Tannin-based Polyurethane Coating for Quality Improvement of Roof Tiles Composite

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Roof tiles come in various forms and are crucial to residential construction. A roof tile composite offers the market a selection of superior roof tile products in terms of strength, low density, and environmental friendliness. This research aimed to improve the surface performance and durability of sorghum bagasse-based roof tile composite (SBRTC) through surface coating with natural polymer. Sorghum bagasse was made into roof tile composite using a mixture of molasses and citric acid adhesives (50:50) with a target density of 0.6 g/cm^3. Furthermore, the SBRTC surface was coated with tannin–polyurethane at different concentrations (10%, 20%, and 30%), and the results were compared with both uncoated and polyurethane-coated samples. The parameters tested included physical and mechanical properties, surface characteristics, and durability against termite and brown-rot fungi. The result showed increasing density, dimensional stability, mechanical properties, and durability. At the same time, the moisture content decreased. Surface performance exhibits a decrease in the average surface roughness (R_a) value, indicating a smoother surface of roof tile composite after surface coating. Furthermore, a high contact angle, low K-value, and low wettability were achieved. It indicates a more hydrophobic surface. The optimal tannin concentration in the coating solution was 20%.

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INTRODUCTION

After Japan and China, Indonesia is one of the three countries with the highest earthquake activity worldwide. Over the past three decades, Indonesia has been hit by catastrophic earthquakes that have killed thousands of lives and destroyed significant infrastructure (Rifa *et al.* 2020; Pribadi *et al.* 2021). Earthquakes often cause severe damage to buildings, including roofs that use heavy tiles such as clay, concrete, and ceramic. Most houses in Indonesia still use roof tiles made from clay, cement, ceramic, and concrete, which, apart from being heavy, also break easily (Tambak *et al.* 2013; Akinwande *et al.* 2021). The heavy weight of traditional tiles can increase the risk of structural collapse during an earthquake, threatening the safety of occupants. Koesmartadi *et al.* (2019) stated that roof collapses are a common cause of casualties. Therefore, safer construction solutions are needed, especially using lightweight roof tiles.

Akinwande *et al.* (2021) found that the best material for roof tile manufacture was a polymer composite made up of 98% high-density polypropylene and 2% granite dust by weight after comparing five different materials (fired ceramic tiles, concrete tiles, concrete clay tiles, polymer tiles, and metal tiles) using the digital logic method (DLM). Polymer roof tiles can have good insulation properties. They are generally characterized by relatively low density, high stiffness, and good durability, although they tend to be expensive. Roof tiles composites have been widely developed by substituting some roof tile materials with biomass or natural fibers. Biomass is renewable, abundantly accessible, low-cost, and eco-friendly compared to synthetic polymer fibers (Hancharoen *et al.* 2024). The biomass that has been used includes the empty oil palm fruit bunches (Hancharoen *et al.* 2024), long coconut fiber (Milawarni 2013; Yusniyanti and Irwansyah 2019), boiler ash waste from the palm oil industry (Maghfirah *et al.* 2018), oil palm broom fiber (Momoh and Dahunsi 2017), sugarcane bagasse (Rambe *et al.* 2017), and coir fiber (Darsana *et al.* 2016). Adding biomass to roof tile composite can reduce production costs and act as reinforcement. Momoh and Dahunsi (2017) reported that cement, concrete, and ceramic are often brittle and perform poorly under tensile and flexural loads.

Because all-natural fibers have superior mechanical and physical properties, they can be used more effectively to create composite materials for a range of construction applications (Barthwal *et al.* 2014). Biomass is mixed in certain proportions to obtain the desired properties of roof tile composite. Darsana *et al.* (2016) reported that up to 15% of coconut fiber was used as a substitute for cement when creating roof tiles. The findings indicate improvements in ductility and breaking load, alongside a significant reduction in material costs. In addition, Momoh and Dahunsi (2017) reported that reinforcing material in fiber mortar roof tiles and oil palm broom fibers will produce a good result with the highest flexural strength at a 1:1 laterite-to-cement ratio, a mesh size of 10 mm, and a 5% fiber volume fraction. Hancharoen *et al.* (2024) developed a natural fiber-rubber-cement (FRC) composite for roofing applications by combining type-I Portland cement, plasticizer, oil palm fibers, and rubber powders in a weight ratio of 100:0.8:10:50. The findings indicated a 10% improvement in flexural strength and a 20% enhancement in the noise reduction coefficient.

