

Journal of Experimental Agriculture International

23(3): 1-11, 2018; Article no.JEAI.41335 ISSN: 2457-0591 (Past name: American Journal of Experimental Agriculture, Past ISSN: 2231-0606)

Quantification and Qualification of Tannins from Residues of Forest Species *Hymenea courbaril* Duke and *Bowdichia virgiloides* Kunth

Felipe Silva Amorim¹, Leandro Calegari², César Henrique Alves Borges^{3*}, Clebson Lima Cerqueira⁴, Maílson Pereira de Souza⁵ and Francisco Tibério de Alencar Moreira³

> ¹Department of Forest Science, Federal University of Piauí, Brazil. ²Department of Forest Science, Federal University of Campina Grande, Brazil. ³Department of Forest Science, Federal Rural University of Pernambuco, Brazil. ⁴Department of Forest Science, Federal University of Paraná, Brazil. ⁵Department of Forest Science, Federal Rural University of Rio de Janeiro, Brazil.

Authors' contributions

This work was carried out in collaboration between all authors. Authors FSA, LC, CHAB, CLC, MPS and FTAM conceived and designed the research. Authors FSA, CHAB and MPS performed the experiments. Authors FSA and LC analyzed the data. Authors CLC and FTAM contributed reagents/materials/analysis tools. Authors FSA, LC and CHAB wrote and edited the manuscript. All authors read and approved the final manuscript.

Article Information

DOI: 10.9734/JEAI/2018/41335 <u>Editor(s):</u> (1) Suleyman Korkut, Professor, Duzce University, Department of Forest Industrial Engineeering, Division of Wood Mechanic and Technology, Turkey. (1) Nebi Bilir, Suleyman Demirel University, Turkey. (2) Aurora Martínez Romero, Universidad Juárez del Estado de Durango, Mexico. (3) Redemtor Awuor Ojwang, University of Nairobi, Kenya. Complete Peer review History: <u>http://www.sciencedomain.org/review-history/24761</u>

Original Research Article

Received 23rd February 2018 Accepted 2nd May 2018 Published 23rd May 2018

ABSTRACT

This research aimed to compare yield and quality of tannic powder extracts from forest residues from jatobá (*Hymenea courbaril* Duke) and sucupira preta (*Bowdichia virgiloides* Kunth.) generated from a joinery. Tannic extract was obtained with an adapted soxhlete extractor. Extract was analysed about its extraction yield (Y%), total solids (TS%), humidity (H%), corrected soluble and insoluble solids (SS%, IS%), tannic and non-tannic contents through formaldehyde and skin powder method (TCf%, nTCf%, TCp%, nTCp%), ashes content (%), density (g cm-3) and pH. The

*Corresponding author: E-mail: cesarhenrique27@yahoo.com.br;

experimental design was entirely casualized, with variables compared through F test. Jatobá presented yield of 15.87%, and black sucupira of 17.09%. For TCf% and nTCf%, jatobá presented 74.66% and 12.66% respectively, and black sucupira presented 64% and 12.66% respectively. Species showed potential for the production of tannic extract, with highlight to jatobá.

Keywords: Hymenea courbaril Duke; skin powder; Brazilian savanna and Caatinga.

1. INTRODUCTION

The furniture industry is known for heating up the wood industry due to its economic feedback. With this production, a great quantity of forest residues is generated. These residues, when in large quantity, impact the environment by polluting the air when incinerated and also the watercourses through leaching. In addition to bringing damage to the environment, forest residues also harm companies' logistics due to its volume, occupying large spaces [1].

Before this generation of forest residues, it is necessary to think of alternative ways for its utilization, considering its impact on the environment. Companies around the world have been reusing these residues (bark, shavings) for decades. Products such as pellets, agglomerated panels and briquettes have been produced for a long time and have won their place in the market.

An alternative way to add value to these residues would be producing tannin for animal tanning and/or adhesive production. Exploitation of tree barks would be one of the ways to extract hydrolysable tannins for production of adhesives for panels, once these give panels better characteristics such as: mechanical resistance and better collage [2]. Some species have been used for tannin production for animal tanning as chestnut tree, acacia and quebracho [3] and also of the forest tree species such as cedrus. For the northeast of Brazil, the Angico vermelho (*Anadenanthera columbrina* (Vell.) Brenan var. *cebil* (Gris.) Alts) is the most used species in animal tanning, for both goats and cattle.

Allied to this great tannin extraction, there is a problem with predatory exploitation. Without a management plan, this species has not been properly explored, which is promoting reduction of its population density in the region.

There is a clear need to use new species for tannin extraction for animal tanning in order to diversify the product and diminish the pressure on *Anadenanthera columbrina*. When using bark residues for this purpose, value addition to this material should also be considered. The present study aims to compare yield and quality of tannin powder extracts from forest residues (bark) of distinct species; *Hymenea courbaril* Duke (*H. courbaril*) and *Bowdichia virgiloides* Kunth (*B. virgiloides*) generated at a joinery located in Piauí extreme south.

