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Efficacy of wollastonite-enriched acrylic coatings against fungal decay in non-treated and thermally-modified wood

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Research Article

Keywords: Compression strength, Fungal resistance, Mass loss, Thermal modification, Wollastonite.

Posted Date: October 13th, 2026

DOI: https://doi.org/10.21203/rs.3.rs-11082162/v1

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Additional Declarations: No competing interests reported.

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Abstract

Wood exposed under adverse environmental conditions must be preserved to provide long service life. While there are a number of highly effective preservative systems, there is a need for more easily applied protective treatments. Thermal modification has some potential in this application, but it is not effective against all decay agents and needs supplemental protection. Wollastonite ($CaSiO_3$) is widely used in a variety of industrial applications including plastics, insulation, and metallurgy. It has also proven to be useful for improving the properties of cellulose-based composites. The potential for a wollastonite-enriched acrylic paint to protect non-treated and thermally-modified wood of three commercial wood species was evaluated against *Coniophora puteana* and *Trametes versicolor*. *C. puteana* caused greater mass losses in poplar and spruce while *T. versicolor* resulted in higher mass loss in beech. Wollastonite was associated with new ionic bonds, and created a physical barrier against hyphal penetration into the wood, ultimately preserving wood. It was concluded that wollastonite-enriched acrylic paint can provide an effective and easy-to-apply protection against both fungi.

***Keywords*:** Compression strength; Fungal resistance; Mass loss; Thermal modification; Wollastonite.

1. Introduction

As a biomaterial, wood is susceptible to degradation by many biological agents, including fungi and insects (Milton 1995; Wegner & Jones 2005; Teng et al. 2018; Piętka et al. 2026). It is also combustible, and vulnerable to both fire and high temperatures. Moreover, it is dimensionally unstable under changing moisture conditions (Tjeerdsma et al. 1998). However, wood is one of our few renewable construction materials and has remarkable properties that make it worth modifying to improve these properties. The most commercially-used non-biocidal modification method on solid wood is thermal modification (Tjeerdsma et al. 1998; Hill 2006; Boonstra et al. 2006a,b& 2007; Borrega M, Kärenlampi 2010). It is considered an environmentally-friendly modification method and therefore, it is preferred by some users over traditional chemical preservatives and chemical modification schemes such as acetylation and furfulysation (Cetin et al. 2009; Dong et al. 2015; de Medeiros et al. 2016; Lykidis et al. 2016; Papadopoulos et al. 2019; Xi et al. 2020; Antov et al. 2023; Kawalerczyk et al. 2023; Savov 2023). Thermal modification involves exposing wood to mild (lower than 190ºC) to rather high (higher than 200ºC) temperatures in different mediums (including atmospheric air, nitrogen, hot water, and water vapor) for predetermined durations and temperature regimes, so that the chemical structure of wood (mainly cell-wall polymers, including cellulose, hemicellulose, and lignin) is altered, or so-called modified (Tjeerdsma et al. 1998; Hill 2006; Boonstra et al. 2006a,b & 2007; Schwarze 2007; Borrega & Kärenlampi, 2010; Cetin et al., 2009; Hao et al. 2018). Identifying methods for reducing the temperatures employed in the thermal modification process while still improving durability would make these processes more attractive.

Thermal modification of wood involves heavy and expensive machinery. Therefore, presenting a low-cost and easy-to-apply method to protect wood against biological deteriorating agents would be highly demanded by the commercial sector. In this connection, different materials were tested, reporting significant improvement (Hanumantha et al. 1998; Parvez et al. 2014; Chan et al. 2020; Majidi et al. 2023). It has been reported that even small amounts of particular minerals (like wollastonite and clay) in epoxy coatings have the potential of making a significant improvement in the superficial behavior and prolonging the service life of the coating film (Aghazadeh et al. 2011; Shirehjini et al. 2016; Sheykhifard et al. 2018; Chan et al. 2020; Yang et al. 2024; Droß et al. 2025).

Wollastonite is another mineral which is considered a functional filler and have been repeatedly studied as reinforcing additives and extender to improve properties in different materials and polymer matrices to be substituted with more expensive reinforcement additive like glass fiber (Švab, 2007; Chan et al. 2020; Choo et al. 2022).

Previous research studies have shown the efficacy of wollastonite as an extender and reinforcing additive to improve shear strength in PVA resin (Antov et al. 2023), and as a fire-retardant to improve fire properties in acrylic paints as well (Kawalerczyk et al. 2023; Savov 2023). However, little study has been done on its potential in improving different types of paints. Based on the above, the objective of this study was to assess the effects of wollastonite-enriched coating on the decay resistance of both non-treated and thermally-modified timbers against two representative wood-decay fungi.

2. Materials and methods

2.1. Specimen preparation and heat treatment process

Beech (*Fagus orientalis* Lipsky), poplar (*Populus nigra* Lipsky), and European spruce (*Picea abies* L.H.Karst.) lumber were obtained locally. The densities for the three species were 0.64, 0.43, and 0.44 $g.cm^{-3}$, respectively, following methods described in ASTM Standard D2395-17.

Sixty defect free specimens (50 × 20 × 20 mm, longitudinal, radial, and tangential directions, respectively) were cut for each species and allocated to be either left non-treated or thermally-modified. Thermal modification was carried out at 185±4 ºC for four hours in a laboratory oven. At the end of the heating period, the specimens were slowly cooled. The specimens were conditioned for one month at 25±2 ºC, and 45% (±3) relative humidity.

The 30 specimens in each group of the unheated and the heat-treated specimens for each of the three wood species were allocated to be left uncoated, coated with an acrylic paint, or coated with a wollastonite amended coating, as described in Table 1. The coded specimens were exposed to either of the wood-decay fungus, namely *C. puteana* and *T. versicolor*. Once the exposure time was up and the mass losses were calculated, the compression strengths parallel to grain were measured on the fungal-exposed specimens.

Separate sets of extra specimens (four replicate specimens) were prepared for comparison, reported as the unexposed specimens.

105 **Table 1.** Description of the coding for each and every treatments in the three wood species,
106 (namely beech, poplar, and spruce) to be exposed for four months to either of the wood-
107 decay fungi (*Coniophora puteana* and *Trametes versicolor*)

Treatments for each of the three wood species (beech, poplar, and spruce)			Description of treatments
Treatments to be exposed to either of the two studied wood-decay fungi (*C. puteana* or *T. versicolor*)	Unheated	Control	Unheated uncoated specimens
		Painted	Unheated specimens coated with plain acrylic paint
		W-painted	Unheated specimens coated with wollastonite-added acrylic paint
	Thermally-modified	TM	Thermally-modified uncoated specimens
		TM-painted	Thermally-modified specimens coated with plain acrylic paint
		TM-W-painted	Thermally-modified specimens coated with wollastonite-added acrylic paint

108

109 The coated specimens with acrylic paint received two coats of ALCO-6510 water-
110 based acrylic paint applied 24 hours apart. The paint contained 38 % solids of which 32 %
111 was a binder mixture of containing methacrylic acid and methacrylate ester derivatives
112 (Alvan Paint and Coatings Inc., Tehran, Iran). The resulting dry coating averaged 210 µm
113 in thickness (SD 10 µm) as determined using Model 121/4 thickness gauge (Elcometer,
114 United Kingdom).

115 A wollastonite suspension (60% solids) prepared by Mehrabadi Mfg. Co. (Tehran,
116 Iran) was added to the acrylic paint at a 15:85 ratio of wollastonite:acrylic paint and
117 thoroughly mixed (Fig. 1). The system was applied in two coats 24 hours apart. Each

specimen received 3.5 g of coating. All of the specimens were then conditioned for four weeks at 25° C and 45 % relative humidity to a final average moisture content of 9% (SD= 0.5 %).

