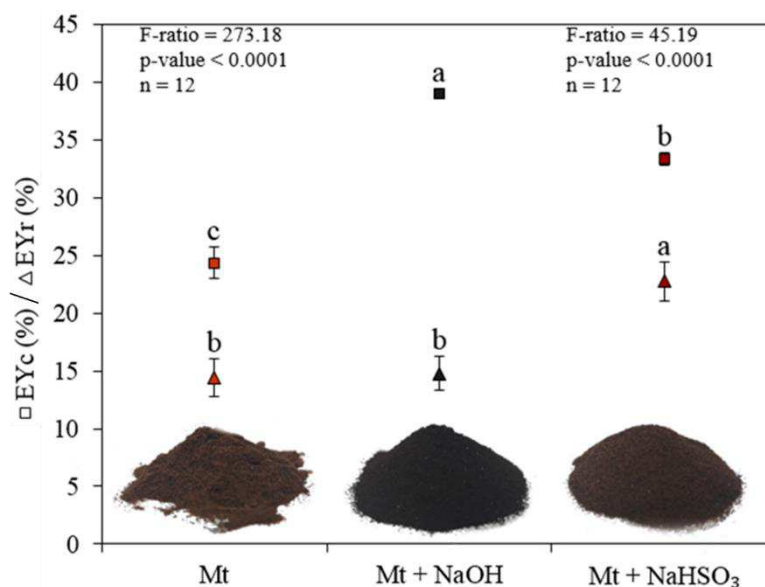
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236 Powder extract yields

237

238 The air-dried particles of the MT bark showed MC and dry mass of 11.25 ± 0.44% and 443.79 g,
 239 respectively. Besides, there were no significant differences between groups. The yields of PTE was the first
 240 parameter to determine the extractive content belonging to the bark, which indicates the efficiency and productivity
 241 of the extractions. Regarding the crude powder extract content (EY_c), the addition of MT + NaOH or MT +
 242 NaHSO₃ favoured a significant increase in the removal of extractives from the raw material. However, the former
 243 extracting agent promoted a higher percentage of extracted solids. Regarding the refined powder extract content
 244 (EY_r), only the treatment with the addition of the latter extracting agent, significantly favoured the conversion into
 245 powder during beneficiation, as shown in Figure 2.

246



247
 248 **Fig. 2** Crude (EYc) and refined (EYr) PTE contents and side view of them. Averages (\pm standard deviations)
 249 followed by a same letter, in a same column, do not differ statistically from each other according to the Fisher test.
 250

251 Based on these yields, the additions of the extracting agents (through hydrolysis and sulfitation) promoted
 252 significant increases in the extraction of solid tannin fractions from the MT bark. Some previous studies also found
 253 similar results using other extracting agents [2, 13, 40-42]. In the present study, the EYc values can be mainly
 254 attributed to the use of basic hydrolysis (NaOH) and sulfitation (NaHSO₃).

255 Regarding the composition of the extracts from the MT bark, several phenolic (like tannins) and non-
 256 phenolic substances (such as carbohydrates, esters, and insoluble inorganic compounds) were detected [6].
 257 According to the literature [13], high purity tannins can be extracted using high levels of pressure and temperature,
 258 which leads to greater solubility of compounds, as well as decreased viscosity and improved movement of the
 259 solvents. However, an over-oxidation of the phenolic compounds may occur if the extraction takes a long time,
 260 which negatively affects the final extraction yield and its quality.

261 Regarding the MT + NaOH, there was a little transformation of the crude extract and, thus, significant
 262 loss (about 24%) during its processing, which was mainly due to the extraction and formation of more rigid
 263 components. This indicates that this solid phase is not easily fragmented into particle sizes less than 60 mesh (0.250
 264 mm). In addition, the grinding of the crude powder extract probably affected the EYr due to a large amount of
 265 solid material being superimposed on the internal sieve of the rotation drum, consequently impairing the passage
 266 to the collection reservoir. This probably hindered the contact between the rotating rods from the mill and the
 267 particles from the powder.