2. MATERIALS AND METHODS

2.1 Study Area Characterization

Bark from *H. courbaril* and *B. virgiloides* stems were collected in Corrente, located in the extreme south of Piauí. Bark from both species was obtained at a rural community called Pau de Terra, 15 km from the municipality seat, using three individuals of each species. The municipality has an area of 3,048.466 km², with a population of 25,407 habitants. It is located in a transition zone between Brazilian savanna and Caatinga biomes, and a small portion of Atlantic forest. It comprises the geographical coordinates of 10°26' 34" S and 45°09' 43" W [4].

The city is bathed by Corrente and Paraim rivers, and also by a few streams. The municipality has a mean altitude of 438 m above sea level in its urban centre, with minimum mean temperatures of 23°C and maximum mean temperature of 39°C, with rain in November to April and mean annual rainfall of 900 mm [4]. Soil in the region is classified as yellow Latosol (allic or distrophic), with mean texture in association with quartz-type sand or red-yellow podzolic concretionary plinthic or non-plinthic [5].

2.2 Sampling and Bark Preparation

Initially, 5kg of bark from stalk of 5 species most used in a joinery located in the city of Corrente, in the state of Piauí. They were: *Tabebeuia alba*, *Daugerbia cearensis* D., Pterodon ermaginatus, *H. courbaril* and *B. virgiloides*. Barks were removed from the stem using a machete. Then, barks went through a preliminary study to determine which had tannin potential, following methodology presented by [6,7,8]. Stiasny number was determined, which corresponds to the content of reactive polyphenols in the tannic

extract (condensed tannin or proanthocyanidins). In this determination, 50 mL of the filtered analytical solution is added with 4 mL of formaldehyde (37%) and 1 mL of concentrated hydrochloric acid. This mixture is placed in a 500 mL volumetric flask on a heating blanket at 100°C for 30 minutes. Under these conditions, flavonoid units (mainly catechin and leucoanthocyanins) form insoluble complexes and are thus filtered off using quantitative medium filter paper. Among them, H. courbaril and *B. virgiloides* had the highest significative content of tannin in their barks, and thus they were used in this research.

Bark samples (5 kg) were stored in plastic bags, hermetically closed and addressed to the Laboratory of Forest Products Technology (LTPF) from Federal University of Campina Grande (UFCG) – Patos – PB campus, Centre of Health and Rural Technology (CSTR).

Bark particles were reduced after going through a forage machine, and hen directed to a milling process in a Willey-type mill. Pauses were performed during this process to guarantee that the samples were not chemically damaged. Then, the material was classified according to its granulometry after passing through a vibrating sieve [9]. After the sieving process, the material that passed through a 40-mesh sieve was selected and some material was retained in the 60-mesh sieve. Finally, each sample was stored separately in closed containers.

2.3 Preparation of Bovine Skin and Preparation of Skin Powder

Bovine skin was obtained from the Rural National Learning Service (SENAR) of Campina Grande -PB. Bovine skin went through the normal processing used by the institution. Arriving from the slaughterhouse, the skin was washed with current water to remove salt and impurities. Then, it was washed with water and detergent for 4 hours in a tannery machine. This machine is used to remove dirt, salt and to slightly hydrate the skin. After, the skin was washed again and put in the tannery machine with 40% water, sodium sulphide and lime for 1.5 hours. Right after this period, the material was left in the tannery machine, turned off, for more 18 hours turning on the machine every 2 hours for 15 minutes. After this step, the skin was washed with current water and flesh and fat were removed. The skin was then put in the tannery machine again where decalcification was

performed, using water and ammonia sulphite during 4 hours. After decalcification, bovine skin was washed, stretched with clips and dried in the shadow. This material was then taken to LTPF, where the skin was cut in little pieces of approximately 2 cm² with a scissor. This material was milled with a Willey-type mill with 40 and 60 mesh, generating skin powder that was stored in hermetically closed containers at room temperature.

2.4 Obtainment of Tannic Powder Extract

Approximately 40 g of air dried sample (336 hours) from species bark (sample 1) was soaked for 24 hours in distilled water and allocated in an extraction chamber in a Soxhlete extractor, which was adapted to avoid the reflux to return to the volumetric flask, directed to a beaker away from the high temperature of the boiling volumetric flask. The process was finished when 1000 ml of solution was generated.

The beaker containing the liquid extract was put in a stove at a constant temperature of 50°C with internal ventilation until liquid evaporation. Then, the anhydrous mass of the residue was calculated by the difference in relation to the mass of the empty beaker. This sample was grounded with a pistil and a mortar and then sieved. This process aimed to make an extract that could be sieved through the 60-mesh sieve.

2.5 Determination of the Extraction Parameters

After obtaining sample 1 (tannic extract), a second sample (sample 2) of 5 g was taken and put in the oven at 100°C to obtain its anhydrous mass, in order to calculate its humidity according to regulation D 6403 [10].