Five blocks from each treatment for each wood species were then exposed to either *Trametes versicolor* (L ex Fr.) Pilat (Isolate 325) or *Coniophora puteana* (Fr.) Karst (Isolate 167) in an EN-113 agar block test. Briefly, the test blocks were sterilized by autoclaving for 20 minutes at 121 °C. The specimens were exposed to either of the fungi species for four months at 25 °C. At the end of the test period, the blocks were removed, scraped clean of adhering mycelium and oven dried at 103 °C for 24 hours before being weighed to determine mass loss.

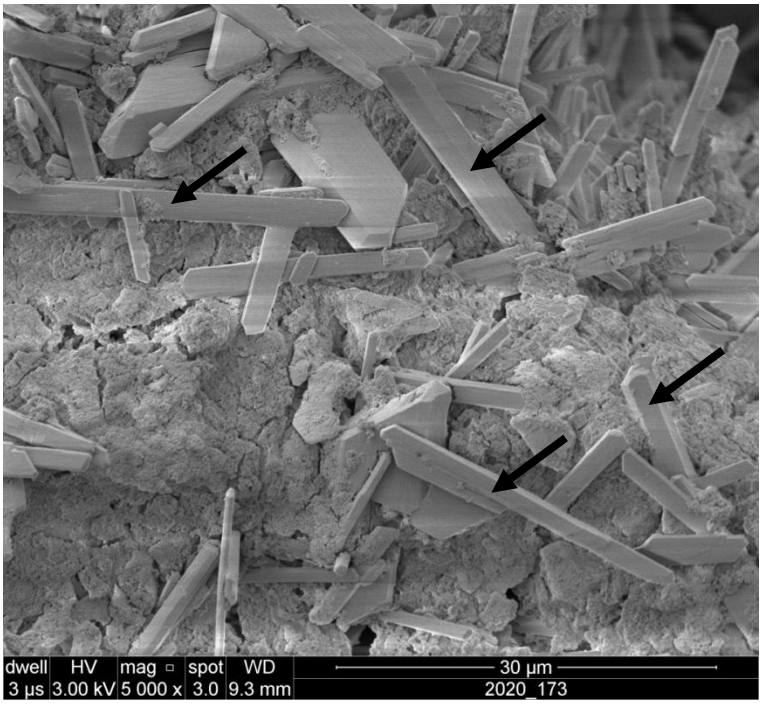


Fig. 1. SEM image of the wollastonite suspension used in the present study, demonstrating the needle-shaped wollastonite (↓)

2.4. Compression strength parallel to grain measurement

Once the exposed specimens were weighed and the mass losses were calculated, the specimens were subjected to compression parallel to the grain as described in DIN 2-185 on a 50 kN Zwick/Roell Z050 Universal Testing Machine. The specimens were loaded at 0.8 $mm.min^{-1}$ to failure. Compression strength parallel to grain was calculated, using Equation No. 1.

$$P_{c\parallel} = \frac{F_{\max}}{A} \quad (MPa) \qquad (1)$$

where $P_{c\parallel}$ represents compression strength parallel to grain value; F_{max} indicates the maximum force as measured for each specimen by the universal testing machine; and A is the cross section of the specimens along which the force was applied.

2.5. SEM imaging

Microscopic assessments of specimen condition were carried using Scanning Electron Microscopy. Small blocks of 10 × 10 × 2 mm were cut from the original specimens, according to the desired side to be imaged and a thin layer was sliced, oven-dried and placed on an aluminium stub and sputter coated with gold. The coated specimens were examined on an FEI Quanta 250 FESEM field-emission scanning electron microscopy.

2.6. Statistical analysis

The resulting data were subjected to an analysis of variance (ANOVA) at a 95% level of confidence, using SPSS software (version 18, 2010). Differences between treatment means were examined using Duncan's multiple range test (α=0.05). Trends among the studied

properties were visually designed as fitted-line, contour, and surface plots by Minitab software (version 16.2.2, 2010). Hierarchical cluster analyses were performed by SPSS to ascertain similarity of treatments by vertical connecting lines. Hierarchical cluster analyses were carried out based on more than one property at a time.

3. Results

3.1. Mass loss

Mass losses of the unheated (control) specimens exposed to *C. puteana* averaged 23.8, 26.1 and 39.8 % for poplar, beech and spruce, respectively (Table 2). Mass losses for similar samples exposed to *T. versicolor* were 13.3, 28.0 and 42.4 % for spruce, poplar and beech, respectively (Table 2). These mass losses suggested that conditions were suitable for aggressive decay by this fungus. The lower mass losses associated with spruce were typical of white-rot fungi under these test conditions. Thermal modification resulted in significantly lower mass losses for all three wood species, illustrating the benefits of this process on resistance to fungal decay.

The application of acrylic paint was also associated with significantly lower mass losses for the unheated and thermally-modified specimens except for the thermally-modified spruce exposed to *T. versicolor*. These data illustrated the benefits of combining thermal modification with the coating to supplement the benefits of wood modification.

The addition of wollastonite to the acrylic paint was associated with significantly lower mass losses for the non-thermally modified woods of all three wood species exposed to *C. puteana* but did not significantly improve protection against *T. versicolor*. The lack of

effect with *T. versicolor* is more likely due to the very high mass losses associated with this fungus with the non-modified acrylic coating.

Table 2. Mass losses (%) in beech, poplar, and spruce wood specimens as a result of exposure to either *Coniophora puteana* or *Trametes versicolor* (Painted=coated specimens with plain acrylic paint; Wollastonite-painted=coated specimens with graphene-added acrylic paint)

Treatments		Un-heated			Thermally-modified at 185ºC		
Wood species	Fungi species	Control (unpainted)	Painted	Wollastonite-painted	Unpainted	Painted	Wollastonite-painted
Beech	*C. puteana*	26.1 (3.5) C*	10.3 (1.7) E	2.8 (.2) FG	11.3 (.7) E	1.7 (.2) G	1.4 (.2) G
	T. versicolor	42.4 (3.2) A	29.9 (2.0) B	2.3 (.2) FG	17.9 (.9) D	4.2 (.4) F	2.0 (.2) FG
Poplar	*C. puteana*	39.8 (2.8) A	17.2 (1.2) C	4.3 (.5) EFG	8.9 (.9) D	4.9 (.4) E	2.9 (.2) EFG
	T. versicolor	28.0 (4.7) B	4.7 (.6) EF	3.5 (.5) EFG	4.9 (.6) E	2.3 (.5) FG	2.2 (.2) G
Spruce	*C. puteana*	23.8 (2.6) A	12.4 (1.7) C	3.2 (.9) EF	20.2 (2.3) B	9.0 (1.0) D	1.8 (.3) F
	T. versicolor	13.3 (1.9) C	3.2 (.5) EF	2.1 (.2) F	4.3 (.4) E	1.5 (.4) F	2.0 (.2) F

* Values represent means of five replicates while figures in parentheses represent one standard deviation.

Values followed by the same letter(s) do not differ significantly by Duncan's multiple range test (α=0.05)

3.1. Compression strength parallel to grain

Compression strength parallel to grain of the thermally-modified beech or poplar was significantly higher than the unheated specimens prior to fungal exposure (Table 3).

185 Compression parallel to grain results were more variable with spruce, but also tended to be
186 higher in the thermally-modified material.

187 Exposure of the unheated beech samples to either *C. puteana* or *T. versicolor* was
188 associated with highly significant decreases in compression strength (Table 3). Losses were
189 significantly greater with non-painted materials and decreased with the two coating
190 systems. However, the losses were still 40 to 60 % of the original capacity for beech, but
191 slightly lower for poplar or spruce. Decline in the compression strength tended to be lower
192 for the specimens with the wollastonite-enriched acrylic coating. In the unheated spruce
193 specimens, addition of wollastonite seemed to eliminate the negative effect of fungal
194 exposure almost completely as the statistical groupings were the same in comparison to the
195 unexposed specimens.