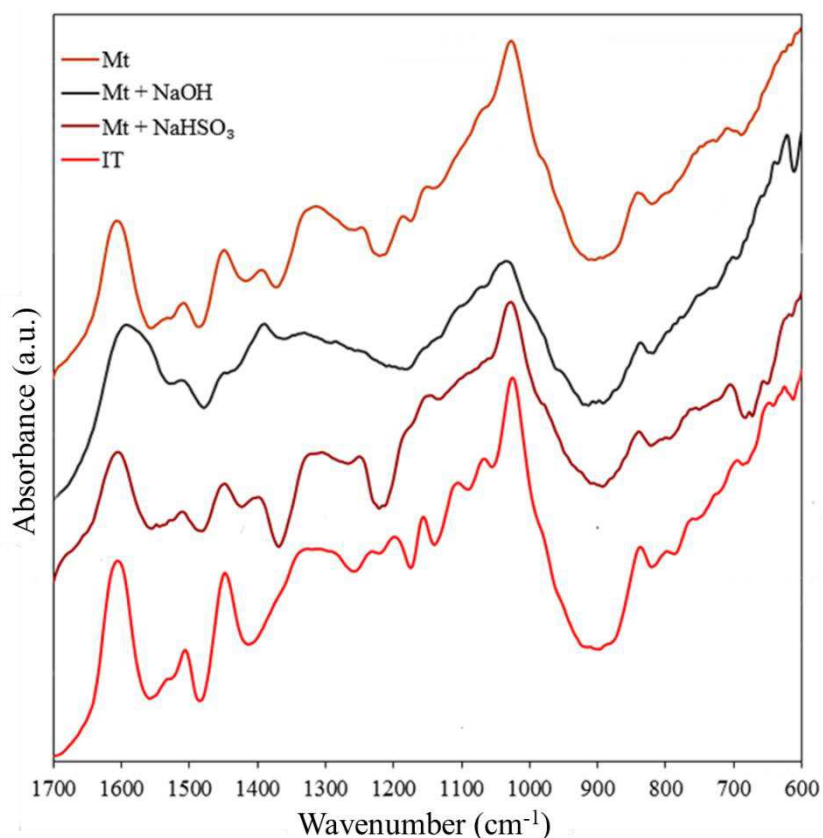
268 This wood is known due to its high calorific value. In this region, there is a density of 65 trees per ha [43]
269 and these thousands of cubic meters of logs are often burned for energy generation, which encompasses both
270 firewood and charcoal consumed every year by residences, commercial establishments, and industrial plants.
271 Lopes et al. [5] studied MT trees from Northeastern Brazil with a mean basal diameter of 11.4 cm and reported a
272 mean dried mass of bark of 3.7-4.0 kg per tree. Therefore, the bark has a non-significant significant influence in
273 the calorific power of the overall tree, even due to its known high content of impurities. In this context, it seems
274 clear that the MT bark should be considered for noble applications, which may be achieved following biorefinery
275 concepts.

276

277 **Analysis of powdered tannin-based extracts (PTE)**

278

279 FTIR spectra indicate the chemical structure of the different PTE and IT (Figure 3). According to Tondi
280 [44], the 1800-600 cm^{-1} range is the most important region for tannins. In general, if compared to each other, the
281 powder extracts have a similar molecular chemical structure since these differences are only related to the
282 intensities of the existing bands, as well as minor displacements in some bands located at the 1630-650 cm^{-1} range.
283 Briefly, the bands at 1800-1650 cm^{-1} , 1650-1400 cm^{-1} , 1400-1100 cm^{-1} , and 1100-600 cm^{-1} refers to C=O
284 stretching, C=C stretching in aromatic rings from phenolic compounds, C-O elongation in aromatic phenolics,
285 and C-H elongation in aromatic compounds [44].



286
 287 **Fig. 3** FTIR spectra of the powdered tannin-based extracts.

288
 289 In comparisons between the tannin-based extracts, the MT + NaOH showed the highest TS and the lowest
 290 MC (Table 3). The higher MC presented by the IT may be due to its higher hygroscopicity ascribed to its smaller
 291 particle size (< 100 mesh, 0.15 mm) when compared to the other tannins (< 60 mesh). This smaller particle
 292 diameter may lead to a greater surface area and, consequently, this tannin probably became prone to form chemical
 293 bonds with the humidity from the environment. These features also lead to a lower amount of solid powder extract.
 294 The inclusion of extracting agents in the extraction process provided significant increases in the overall solubility
 295 of the PTE extracted from the MT bark. These results are comparable to those obtained for the CT. The use of the
 296 extracting agent (NaOH and NaHSO₃) also yielded an increase in SS from PTE and a decrease in the IS extracted
 297 from the MT bark.

298 In this study, only the properties obtained for the MT + NaOH PTE meet those parameters proposed by
 299 the IS6199-1971 [45]. This standard procedure regards a powdered black wattle extract and indicates that MC, IS,
 300 and tannic SS shall be of at least of 6.77%, 1.22%, and 67.29%, respectively. Compared to the unmodified MT,
 301 the MT + NaOH showed a significant increase in CT and a decrease in NT, which yielded improvements in the
 302 PTE extracted under neat water. The use of MT + NaHSO₃ also resulted in greater CT if compared to the

303 unmodified MT, although the NT remained similar in this case. These results are different from those reported in
 304 a previous study on MT bark and some extractions done under hot water [46]. These divergences can be explained
 305 by some differences in extracting procedures, as well as differences in the used raw materials (like particle size of
 306 and extraction mechanism).

307

308 **Table 3 Characterization of the powdered tannin-based extracts (PTE) and Phenotan M® (IT)**

| Powder (Sample) | TS | MC | SS | IS | CT | NT |
|----------------------------------|----------|----------|--------|--------|----------|--------|
| | (%) | | | | | |
| MT (8.30 g) | 91.62 b | 8.38 b | 87.27 | 12.78 | 53.88 d | 33.38 |
| | ± 1.24 | ± 1.24 | ± 2.28 | ± 2.18 | ± 1.39 | ± 1.98 |
| MT + NaOH (6.50 g) | 93.19 a | 6.81 c | 98.78 | 1.40 | 68.31 b | 31.29 |
| | ± 0.78 | ± 0.78 | ± 0.58 | ± 0.48 | ± 1.68 | ± 0.99 |
| MT + NaHSO ₃ (7.10 g) | 92.01 ab | 7.98 bc | 97.16 | 2.84 | 63.49 c | 34.79 |
| | ± 1.51 | ± 1.51 | ± 1.36 | ± 1.36 | ± 1.84 | ± 1.88 |
| IT (6.00 g) | 88.45 c | 11.55 a | 96.55 | 3.63 | 77.73 a | 20.05 |
| | ± 1.47 | ± 1.47 | ± 1.58 | ± 1.07 | ± 0.65 | ± 0.73 |
| F-ratio | 17.21 | 17.21 | - | - | 282.28 | - |
| p-value | < 0.0001 | < 0.0001 | - | - | < 0.0001 | - |
| n | 34 | 34 | 30 | 31 | 29 | 32 |

309 Where: Averages (± standard deviations) followed by a same letter, in a same column, do not differ statistically
 310 from each other according to the Fisher test. TS = Total solids; MC = Moisture Content; SS = soluble solids; IS =
 311 insoluble solids; CT = Condensed tannin; NT = non-tannins.