With the dry mass from sample 1 and humidity of sample 2, the anhydrous mass of the sample used in the extraction was calculated using the following Equation 1:

$$Ma_e = Mu_e (1 - (U\%/100))$$
(1)

Where:

- Ma_e = Anhydrous mass of the primary sample (g);
- Mu_e = Air dried mass of the sample used, 40g (g);

U% = Humidity of the secondary sample (%). With the anhydrous mass, the yield of the tannic extract was calculated using Equation 2:

$$Y\% = (Me/Ma_e) \times 100 \tag{2}$$

Where:

- Y% = yield of the tannic powder extract (%);
- Me = anhydrous mass of the extract in the container (g);
- Ma_e = anhydrous mass of the particles used in the extraction (g).

During the extraction process, a thermometer was used to obtain the temperature of the sample submitted to extraction in standard intervals (every 45 minutes). To complement the analysis, the total time of the process was registered.

2.6 Quantification of the Tannic Powder Extract

2.6.1 Preparation of the analytical solution

Tannic powder extract (3 g) was deposited in a Petri dish on an analytical balance with 0.1 mg precision to register its exact mass. With a funnel, 400 ml of distilled water at 80°C was transferred to a 500 ml volumetric flask, washing the Petri dish with the extract and the funnel carefully. The mixture was agitated vigorously until homogenised. Solution was then cooled until room temperature gradually for 12 hours in order to avoid temperature shocks. Finally, volume was completed to 500 ml with distilled water at room temperature.

2.6.2 Total solids and humidity content

Air dried tannic extract (1 g) was deposited in a Petri dish and its exact mass was registered. It was taken to the oven at $103\pm2^{\circ}$ C for 12 hours and then cooled in the desiccator until room temperature (15 minutes). The anhydrous mass of the residue was measured using an analytical balance. The total solid content was calculated using Equation 3, and humidity according to Equation 4:

$$TS\% = (Ma/Mde) \times 100 \tag{3}$$

Where:

TS = total solids of the tannic powder extract (air dried), in %;

Ma = mass of extract after dry (g); Mde = mass of dried extract (1 g), in g.

$$U\% = 100\% - TS\%$$
(4)

Where:

U% = extract humidity; ST = total solids (%);

2.6.3 Soluble and Insoluble solids

Analytical solution was submitted to a filtration sequence. The first filtration was performed with quantitative paper filter no. 40. From the filtered solution, a second filtration was performed using porous melting pot n° 2 (mean porosity 40-100 μ m).

After filtrations, two aliquots of 50 ml each were taken with a pipette. One was deposited in a beaker and the other on a volumetric flask. The beaker was transferred to the oven for 12 hours and then cooled in the desiccator until room temperature (15 minutes), then the total anhydrous mass was registered using an analytical balance.

Soluble solids content was calculated according to Equation 5, and insoluble solids according to Equation 6.

(5) $SS\% = [(Mrs/Mda) \times 100]$

Where:

- Mrs = anhydrous mass of the residue obtained after filtrations, extrapolated for the volume of the solution (g);
- Mda = airdried mass of the powder extract used to prepare the analytical solution (g).

$$IS\% = TS\% - SS\%$$
 (6)

Where:

IS = insoluble solids (%);

ST = total solids (%);

SS% = soluble solids (%).

2.6.4 <u>Tannic and non-tannic compounds by</u> <u>formaldehyde method</u>

The analytical solution (80 ml) filtered through a melting pot with porosity 2 was used. From this filtered, an aliquot of 50 ml was taken with a pipette and transferred to a 500 ml flat-bottomed flask, and 4 ml of 40% formaldehyde and 1 ml of concentrated hydrochloric acid were added. The

mixture was kept at 100°C using a heating mantle during 30 minutes under constant agitation. Filtration was performed using a quantitative paper filter (medium filtration) using 100 ml of distilled water. After filtration, the retained material was taken to an oven to obtain the constant mass of the precipitate. The calculation was performed according to Equations 7 and 8.

$$CTC\% = \left((Mp/g_1) \times 100 \right) \tag{7}$$

Where:

- CTC = condensed tannin content on powder extract (%);
- Mp = anhydrous mass of the precipitate (g);
- g_1 = anhydrous mass of the powder extract (g).

$$CnT\% = SS\% - CT \tag{8}$$

Where

CnT = condensed non-tannic content (%);

SS% = soluble solids

CTC = condensed tannin content on powder extract (%).

2.6.5 <u>Tannic and non-tannic compounds by</u> <u>filter method</u>

After putting 5g of skin powder in the Procter campanula, this portion was immersed in non-filtered analytical solution until the powder was completely damp. After 24 hours, the dripping process was initiated (8-10 drops per minute) during two hours, collecting 90 ml of the solution. The first 30 ml were eliminated. From the remaining 60 ml, 50 ml was taken with a pipette and overdried at 100°C to obtain the anhydrous mass of the residue.

