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Original article

Evaluation of decay resistance of wood products made from borax-impregnated wood and bonded with a formaldehyde-free cornstarch and tannin adhesive

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Abstract

• At present, the production of wood composites mainly relies on the petrochemical-based and formaldehyde-based adhesives such as phenol-formaldehyde (PF) resins and urea- formaldehyde (UF) resins, which are non-renewable and therefore ultimately limited in supply.

• This paper concerns the decay resistance of wood products bonded with a new, environment-friendly adhesive derived from abundant and renewable cornstarch and tannin. To improve the total resistance of the composite against both *Coriolus versicolor* and *Coniophora puteana* rot fungi, borax (di-sodium tetraborate) was added in proportions of 0.5%, 1% and 2% (w/w) to the cornstarch-tannin adhesives.

• The results show that increasing the concentration of borax in the adhesive decreased the mechanical properties of the composite. The best way to avoid this problem was to use wood impregnated with borax.

• Biodegradation studies were conducted on new composites, first without any treatment, followed by borax at 0.5% aqueous solution treatment. The results show that wood impregnated with borax, in the presence of tannin and sodium hydroxide in the adhesive improves the total resistance of the wood composite against both *Coriolus versicolor* and *Coniophora puteana* rot fungi.

Mots-clés :
borax /
amidon de maïs /
durabilité /
tannin /
adhésif

Résumé – Évaluation de la durabilité des produits en bois imprégnés avec du borax et collés avec un adhésif naturel à base d'amidon de maïs et de tannin.

• Actuellement, la production des composites à base de bois nécessite l'utilisation d'adhésifs d'origine pétrochimique et à base de formaldéhyde, tels que le phénol-formaldéhyde (PF) et l'urée-formaldéhyde (UF), qui sont non renouvelables et limités dans l'approvisionnement.

• Ce travail a pour objectif d'étudier la durabilité du bois collé avec une nouvelle colle naturelle à base d'amidon et tannin. Pour améliorer la résistance des composites vis-à-vis des deux types principaux de dégradation fongique, *Coriolus versicolor et Coniophora puteana,* nous avons ajouté à l'adhésif, différentes concentrations de borax (di-sodium tetraborate) 0,5 %, 1 % et 2 % (m/m).

• Les résultats obtenus montrent que l'augmentation de la concentration du borax dans l'adhésif diminue les performances mécaniques des composites. La meilleure façon d'éviter ce problème est de traiter le bois avec du borax, avant collage.

• L'étude de la biodégradation a été réalisée sur les nouveaux composites, d'abord sans traitement, et après traitement à 0,5 % (m/m) avec du borax. Les résultats montrent que le bois traité au borax associé à la présence des tannins et de NaOH dans l'adhésif améliore la résistance totale des composites vis-à-vis de *Coriolus versicolor* et *Coniophora puteana.*

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1. INTRODUCTION

Formaldehyde-based adhesives such as phenol-formaldehyde (PF) and urea-formaldehyde (UF) resins presently predominate in the production of wood composites. In 1998, according to the European Panel Federation (EPF), the quantity of the adhesives used in Western Europe was estimated at approximately 3.4 million tons (Mansouri and Pizzi, 2007; Mansouri et al., 2006). Formaldehyde can be emitted in the production and use of wood products bonded with UF or PF resins. These problems push the wood industry to investigate in the framework of new environment-friendly adhesives derived from renewable resources.

Starch is the naturally occurring polymer of D-glucose stored in granules as a food source in most plants. Its low cost and versatility for chemical manipulation makes it an attractive material for use as a substitute for synthetic polymers (Imam et al., 2001). Starch has been used as an adhesive in a wide range of products, including binders, sizing material, glues and pastes (Imam et al., 1999). More recently, the development of a starch-based wood adhesive in interior applications has been described (Imam et al., 1999). Starch yields adhesives with excellent affinity for polar materials such as cellulose. Ideally, the contact angle between the adhesive and the substrate should be small. This allows the adhesive to wet the surface and spread uniformly in a thin film with a minimum of voids. In this regard, starch-based adhesives wet the polar surface of cellulose, penetrate crevices and pores and, thus, form adhesive bonds. The bonding is the result of both mechanical interlocking and Van der Waals forces (Imam et al., 1999).