312

313 For the PTE, high values of both SS and CT are desired. In this sense, the use of MT + NaOH and MT +
 314 NaHSO₃ yielded higher values of these parameters. Low values of IS and NT are surely desired. However, with
 315 sulfitation, the same behaviour occurred for the CT and without the influence reducing of NT compounds. It is
 316 well known that sulfitation promotes the opening of heterocyclic rings of tannins and introduce sulfonic groups,
 317 which also lead to a reduction in the viscosity and increases in both extraction yield and solubilisation of tannins
 318 [2, 26]. The alkaline hydrolysis process promotes rearrangement of the tannins through the rupture of the

319 interflavonoid bond (C4-C8) and the opening of the heterocyclic ring connected to the ring of the flavonoid unit,
320 which induce an increase in the solubility of the tannins, especially CT [2].

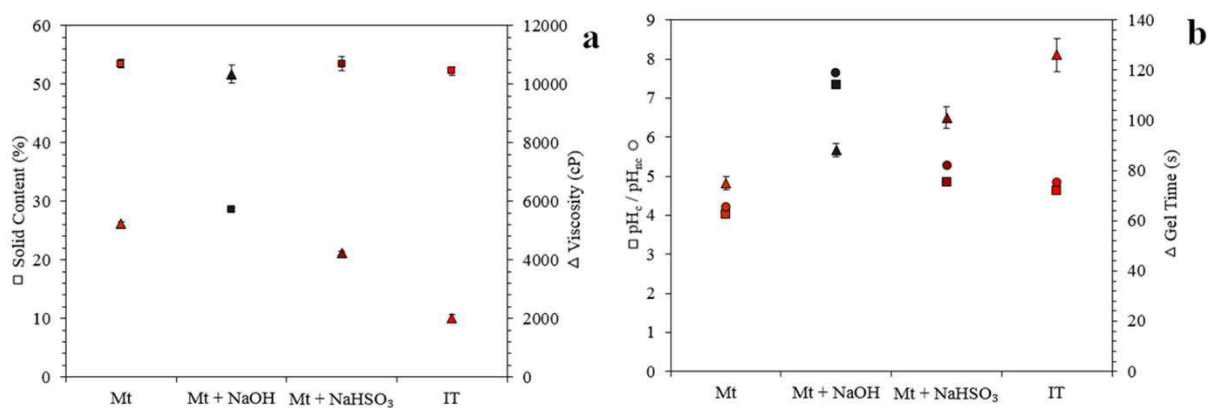
321

322 **Properties of the tannin-based adhesives**

323

324 The SC of the MT + NaOH-based adhesive was lower than the others (Figure 4a). This adhesive was
325 formulated with a lower concentration of the base agent in the PTE and a greater amount of solvent (distilled
326 water), which explains the lowest SC and the highest viscosity attributed to this adhesive. In this study, the
327 modification at the time of hydration and solubilization of the PTE was performed to avoid reactivations of the
328 solid compounds. When carried out in very alkaline conditions (like in the present study), the extraction of tannins
329 produces high viscosity adhesives, which can hinder their subsequent use [47]. The reduction in viscosity is a
330 common characteristic of the sulfitation of the LE and PTE, as it favours the breakdown of chemical bonds from
331 the extract and, thus, leads to an increase in the viscosity of the adhesives. In addition, this may promote increases
332 in both pH and gel time, which explains these results displayed in Figure 4b.

333



334 **Fig. 4** Averages and standard deviations for the solid contents and viscosity (a) and potential hydrogens with (pH_c)
335 and without (pH_{nc}) catalyst and gel time (b) for the tannin-based adhesives.

336

337 Adhesives with viscosity means greater than 1500 cP are not able to be applied via microparticles [48],
338 and those above 6000 cP are not suitable to be applied on solid woods [49]. However, they can be applied directly
339 and manually on the wood using spatulas or rollers. The higher viscosity of adhesives can yield the formation of
340 a glue line thicker than that ideal due to an uneven distribution on the wood surface and inadequate wetting of the
341 adhesive [16].

342

343 The MT + NaOH adhesive presented a basic pH, whereas the others have acidic levels. The pH of the
344 adhesives must not exceed the 2.5-11.0 range [50], although the pH of the wood is also important since affects that
345 time required for total curing and hardening of the glue. There were decreases in the pH of all the adhesives due
346 to the addition of the catalyst (CH₂O). After mixing, chemical reactions started in the adhesive solution (with the
347 release of H⁺ ions), which ended above 90 °C and caused a phase change to form the adhesive gel (by a complete
348 cure of the resin). The extracting agents significantly increased the gel time (phase change), which is desired since
349 adhesives with a very short gel time hinder an adequate spreading over the wood surface since a rapid
350 polymerization may occur leading to decreases in the glue line strength.