During this process, particles of substances from the skin powder were transported so it was necessary to perform a black test using the previously described methodology, replacing the analytical solution with distilled water and then determining the dry residue of the filtered.

After this step, the content of non-tannic compounds was calculated according to Equation 9 and the content of tannic compounds according to Equation 10:

$$nTS\% = (g_3/P_1) \times 10 \tag{9}$$

Where:

- nTS% = non-tannic solids from powder extract (%);
- g₃ = anhydrous mass of the residue without tannins minus the black value, extrapolated for the volume of the solution (g);
- P₁ = airdried mass of the powder extract used to prepare the analytical solution (g).

Assuming that part of the tannins in solution will be retained in the powder skin, tannin content was calculated by the difference between soluble and insoluble non-tannic solids (Equation 10).

$$TC\% = SS\% - nTS\%$$
 (10)

Where:

- TC% = tannin content in the powder extract (%); SS% = soluble solids, Equation 6 (%);
- nTS = non-tannic solids, Equation 8 (%).

2.6.6 Correction for 0% humidity

A correction of humidity was performed, as most values related to tannic extract correspond to humidity content of the airdried extract. Therefore, it was necessary to know the anhydrous mass of the extract, through Equation 11, in order to calculate the mass of the extract in the desired humidity conditions, through Equation 12.

$$M_o = M_{ad} \times \left(1 - (U\%/100)\right) \tag{11}$$

Where:

- M_o = anhydrous mass of the extract used to prepare the analytical solution (g);
- M_{ad} = airdried mass of the extract used to process the analytical solution (%);

U% = humidity of the extract.

$$MU_x = M_{0\%} \div \left(1 - (U_{x\%}/100)\right)$$
(12)

Where:

- MU_x = anhydrous mass of the extract used to prepare the analytical solution in a certain humidity (g);
- $M_{0\%}$ = anhydrous mass of the extract used to prepare the analytical solution (g);
- $U_{x\%}$ = correction humidity (%).

In this way, from the previous equations, unit P_1 was replaced by the mass of the extract on $MU_{x\%}$, for correction of results considering the anhydrous mass of the powder extract.

2.6.7 <u>Determination of tannin content in the</u> residue

Knowing the extraction yield for each analysed species, the tannic content and considering both methodologies (formaldehyde and skin powder), the tannin content in the bark was calculated (Equation 13).

$$TC_r\% = (Y\% \times TC\%/100)$$
 (13)

Where:

TCr = tannin content in the residue (%);

Y% = powder extract yield (%);

TC% = tannin content in the powder extract.

2.6.8 <u>Determination of apparent density, pH</u> and ashes

The NBR 14449 [11a] was used to determine the apparent density of the powder extract using a graduated cylinder. Using NBR 11119 [11b], pH of the analytical solution was determined using a pH meter model LUCA 2000.

Determination of ashes content was performed according to norm NBR 11042 [12]. A mass of 1g of tannic extract was placed in a previously weighted melting pot in a balance with a 0.001 g precision. Samples were calcined in a Bunsen burner and then placed in a muffle furnace at 800°C for 3 hours.

2.6.9 Experimental design and statistical analysis

The experimental design was entirely casualized, with treatments corresponding to the two species, each submitted to five extractions (repetitions) that were analysed in duplicates (subrepetitions). Main variables analysed were extraction temperature (°C), extraction time (hours:minutes), tannic extract yield (Y%), total solids of the extract (TS%), extract humidity (U%), corrected solublr solids (SSc%), corrected insoluble solids (ISc%), tannic content by formaldehyde method (TCf%), non-tannic content by formaldehyde method (nTCf%), tannic content by skin powder method (TCp%), non-tannic content by formaldehyde method (TCp%), non-tannic content by formaldehyde method (nTCp%), ashes content (%), density (g cm⁻³) and pH. These variables were compared using an F-test. For the data set, assumptions of normality and homoscedasticity were verified. In all cases, a 5% error probability was considered.

3. RESULTS AND DISCUSSION

3.1 Parameters of the Generation of Tannic Extract

Humidity of the bark of both studied species did not differ from each other (Table 1). This shows that experimental conditions were the same in both cases and that species have similar behaviour regarding humidity loss and absorption from the environment.

No significant difference also occurred for time and temperature when generating the analytical solution from tannic extract, therefore the experiment was standardized regarding extraction conditions and thus the only factor that caused alterations on yield was the species.

[6] cited mean values of humidity of 7.93% for *Anadenanthera columbrina*, 9.72% for cashew tree, 8.96% for *Mimosa teuiflora* and 9.75% for red jurema. [7] cited mean values of humidity for *Mimosa teuiflora* of 9.6% and for black acacia of 8.3%.