Roof tile composite products offer a cost-effective alternative to other pricey fibrous materials, contributing to affordable housing costs and lightweight roof tile composite materials (Momoh and Dahunsi 2017). The entire use of biomass in the manufacture of roof tile composites was carried out by Syahfitri *et al.* (2024) using sorghum bagasse with molasses adhesive. The density, water content, and internal strength met the requirements of the Japanese Industrial Standard (JIS) A 5908 (2003). Moreover, this roof tile was found to be lighter. Molasses as a natural adhesive has been optimized by Sutiawan *et al.* (2024). Combining it with citric acid increased the bond strength by a ratio of 50:50 to provide the best results. Combining biomass and natural adhesive produces strong, durable composite products and reduces waste and carbon emissions. Lightweight roof tile composite made from sorghum bagasse can be the ideal choice for earthquake-resistant roof elements. Although biomass composite tiles with natural adhesives offer many advantages, challenges, such as dimensional stability, resistance to weather, and biological attack, still need to be thoroughly addressed (Darsana *et al.* 2016; Syahfitri *et al.* 2024). Further research is needed to enhance surface performance and improve resistance to biological degradation.

Surface coating is a simple and efficient method used for protection of roof tiles composite from environmental change. Solid and liquid surface coating materials are frequently applied to particleboards to improve their mechanical, physical, and appearance (Özlüsoylu 2023). Coating significantly increases wood's resistance to weather factors and reduces surface deterioration when exposed to outdoor conditions and anti-corrosion (Payra *et al.* 2015). Considering environmental and health issues, biobased coating material appears to be the right choice for coating. Tannin used as preservation and first layer on the surface properties of wood followed by coating with water-based and polyurethane varnishes was conducted by Yasar *et al.* (2024). Thébault *et al.* (2015) coated with non-isocyanate polyurethanes (NIPU) based on tannin on a medium-density fiberboard (MDF) surface. The result showed that the coated surfaces had increased hydrophobicity compared to uncoated MDF. A NIPU coating based on THEIC–ester of fatty acid was formulated and applied on mild steel panels by Pathak *et al.* (2015). It can serve as a viable substitute for traditional polyurethane-based coatings with different amines that will determine the curing process and properties of the NIPU coating. It is known that polyurethane (PU) is toxic. As a consequence, there has been a lot of study focuses on substituting and replacing this material with harmless or low-hazard materials (Gholami *et al.* 2021; Aristri *et al.* 2023).

As a bio-source of polyols, tannin contains numerous OH groups that react with polymeric isocyanates to form polyurethanes through polycondensation and exothermic reaction (Aristri *et al.* 2023; Thébault *et al.* 2015). Tannin was used directly as a substitute for polyols which are reacted with dimethyl carbonate and diamine (Thébault *et al.* 2015). Meanwhile, Yasar *et al.* (2024) used tannin as first protector layer before the coating layer (water-based and varnish). In the present work, tannin-based polyurethane was developed by mixing methylene diphenyl diisocyanate (MDI), polyol, and tannin extract as coating materials in various combinations. The tannin-based polyurethane coating was applied to sorghum bagasse-based roof tile composites (SBRTC) surface and the results were compared with both uncoated and polyurethane-coated samples. The environmentally friendly materials in this research (sorghum bagasse tile composite products using natural adhesive and coated with biobased-coating) align with the global trend to create sustainable and eco-friendly products. The type of coating material and the quality of the bagasse-based panel both play a crucial role in determining the performance of the coated panels.

EXPERIMENTAL

Materials

The sorghum bagasse was sourced from the BRIN experimental garden in Cibinong, Bogor, Indonesia. According to previous study, a drum chipper (Pallmann, German) and ring flaker machine (Pallmann, German) were used to create the sorghum bagasse particles (Sutiawan et al. 2023). The sorghum bagasse was converted into particles and sieved to obtain the desired size range of 4 to 20 mesh. After that, it was dried at 60 °C in the oven until the moisture content was less than 10%. The adhesive comprises molasses (MO) and citric acid (CA). Adhesive preparation refers to Syahfitri *et al.* (2024) and

Sutiawan *et al.* (2024). For MO and CA, each adhesive was dissolved in water until it reached a solid content of 59% at room temperature and temperature of 60 °C under stirring of 200 rpm for 25 min, respectively. Coating materials are a mixture of diphenylmethane diisocyanate (MDI), polyol, and tannin. The tannin extract used was condensed tannin derived from *Acacia mangium,* as described in a previous study (Lubis *et al.* 2024).

The characteristics of the tannin extract, in comparison to commercial tannin, were analyzed using Fourier transform infrared spectroscopy (FTIR), pyrolysis gas chromatography mass spectrometry (Py-GCMS), X-ray diffraction (XRD), and thermogravimetric analysis (TGA). Functional group analysis was conducted using FTIR (Perkin Elmer Spectrum Two) with an average of 16 scans, and the spectra were captured in areas 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1}. Furthermore, Py-GCMS (Shimadzu GCMS-QP2020 NX, Japan) was used to analyze chemical compounds. Meanwhile, XRD and TGA were utilized to observe variations in the degree of crystallinity and thermal characteristics.

