

EVALUATION OF FUNGI TOXIC ACTIVITY OF TANNINS AND A TANNIN–COPPER COMPLEX FROM THE MESOCARP OF *COCOS NUCIFERA* LINN

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Abstract. The fibrous envelope of the coconut tree is an agroindustrial waste product that possesses relevant properties, such as a high resistance to biological degradation, because of its phenolic compound composition. In this study, the fungitoxic activity of tannins and a tannin–copper complex from the mesocarp of *Cocos nucifera* Linn was conducted. An extract from the coconut mesocarp was obtained in acidic media using a 2.0 wt % solution of NaHSO₃. Bioassays were conducted on alder wood (*Alnus acuminata* ssp. *glabrata* H.B.K.) using two types of aqueous solutions: 1) crude extract from the coconut mesocarp diluted at several concentrations (0.5, 1.0, 2.0, and 4.0 wt %); and 2) a tannin–copper complex formed in two stages by impregnation of the alder wood samples, first with the coconut mesocarp extract solutions followed by addition of a CuCl₂ solution. The fungitoxic capacity of the tannin and tannin–copper complex solutions was evaluated by means of bioassays with *Trametes versicolor* (L. ex. Fr.) Pilát according to ASTM D 1413-07. The bioassays showed poor fungal inhibition for the wood samples impregnated with the crude extract solutions. However, the tannin–copper complex solutions showed greater fungal inhibition. Retention values of 4.03 and 1.76 kg m⁻³ were obtained. These values revealed high fungal inhibition for *Trametes versicolor*.

Keywords: *Cocos nucifera* Linn, tannin, tannin–copper complex, decay, xylophagus fungi.

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INTRODUCTION

The understanding of the survival mechanism of trees in nature and attempting to mimic these processes is a topic of interest for most wood preservation specialists. It is highly desirable to overcome some of the triggers for wood deterioration such as exposure to outdoors, soil, high humidity, and micro-organisms. In nature, only a limited number of wood species can exhibit long-lasting durability. It has been reported that this capability is caused by wood extractives, mainly from heartwood, such as terpenoids with a small number of isoprene units, resinic acids, and phenolic compounds (Sjöström 1993; Binbuga et al 2008). Several authors stated that most extractives with antifungal properties appear to have monomeric structures (Malterud et al 1985; Hart 1989; Kai 1991). However, other wood extracts such as condensed tannins (Harun and Labosky 1985; Hemingway 1989; Eberhardt and Young 1994), elagic tannins (Hart 1989; Kai 1991), lignin (Fitzgerald and Line 1990), as well as elagic tannins and lignin interactions (Helm et al 1997), have also been reported as molecules with some antifungal activity. These polyphenolic compounds also have the ability to form metal complexes with cations such as copper, zinc, or chromium (Yamaguchi and Yoshino 2001; Ratajczak et al 2008), because of their catechol structure (Bariska et al 1986; Pizzi et al 1986; Scalbert et al 1998; Humar et al 2002).

In literature, there are several evaluations of the antifungal effects of extracts from long-lasting woods. The existence of biocides made from wood extracts and copper salts is also reported. For example, Smith et al (1989) reported that black locust heartwood extracts had the same antifungal resistance as pentachlorophenol, and Kamdem (1994) found that wood extracts from *Robinia pseudoacacia*, *Maclura pomifera*, and *Intsia bijuga* exhibited high resistance to fungi stasis. Compounds identified in these wood samples were robinetin and dihydrorobinetin (Hart 1989), penta- and tetra-hydroxystilbenes (Wang and Hart 1983), as well as condensed tannins (Kamdem 1994). Laks et al (1988) reported that extracts from Loblolly pine (*Pinus*

taeda) complexed with copper (II) ions showed good antifungal activity against *Gloeophyllum trabeum* but had considerably less preservation effect for wood decay caused by white-rot fungus *Trametes versicolor*. Similar results were reported by Pizzi et al (1986), who evaluated complexes obtained from copper and chestnut extracts.