Tannin is another example of such a renewable material. These tannins, consisting of flavonoid units which have undergone varying degrees of condensation, could react with hexamethylenetetramine (hexamine) as a hardener, hence tannin-hexamine adhesives can be used as wood adhesives. ^{13}C-NMR has confirmed (Kamoun et al., 2003; Pichelin et al., 1999) that in the presence of chemical species with very reactive nucleophilic sites, such as condensed flavonoid tannins, hexamine is not at all a formaldehyde-yielding compound. The very reactive imines and iminoaminomethylene intermediates initially formed in its decomposition do react with the phenolic species present without ever passing through the formation of formaldehyde (Kamoun et al., 2003; Pizzi and Tekely, 1995; 1996; Pizzi et al., 1996).

In this work we particularly studied the decay resistance of a wood composite treated with 0.5% of borax and bonded with the cornstarch-tannin adhesive against the rot fungi *Coriolus versicolor* and *Coniophora puteana*.

2. EXPERIMENTAL METHODS

2.1. Materials

Unmodified commercial grade cornstarch (extra pure) was obtained from ACROS ORGANICS; the moisture content was in the range of 10 to 12%. Sodium hydroxide (M_w = 40.00 g/mol) was purchased from VWR Prolabo. Commercial flavonoid mimosa tannin (Mimosa OP) was provided by SILVATEAM. The hardener, hexamethylenetetramine 99% (hexamine), was supplied by Aldrich. Borax (di-sodium tetraborate) was used as the boron compound. It was purchased from Fisher Scientific.

Maritime pine (*Pinus pinaster*) veneer was provided by the sawmill Ets. Labadie (Roquefort, France) Sd (moisture content around 8–10%).

2.2. Preparation of cornstarch-tannin adhesives

To prepare 500 g of adhesive, cornstarch water solution was prepared at 65% (p/v) concentration, by dissolving 130 g of cornstarch in 200 mL of deionised water and stirring at room temperature, to which 13 g of mimosa tannin were added. The hardener content used was 5% hexamine by weight on tannin extract solids content. The hexamine was dissolved in water to yield a 30% concentration solution in water before being added to the cornstarch-tannin solution. The solution was mixed and 100 mL of sodium hydroxide (33%) was added. The resulting adhesives were mixed for 45 min at room temperature then used to bond wood veneer.

2.3. Two-ply wood composites' preparation and testing

Maritime pine veneer with a thickness of 3 mm was cut into rectangular specimens 2.5 × 11.5 cm^2 (according to British Standard 1204, 1965, part 2, for synthetic resin adhesives). The adhesive was applied to one side of each piece of veneer. The application area was 2.5 × 2.5 cm^2 for each veneer. The spread rate of cornstarch-tannin adhesive was 120–150 g/m^2 on a dry weight basis (Nihat et al., 2002; Pizzi, 1977). Two adhesive-coated veneer boards were lapped together with the grain parallel to each other and then pressed at 0.45 MPa (Fig. 1). The press temperature and press time were fixed at 170 °C and 4 min, respectively. After bonding, the assemblies were conditioned in a Vötsch climate room (25 °C and 65% relative humidity) for 24 h. Seventy samples were prepared for the mechanical tests.

2.4. Evaluations of mechanical properties of the two-ply wood composites

The shear strength of the two-ply wood composites was determined on an Instron testometric M500-50 AT testing machine. The crosshead speed was 1 mm/min. The load at fraction was measured, and reported as shear strength. All the experiments were repeated; at least ten replicates and their standard deviations were calculated.

2.5. Biological properties

The determination of biological durability of wood is an issue requiring sufficient reliability regarding end-use-related prediction of performance.