196 Exposure of thermally-modified blocks with an acrylic coating with or without
197 wollastonite to the two fungi was associated with much lower losses in compression
198 strength. The effects were most pronounced with beech but were also evident with polar
199 and spruce. As with the non-thermally modified samples, the wollastonite-enriched paint
200 was associated with smaller reductions in compression strength.

201 The results indicated that coatings enhanced performance for all three species, but
202 the effect was greatest when thermally-modified wood was supplemented with a coating.
203 Wollastonite addition was associated with enhanced protection in the unheated specimens.

204 **Table 3.** Compression strength parallel to grain (MPa) in beech, poplar, and spruce wood
205 specimens as a result of exposure to either *Coniophora puteana* or *Trametes versicolor*

(Painted=coated specimens with plain acrylic paint; Wollastonite-painted=coated specimens with graphene-added acrylic paint)

Treatments		Un-heated			Thermally-modified at 185ºC		
Wood species	Fungi species	Control (unpainted)	Painted	Wollastonite-painted	Unpainted	Painted	Wollastonite-painted
Beech	Unexposed	52.1 (3.5) DEF*	53.1 (3.1) DE	53.4 (2.5) DE	62.1 (3.7) AB	59.0 (2.3) BC	63.6 (1.5) A
	C. puteana	16.8 (1.0) J	21.5 (1.5) I	34.0 (1.7) G	27.0 (2.0) H	51.2 (1.7) EF	55.7 (2.2) CD
	T. versicolor	13.0 (2.7) J	23.1 (2.4) HI	24.1 (2.2) HI	34.1 (3.5) G	48.5 (4.8) F	55.6 (3.1) CD
Poplar	Unexposed	39.4 (6.0) A	40.7 (3.2) A	39.6 (2.7) A	44.0 (2.4) A	42.9 (3.1) A	43.6 (2.2) A
	C. puteana	7.2 (2.7) F	22.6 (.7) D	26.5 (1.6) CD	32.2 (1.3) B	33.7 (1.5) B	29.3 (1.4) BC
	T. versicolor	17.7 (3.5) E	30.7 (5.1) BC	29.5 (1.6) BC	41.1 (5.1) A	41.2 (6.5) A	39.3 (2.0) A
Spruce	Unexposed	34.8 (3.6) CD	35.8 (2.1) BCD	38.4 (2.8) ABC	37.9 (1.9) ABC	39.3 (2.6) AB	40.8 (2.2) A
	C. puteana	11.9 (2.1) H	25.3 (2.4) F	37.5 (1.3) ABC	17.8 (2.1) G	35.5 (1.6) BCD	29.9 (2.5) E
	T. versicolor	20.1 (1.9) G	30.5 (3.6) E	35.6 (1.2) BCD	32.8 (1.9) DE	37.0 (2.7) BC	37.2 (1.2) ABC

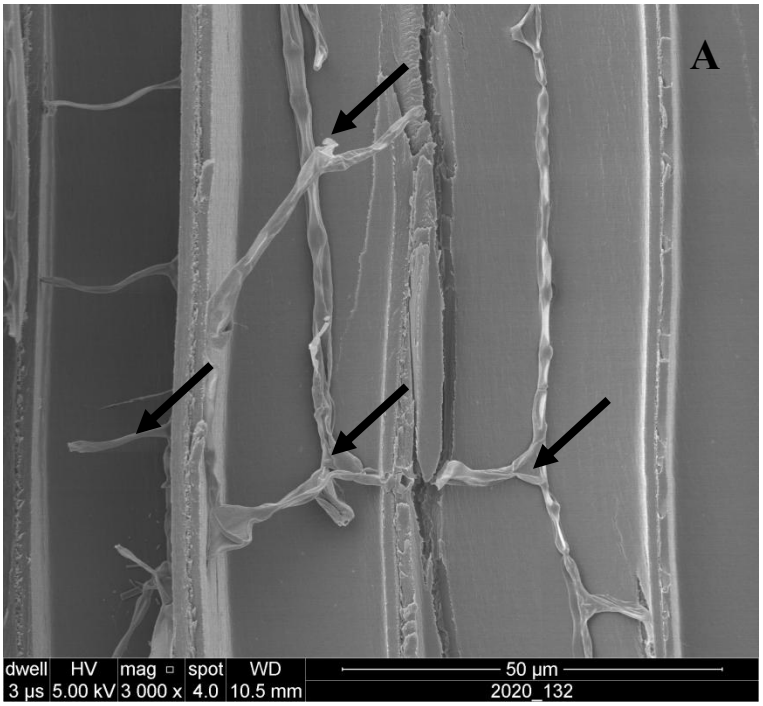
* Values represent means of five replicates while figures in parentheses represent one standard deviation.

Values followed by the same letter(s) do not differ significantly by Duncan's multiple range test (α=0.05)

4. Discussion

The severe MLs occurred in the control specimens were attributed to the easy penetration of fungi mycelia deep into the specimens through the wide-open pits and vessel openings (Table 2; Figs. 2A & 3A & 4A). SEM images revealed that the mycelia were spread in the innermost cell-wall cavity spaces (Fig. 2A,B). Comparison of the MLs revealed highly

significant decreases in the ML values of the coated (both the unheated and the thermally-modified) specimens in all three wood species (Table 2).



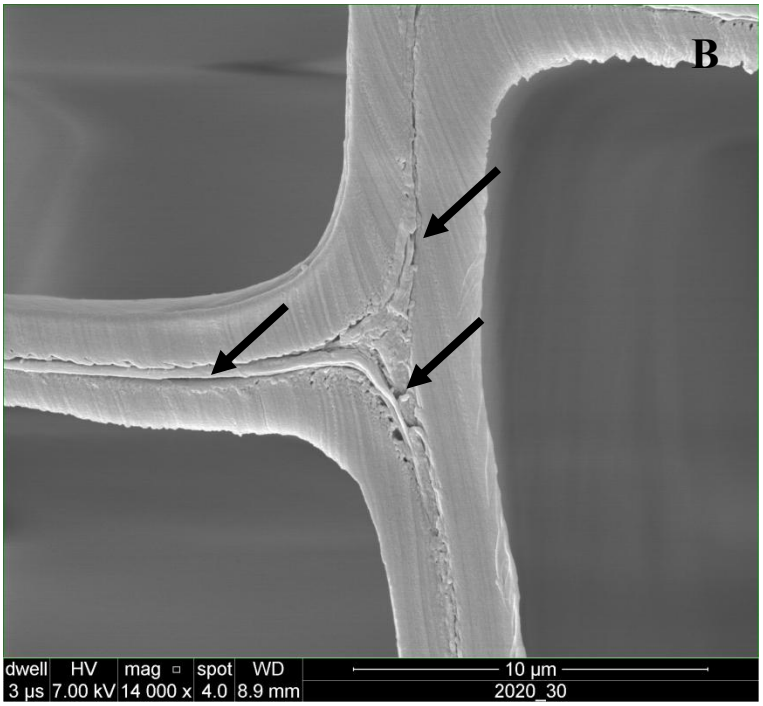


Fig. 2. SEM images of the longitudinal (A) and cross-sectional (B) views in spruce specimens exposed to *Coniophora puteana* for four months, showing the spread of fungal mycelium in the cell-wall cavity (A↓) and the decayed cell wall (B↓)