351 The condensation of tannins due to the presence of formaldehyde under heating (95-100 °C) is strongly
352 influenced by the pH of the medium, as there is an increase in the reaction speed if there is a decrease in the pH
353 [51]. According to Tondi [44], an amount of about 4% of this catalyst is enough to promote the polymerization of
354 tannin-based resins.

355 Azevêdo et al. [49] produced sulfited (Na₂SO₃) tannin adhesives extracted from the MT bark cut in the
356 dry period (August-December) and reported the following parameters: pH values from 5.69 to 5.76), SC from 46.8
357 to 46.9% and viscosity from 2067 to 10000 cP, which are similar to the present study. Some minor differences in
358 a comparison with the present study are probably associated with the acid sulfitation process after extracting the
359 bark, which was not done in the present study. The IT resin (Phenotan M[®]) extracted from the black wattle bark is
360 commercialized with an MC of 5-8%, a viscosity (at 25 °C) from 1000 to 1600 Cp, and a pH ranging from 4.0 to
361 5.0 [17]. The IT adhesive showed the lowest viscosity among those analysed here, as well as the greatest fluidity
362 and, consequently, this adhesive was easily spread on the wood surface.

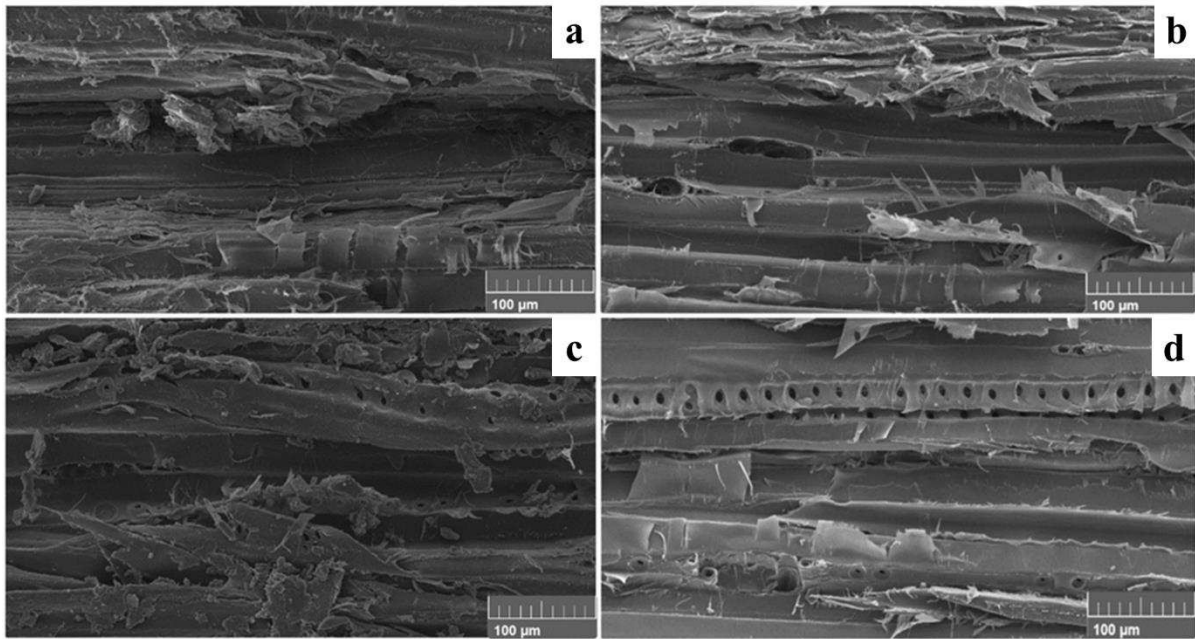
363

364 **Bonding performance of the tannin-based adhesives**

365

366 The different tannin-based adhesives used for bonding the wood joints promoted uniform glue lines as
367 shown in Figure 5. All the MT adhesives promoted thick glue lines in the union between the wood surfaces, which
368 is mainly due to their high viscosity, as aforementioned. In general, this may impair the adhesive-wood connection
369 but may positively contribute to the union in the internal layers of the adhesive. This fact may limit its full capacity
370 of undergoing external loads on a large scale, such as shear forces.

371



372
 373 **Fig. 5** Side view of the glue lines promoted by the bonding with the tannin-based adhesives. Where **a**, **b**, **c**, and **d**
 374 refer to MT, MT + NaOH, MT + NaHSO₃, and Phenotan M[®] (IT), respectively.

375
 376 The shear strengths of the MT and MT + NaOH adhesives overcame that of the IT (Table 4). The same
 377 behaviour was demonstrated in the evaluation of the percentage of wood failure on the shear plane. According to
 378 a previous study [52], the bonding of wood joints with tannin-based adhesives often do not reach a minimum safety
 379 rate due to the low viscosity of the tannin resins and, consequently, high leakage lines are normally formed. These
 380 results are contrasting with those of the present study, which is probably due to the viscosity of the adhesives and
 381 their chemical composition.

