Scientific Article

PROPERTIES OF CROSS-LAMINATED TIMBER BONDED WITH AN ADHESIVE BASED ON TANNINS FROM THE BARK OF *Mimosa tenuiflora* TREES

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ABSTRACT – This study aimed to determine the concentration of condensed tannins in the bark of *Mimosa tenuiflora* (Willd) Poir trees. Additionally, the physical-mechanical properties of cross-laminated timber (CLT) panels bonded with an adhesive based on *M. tenuiflora* tannins were assessed. Bark was collected from five trees. The adhesive formulation was synthesized by mixing powdered tannins, paraformaldehyde, and water at 50 °C under continuous stirring. Bark tannins extract in its pristine state without any previous chemical treatment or modification was employed to synthesize the adhesive. Viscosity, pH, solids content, and gel time of the adhesive formulation were determined. CLT panels were manufactured with *Eucalyptus* spp. wood. The bonding performance was assessed by determining the physical (apparent density and water absorption) and mechanical (modulus of elasticity – MOE, modulus of rupture – MOR, and shear strength) properties of the CLT panels. Condensed tannins content in the *M. tenuiflora* bark was 23.4%. Adhesive properties were pH = 3.93, TS of 50.64%, gel time of 460 s, and 6,000 cP for viscosity. Values of MOE, MOR and shear strength were 4,411, 16.18, and 1.06 MPa, respectively. The *M. tenuiflora* tannins are promising for the formulation of adhesives to bond CLT.

Keywords: Polyphenols; Cross-laminated timber; Brazilian dry forest.

PROPRIEDADES DA MADEIRA LAMINADA CRUZADA COLADA COM ADESIVO À BASE DE TANINOS DA CASCA DAS ÁRVORES DE Mimosa Tenuiflora

RESUMO – Este trabalho teve como objetivo determinar a concentração de taninos condensados na casca da árvore ***Mimosa tenuiflora*** *(Willd) Poir. Adicionalmente, foram avaliadas as propriedades físico-mecânicas de painéis de madeira laminada cruzada (MLC) colados com um adesivo à base de taninos de* ***M. tenuiflora****. A casca foi coletada de cinco árvores. A formulação adesiva foi sintetizada pela mistura de taninos em pó, paraformaldeído e água a 50°C sob agitação contínua. Extrato de taninos da casca em seu estado primitivo sem qualquer tratamento químico prévio ou modificação foi empregado para sintetizar o adesivo. Foram determinados a viscosidade, pH, teor de sólidos e tempo de gel da formulação adesiva. Os painéis foram fabricados com madeira de* ***Eucalyptus*** *spp. O desempenho de colagem foi avaliado determinando as propriedades físicas (densidade aparente e absorção de água) e mecânicas (módulo de elasticidade – MOE, módulo de ruptura – MOR e resistência ao cisalhamento) dos painéis de MLC. O teor de taninos condensados na casca de* ***M. tenuiflora*** *foi de 23,4%. As propriedades adesivas foram pH = 3,93, TS de 50,64%, tempo* *de gel de 460 s e viscosidade de 6.000 cP. Os valores de MOE, MOR e resistência ao cisalhamento foram 4.411, 16,18 e 1,06 MPa, respectivamente. Os taninos de* ***M. tenuiflora*** *são promissores para a formulação de adesivos para unir MLC.*

Palavras-Chave: Polifenóis; Madeira laminada cruzada; Floresta seca brasileira.

1. INTRODUCTION

The development of synthetic adhesives resulted in the consolidation of the wood panel industry, with urea-formaldehyde (UF) and phenol-formaldehyde (PF) being the most commonly used adhesives for wood bonding. However, these synthetic adhesives are based on petrochemicals and will become more expensive if oil prices increase significantly in the future. Of the two types of adhesives, PF only can be used to produce water-resistant materials. Additionally, PF adhesives are expensive, so the search for renewable compounds able to replace phenol in synthesizing phenolic wood adhesives has increased in the past two decades (Hemmilä et al., 2017; Norström et al., 2018). Among the renewable resources employed to synthesize wood adhesives, vegetable tannins stand out as a technologically viable alternative, mainly due to the formation of a polymer with a rigid structure after reacting with formaldehyde, forming the well-known tannin-formaldehyde class of adhesives (Carvalho et al., 2014a). In this material, less formaldehyde is used for reaction compared to the classic phenolic resins, which consequently reduces the demand for this fossil resource to make wood adhesives (Gonçalves and Lelis, 2009).