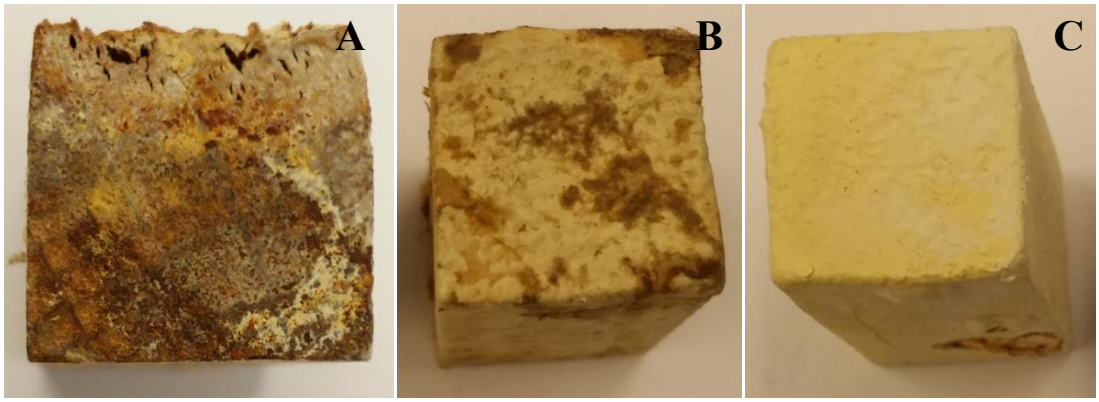


Fig. 3. Comparison of the cross-sectional views in specimens exposed to a wood-decay fungus for four months showing severely decayed wood in the control (uncoated) specimens (A), deteriorated paint film in the specimens coated with plain acrylic paint (B), and nearly intact paint film in the specimens coated with wollastonite-enriched acrylic paint (C)



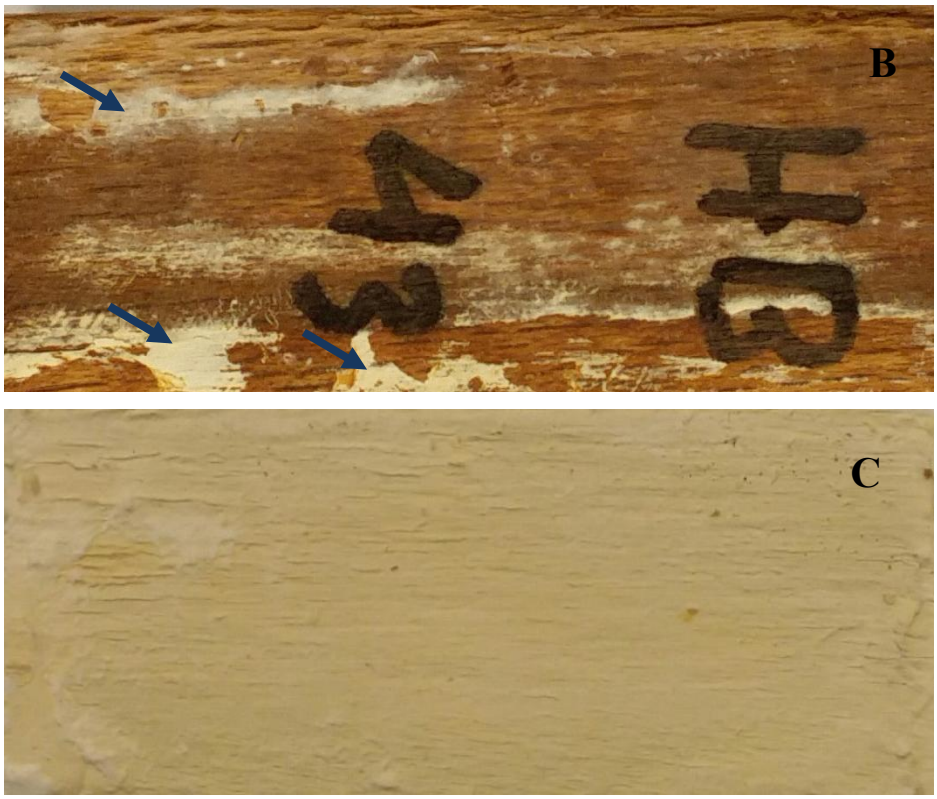


Fig. 4. Control beech specimen (longitudinally cut in half) exposed to *T. versicolor* for four months (A), thermally-modified beech specimen coated with plain acrylic paint showing wiped out paint film in most parts of the specimens (B), the remnant paints are marked (↓), and a specimen coated with wollastonite-enriched acrylic paint with an almost intact paint film after four months of exposure (C)

Though the coated specimens with the plain acrylic paint demonstrated significant lower MLs in all three wood species, the extent of protection seemed to be much lower in the beech specimens exposed to *T. versicolor*. In this connection, beech wood was reported to have a significantly higher swelling coefficient in comparison to poplar (82% in comparison to 14%, respectively) (Taghiyari et al. 2013). The higher swelling coefficient caused ultra small checks and microcracks in the paint film through which the delicate and microscopic mycelia could easily penetrate into the wood substrates. Both the unheated and

246 the thermally-modified beech specimens coated with the plain acrylic paint revealed severe
247 damages on the paint film; in many parts the film was completely removed (Fig. 3B & Fig.
248 4B). The direct relationship between an increase in the moisture sorption property and an
249 enhanced fungal decay has also been positively proved in wood-plastic composites (WPCs)
250 (Kim et al. 2008).

251 Comparison of the beech specimens coated with the plain and the wollastonite-
252 enriched acrylic paints showed a highly significant impact of wollastonite in decreasing the
253 ML (Table 2). The decreased ML in the wollastonite-enriched acrylic paint was attributed
254 to multiple factors and can be discussed from different perspectives. Firstly, based on the
255 chemical structures of wollastonite and acrylic polymer (shown in Figs. 5&6), multiple and
256 different types of chemical interactions can occur between them, improving the cohesion in
257 the paint film. The first would be the ionic coordination of the positively charged Ca^{2+} of
258 wollastonite molecule linked to the negatively charged C=O of the methacrylic acid
259 component (polymethylmetacrylate structure was taken as a typical example); this can
260 create strong new bonds. The abundance of calcium ions (Ca^{2+}), being typical of
261 wollastonite chemical structure, significantly improved the strength and mechanical
262 resilience in the paint film. The formation of the bonds is consistent with previous studies
263 on other ionic bonds, such as metal cations (like chrome as in Cr^{3+} and Cr^{6+}, and iron as in
264 Fe^{3+}) and organic molecules (Kenny & Nicolais 1989; Pizzi 1990a,b; Ntsihlele et al. 1994;
265 Hagita et al. 2016; Bustamante-Torres et al. 2021). In addition, electrostatic interactions
266 and van der Waals forces, acting as secondary forces, were likely to contribute to the
267 overall improved integrity and strength of the film (Kickelbick 2003; Smith et al. 2003).

Moreover, ionic interactions were possibly formed between the calcium ions (in wollastonite) and the wood substrates (possibly with the oxygen atoms in the xylan). Furthermore, based on the chemical structure of methylglucuronoxylan (Fengel & Wegener 1984), formation of hydrogen bonds between the positively-charged hydrogen atoms (in the hydroxyl groups of xylan) and the charged oxygen atoms (in wollastonite molecules) were probable (Fig. 7). There were also potential electrostatic and van der Waals interactions in large number that can be instrumental in both the cohesion and the adhesion improvements of the wollastonite-enriched acrylic paint. Studies on addition of wollastonite as filler in thermoplastic polymer composites have also reported that wollastonite had adhesion interactions with the polymer matrices, resulting in improved tensile and flexural strength of the composites (Švab, 2007; Choo et al. 2022). The formation and the extent of involvement of each type of bond and interaction, as discussed above, can be the main objective of further research studies.