Tannic extract yield of *H. courbaril* and *B. virgiloides* were of 15.87% and 17.09%, respectively. [13] worked with quantification of tannin contente in bark an wood of five tree legume species. They found yield values varying from 4 (*Acacia mangium*) to 16% (*Piptadenia gonocantha*), which is similar to the values found in this work. Analysing total tannins in aromatic and medicinal plants, [6] found value of tannic extract yield varying from 12.9% for

 Table 1. Parameters of the generation of the tannic extract analytical solution from *H. courbaril* and *B. virgiloides* barks

Species	U (%)	Temperature (C°)	Time (h:min)	Yield (%)
H. courbaril	4.32 ^{ns}	67.8 ^{ns}	6:46 ^{ns}	15.87 ^s
B. virgiloides	4.72	68.2	6:48	17.09
p value (%)	28.30	79.33	87.32	0.09

U= humidity; Temperature = sample temperature during extraction; Time = extraction time;b Yield = extraction yield (%); ns = non-significant statistical difference; s = significant statistical difference. Thymus vulgaris to 23.3%, for Acacia mearnsii. Tannin extraction was performed using 5 g of bark from each species and 50 ml of ethanol in an erlenmeyer for 10 days. [14] performed tannin extraction of bark from four Pinus species with water and different concentrations of sodium peroxide. They found mean tannic extract yield of 13.19% for Pinus caribea var. caribaea; 15.38% for Pinus caribaea var. hondurensis; 18.39% for Pinus caribaea var. bahanmensis; and 21.51% for *Pinus oocarpa*. Therefore, the values found by these authors are higher than the ones found in this work. This can be explained by the fact that each species has a determined tannin content in its composition, considering that this content can reach up to 40% in the bark of some forest species [14].

3.2 Tannic Extract Quantification

For total solids (TS%), there was no significant difference between species, as *H. courbaril* presented 93;16% and *B. virgiloides* 92.40% (Table 2). It can be concluded that their hygroscopicity is similar, considering that such parameters are related to humidity and they were stored under the same conditions.

[7] found mean values of total solids of 39;9% for Mimosa teuiflora, and 56.8% for black acacia, with significative difference between species. In different parts of Anadenanthera columbrina, [15] found a content of 23.30% of total solids in the bark. [12] analysed the viability of tannins for leather tanning in four species of the Brazilian semiarid region. They found total solid content of 22.46% for red angico; 33.36% for cashew tree; 26.32% for black jurema and 24.64% for Mimosa teuiflora. In a study qualifying plant tannin content in some indigenous species from Sudan, [8] cited mean values varying from 18.9% (Zizyphusspina-christi) 51.8% to (Acacia mearnsii). [16] found a mean value of total solids of approximately 54.16% for barbatimão. However, the methodology used by these authors differ from the one used in this study, as they performed quantification directly from the

material and not from the tannic powder extract obtained from the material.

Humidity of tannin extract from species was 6.84% for H. courbaril and 7.60% for B. virgiloides, with no significant difference between them. The main factor that allowed this result was the same environmental condition provided to the extracts from both species. Studies species showed low humidity, which is desirable because the lower the humidity, the higher the anhydrous mass and higher the vield. Regarding to the parameters of the tannic extract, norm IS 6199 (2006) determined for black acacia extracts a maximum value of 6% of humidity. In the technical file of TANAC company [17], a mean humidity of 6.5% was found for the product Weibull® (extract from natural mimosa, indicated for tanning and re-tanning of regular leather). In the technical file of Seta company [17], a value of 7% was found for an extract of acacia of universal use.

There was no significative difference between insoluble solids. *H. courbaril* presented a content of 5.82% and B. virgiloides of 13.39%. These values are low, especially or *H. courbaril*, which is preferable as this fraction of total solids is not soluble in water. For soluble solids, H. courbaril presented 87.33% and B. virgiloides presented 79%. Content of soluble solids was high in this study, especially for H. courbaril, which is also preferable as this parameter corresponds to the portion of total solids that is soluble in water. [8] cited a mean value of 48.7% f soluble solids for Acacia mearnsii in it tannic content. Norm IS 6199 (2006) determined a maximum value of 2.5% of insoluble solids for black acacia extract. The product from SETA company, Seta Sun ®, presented a value of 1% [17].

There was a significative difference in tannic content by formaldehyde method between species, especially for *H. courbaril* that presented a content of 74.66%, while *B. virgiloides* presented 64%. There was also a significative difference for tannic content by skin powder method, especially for *H. courbaril*.

 Table 2. Comparisons between total solids, humidity content, soluble and insoluble solids of tannic extracts of barks from *H. courbaril* and *B. Virgiloides*

Species	TS(%)	U(%)	SSc(%)	ISc(%)
H. courbaril	93.16 ^{ns}	6.84 ^{ns}	87.33 [°]	5.82 ^s
B. virgiloides	92.40	7.60	79.00	13.39
p-value (%)	5.16	5.16	0.01	0.06

TS (%) = total solids; U (%) = humidity; SS(%) = corrected soluble solids; ISc(%) = corrected insoluble solids; ns = non-significant statistical difference; s = significant statistical difference To determine tannic and non-tannic content, the skin powder method reacts with condensed and hydrolysed tannins, while the formaldehyde method reacts only with condensed tannins. This implies that values from skin power method should be higher, which was not observed in this study. This can be explained by the fact that the powder skin used was the one produced in laboratory, instead of the commercial standard.