Condensed tannins area class of polyphenolic substances that can be found in various parts of the higher plants, where the concentration may vary depending on age, plant structure, and collection site (Sartori et al., 2014), plant phenophase (Azevêdo et al., 2017) and plant part collected. Condensed tannins, mainly due to their biological functions (Sartori et al., 2014), are the most common type of tannic substances sold. They are employed for leather tanning (Guo et al. 2020), treatment of wastewater and drinking water (Bongiovani et al. 2015; Grehs et al. 2019), as dispersing agents to control the viscosity of clays in drilling oil wells (Rutledge and Anderson 2015), and in the pharmaceutical and food industries. They are also used in the synthesis of adhesives for bonding wood and wood products (Antov et al., 2020), especially because of their fast reaction with formaldehyde, making them suitable to make wood panels without significant changes in the usual bonding and pressing conditions.

Several types of forest species have been mentioned in the literature as potential sources of condensed tannins to synthesize adhesives for wood bonding. For instance, adhesives based on tannins from the bark of *Stryphnodendron adstringens* trees were successfully employed by Carvalho et al. (2014b) to bond plywood panels. Likewise, Vieira et al. (2014) found the same potential for adhesives based on *Pinus oocarpa* bark tannins. Currently, the only tannin-producing species widely used as raw material for wood adhesive production is *Acacia mearnsii*. However, the bark tannins of other forest species can be employed for the same purpose, especially considering that in some places the introduction of exotic species may not be desirable. Thus, it is important to research to find a greater variety of local species able to supply condensed tannins that are adequate to synthesize adhesives for wood bonding.

In this respect, *Mimosa tenuiflora* is a native species widely found in the dry forests of the Brazilian semiarid region. It is a pioneer and fast-growing species and is considered a highly aggressive invader since after cutting it sprouts at any time of the year (Pereira Filho et al., 2005). In Brazil's Northeast region, the species is widely used for the production of firewood and charcoal (Silva et al., 2012), and its bark can be used as a source of vegetable tannins (Paes et al., 2006). Due to the species' easy growth in a region of difficult plant production, it has high potential. Since the introduction of exotic species in the region has resulted in serious environmental impacts, like the case of *Prosopis juliflora*, which aggressively suppresses a significant part of the local flora where it is cultivated, attention is now focused on finding native tree species for making timber and non-timber products. In this sense, native species from both natural forests and forested areas have been assessed.

Here we report the viability of synthesizing a renewable adhesive for wood bonding that is environmentally friendly and technologically efficient, using as raw material the condensed tannins extracted from the bark of *M. tenuiflora* trees. More specifically, we aimed to i) determine the concentrations of tannins in the bark of *M. tenuiflora* trees; and ii) evaluate the physical-mechanical properties of cross-laminated timber panels bonded with a tannin-formaldehyde adhesive formulation.

2. MATERIAL AND METHODS

Depicts the main steps employed for the production of the CLT panels using the *M. tenuiflora* tannin-formaldehyde adhesive formulation (Fig 1).

2.1 Bark collection and preparation

Five *M. tenuiflora* trees with age 5 years and in good phytosanitary condition were harvested and debarked, from a forest plantation of the Agricultural Sciences Academic Unit located in the municipality of Macaíba, Rio Grande do Norte State, Brazil – 5°51'30" S and 35°21'14" W. The local climate, according to the Köppen classification, is characterized as tropical rainy (transition between types As and BSw), presenting a rainy season in autumn and winter, with an average temperature of 27 °C, with a maximum of 32 °C and a minimum of 21 °C, average annual relative humidity of 76% and rainfall ranging between 863.7 and 1,070.7 mm (Alvares et al., 2014). The diameter at breast height (DBH) of the harvested trees had an average of 12.6 cm. The bark was dried in a solar oven (40 ± 5ºC) and ground in a Wiley mill. The ground material was sifted and the fraction that passed through a 2.00mmmesh sieve was stored for further processing.