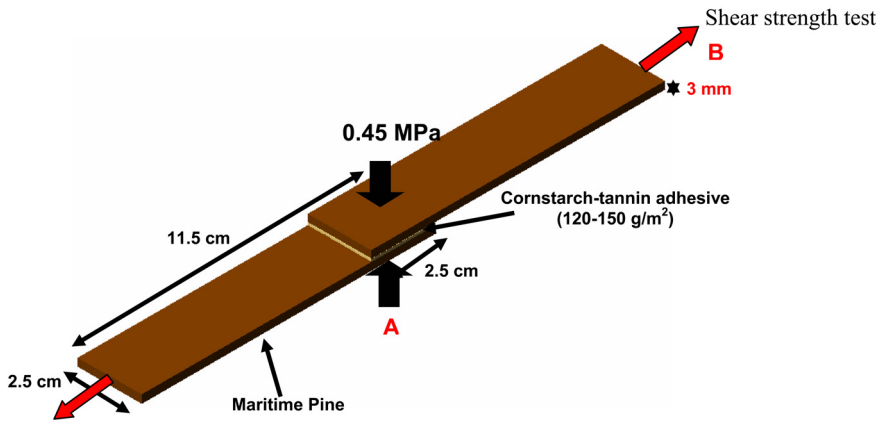

Figure 1. Schematic representation of wood sample preparation using cornstarch-tannin crosslinked adhesives. Step A: pressure at 170 °C, 4 min and 0.45 MPa; step B: shear strength test after 24 h in Vötsch climate room (25 °C and 65% humidity).

2.5.1. Decay test of the two-ply wood composites

Two-ply wood composites measuring 25 × 25 × 3 mm^3 were prepared for a decay test, from Maritime pine (*Pinus pinaster*). The wood was impregnated in laboratory conditions with borax at 0.5% (w/w) distilled water solution in a small-scale impregnation container, applying 0.7 kPa vacuum for 15 min. After the treatment the wood samples were removed from the treatment solution, and wiped gently to remove the rest of the solution from the wood surface. The samples were conditioned at 25 °C and 65% relative humidity for four weeks prior to decay testing. A fungal decay test was done according to an adaptation of the norm NF EN 113 (AFNOR 1996) using a brown rot fungus, *Coniophora puteana* (BAM Ebw.15) and the white rot fungus, *Coriolus versicolor* (CTB 863A). A culture medium was prepared in Roux flasks. In each one, a nutritive medium, made up of a malt-agar mixture (40 g of malt and 20 g agar dissolved in 1000 mL distilled water), was placed. After sterilisation (121 °C for 20 min at 1 bar), the *Coriolus versicolor* and *Coniophora puteana* were inoculated on culture medium in Petri dishes under sterile conditions. Then they were placed in constant conditions of 25 °C in 65% relative humidity for 3 weeks to favour the fungal development. After being conditioned at 25 °C and 65% air moisture content, the two-ply wood composite samples were put into Petri dishes and exposed to *Coriolus versicolor* and *Coniophora puteana* for 16 weeks in darkness. Finally, test results were expressed as percentage of weight loss of composite panels due to fungal attacks after decay test using Equation (1) (The measuring was done after the drying process of each sample). Ten replicates were used for each decay fungus.

$$\text{Weight loss}(\%) = \left(\frac{m_i - m_f}{m_i}\right) \times 100 \qquad (1)$$

where m_i and m_f are the oven-dry weights of the sample before and after the decay test.

2.5.2. Decay test of wood adhesives

In order to confirm the fungicidal effect of cornstarch-tannin adhesives, we added polymerised adhesive to the culture medium. Each polymerised adhesive was ground and mixed at different concentrations (from 0 to 7%, p/v) in malt-agar culture medium. After sterilisation, the *Coriolus versicolor* and *Coniophora puteana* were inoculated in the centre of the culture medium under sterile conditions. Then they were placed in constant conditions of 25 °C in 65% relative humidity for 10 days to favour the fungal development. The mean radius of the fungal development was measured. Test results were expressed as percentage of fungal development using Equation (2). Ten replicates were used for each fungal development.

$$\text{Percentage of fungal development}(\%) = \frac{R_1}{R_0} \times 100 \qquad (2)$$

where R_1 is the mean radius of the fungal development after 10 days (adhesives present in Petri dishes); R_0 is the mean radius of the fungal development after 10 days (control: without the presence of adhesive).