[18] analysed tannic extracts of *Pinus oocarpa* bark aiming its utilization as adhesives for wood. They found a tannic content of 15.06% using the formaldehyde method. [19] evaluated the productivity of condensed tannins in bark and found values for tannic content varying from 63% to 71.2% for *Mimosa tenuiflora* using formaldehyde method. [18] analysed adhesives made from tannins from eucalypt barks and found tannic content of 73.20% for *Eucalyptus grandis* and 72.54% for *Eucalyptus pellita*.

Regarding tannic content by skin powder method, there was a significative difference between the analysed species. *H. coubaril* presented a content of 54.53% and *B. virgiloides* of 43.06%.

[7], in a work to quantify tannins, found for *Mimosa tenuiflora* and black acacia values of 24.1% and 47.8%, respectively, using the skin powder method.

Weibull®, a product from TANAC, presented a value of 72% for content of tannic substances [17], while Seta Sun®, a product from SETA, presented a value of 72.5% [17].

According to the results from the formaldehyde method, there was no significant difference on non-tannic content between species. *H. courbaril* presented a content of 12.66% and *B. virgiloides* of 14.99%.

[15] analysed the content of tannic substances in different parts of *Anadenanthera columbrina* tree,

and found values close to 9.35% using the formaldehyde method. However, in this study, this value refers to the bark itself and not to the tannic powder extract obtained from the bark.

[20] determined the tannin content of *Mimosa tenuiflora* for production of formaldehyde tannic adhesive and found values between 7.60% and 9.60% of non-tannics in the bark from stem of this species. [21] extracted tannin from acacia bark and found a mean non-tannic content of approximately 4.59%.

Regarding non-tannic content by skin powder method, the studied species presented mean values of 32.78% for *H. courbaril* and 34.80% for *B. virgiloides*, with no significative difference between species. Seta Sun®, from SETA company, showed a value of 26.5% for non-tannic substances content [17]. Norm [22] establishes tannic and non-tannic contents of 72% and 27% respectively, for black acacia tannic extract.

It is observed that the values found in this work strongly differ from the results of the authors cited above. This is mainly due to the methodology used and the species. Another factor that can be considered is the period when samples were collected, as in different seasons tree tend to produce certain substances in distinct quantities.

Ashes content, density and pH from both studied species presented statistical difference between each other (Table 4).

Ashes represent the content of inorganic substances in the material, constituted by aluminium, iron, potassium, calcium, copper and manganese ions. In other words, ashes are the inorganic part left of the material after calcinated and with its organic matter transformed in CO₂. In Seta Sun®, a mean value of 3.5% of ashes content was found [17].

Table 3. Comparisons between tannic and non-tannic contents using formaldehyde and skin powder for tannix extracts from *H.courbaril* and *B. virgiloides* barks

Species	TCf(%)	nTCf(%)	nTCp(%)	TCp(%)
H. courbaril	74.66 ^s	12.66 ^{ns}	32.78 ^{ns}	54.53 ^s
B. virgiloides	64.00	14.99	34.80	43.06
P value (%)	0.03	24.65	33.83	0.01

TCf(%) = tannic content by formaldehyde method; nTCf(%) = non-tannic content by formaldehyde method; nTCp(%) = tannic content by skin powder method; TCp(%) = tannic content by skin powder method; ns = nonsignificant statistical difference; s = significant statistical difference.

Species	Ashes content (%)	Density (g cm⁻³)	рН
H. courbaril	4.06 ^s	0.788 ^s	6.34 ^s
B. virgiloides	1.82	0.720	5.57
P value (%)	0.01	1.24	0.65

 Table 4. Ashes content, apparent density and pH of tannic extract from bark of the studies

 species

ns = non-significant statistical difference; s = significant statistical difference.

Ashes are an undesirable part in the tannic extract, that is, the lower the ash content in the material, the better its utilization and, consequently, its quality. The *B. virgiloides* stands out for its low quantity of this parameter in its composition (1.82%).

Regarding density of tannic extract of the studied species, *H. courbaril* presented apparent density of 0.788 g cm⁻³, and *B. virgiloides* of 0.720 g cm⁻³. Density expresses the degree of ass concentration in a determined volume, that is, it defines the quantity of tannic extract present in a given place and/or container. In this work, *H. courbaril* stands out.

The pH determines how acid, basic or neutral a substance is. In this aspect, the studied species were statistically different between each other. *H. courbaril* presented pH of 6.34, and *B. virgiloides* of 5.57.