2.2 Tannins extraction and quantification

The ground bark was sieved again, this time collecting the fraction retained with 0.25 mm mesh. The moisture content of the bark was determined after drying in a laboratory oven at 103 + 2 °C with two replicates of 5 g each. The bark was subjected to hot-water extraction with 250 ml of distilled water and 25 g of the dry material mixed in a 1,000 mL flask (10:1 v/m ratio of water and bark). The material was kept boiling for two hours under reflux. Then the material was subjected to a new extraction in the same conditions to remove the maximum quantity of extractable solids possible. After the hot-water extraction, the resulting liquid from both extraction steps was mixed and then filtered with a porous sintered glass crucible (porosity grade 2). The material's volume was concentrated to 250 mL by

Figure 1 – Schematic representation of the stages for assessing the quality of cross-laminated timber panels bonded with an adhesive based on tannins from *M. tenuiflora* bark.
Figura 1 *– Representação esquemática das etapas de avaliação da qualidade de painéis de madeira laminada cruzada colados com adesivo à base de taninos da casca de M. tenuiflora.*

evaporating the water using a Soxhlet apparatus. After concentration, four 50 mL aliquots were used for each extract. Two aliquots were oven-dried at 103 ± 2 ºC for 48 h to obtain the total solids content (TS) and the other two were assayed to determine the condensed tannins content (CT). The CT was determined by using the Stiasny method described by Guangcheng et al. (1991). For each aliquot, 4 mL of formaldehyde (37% w/w) and 1 mL of concentrated hydrochloric acid were added. Each mixture was boiled under reflux for 30 minutes. The material was separated by filtration with a porous sintered glass crucible (porosity grade 2). The retained fraction was oven-dried at 103 ± 2 ºC for 48 hours and the Stiasny index was calculated.

2.3Adhesive synthesis and determination of properties

To produce the hot-water extract for adhesive synthesis, 1 kg of ground bark and 10 L of water were employed (1:10 m/v ratio). The samples were boiled in an autoclave, with a capacity of 100 L, for two hours. Each bark sample was subjected to two extractions, so the final bark/water ratio was 1:20. The water extract was homogenized, transferred to aluminum trays, and dried in a solar oven (40 ± 5 ºC) until the tannins became completely solid. The dry material was powdered, sieved (0.25 mm granulometry), and stored under refrigeration until further synthesis of the adhesive formulation. The adhesive was synthesized by mixing 156 g of tannin powder, 170 mL of distilled water, and 14 g of paraformaldehyde in a 500 mL double-necked glass flask. The mixture was heated to 50 °C and kept under continuous stirring (500 rpm) until the complete mixing of the components. Then the adhesive formulation was cooled to room temperature and immediately employed for CLT bonding. There was no addition of any extender or filler to the adhesive formulation. The following properties were determined: pH, viscosity, gel time, and solids content. Viscosity was measured by using a Brookfield viscometer, according to the procedures described in the ASTM 1084-1997 standard. Gel time was determined using a Marconi MA560 gel timer (Piracicaba, SP, Brazil) with a glass tube dipped in glycerin (120 °C) with an adhesive sample of 6.0 g. The solids content was determined according to the ASTM 1490-01-2006 standard. All determinations were carried out in triplicate.

2.4 Production and quality assessment of cross-laminated timber (CLT) panels

Three-layered CLT panels with dimensions of 20 cm x 21 cm were produced with battens of Eucalyptus spp. wood measuring 7 cm(width)x 1 cm(thickness) x 20 cm(length). Five panels were bonded with the tannin-formaldehyde adhesive applied on only one side of the battens in the amount of 250 g m-2. After adhesive spreading, the panels were left for 15 min so the adhesive could be properly absorbed by the wood. For the pressing cycle, the following parameters were adopted: the pressure of 10 kgf cm-2, time of 10 min, and temperature of 150 ºC. After pressing, the panels were conditioned in a chamber at 20 ± 2 °C and relative humidity of 65 ± 5%. After conditioning, the panels were sawn to form the test specimens for the physical-mechanical assays. The moisture and apparent density were determined according to the standard NBR 7190 (ABNT, 1997). The water absorption (after 2 and 24 h of immersion) was determined as recommended by ASTM D1037: 2012. The shear strength of the glue line, modulus of elasticity (MOE), and modulus of rupture (MOR)from the static bending assay were determined according to ASTM D198: 2009. The Janka hardness was determined according to NBR 7190 (ABNT, 1997). For the analysis of the physical