382
 383 **Table 4 Physical-mechanical properties of the wood joints glued with different tannin-based adhesives**

| Adhesive Sample | MC (%) | Shear strength (MPa) | Failed wood (%) |
|-------------------------|-------------|-------------------------|-----------------|
| MT | 7.03 ± 0.29 | 1.41 b ± 0.54 | 41.09 a ± 7.26 |
| MT + NaOH | 7.26 ± 0.23 | 1.51 b ± 0.73 | 44.13 a ± 3.44 |
| MT + NaHSO ₃ | 7.43 ± 0.21 | 1.27 ab ± 0.53 | 40.70 a ± 4.55 |
| IT | 7.35 ± 0.23 | 0.76 a ± 0.37 | 35.11 b ± 6.03 |
| F-ratio | - | 3.36 | 5.36 |

| | | | |
|---------|----|--------|--------|
| p-value | - | 0.0269 | 0.0033 |
| n | 48 | 48 | 45 |

384 Where: MC is moisture content of the glued wood sample (dry basis). Averages (\pm standard deviations) followed
385 by the same letter, in a same column, do not differ statistically from each other according to the Fisher test.

386

387 According to the NBR 7190 [53], the shear strength parallel to the fibres in softwoods varies from 4 MPa
388 to 6 MPa for a 0.4-0.5 g cm⁻³ basic density range. On the other hand, according to the ASTM D5751 [54], the
389 shear strength mean for glued joints must reach at least 60% of shear strength belonging to the respective solid
390 wood. Based on these different standards, those shear strengths displayed in Table 4 should range between 2.4 and
391 3.6 MPa.

392 The ASTM D2559 [55] also indicates that, for a satisfactory bonding with structural adhesives (designed
393 for outdoor uses), a minimum average value of 75% of wood failure must be reached. For dried laminated joints
394 and other non-structural joints, the minimum percentage of failed wood is 60%, which is visualized on the shear
395 plane [54]. In this evaluation, the higher the percentage of wood failure, the higher the bonding quality, which is
396 mainly associated with characteristics of both adhesive-adhesive and adhesive-wood interfaces. None of the
397 adhesives reached these levels proposed in ASTM D2259. This may be related to the high viscosity of the different
398 adhesives from MT bark. This factor probably impaired the spreading and wetting of the glue on the wood surfaces,
399 thus, negatively affecting the cohesion between adhesive molecules and the overall adhesive-wood interface.
400 Moreover, although the IT resin is commonly used for the manufacture of plywood and other structural wood parts
401 [17], in this study, its use was adapted for lateral bonding of wood joints, which probably impaired its mechanical
402 performance.

403 The phenomenon of connections at the adhesive-wood and adhesive-adhesive interfaces depends on
404 several simultaneous factors, such as anatomy, physics and chemistry of the wood, physicochemical properties of
405 the adhesive, and the bonding process. In further studies, a decrease in the viscosity of the adhesive may favour
406 an improvement of the bonding quality. This would provide an improved penetration of the wood and thus increase
407 in both the shear strength of the glue line and the percentage of wood failure on the shear plane.

408

409 **Conclusions**

410

411 Some valuable compounds from the *M. tenuiflora* bark have been wasted since this vegetable is

412 traditionally underutilized. By using a biorefinery concept, integrated and sustainable processing applied to this
413 biomass may yield marketable chemicals, like a wood adhesive. In the present study, aqueous solutions containing
414 0.5% of NaOH and 0.5% of NaHSO₃ were successfully used as solvents to extract tannin-based compounds. Then,
415 tannin-based adhesives were produced and yielded glue wood joints with promise mechanical performances,
416 although the tested glue lines did not yield wood joints able to meet those parameters proposed in known standards.
417 Nevertheless, the novel tannin-based adhesives showed better performances if compared to a commercial structural
418 adhesive (IT). Further studies are strongly encouraged and may address different pressing conditions and the
419 influence of the adhesive viscosity.

420 **List of abbreviations**

421 MT: Mimosa tenuiflora; NTFPs: non-timber forest products; IT: industrial tannin; PTE: tannin-based powdered
422 extract; LE: tannin-based liquid extract; MC: moisture content; TS: Total solids content; SS: soluble solids content;
423 IS: insoluble solids content; CT: condensed tannins content; SC: solid content; EYc: crude powder extract content;
424 EYr: refined powder extract content.

425 **Availability of data and materials**

426 All relevant data is presented in the manuscript, and additional information can be made available on request if
427 necessary.

428 **Competing interests**

429 The authors declare that they have no known competing financial interests or personal relationships that could
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433 **Authors' contributions**

434 PJGL, WAMS, PNMN, and IAB developed the experiments and analysed the results. PJGL, RAD, and ALM
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Figures