The effect of condensed tannins as natural wood preservatives is a widely studied field. It has been reported that the remarkable fungi static activity shown by these compounds is directly related to the ability of condensed tannins to form complexes. Several theories have been proposed, including tannin complexation with proteins (Laks et al 1988) and tannin complex formation with ions capable of decreasing the iron available for micro-organisms (Scalbert 1991). Some authors have reported strategies to improve the antifungal efficiency of tannins, ie modification of the tannin structure with sulphites to obtain a more reticulated copper-tannin complex (Laks et al 1988), control of the tannin-metal complex size by adjusting pH in the reaction media (McDonald et al 1996), use of resorcinol and trichloroacetic acid to modify the tannin structure and improve the antifungal efficiency of such chemically modified tannin complexes (Yamaguchi and Yoshino 2001), and synthesis of polymerized tannin resin-boric compounds by mixing tannins with boron and hexamine (Thévenon et al 2010).

Copper has been widely used for tannin complexation. It has proven efficiency as a wood preservative. However, copper and tannin-copper complex concentrations in treated woods are decreased by copper lixiviation (Stilwell and Gorny 1997; Freeman and McIntyre 2008). Recently, several strategies used against copper lixiviation were proposed: formulation of novel amine-copper complexes (Zhang and Kamdem 2000; Humar et al 2002, 2005, 2007) and the synthesis of water-soluble quelates from ions reacted with carboxylic acids, natural and synthetic amino acids, hydroxy acids, peptides, acetylacetone, and sodium tripolyphosphate (Mamers and McCarthy 1998).

In Mexico, coconut mesocarp is an abundant byproduct from copra. This byproduct has 29.2 wt % lignin (Hart 1989) and 12.9 wt % tannin content (Gutierrez 1992). A remarkable property of coconut mesocarp is that when left outdoors, it is resistant to biodegradation. The aim of this study was to evaluate coconut mesocarp extract as a fungitoxic agent. The effects of crude extract and copper salt complexes, retention levels, and fixation of specific compounds into test specimens of alder wood are reported.

MATERIALS AND METHODS

Isolation of Tannins

Coconut mesocarp was cut in small pieces and milled in a Wiley mill (Wiley Retsch GmbH, Haan, Germany; Model 5657) to pass a 40-mesh screen. The obtained material was dried in an oven at 60°C to constant weight. Mesocarp extract was obtained using a 2 wt % aqueous solution of NaHSO₃ (J.T. Baker, analytical grade) in an autoclave at 120°C, for 60 min, at a 10:1 solution-to-mesocarp ratio. The extracted material was concentrated in a rotary evaporator (Büchi Labortechnik AG, Flawil, Switzerland;

Model R-15) at 40°C and 73.15 kPa. Residual humidity was eliminated from the sample by means of spray drying (Niro atomizer, BSLA; GEA Process Engineering, Columbia, MD). Tannins were isolated by solubilizing them with ethanol (J.T. Baker, analytical grade).

Solutions Used for Bioassays

For the bioassays, two types of aqueous solutions were prepared. The first was made from tannins with 0.5, 1.0, 2.0, and 4.0% (w/v) concentrations, and the second was made of CuCl₂·2H₂O ("ANALIT", reactive grade) at 0.25, 0.50, 1.0, and 2.0% (w/v). Each solution used was labeled, and the codes used are presented in Table 1.

Test Specimen Preparation

Bioassays test specimens were made from alder wood (*Alnus acuminata* ssp. *glabrata* H.B.K.) because of its high susceptibility to fungi attack (Lomelí and Fuentes 1997). Alder wood test specimens were cut into 20-mm cubes. All test specimens were dried at 60°C until constant weight. Wood test specimens were used as follows: eight for bioassays, four for lixiviation

Table 1. Soil block tests using tannins, copper salt, and tannin–copper complexes with the white-rot fungus *Trametes versicolor*.^a

Treatment	Key	Conc. in solution (g·L ⁻¹)		Mean retention ^b (kg·m ⁻³)		Weight loss ^c (%)
		Tannins	Copper salt	Tannins	Copper salt	
Tannins	T1	0.5		2.02 (0.06)		71.04 (3.20)
	T2	1.0		3.41 (0.13)		61.62 (4.15)
	T3	2.0		7.00 (0.24)		57.71 (4.66)
	T4	4.0		13.76 (0.38)		57.09 (5.07)
Cu Cl ₂	C1		0.25		0.91 (0.02)	16.15 (1.26)
	C2		0.5		1.42 (0.03)	13.68 (1.52)
	C3		1.0		3.22 (0.08)	11.97 (0.46)
	C4		2.0		6.84 (0.19)	14.24 (0.73)
Tannins + Cu Cl ₂	TC1	0.5	0.25	2.11 (0.07)	0.83 (0.02)	14.33 (1.01)
	TC2	1.0	0.5	4.03 (0.16)	1.76 (0.07)	11.81 (0.27)
	TC3	2.0	1.0	7.30 (0.29)	2.98 (0.14)	12.14 (0.29)
	TC4	4.0	2.0	14.03 (0.42)	5.17 (0.21)	13.75 (0.90)
Controls	T					73.00 (4.98)

^a Numbers in parentheses represent 1 standard deviation.