TS = total solids content; I = Stiasny index; CT = condensed tannins content
*Percentage based on the dry weight of bark.
TS = teor de sólidos totais; I = índice de Stiasny; CT = teor de taninos condensados
**Porcentagem baseada no peso seco da casca.*

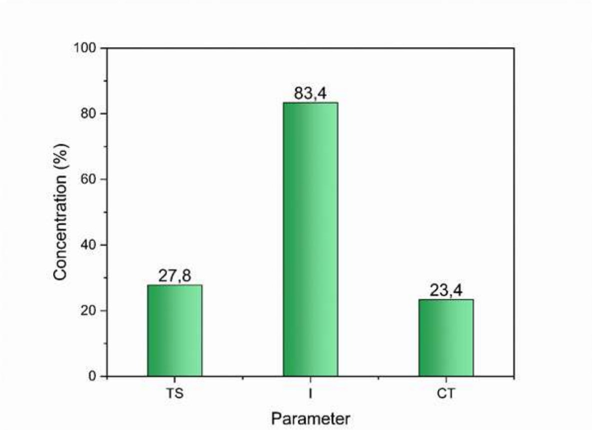

Figure 2 – Total solids content (%), Stiasny index (%), and condensed tannins content (%) of the hot-water extract from *M. tenuiflora*.
Figura 2 – *Teor de sólidos totais (%), índice de Stiasny (%) e teor de taninos condensados (%) do extrato em água quente de M. tenuiflora.*

Table 1 – Properties of the adhesive formulation based on tannins from *M. tenuiflora* bark
Tabela 1 *– Propriedades da formulação adesiva à base de taninos da casca de* ***M. tenuiflora****.*

Adhesive Property			
pH	**Solids content(%)**	**Gel time(s)**	**Viscosity(cP)**
3.93	50.6	460	>6.000

Table 2 – Physical properties of cross-laminated timber (CLT) panels bonded with the adhesive formulation based on tannins from *M. tenuiflora* bark.
Tabela 2 *– Propriedades físicas de painéis de madeira laminada cruzada (CLT) colados com a formulação adesiva à base de taninos da casca de* ***M. tenuiflora****.*

	Moisture content (%)	**Apparent density (kg m⁻³)**	**Water absorption(%)**	
			2 h	24 h
Mean	4.69	579	14.7	35.3
CV (%)	9.1	6.7	36.1	19.1

Table 3 – Mechanical properties of cross-laminated timber (CLT) panels bonded with the adhesive formulation based on tannins from *M. tenuiflora* bark.
Tabela 3 *– Propriedades mecânicas de painéis de madeira laminada cruzada (CLT) colados com a formulação adesiva à base de taninos da casca de* ***M. tenuiflora****.*

	Janka hardness (MPa)	**Shear strength (MPa)**	**Static bending (MPa)**	
			MOE	MOR
Mean	44.3	1.06	4,411	16.2
CV (%)	18.8	13.0	10.4	8.8

and mechanical properties, descriptive statistics were calculated from the data, using the *R software*.

3. RESULTS

In Figure 2 it is possible to observe the values of total solids content (TS), Stiasny index (I), and condensed tannins content (CT) of *M. tenuiflora* bark extracted in hot water.

The properties determined for the adhesive formulation produced with tannins from the *M. tenuiflora* bark are displayed in Table 1.

The data on the physical properties of the panels produced with the tannin-formaldehyde adhesive are shown in Table 2. The values of the mechanical properties of the panels are shown in Table 3.

4. DISCUSSION

The value of the total solids content (TS), which expresses the quantity of total extractable compounds present in the bark samples, was lower than that found by Calegari et al. (2016) for the bark of the same species assessed in this study (39.9%). The amount of bark and the time used in the tannin extraction process were higher than those used in the methodology applied in the present study, factors that may have influenced the difference in the results found. The Stiasny index (I) is related to the purity of the extract, and the higher the percentage, the lower the content of other compounds will be, such as gums, sugars, and hemicelluloses, which do not react with formaldehyde. The index found in this study for the *M. tenuiflora* bark was 83.4%, higher than that found by Lopes et al. (2015) and Azevêdo et al. (2015), respectively 71.02 and 71.12%. The higher values found here for the Stiasny index may be related to variations in the condensed tannins content in the bark due to different growing conditions (Paes et al. 2013).