3. RESULTS AND DISCUSSION

Biocides intended for indoor applications must be non-toxic, non-volatile, odourless, hypoallergenic, and able to provide long-term protection under conditions of high humidity. In this work we used borax (di-sodium tetraborate).

3.1. Effects of borax concentration on shear strength

First of all, in order to improve the total resistance of the two-ply wood composites, we added borax in proportions of 0.5%, 1% and 2% to the cornstarch-tannin adhesives. The results (Tab. I) show that the mechanical properties (stress at the crack sample) of cornstarch-tannin adhesive decreased significantly when the concentration of borax was increased. This may have been due to a decrease in pH value of the adhesive or to an effective lowering of the molar ratio due to the presence of reactive material, like the tannin (Gökay et al., 2006), or to the well-known marked effect of decrease in viscosity that borax has in all starch adhesives (Custers et al., 1979). The test revealed that in most cases, the rupture is not adhesive but

Table I. Effect of borax concentration on shear strength of plywood composite bonded with formaldehyde-free cornstarch-tannin wood adhesives. Ten replicates were used for each mechanical test. SD: Standard deviation.

Type of treatment	Borax concentration, %	Shear strength, MPa Mean ± SD	Veneer failure, % %
Treatment of cornstarch-tannin adhesives (untreated wood)	0	5.22 ± 0.06	85
	0.5	4.70 ± 0.15	75
	1	4.50 ± 0.19	50
	2	4.07 ± 0.16	50
Treatment of wood (untreated adhesives)	0	5.22 ± 0.06	85
	0.5	5.19 ± 0.08	85
	1	5.11 ± 0.17	75
	2	5.07 ± 0.10	62

Table II. Weight loss of wood composites pretreated with borax after exposure to both decay fungi (*Coriolus versicolor* and *Coniophora puteana*) for 16 weeks. Ten replicates were used for each decay fungus. SD: Standard deviation.

Treatment	Weight loss (%)	
	Coriolus versicolor Mean ± SD	*Coniophora puteana* Mean ± SD
Control, Maritime pine (untreated)	38.0 ± 5.2	43.2 ± 3.7
Maritime pine (treated; 0.5% (w/w) borax)	6.6 ± 0.7	10.1 ± 3.4
Plywood (untreated wood and cornstarch-tannin adhesives)	23.5 ± 4.6	27.3 ± 5.0
Plywood (treated wood (0.5%) and urea-formaldehyde adhesives)	6.1 ± 2.3	7.6± 0.6
Plywood (treated wood (0.5%) and cornstarch-tannin adhesives)	7.0 ± 1.6	9.3 ± 2.5

interfacial and more precisely in the wood side. On the other hand, the impregnation of wood with borax does not have an effect on the mechanical properties of composites (Tab. I). The samples made from wood impregnated with borax at 0.5% and 1% (w/w) aqueous solution have the same shear strength values.

Wood impregnated with borax at 0.5% (w/w) was selected for two reasons. Firstly, the effectiveness threshold of borax against decay fungi *Coriolus versicolor* and *Coniophora puteana* attacks is 0.26% (Drysdale, 1994; Knudson, 1990). Secondly, the cornstarch-tannin adhesive is intended for indoor applications; hence, a relatively lower retention load is needed.