In Weibull® and Seta Sun®, values of 4.5 to 5.0 and 4.3 to 4.7 were cited, respectively [17]. In a study evaluating tannins reactivity from barbatimão bark, [16] found a mean pH value of 4.9 in the tannic extract. Analysing parameters of tannic extract from black acacia through norm IS 6199 (2009), a pH value varying from 4.8 to 5.4 was found. [14]) verified tannin content from bark of four Pinus species and found pH values of 3.45 for Pinus caribaea var. bahamensis, 3.27 for Pinus caribaea var. caribaea, 3.21 for Pinus caribaea var. hondurensis and 3.35 for Pinus oocarpa. [18] studied chemical propertis of Pinus oocarpa bark extract in order to use it as adhesive and found pH of 3.14 for its tannic extract. [8] analysed tannic content of many species in Sudan ans found mean pH of 6 for Acacia mearnsii e Azadiratcha indica.

3.3 Tannic Content in the Residue

Using the formaldehyde method, the tannic content in material from *H. courbaril* and *B. virgiloides* was 11.85% and 10.94%, respectively. Using the skin powder method, values were 8.65% and 7.36%, respectively (Table 5).

Table 5. Tannic content of the material, forboth species and methods

Species	Tannic content (%)		
	Formaldehyde	Skin powder	
H. courbaril	11.85	8.65	
B. virgiloides	10.94	7.36	

In order to determine whether a forest species is considered viable in tannin production, regardless of the method used, the tannin content in the material must be at least 10% [23,8]. *H. courbaril* and *B. virgiloides* presented higher values using formaldehyde method. This implies that both species studied have the potential to be exploited commercially. Also, there is the advantage that the raw material originates from waste, that is, free material, which minimizing production costs.

4. CONCLUSIONS

Bark residues from *H. courbaril* and *B. virgiloides* were shown to have potential for tannic extract production due to its considerable values of yield and tannic extract quality.

In general, *H.* courbaril was the one that stood out because it presented better values in the majority of the evaluated parameters.

COMPETING INTERESTS

Authors have declared that no competing interests exist

REFERENCES

- Protásio TP, Alves ICN, Trugilho PF, Silva VO, Baliza AER. Compactação de biomassa vegetal visando à produção de biocombustíveis sólidos. Pesquisa Florestal Brasileira. 2011;31(68):273-283. Available:<u>http://pfb.cnpf.embrapa.br/pfb/ind ex.php/pfb/article/viewFile/288/227</u>
- Carneiro ACO, Vital BR, Frederico PGU, Santos RC, JÚNIOR WPS. Efeito da hidrólise ácida dos taninos de Eucalyptus

grandis. Hill ex maiden nas propriedades dos adesivos tânicos. Revista Árvore, 2009;33(4):733-799.

Available:<u>http://www.scielo.br/pdf/rarv/v33n</u> <u>4/v33n4a16</u>

- Jorge FC, Brito P, Pepino L, Portugal A, Cota RP. Aplicações para as cascas de árvores e para os extractos taninosos: Uma revisão. Silva Lusitana. 2001;9(2): 225-236. Available:<u>https://bibliotecadigital.ipb.pt/bitst</u> ream/10198/1273/1/2001 sl Jorge et al.p df
- IBGE Instituto Brasileiro de Geografia e Estatística. Índices de nomes geográficos, Rio de Janeiro; 2011. Available:<u>ftp://186.228.51.40/documentos/</u> cartografia/indice nomes geograficos.pdf
- Jacomine PKT, Camargo MN, Carvalho AP, Pessoa SCP, Cavalcanti AC, Mélo Filho HFP, Medeiros LAR, Burgos N, Lopes OF, Formiga RA. Levantamento exploratório – reconhecimento de solos do Estado do Piauí. Rio de Janeiro, RJ: Embrapa Solos. (Embrapa Solos. Boletim Técnico, 63). 1986;234.
- Paes JB, Diniz CEF, Marinho IV, Lima CR. Avaliação do potencial tanífero de seis espécies florestais de ocorrência no semiárido brasileiro. Cerne. 2006;12(3):232-238.

Available:<u>http://www.dcf.ufla.br/cerne/artig</u> os/10-02-

20094298v12_n3_artigo%2004.pdf

- Calegari L, Lopes PJG, Oliveira E, Gatto DA, Stangerlin DM. Quantificação dos taninos das cascas de jurema-preta e acácia-negra. Pesquisa Florestal Brasileira. 2016;36(85)61-69. Available:<u>http://pfb.cnpf.embrapa.br/pfb/ind ex.php/pfb/article/download/986/472</u>
- Haroun M, Khirstova P, Covington T. Evaluation of vegetable tannin contents and polyphenols of some indigenous and exotic woody plant species in Sudan. Journal of Forest Products and Industries. 2013;2(4):48-54. Available:<u>http://researchpub.org/journal/jfpi</u>

/number/vol2-no4/vol2-no4-8.pdf

- Makkar HPS. Quantification of tannins in tree and shrub foliage: A laboratory manual. Kluwer Academic Publishers, Massachusetts. 2003;102.
- American Society for Testing and Materials (ASTM). D 6403-99: Standard test method for determining moisture in raw and spent materials. Pennsylvania: ASTM. 2004b;3.