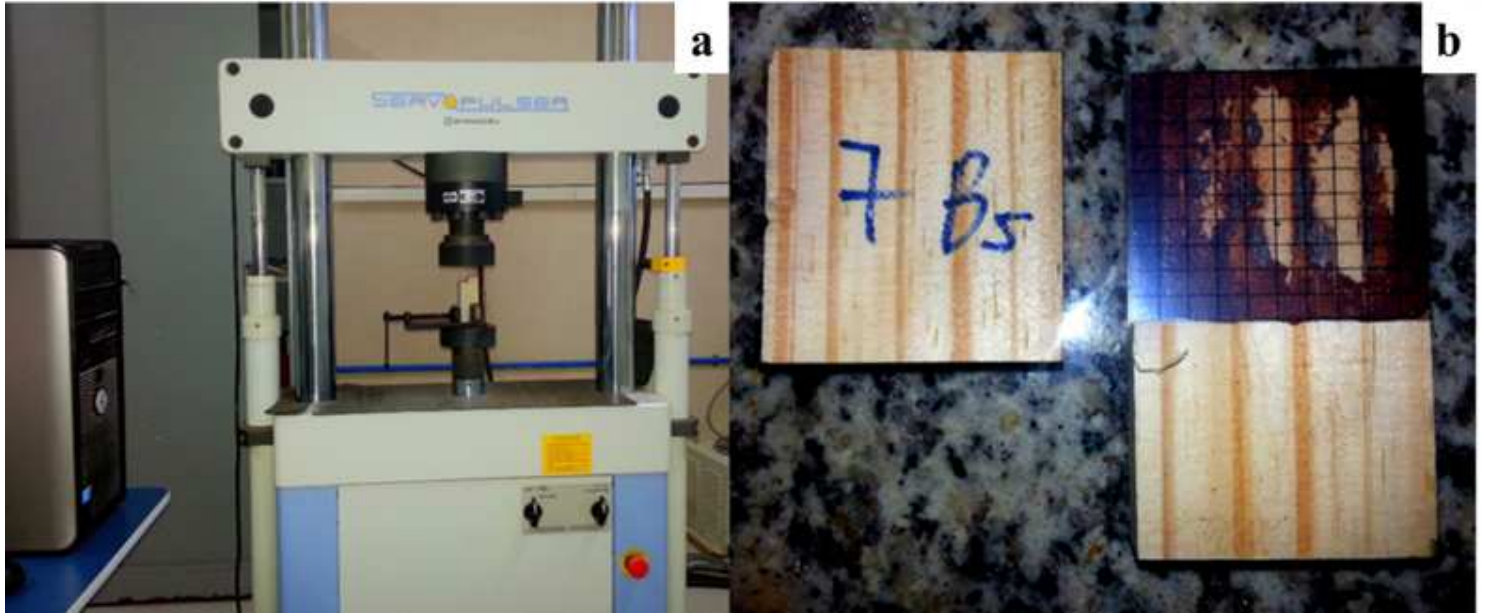


Figure 1

Photographs of the shear test setup (a) and the glued plane of a failed pine wood sample (b).

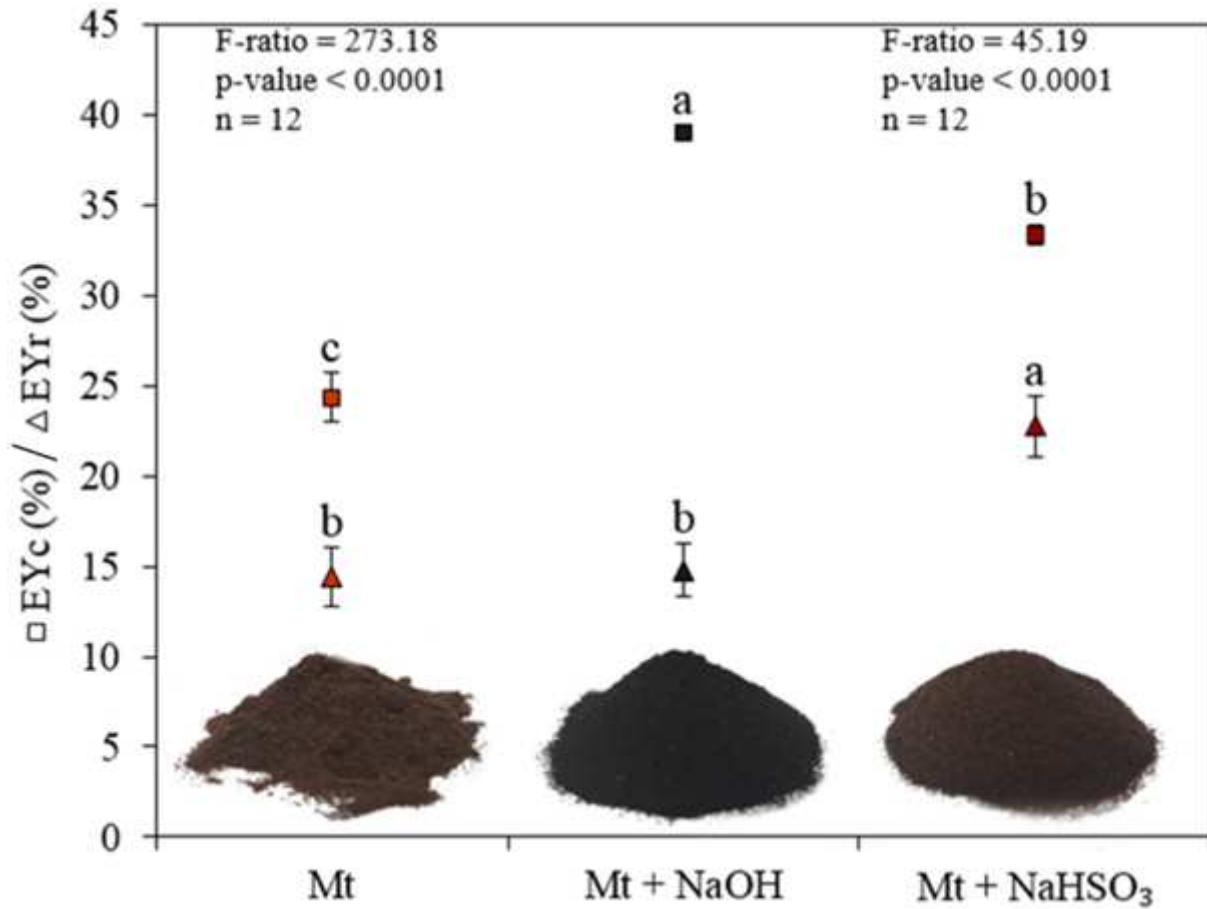


Figure 2

Crude (EYc) and refined (EYr) PTE contents and side view of them. Averages (\pm standard deviations) followed by a same letter, in a same column, do not differ statistically from each other according to the Fisher test.