According to Lopes et al. (2015), the condensed tannins content is the main parameter to evaluate the quality of hot-water extract from a given type of bark, because it represents the percentage of pure tannins in the sample. The CT obtained for the *M. tenuiflora* bark in this work was higher than those found by Lopes et al. (2015), Paes et al. (2006), and Azevêdo et al. (2017) for the same species (respectively, 14.1%,17.74% and from 6.04% to 21.90%). The *M. tenuiflora* also had higher CT values than those usually obtained for *Anadenanthera colubrina*, a native forest species traditionally used as a source of tannins in Northeastern Brazil, in which Paes et al. (2013) and Lima et al. (2014) found CT values of 15.98% and 11.89%, respectively. This comparison shows that *M. tenuiflora* has real potential as a source of tannins, so it can help reduce the overexploitation of *A. colubrina* in Brazil. It is important to note that the differences found between the values in the studies mentioned here for TS, I, and CT in the bark also can be related to differences in the age of the trees, time of the year when the bark was collected, the position of collection in the trees and mainly the site quality (Paes et al., 2010; Souza et al., 2021).

pH is a property of great importance for making adhesives because it influences the reactivity and viscosity of the tannin reactions. If the pH is not in the proper range (3 – 4), acceleration of the polymerization may occur, causing premature curing of the adhesive (Vieira et al., 2014). Also, very acidic adhesives can damage the glue line, causing corrosion and degradation of the panel over time (Zhang et al., 2010). Santiago et al. (2018), working with an industrial adhesive made from powdered tannins of *A. mearnsii*, observed a pH of 3.35, which is close to the value determined here for the *M. tenuiflora* adhesive formulation. Therefore, the pH of 3.93 presented here for the adhesive formulation is in the suitable range for wood bonding.

The solids content reflects the percentage of the binding agent in the adhesive formulation. Generally, a higher solids content is more appropriate for forming a more resistant glue line. However, very high solids content makes it difficult to apply the adhesive via spraying, impairing the spreading and penetration of the adhesive in the wood due to the increase in its viscosity (Carvalho et al., 2016). Further according to those authors, to produce good quality panels, the solids content should remain around 50%. The value found in this study fits the technical recommendation for tannin-based adhesives. For instance, the adhesives based on black wattle tannins, which are used commercially, have solids content close to 50% (Santiago et al., 2018).

The result of the gel formation time, which indicates the reactivity of the tannin molecules with formaldehyde, showed high reactivity. In a study by Santiago et al. (2018), the average gel time of an adhesive with tannins from *A. mearnsii* was 297.48 s, which is faster than we found. According to Carvalho et al. (2014a), these differences are important since they can positively or negatively affect the bonding process by influencing the operational time of using these tannic extracts for bonding at the industrial level.

During the manufacture of wooden panels, drying of the pieces is one of the fundamental processes for successful production. According to Amer et al. (2019), moisture of wood directly influences its elastic properties. In addition, it can cause internal stresses in the piece and make it difficult to anchor the adhesives in the wood cells. In the present study, the moisture content was below the fiber saturation point, to obtain a product with low dimensional variation (Faria et al., 2019) and better bonding characteristics.

The apparent density of 579 kg m-3 determined for the wood used in this study was higher than that reported by Talgatti et al. (2018), who assessed this parameter of six eucalyptus clones, finding a mean value of 538 kg m-3. The authors also reported that apparent density is a complex character that varies according to age, species, spacing, and planting location. It is also important to note that density directly influences the penetration of adhesives in the wood, so that wood with lower density can result in a hungry glue line, while denser wood can make penetration difficult. The water absorption periods tested in this study indicated that the longer the period of immersion in water, the greater the absorption was. According to Ferro et al. (2018), the lower the density of the wood, the greater will be the water absorption, and as a result, the greater the likelihood of shrinking and swelling compared to denser woods. Another interesting factor observed by Silva et al. (2012) was that water absorption occurs more quickly in panels bonded with tannin-based adhesives than in panels bonded with PF adhesives, indicating that a stronger interaction between the surface of those adhesives and water occurs.