- A Associação Brasileira De Normas Técnicas (ABNT). NBR 14449: Insumos -Tanantes - Determinação da densidade. Rio de Janeiro: ABNT. 2008a;3.
 B - Associação Brasileira De Normas Técnicas (ABNT). NBR 11119: Insumos -Tanantes - Determinação do pH. Rio de Janeiro: ABNT. 2008b;1. Associação Brasileira De Normas Técnicas (ABNT). NBR 11042: Insumos - Tanantes -Determinação do teor de cinza. Rio de Janeiro: ABNT. 2013;4.
- Gonçalves CA, Lelis RCC. Teores de taninos da casca e da madeira de cinco leguminosas arbóreas. Floresta e Ambiente. 2001;8(1):167-173. Available:<u>http://www.floram.org/files/v8n%</u> <u>C3%BAnico/v8nunicoa21.pdf</u>
- Pansera MR, Santos ACA, Paese K, Wasum R, Rossato M, Rota LD, Pauletti FF, Serafini LA. Análise de taninos em plantas aromáticas e medicinais cultivadas no nordeste do Rio Grande do Sul. Revista Brasileira de Farmacognosia. 2003;13(1): 17-22.

Available:<u>http://www.scielo.br/pdf/rbfar/v13</u> n1/a02v13n1.pdf

 Paes JB, Santana GM, Azevedo TKB, Morais RM, Júnior JTC. Substâncias tânicas presentes em várias partes da árvore angico-vermelho (Anadenanthera colubrina (Vell.) Brenan. var. cebil (Gris.) Alts.). Scientia Forestalis. 2010;38(87): 444-447.

Available:<u>http://www.ipef.br/publicacoes/sci</u>entia/nr87/cap11.pdf

15. Almeida JE, Logsdon NB, Jesus JM. Painéis de madeira aglomerada produzidos com serragem e poliestireno expandido. Floresta. 2010;42(1):189– 200.

Available:<u>http://200.129.241.80/ppgeea/sis</u> tema/projetos/3.pdf

- 16. Taninos Da Acácia Ltda. Produtos; 2016. Available:<u>http://www.tanac.com.br/pt-br/</u>
- Vieira MC, Lelis RCC, Rodrigues DD. Propriedades químicas de extratos tânicos da casca de Pinus oocarpa e avaliação de seu emprego como adesivo. Cerne. 2014; 20(1):47-54. Available:<u>http://www.redalyc.org/pdf/744/7</u> 4430342006.pdf
- Lopes PJG, Calegari L, Calegari CCA, Oliveira E, Stangerlins DM, Gatto DA. Produtividade em casca e taninos condensados de jurema-preta. Nativa. 2015;03(02):95-101.

Available:<u>http://periodicoscientificos.ufmt.b</u> r/ojs/index.php/nativa/article/viewFile/2015/ pdf

 Azevedo TKB, Paes JB, Calegari L, Nascimento JWB. Qualidade dos taninos de jurema-preta (*Mimosa tenuiflora*) para a produção de adesivo de tanino formaldeído. Ciência Florestal. 2015;25(2): 505-512.

Available:<u>http://www.redalyc.org/pdf/534/5</u> 3439559023.pdf

20. Carneiro ACO, Vital BR, Frederico PGU, Moreira AB, Dambroz CS. Efeito do sulfito de sódio na extração de tanino da casca de Anadenanthera peregrina. Floresta e Ambiente. 2007;14(1):65-69. Available:<u>https://www.researchgate.net/pro file/Andrea Moreira3/publication/24274325</u> <u>8 Efeito do sulfito de sodio na extraca o de tanino da casca de Anadenanther a peregrina/links/0deec538175a49481e00 0000/Efeito-do-sulfito-de-sodio-na-</u> extracao-de-tanino-da-casca-de-Anadenanthera-peregrina.pdf

- 21. INDIAN STANDARD (IS). IS 6199-1971: Specification for wattle extract. New Delhi: IS. 2006;13.
- Vázquez G, González-alvarez J, Santos J, Freire MS, Antorrena G. Evaluation of potential applications for chestnut (*Castanea sativa*) shell and eucalyptus (*Eucalyptus globulus*) bark extracts. Industrial Crops and Products. 2009;29(2): 364–370. Available:http://www.sciencedirect.com/sci

ence/article/pii/S0926669008001544

 Yazici N, Billir N. Aspectual fertility variation and its effect on gene diversity of seeds in natural stands of Taurus cedar (*Cedrus libani* A. Rich.). International Journal of Genomics. 2017;2960624:1-5. Available:<u>http://downloads.hindawi.com/jou</u> rnals/ijg/2017/2960624.pdf

© 2018 Amorim et al.; This is an Open Access article distributed under the terms of the Creative Commons Attribution License (http://creativecommons.org/licenses/by/4.0), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

Peer-review history: The peer review history for this paper can be accessed here: http://www.sciencedomain.org/review-history/24761