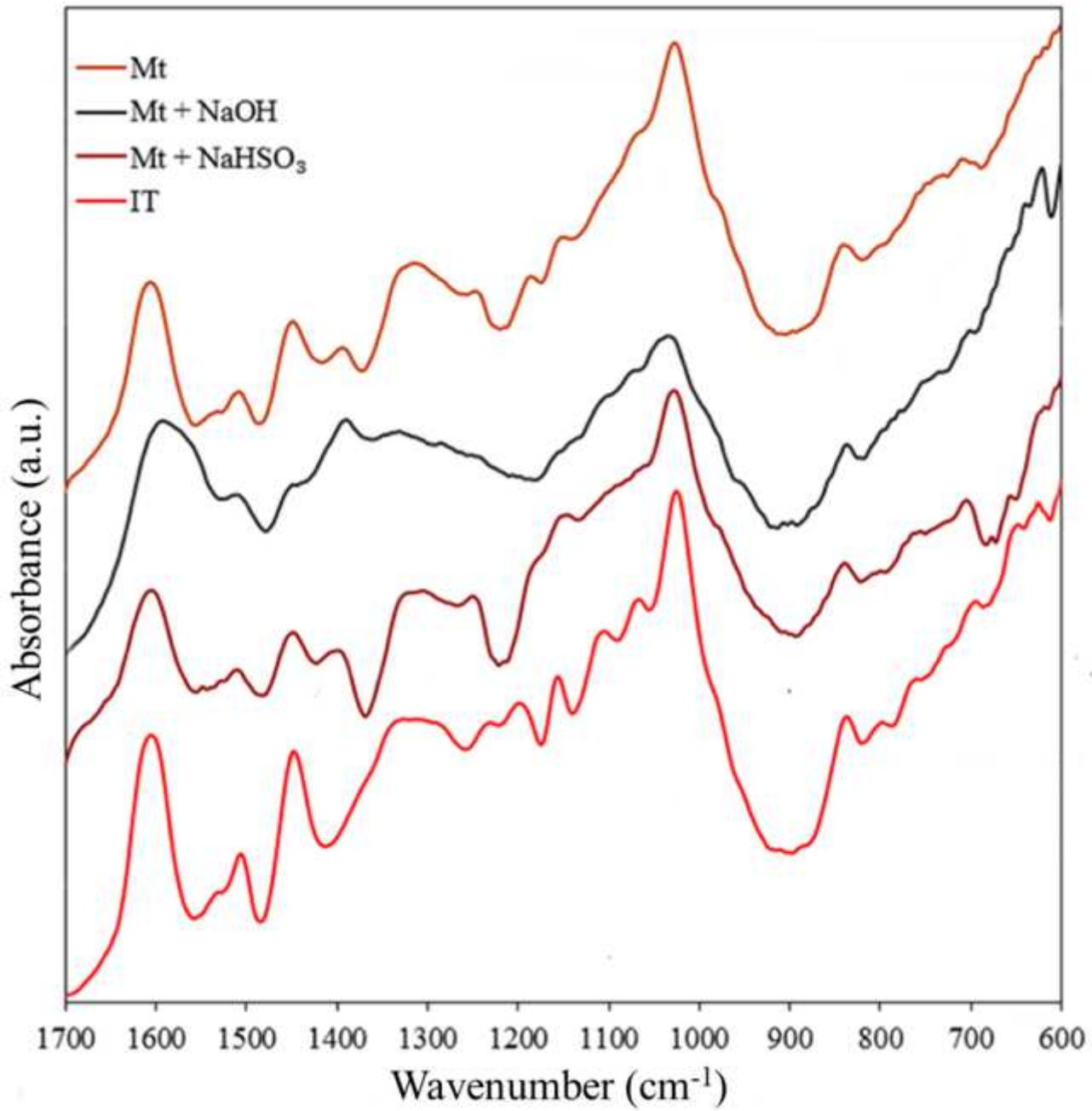


Figure 3

FTIR spectra of the powdered tannin-based extracts.