^b Average of 16 replicates.

^c Average of 8 replicates.

tests, and four as controls. The same number of evaluations was used for wood samples treated with tannin only, copper solution, and tannin–copper complex.

Specimen Impregnation with Tannin Extract

Test specimens were impregnated with tannins extract in one step using two vacuum cycles of 30 min at 39.9 kPa. After the impregnation treatment, the excess solution was removed by drying the wood samples, under controlled conditions, for 21 da. Finally, test specimens were dried at 60°C until constant weight. The impregnated cubes were divided in three sets, one for bioassays, one for blanks, and the other for copper complexing for the fungal inhibition studies.

Tannin–Copper Complex Formation

The tannin–copper complex was obtained as follows. Wood specimens were impregnated with the tannin extract solution and dried as previously described. Then, the treated samples were impregnated with $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ solutions using two vacuum cycles of 30 min at 39.9 kPa. This process allows the formation of the tannin–copper complex. Finally, wood specimens treated with the tannin–copper complex were dried until constant weight.

Leaching Tests

The amount of biocide leaching in the specimens was evaluated according to AWWA (2006). Treated wood samples were impregnated with distilled water using a vacuum cycle of 30 min and 39.9 kPa. After treatment, specimens were submerged in water for 2 wk. The water was changed every 24 h.

Bioassays

The bioassays on the treated alder wood samples were made using a white-rot fungus, *Trametes versicolor* (L. ex. Fr.) Pilat = (*Coriolus versicolor*). The strain (CFNL 01760) was kindly provided by the Forestry Department of the

Autonomous University of Nuevo León, México. The fungus was propagated on malt extract agar at $27 \pm 1^\circ\text{C}$. Five hundred-milliliter glass jars half-filled with garden soil were used as decay chambers. Moisture content of the soil was adjusted to about 130% by adding distilled water. Then the decay chambers were sterilized at 121°C, 103 kPa gauge, for 30 min. The chambers were cooled to room temperature. After cooling, the fungus test was inoculated and the decay chambers were incubated in the dark at $27 \pm 1^\circ\text{C}$ and 70% RH for 21 da. Finally, one cubic specimen per decay chamber was placed and incubated in the same conditions for 12 wk. The degree of attack caused by the fungus on the wood specimens was determined by weight loss according to ASTM (2007).

RESULTS AND DISCUSSION

Chemical Analysis of Coconut Extract

Crude extracts content of coconut mesocarp extract was evaluated according to the skin by hide powder method of the American Leather Chemist Association (ALCA 1970). Chemical composition of the mesocarp extract was 42.0 wt % tannins, 49.0 wt % nontannins, and 9.0 wt % insoluble material. Other determinations were ash content (27.5 wt %) and extract yield (13.5 wt %) weight of material. The tannin content in coconut mesocarp extract was higher than the value reported by Tamolang (1976), who analyzed tannin and nontannin content in a hot water extract from coconut coir dust. He found that the concentrations in the coconut coir dust were 28.46% for tannins and 59.72% for nontannin compounds. Also, Tamolang (1976) reported that tannin and nontannin concentrations in the coconut coir dust corresponded to a ratio of 0.5 and was lower than the mimosa tannin extract (ratio of 2.5). Our results show that, with the use of a 2 wt % solution of NaHSO_3 as a solvent, the tannin/nontannin ratio in the coconut mesocarp extract was 0.876.

Tamolang (1976) also stated that tannins extracted from coconut coir were compounds related to catechol-type substances, and according to Bariska

et al (1986), Scalbert et al (1998), and Humar et al (2002), the catechol structure gives the tannins the ability to form metal complexes with various cations.