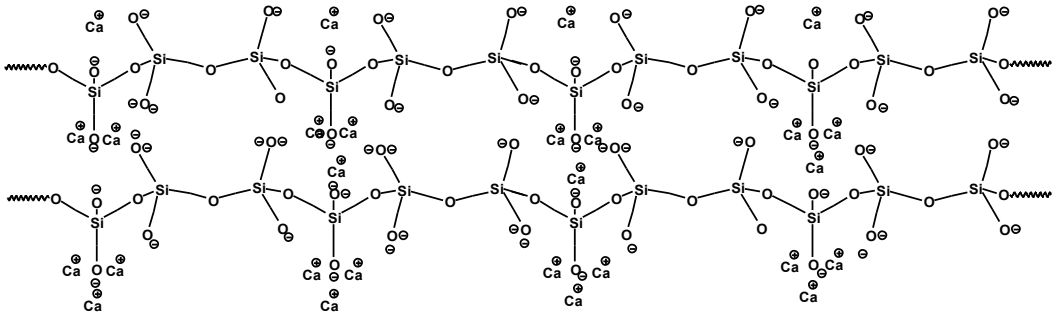


Fig. 5. The chemical structure of the wollastonite chain with its load of coordinated Ca^{2+} (Hill 2006; Taghiyari et al. 2026)

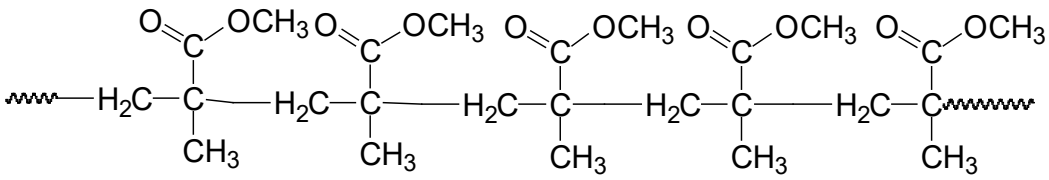


Fig. 6. The chemical structure of the acrylic polymer chain (Polymethylmetacrylate was taken as a typical example of the structure) (Hill 2006; Taghiyari et al. 2026)

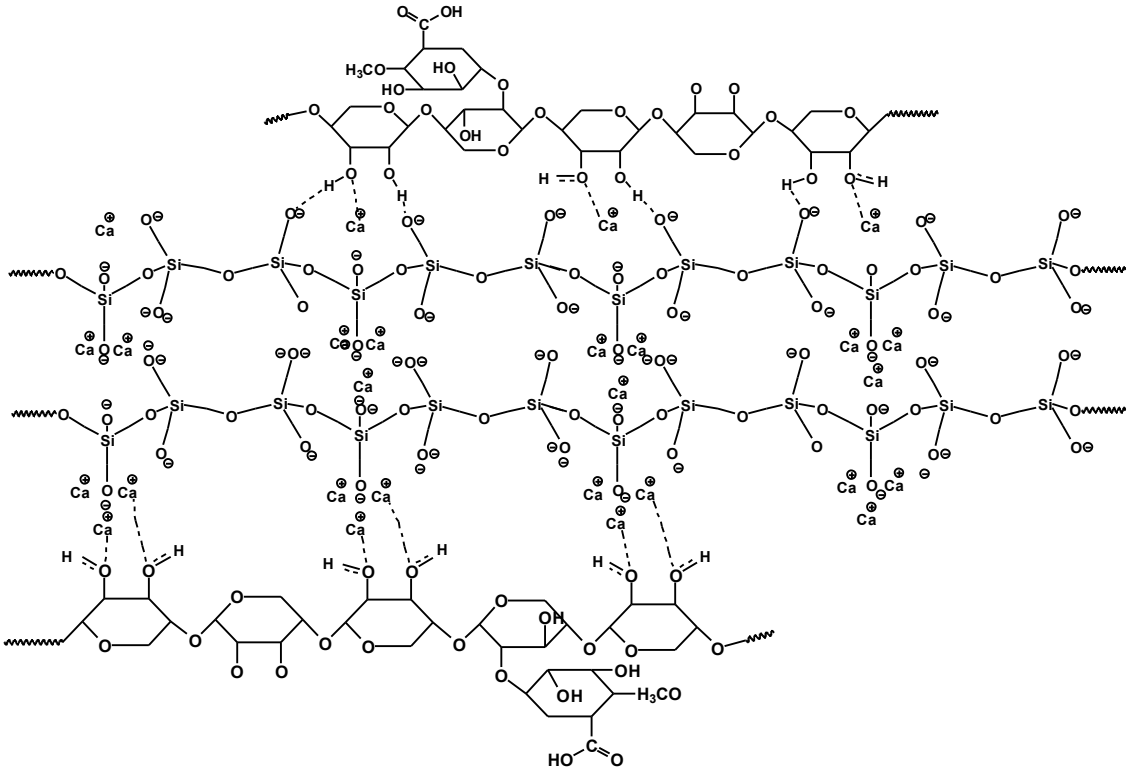


Fig. 7. Potential formation of hydrogen bonds between the positively-charged hydrogen atoms (in the hydroxyl groups of xylan) and the charged oxygen atoms (in wollastonite molecules)

Secondly, due to the high aspect ratio and hardness of wollastonite, the needle-shaped particles acted as a reinforcing additive in the paint film, physically improving its strength. In this connection, previous studies have reported that fillers, dispersed in polymer matrices, can spread the applied stresses over the whole polymer film, improving the

structural stability under multi-component loads and multi-axial stresses (Shen et al. 1993; Li et al. 2003; Meng et al. 2008; Majidi et al. 2023a). Combination of the physical and chemical enforcement of the paint film as a result of the addition of wollastonite made it to be nearly intact, even after four months of fungal exposure in the preserving jars (Fig. 3C & Fig. 4C).

The formation of an impregnable physical barrier had a second function; it acted as a maze-like route, impeding the transmission of oxygen and water molecules (Parvez et al. 2014). This ultimately delayed the growth and progression of fungal mycelium, decreasing the extent of damage.

Wollastonite possibly had another function: acting as an antifungal material and thus, impeding the growth of mycelia on the surface of the specimens (Fig. 8A,B) (Aitken 2010; Abd Rashid et al. 2014; Abd El-Aty et al. 2018; Mardin et al. 2023; Ragab et al. 2026). In this connection, visual examination of the specimens (just after the four-month incubation period) revealed that all around the specimens coated with the plain acrylic paint was covered with *T. versicolor* and *C. puteana* mycelia, in a way that the coding written on the specimens were invisible (Fig. 8A&B-left photos). However, only a very thin layer of mycelium (in some parts even not visible) could be seen on the specimens coated with wollastonite-enriched acrylic paint (Fig. 8A&B-right photos). This was attributed to the wollastonite particles interfering with the fungal enzymatic activity of the fungi, and altering the surface moisture dynamics of specimens as well, reported as the antifungal property of wollastonite (Aitken 2010; Abd Rashid et al. 2014; Abd El-Aty et al. 2018; Mardin et al. 2023; Ragab et al. 2026). Wollastonite antifungal property was translated into

317 the decrease of ML. In fact, ML values in the specimens coated with the wollastonite-added
318 paint were (in all treatments and for all three species) below 2.8%, and they were lower
319 than their counterparts coated with the plain paint. Only the thermally-modified spruce
320 specimens coated with the plain paint and exposed to *T. versicolor* had an insignificantly
321 lower ML in comparison to the wollastonite-added paint. This can be attributed to the high
322 within-group variation which is inherent to all solid wood species. Similar antifungal
323 property was reported for graphene, creating oxidative stress and disrupting fungal cell
324 membranes (Liu et al. 2018; Yaragalla et al. 2021).