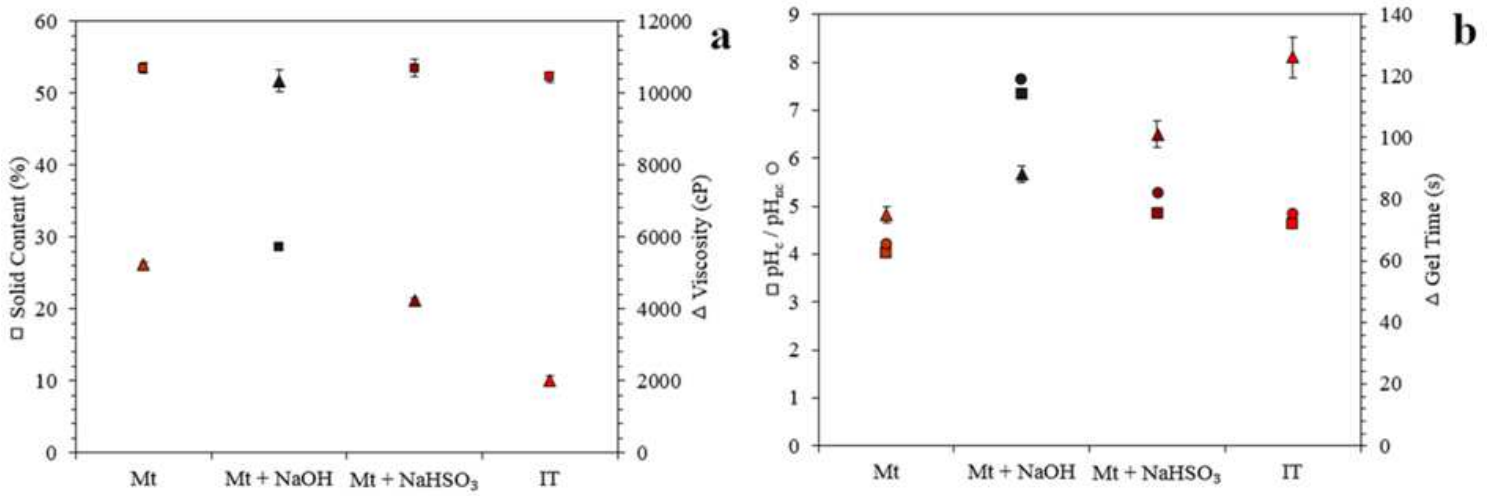


Figure 4

Averages and standard deviations for the solid contents and viscosity (a) and potential hydrogens with (pH_c) and without (pH_{nc}) catalyst and gel time (b) for the tannin-based adhesives.

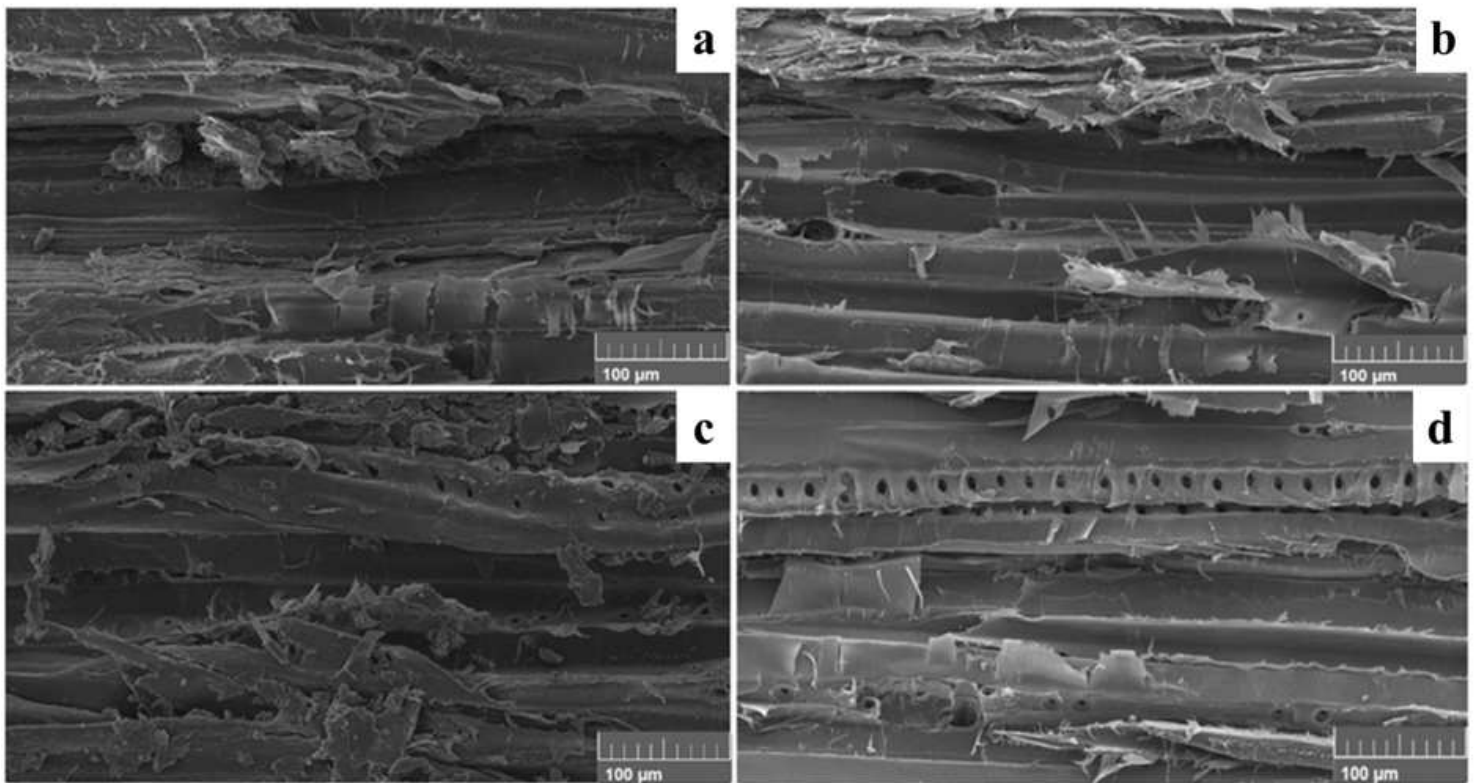


Figure 5

Side view of the glue lines promoted by the bonding with the tannin-based adhesives. Where a, b, c, and d refer to MT, MT + NaOH, MT + NaHSO₃, and Phenotan M® (IT), respectively.