Wood Specimen Impregnation

In general, all solutions of the different treatments exhibited extensive penetration into the wood. This was confirmed by visual inspection of the alder wood specimens. As shown in Table 1, retention rate was proportional to the applied solution concentration. This trend was observed in all wood specimens impregnated with either the aqueous tannin extract or the tannin–copper complex solution. Both kind of samples reached similar retention values (about $14.0 \text{ kg}\cdot\text{m}^{-3}$).

Bioassay Analysis

The bioassays made on the alder wood specimens impregnated with the tannin extract exhibited poor antifungal effect (Table 1). This behavior is similar to the results reported by Laks et al (1988), who found that extracts from loblolly pine bark did not decrease wood decay caused by white-rot fungi *Trametes versicolor*. The results shown in Table 1 indicate that, at a retention rate of $2.0 \text{ kg}\cdot\text{m}^{-3}$, weight loss of the wood samples was very similar to the untreated wood specimens. The greatest antifungal effect within the wood samples impregnated with the tannin extract was reached on the samples treated with a 4.0 wt % solution. The weight loss of the wood specimen treated with the latter solution was decreased only about 22.0% compared with the untreated samples. However, this effect can be considered very poor if compared with the decrease in weight loss reached by the specimens treated with the tannin–copper complex (about 80%).

The small inhibition exhibited by coconut mesocarp extract may be related to issues such as diffusion, selectivity, and purity of the substances contained. Kai (1991) reported that the antifungal effect of the extracts from the cell lumen was smaller than the effect within the cell

wall through which the fungus hyphae must diffuse. It is possible that the treatment applied to the extract was inadequate to impregnate the wood cell walls and was only distributed within the pores and lumens. Conversely, some authors stated that the polyphenol selectivity to fungal inhibition has been linked to factors such as molecular weight, chemical structure of enzyme, and polyphenol origin, among others (Harun and Labosky 1985). Chemical analysis of the mesocarp extract revealed that about half the content was nonphenolic. Thus, it cannot inhibit fungal growth and might require a purification process to show a greater inhibitory effect.

Laks et al (1988) reported that when wood was pressurized with *Pinus taeda* bark extracts, weight loss of test specimens and deterioration caused by attack of micro-organisms were not prevented. In this study, when wood treatment was done in two stages (coconut mesocarp extract + $\text{CuCl}_2\cdot 2\text{H}_2\text{O}$ aqueous solutions), the inhibitory effect for fungus growth was drastically increased. The greatest inhibitory effect was achieved in the wood samples treated with solutions of 1.0 wt % tannin extract and 0.5 wt % copper (Fig 1). Weight loss was 11.81 wt % (control-corrected, 0.49 wt %). Wood specimens treated under such conditions exhibited retention values of $4.3 \text{ kg}\cdot\text{m}^{-3}$ for tannins and $1.76 \text{ kg}\cdot\text{m}^{-3}$ for CuCl_2 . This behavior is in accordance with reports published elsewhere about the enhancement of wood preservation by promoting the formation of insoluble complexes of tannins and copper salts (Bariska et al 1986; Humar et al 2002).

Table 1 also shows the inhibitory effect obtained in the wood specimens treated only with $\text{CuCl}_2\cdot 2\text{H}_2\text{O}$. As reported, maximum wood protection was achieved when a 1.0 wt % copper (II) solution was used. Average weight loss in wood specimens treated with such solution was 11.97 wt % with a retention value of $3.22 \text{ kg}\cdot\text{m}^{-3}$. As mentioned before, similar weight loss was obtained in the wood samples treated with the tannin–copper complex. However, the retention value for copper salt in wood specimens impregnated with the complex was about half that of