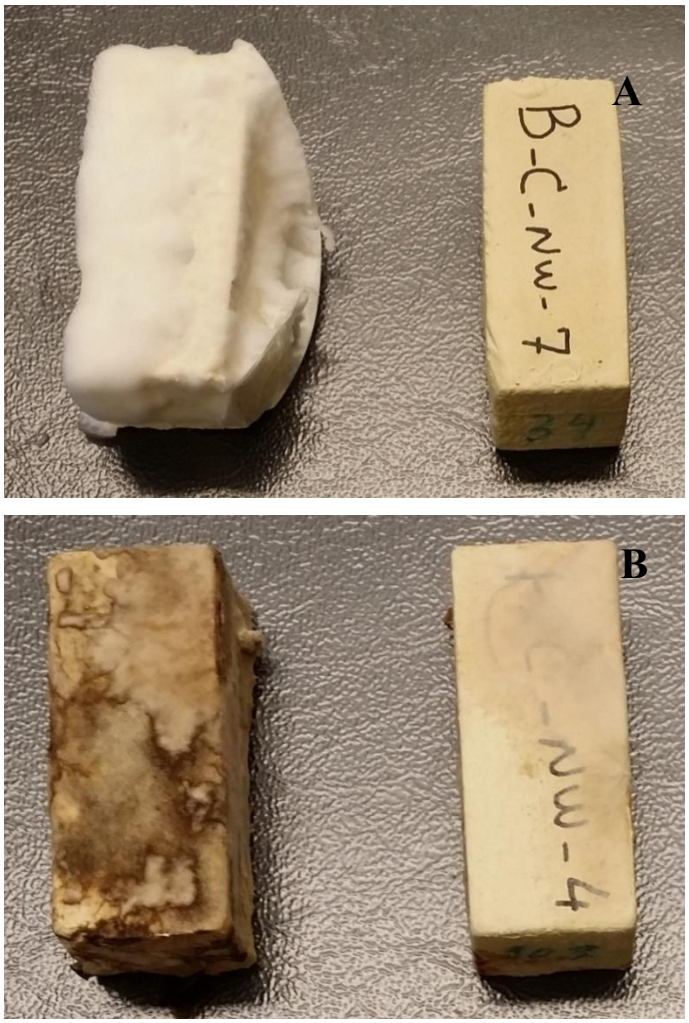


Fig. 8. Beech specimen coated with plain acrylic paint and fully covered with *T. versicolor* mycelium (A-left), beech specimen coated with wollastonite-enriched acrylic paint with a very thin layer of mycelium on the corners and bottom side, being invisible in some parts to the naked eyes (A-right), poplar specimen coated with plain acrylic paint and fully covered with *C. puteana mycelium* (B-left), and poplar specimens coated with wollastonite-enriched acrylic paint with a thin layer of *C. puteana* mycelium (B-right)

[Photos were taken immediately after the specimens were brought out of the incubation jars after four months of fungal exposure to either of the fungi species.]

Regardless of being uncoated or coated (with either the plain or the wollastonite-added paints), all thermally-modified specimens revealed lower ML values in comparison

337 to their unheated counterparts, as a result of exposure to either of the wood-decay fungi
338 (Table 2). This was attributed to the chemically-modified cell-wall polymers caused by
339 thermal modification (Hatakeyama et al. 1982; Boonstra et al. 2006a,b & 2007; Hill 2006;
340 Phuong et al. 2007; Borrega & Kärenlampi 2010). The changes in the modified wood made
341 it hard for the enzymes secreted by the wood-decay fungi to break down wood polymers to
342 their monomer sugar molecules; consequently, MLs decreased substantially. Thermal
343 weight losses were calculated to be 13.25%, 23.98%, and 12.3% in beech, poplar, and
344 spruce, respectively. These weight losses showed high correlation with the decreasing
345 fungal ML% between the unheated and thermally-modified specimens that were caused by
346 both *C. puteana* (R-squared value of 0.80) and *T. versicolor* (R-squared of 0.88). Moreover,
347 the increases in the compression strengths parallel to grain in the unexposed thermally-
348 modified specimens can be considered a corroborating evidence to the above mentioned
349 chemical changes which will be discussed further in this section (Table 3).

350 Among the unexposed treatments, the control beech specimens demonstrated the
351 highest compression strength values, which were attributed to their higher woody mass and
352 density (Table 3). Thermal modification increased the compression strength values in all
353 unexposed specimens compared to their unheated counterparts. The increase was partially
354 attributed to a phenomenon called glass transition of lignin, during which new cross-linking
355 bonds are formed (Hatakeyama et al. 1982; Weiland and Guyonnet 2003; Boonstra et al.
356 2007; Phuong et al. 2007). As a result of thermal modificaion, the normal covalent bonds
357 between lignin and hemicellulose are broken, forming lignin fragments with lower
358 molecular weight, but having higher reactivity (Weiland and Guyonnet 2003; Boonstra et

359 al. 2007). After thermal modification, these fragments will be repolymerized and condensed
360 with some newly-formed cross-linking bonds between lignin and hemicellulose, eventually
361 forming a rearranged structure. The new structure has higher mechanical strength, partially
362 because of the formation of the above-mentioned new bonds.

363 Moreover, another process occurs simultaneously, generally referred to as
364 "hornification" caused by the "irreversible hydrogen bonding" (Fahlén and Salmén 2003;
365 Hakkou et al. 2005; Borrega & Kärenlampi 2010). During this process, the bonds between
366 water molecules and cell-wall polymers are broken and some water molecules are
367 irreversibly removed, providing a potential for formation of new bonds between the
368 polymers, eventually leading to the "hornification" (Kato and Cameron 1999; Matsuda et
369 al. 1994; Weise 1998; Borrega & Kärenlampi 2010). Simultaneous occurrence of these two
370 phenomena (glass transition of lignin, and hornification) resulted in the increased
371 compression strength values of all unexposed heat-treated specimens.

372 R-squared values between MLs versus their counterpart compression strengths after
373 having been exposed to the fungi were calculated to be 53.8%, 78.3%, and 83.9% in the
374 beech, poplar, and spruce treatments, respectively (Fig. 9). This indicated that although
375 compression strength would be closely correlated to the extent of decay caused by either of
376 the fungi, there were other factors involved simultaneously, like the wood species (being a
377 hard- or softwood), the density of the wood substrate, and the protection method, including
378 thermal modification and superficial protection (with either of the paint types). The
379 interactions of the above mentioned factors can also be observed in the fluctuations and
380 inconsistencies found in the contour and surface plots (Fig. 10A,B), though the general

trend can be distinguished between ML values versus compression strengths parallel to grain with or without the fungal exposure. Based on the different impact of the studied variables, the final decision on using the plain or the wollastonite-enriched paints would depend on each species and the desired degree of protection planned by the end-users.