Janka hardness presented the highest variability (18.8%). The result obtained in this study of 44.3 MPa for Janka hardness was lower than that found by Araújo et al. (2012) for Eucalyptus spp., which was 62.70 MPa. However, this value may have been lower because the panel was subjected to high temperatures at the time of pressing since the authors also performed the hardness test on heat-treated wood at 180 ºC and obtained a result of 47.27 MPa, close to the value found in the present study. The shear strength of 1.06 MPa observed in our study is lower than the minimum requirement of ANSI/APA PRG 320 (APA, 2019), which is 1.30 MPa. The result was still lower than those found by Lu et al. (2018) testing CLT panels of a hybrid of Eucalyptus variously bonded with epoxy, isocyanate, phenol resorcinol formaldehyde, and a polyurethane component, in which the values found were 3.49, 3.01, 3.41 and 3.51 MPa, respectively. This unsatisfactory resistance of the glue line found here can perhaps be attributed to the fact that neither previous chemical treatment nor modification of the tannins was performed, suggesting that further studies should be conducted to improve the bonding quality of adhesives formulated with *M. tenuiflora* bark tannins.

Another issue to mention here is the possible negative effect of the high viscosity of the formulation synthesized in this work, which was 6,000 cP. Improper viscosity of adhesives can impair penetration in the wood. According to Vieira et al. (2014), the viscosity values of the tannin adhesives are usually high, because of the high molecular weight of the phenols of the tannin molecules and the molecular aggregates, formed by hydrogen bridges of the reactive condensed tannins. This may have contributed to increasing the viscosity of the *M. tenuiflora* adhesive. Azevêdo et al. (2015), also investigating the tannin-based adhesive of *M. tenuiflora*, found viscosity values ranging from 2,067 to 6,000 cP, similar to the value found in this study. So, in future studies dealing with adhesives formulated with *M. tenuiflora*, a reduction in the final viscosity of the adhesive is advisable. Sulfitation during the hot-water extraction step can be used for this purpose. According to Bianche et al. (2017), greater wood density means less porosity and hence less penetration of the adhesive in the wood structure. When the adhesive penetration is adequate, the contact surface area between adhesive and wood is also suitable for good mechanical anchoring of the adhesive (Frihart and Hunt 2010). Therefore, less viscous adhesives penetrate more deeply into the wood, so using wood from coniferous species is preferable since adhesion is more difficult in denser wood.

According to the ANSI/APA PRG 320 (APA, 2019) standard, the minimum values of MOE and MOR for CLT structural panels should not be less than 8,300 and 10 MPa, respectively. Thus, the value of MOE in the static bending test in this work is lower than that recommended by the standard (APA 2019), while the MOR value is adequate, but it did not satisfy any class established by the standard. Alencar and Moura (2019), in a study analyzing the structural performance of CLT panels made of Eucalyptus grandis and Pinus taeda L. wood using melanin-urea-formaldehyde as adhesive, obtained MOE of 10,270.1 MPa and MOR of 24.4 MPa for the eucalyptus panels, higher than we found. These differences can be related to defects in the wood and also the differences in the methods applied.

Although some results obtained were lower than those required by the standard, it was possible to identify the negative characteristics of the tannin-formaldehyde adhesive and to verify that the Eucalyptus spp. wood is not suitable for the production of CLT panels using this adhesive, because of the high density, which may have hindered the penetration of the glue in the wood pores, thus reducing the anchoring potential. Therefore, further studies are needed for the application of this adhesive in low-density wood and adjustment of its viscosity.

5. CONCLUSIONS

Bark tannins of *M. tenuiflora* are promising for bonding CLT and possibly other wood products. However, the mechanical properties of the CLT panels (MOE, MOR, and shear strength) were below the values required by the quality standards, most likely due to the high viscosity of the adhesive formulation and the high density of the eucalyptus wood used. Therefore, more research should be carried out to adjust the properties of the adhesive formulation based on this type of tannins.

AUTHOR CONTRIBUTIONS

BRFS: research project design, methodology design and execution, material and data collection, data analysis and interpretation, and writing; **JGMUF**: methodology design, data analysis and interpretation, and writing; **ECS**: data analysis and interpretation, and writing; **TLNC**: material and data collection; **TKBA**: research project design, methodology design and critical review; **FAM** and **ASP**: critical review.

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