3.2. Decay resistance of the two-ply wood composites

Weight loss of the two-ply wood composites caused by fungal decay after 16 weeks is shown in Table II. We can see that the untreated maritime pine wood was attacked by both decay fungi (*Coriolus versicolor* and *Coniophora puteana*) with weight loss close to 40%. This proves that the fungus was virulent and the test of durability was valid according to the relevant European Norm EN 113 (EN 113, 1996). The values of weight loss for all treated specimens decreased. Laboratory tests have shown that 0.5% (w/w) borax treatment of wood is very effective in preventing attack by both decay fungi. The results in Table II seem to indicate that *Coniophora puteana* could be more efficient in wood degradation than *Coriolus versicolor*. This tendency has been observed by many authors (Nemli et al., 2006; Pandeya and Pitmanb, 2003). This may be due to the differences between their decay mechanisms. One of the distinguishing biochemical features of brown rot fungi is that many brown rot fungi produce oxalic acid. The production of oxalate by brown rot fungi has been investigated by many authors (Bech-Anderson, 1987; Eduardo and Eduardo, 1991; Green and Clausen, 2003; Illman et al., 1988; Sutter et al., 1983) reported that since lignin and cellulose are relatively resistant to acids, while hemicellulose hydrolyses fairly easily in acid conditions, oxalic acid is the agent by which brown rot fungi hydrolyse hemicelluloses and increase the accessibility of cellulose to wood decay enzymes and yield water-soluble sugars, leaving only a lignin skeleton. Dutton et al. (1993) found that white rot basidiomycete *Coriolus versicolor* also produced oxalate in liquid culture; however, little oxalate was accumulated during active growth. In addition, they reported that oxalate accumulation did not lower pH of the medium during growth, unlike brown rot fungi. The degradation of cellulose, hemicelluloses and lignin polymers is also due to extracellular enzymes. Cellulases and hemicellulases are involved in polysaccharide degradation, whereas peroxidases and laccases are mainly devoted to lignin breakdown, the production of these extracellular enzymes being regulated essentially at the transcriptional level (Cullen and Kersten, 2004). We also observed that the two-ply wood treated (0.5% borax) and glued with cornstarch-tannin adhesives showed considerable resistance to both the decay fungi compared with that of the control two-ply wood treated (0.5% borax) and glued with urea-formaldehyde adhesives. The two-ply untreated wood glued with cornstarch-tannin adhesives showed considerable resistance to both types of decay fungi compared with untreated control specimens. This result may

Table III. Effect of cornstarch-tannin polymerised adhesive concentration on fungus development. Ten replicates were used for each fungus's development.

Cornstarch-tannin polymerised adhesive concentration	Percentage of fungus development, %	
	Coriolus versicolor Mean	*Coniophora puteana* Mean
0	100	100
2	19	22
5	1	7
7	0	1.6

indicate that cornstarch-tannin adhesive imparted further resistance to the two-ply wood composite.

3.3. Fungicidal effect of cornstarch-tannin polymerised adhesives

Table III shows the effect of cornstarch-tannin adhesive concentration on fungal development. A decrease in fungal development for both decay fungi *Coriolus versicolor* and *Coniophora puteana* is observed on incorporation of cornstarch-tannin adhesives into the culture medium. The results in Table III confirm the fungicidal effect of cornstarch-tannin adhesives, that contributes to improving the total resistance of the two-ply wood composites.

Hart and Hillis (1972) have demonstrated that the presence of tannin improves the durability of wood. Further studies have evaluated the fungicidal effect of tannin (Aloui et al., 2004; Charrier et al., 1995; Cornelius et al., 2004; Pizzi and Conradie, 1986; Zucker, 1983).

4. CONCLUSIONS

A cornstarch-tannin adhesive is easy to prepare. A simple mixing of a tannin solution, cornstarch, hexamine and NaOH for about 45 min provides a high strength wood adhesive. The results show that two-ply wood composite (wood treated (0.5%) and cornstarch-tannin adhesives) proved to be biologically resistant against decay fungi. Mass loss caused by decay fungi was 7.0 and 9.3% for *Coriolus versicolor* and *Coniophora puteana*, respectively. These levels are close to those specified as acceptable in relevant standards (according to hazard class 2). The environment-friendly cornstarch-tannin adhesive improves the total resistance of the two-ply wood composites.

In conclusion, according to our experimental conditions, wood veneers treated (0.5%) and glued with cornstarch-tannin adhesives may have sufficient properties to be used as wood panels for interior applications.

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