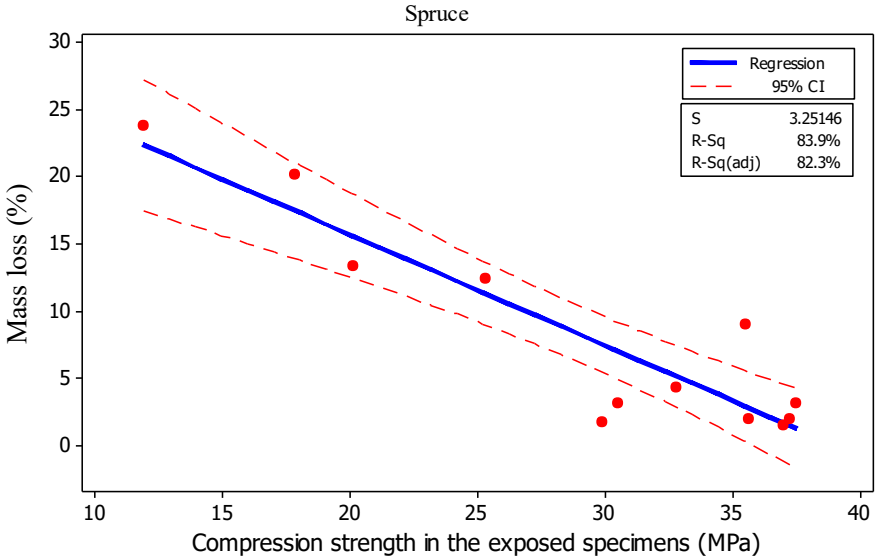
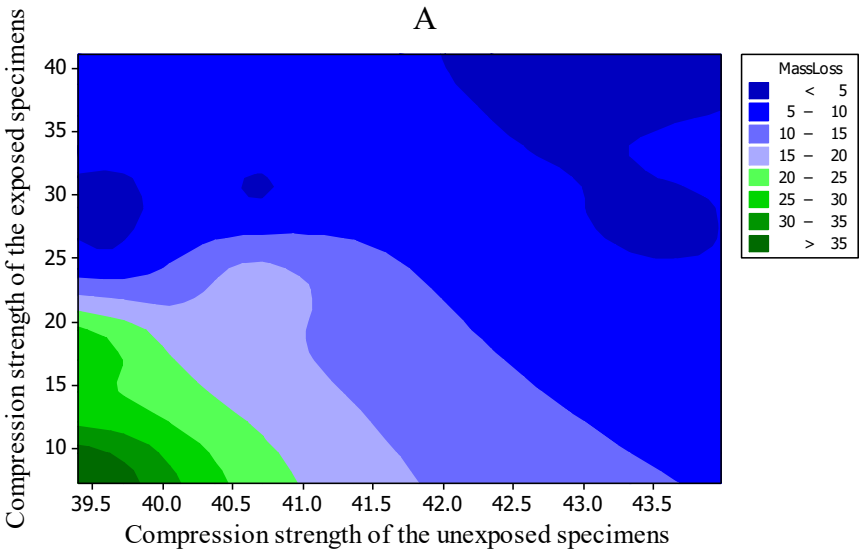
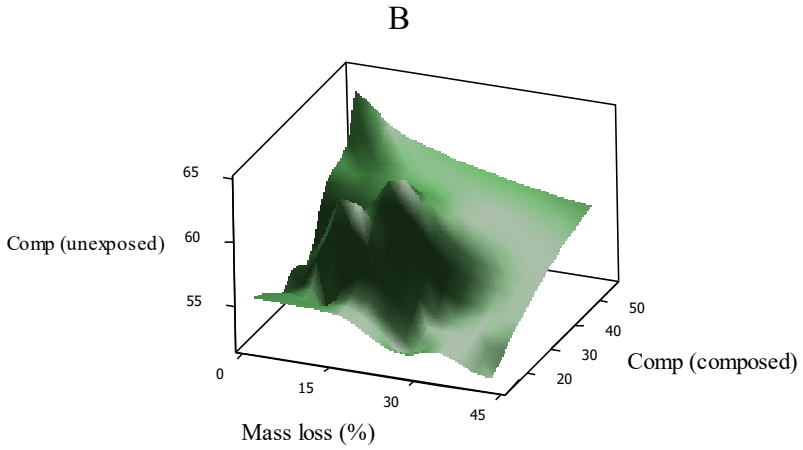


Fig. 9. Fitted-line plot between mass losses (%) versus compression strength (MPa) after the fungal exposure to *Coniophora puteana* and *Trametes versicolor* in the spruce specimens



389



390

391 **Fig. 10.** Contour (A) and surface (B) plots between mass losses (%) versus compression
392 strength parallel to grain values (MPa) in the unexposed and exposed specimens to either of
393 the two fungi (*Coniophora puteana* and *Trametes versicolor*)

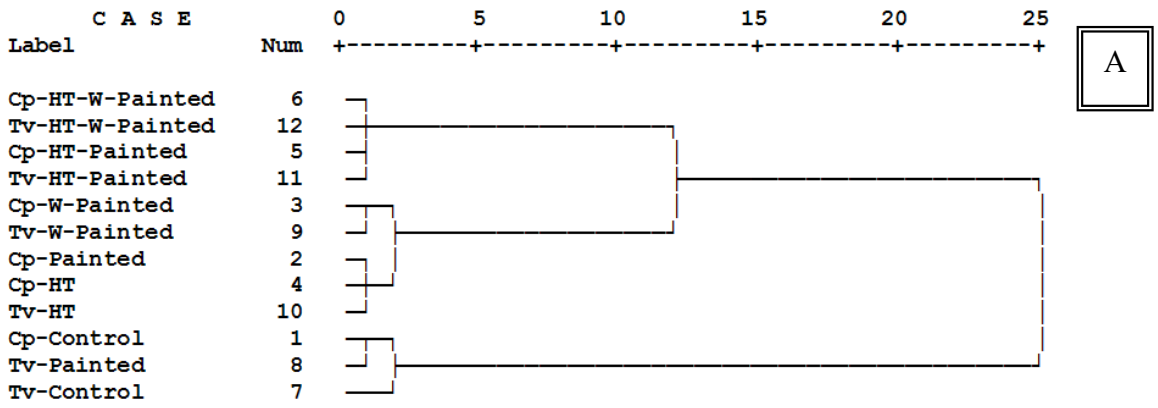
394 Based on the results of the tests as discussed above, and in order to have a more
395 comprehensive outlook on both differences and similarities among the treatments in each

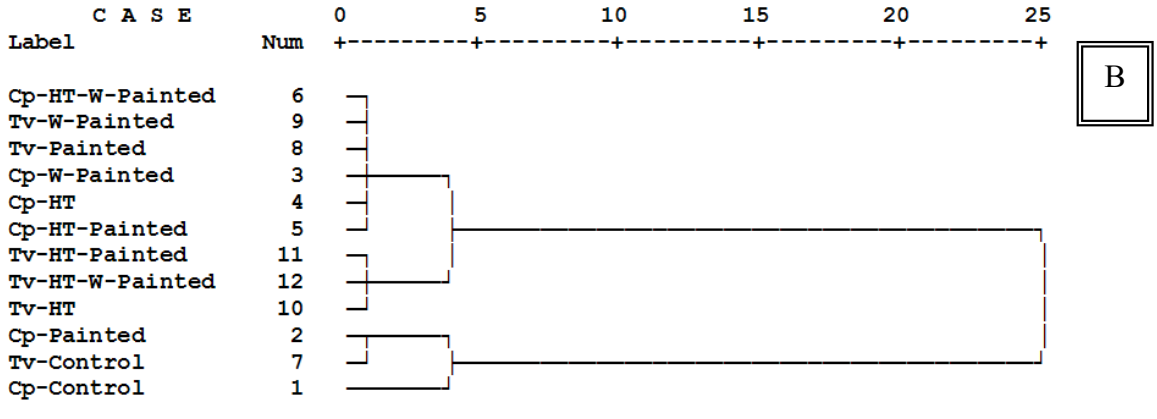
wood species depending on multiple factors, separate cluster analyses were carried out for each of the three species. In all three cluster analyses, the control (the unpainted and unheated) specimens were clustered remotely from the majority of other treatments, regardless of being exposed to either of the wood-decay fungi (namely *C. puteana* and *T. versicolor*) (Fig. 11A,B,C). This showed the significant effects of both the thermal modification and the coatings (either with or without wollastonite) on the overall properties in each treatment. In beech cluster (Fig. 11A), the unheated specimens coated with the plain acrylic paint and exposed to *T. versicolor* were clustered very closely to the control specimens, indicating that a mere coating cannot sufficiently protect beech specimens to a desirable degree. Similarly, the unheated poplar and spruce specimens coated with the plain paint and exposed to *C. puteana* were closely clustered to the control specimens, demonstrating that in these two lower-density species, a mere coating with plain acrylic paint cannot be considered an effective means of protection. However, all unheated specimens of the three species coated with the wollastonite-enriched acrylic paint were remotely clustered from the control specimens, showing the effectiveness of wollastonite in protection of the unheated substrates against both wood-decay fungi.