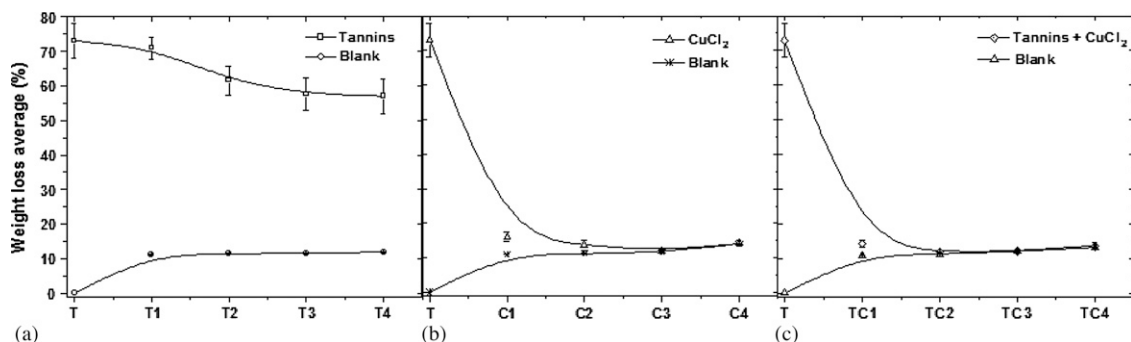


Figure 1. Weight loss average of wood specimens exposed to white-rot fungus *T. versicolor* and treated with solutions of (a) coconut extract, (b) copper salt, and (c) tannin-copper complex. For each solution, different concentrations were evaluated.

copper-only-treated wood. The latter observation suggests a synergistic effect in the tannin-copper complex to prevent fungi growth.

Figure 1 shows weight loss of wood specimens exposed to *T. versicolor* as a function of concentration of different solutions tested. As shown in Fig 1, when the concentration of each solution increased, average weight loss in evaluated wood specimens decreased. Wood samples treated with tannins (Fig 1a), copper (Fig 1b), and tannin-copper complex (Fig 1c) solutions exhibited greater protection against wood decay. The latter solution exhibited a similar preservative effect to the copper-only solutions but with less copper concentration.

Solution Leaching Analysis

Loss of preservative in the wood specimens caused by lixiviation was evaluated. Figure 2 shows leachate behavior of the tannin, copper, and tannin-copper complex solutions in the treated wood specimens. As illustrated, wood samples treated with copper solution exhibited the lowest retention values. These samples also present the greatest rate of leachate accumulation. As reported by others, copper is a good wood preservative agent. However, it has poor fixation in the treated woods (Laks et al 1988; Mamers and McCarthy 1998; Scalbert et al 1998; Humar et al 2007; Stevanovic et al. 2001; Townsend et al. 2003; Robinson et al 2006). Similar behavior

was exhibited by wood samples treated with coconut extract. These solutions also showed very poor capacity to interact with the cell wall in treated wood specimens. Conversely, the solutions of the tannin-copper complex showed a different behavior. Wood samples treated with the tannin-copper complex solutions had good retention values and the lowest amount of leachate. This trend indicates that there was a synergistic effect exhibited by the tannin-copper complex, which allowed fixation of the preservative agent to the wood. This behavior is in agreement with Scalbert et al (1998), who evaluated the fixation ability of tannins from

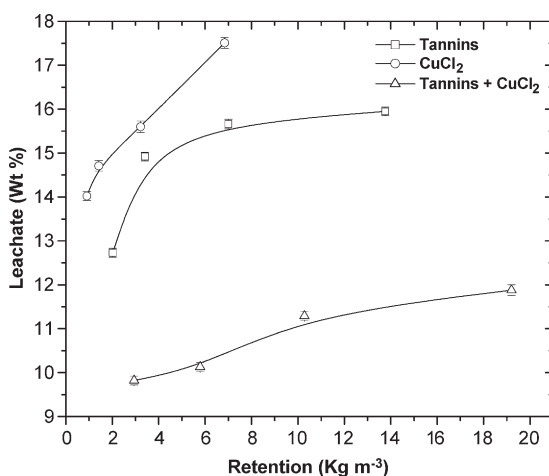


Figure 2. Leachate as function of retention values of coconut extract, copper, and tannin-copper complex used as wood preservatives in alder wood specimens.

chestnut and a tannin–copper complex. They reported an increase in retention for the tannin–copper complex of up to 85% and stated that such fixation was caused by formation of a water-soluble complex.

CONCLUSIONS

In this study, the tannin extract obtained from the coconut mesocarp showed a poor inhibitory effect against growth of the *Trametes versicolor* white-rot fungus. It could require greater retentions to prevent fungus growth, which could render it unprofitable.

The fungus tested was intolerant to copper (II) salts. However, when the tannin extract from the coconut mesocarp was combined with CuCl_2 solutions, a synergistic effect was observed, promoting good levels of wood preservation. The greatest inhibitory effect was achieved in wood samples treated with solutions of 1.0 wt % tannin extract and 0.5 wt % copper.

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