In terms of the effects of thermal modification, all thermally-modified specimens of beech and poplar (as the two hardwoods studied) were remotely clustered from the control specimens, regardless of having been exposed to either of the fungi (Fig. 11A,B). This indicated the degree of effectiveness of this mild modification method (thermal modification at 185ºC) to be used for hardwoods against exposure to both white- and brown-rot fungi. However, thermally-modified spruce specimens exposed to *C. puteana*

were closely clustered to the control specimens (Fig. 11C). This implied that a mere thermal modification at a mild temperature of 185ºC cannot provide an efficient protection against the brown-rot fungus.

Thermally-modified beech specimens coated with either of the paint types (plain and wollastonite-enriched) were very closely clustered together, regardless of the fungus species (Fig. 11A). The same similarity was observed, to a lesser extent, in the clustering of the thermally-modified coated spruce and poplar specimens. This indicated that addition of wollastonite in the acrylic paint had a least impact on protecting thermally-modified beech, poplar, and spruce specimens against either of the fungi species. In the unheated specimens, however, the similarities were not so close and varied depending on each species. It can be concluded that addition of wollastonite to the acrylic paint had a more significant improving impact in the unheated wood species, being dependant on the fungus species.





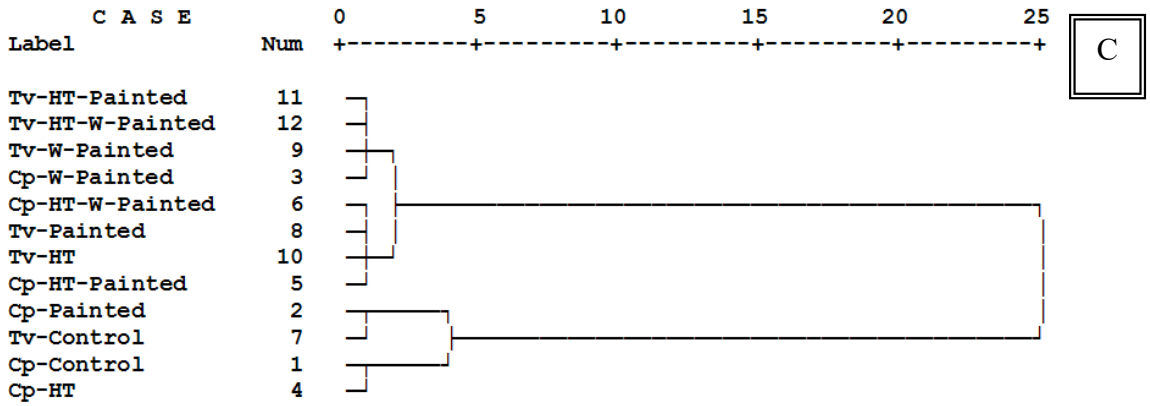


Fig. 11. Cluster analyses of the treatments of each of the three wood species separately carried out, comprising of beech (A), poplar (B), and spruce (C) treatments; the analyses were conducted based on mass losses and compression strength parallel to grain after being exposed to either *Coniophora puteana* or *Trametes versicolor* wood-decay fungi (Cp=exposed to *Coniophora puteana* fungus; Tv=exposed to *Trametes versicolor* fungus; HT=Thermally-modified specimens; W=wollastonite-enriched acrylic paint)

Based on the findings in the present study and the facts in the literature, wollastonite-enriched acrylic paint significantly decreased the mass losses caused by exposure to both wood-decay fungi (*C. puteana* and *T. versicolor*) in all three wood species (beech, poplar, and spruce). It also maintained the compression strength parallel to grain to a highly significant extent. Exposure of beech specimens to *T. versicolor* resulted in more

444 severe damages and therefore, heat treatment as an additional protecting measure would be
445 recommended to be incorporated with the coating with wollastonite-enriched paint. The
446 functions of wollastonite can be discussed from different perspectives, all partly
447 contributing to in the significant improvement of strength and mechanical resilience of the
448 paint film, including: (1) forming ionic bonds (as primary force) and electrostatic
449 interactions and van der Waals forces (as secondary forces) to chemically improve the
450 cohesion in the paint film; (2) acting as a reinforcing additive in the acrylic polymer matrix,
451 physically improving its cohesion and strength; (3) acting as an antifungal material,
452 limiting the growth and penetration of fungal mycelia in to the wood substrate; (4)
453 functioning as an impermeable barrier for the transmission of oxygen and water molecules
454 to the wood substrate; and (5) possibly forming bonds with the cell-wall polymers on the
455 surface of xylan substrates, improving the adhesion between the wollastonite-enriched paint
456 film and the wood substrate. The collective result of the above-mentioned functions
457 eventually increased both the cohesion and adhesion of the paint film, ultimately
458 ameliorating the negative effects of the fungal exposure on both ML and compression
459 strength parallel to grain. It was concluded that wollastonite-enriched acrylic paint has
460 significant efficacy in protecting solid wood substrates against wood-decay fungi;
461 therefore, it is recommended to the commercial sector as an easy-to-apply and reliable
462 protection and preservation of both hard- and softwoods.

463 Still, more concerns on the utilization of wollastonite as a reinforcement additive
464 and extender should be addressed to, focusing on its content to achieve the optimum results
465 for collective improved properties (like improved fire properties, biological resistance, and

aesthetic aspects). Furthermore, its mixing methods in different paint types should be investigated to provide a uniform dispersion, in which wollastonite particles are consistent and settlement does not occur, and also its sustainable application for the environment is considered. Field studies on commercial scale wood bodies in service can provide a better outlook on the extent of protection in practice. A cost-benefit feasibility study would clarify if the improved properties are justified from an economical point of view. Moreover, further studies are still needed to clarify if the antifungal property of wollastonite alone (without a polymer matrix to be dispersed in) is sufficient in impeding the growth of wood-decay fungi, and if coating on wood bodies with wollastonite alone would be effective in protecting the substrates against various wood-decay fungi species. Clarification on the above mentioned points would accelerate the process for the industrial sector to use wollastonite more effectively in their coating formulations.

Acknowledgements

The first author would like to pay his tribute to the Alexander von Humboldt Stiftung (Germany) to finance his stay in Germany and to facilitate doing the tests at Hamburg University. He is also grateful to the departed Prof. Olaf Schmidt for his guidance on the fungal culture and laboratory procedures.

Competing Interests: The authors declare no conflicts of interest.

Author Contributions: Conceptualization, H.R.T., A.P. and O.S.; methodology, H.R.T., A.N.P., J.J.M.; software, H.R.T. and E.N.; formal analysis, H.R.T., E.N., J.J.M., J.K., I.A.P. and O.S.; investigation, H.R.T., E.N., and O.S.; resources, H.R.T., E.N., M.H.S., and O.S.; data curation, H.R.T., E.N., I.A.P., and O.S.; writing original draft preparation, H.R.T., A.P., A.N.P., J.J.M., and J.K.; writing review and editing, H.R.T., A.P., A.N.P. and J.J.M.;

visualization, H.R.T., E.N., and I.A.P.; supervision, H.R.T., A.P., and A.N.P.; validation, H.R.T., A.P., and M.H.S.; legal consultation, M.H.S.; project administration, H.R.T.; funding acquisition, H.R.T., E.N., and O.S.;

Data Availability Statement: Data are contained and presented within the article.

Funding: This research received no external funding.

Ethics statement: Not applicable.

Declaration of generative AI use: No generative AI has been used in the present research study.

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