Manufacture of Roof Tile Composite

Sorghum bagasse (SB) was used to manufacture roof tile composite using a mixture of MO and CA adhesive with a target density of 0.6 g/cm^3 (Syahfitri *et al.* 2024). Bio-based adhesive of MO was modified with CA, following the procedures of Sutiawan *et al.* (2024b) with an optimum ratio of 50:50. Oven-dried SB particles were mixed with adhesive in a mixer using a spray gun (Meiji, Japan). The particles are then placed into a 30 cm × 30 cm mold with a Teflon base and sides of a 0.6-cm-thick steel bar. The samples were hot-pressed (Shinto, Japan) at 200 °C for 10 min at a maximum specific pressure of 5 MPa (Sutiawan *et al.* 2024a). Two weeks were then spent for the roof tile composite to be conditioned at room temperature and ± 60% relative humidity before coating application.

Coating Application

Coating materials consist of MDI, polyol, and tannin extract. The formulation is shown in Table 1. The unsanded sample, measuring of 30 cm × 30 cm, was manually coated on the surface using a brush, excluding the edges of the board (Fig. 1). The targeted application rate was 110 g/m^2 (Özlüsoylu 2023) then dried and conditioned for 2 weeks before being used for surface characterization and other tests.

Table 1. Formulation of PU-tannin Coating with Different Compositions

Formulas	Materials		
	MDI (%)	Polyol (%)	Tannin (%)
No coating	-	-	-
PU + 0% T	50	50	0
PU + 10% T	50	40	10
PU + 20% T	50	30	20
PU + 30% T	50	20	30

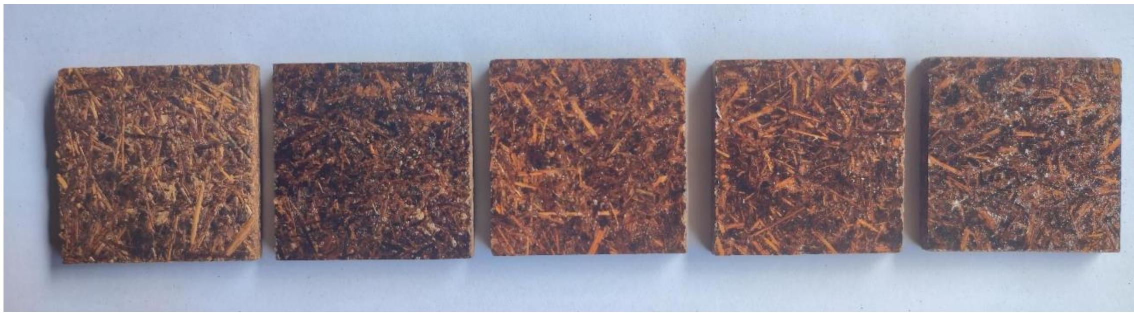


Fig. 1. Coating appearances on particleboard in different formulations (a) No coating, (b) PU + 0% T, (c) PU + 10% T, (d) PU + 20% T, (e) PU + 30% T

Determination of Roof Tile Composite Quality

Physical properties

Physical properties determined in this study were density (ρ), moisture content (MC), water absorption (WA), and thickness swelling (TS) according to the Japanese industrial standards for particleboards JIS A 5908 (2003). A sample size of 50 mm x 50 mm x 6 mm cm was used for density, MC, WA, and TS. The density and MC of control samples were measured after the samples were conditioned for 2 weeks. Density was measured as the ratio of the mass and volume of a sample. Following 24 h of drying at 103 °C ± 2 °C in an oven, the masses before and after were used to determine the moisture content test and density.

Water absorption and thickness swelling represent the dimensional stability of SB roof tile composite. The mass was weighed, and the dimensions of the samples were measured before and after being immersed in water for 24 h at 20 °C. Sample weight was used to calculate water absorption, while thickness swelling based on dimensional changes.

Surface characterization

Surface roughness and wettability were evaluated to show the surface performance of SB roof tile composite. The roughness measurement was carried out using a diamond tip radius of 5 µm, tracing length of 15 mm, cut-off of 2.5 mm, and speed of 0.5 mm/s. The 50 mm x 50 mm x 6 mm samples were used to measure at five different positions of each sample using a Mitutoyo-type SJ-210 tester. The arithmetical mean roughness for 15 readings (R_a) was used to express roughness (Hanifah *et al.* 2023).

Wettability describes how a liquid interacts with a substrate surface, which was evaluated by measuring the dynamic contact angle. The dynamic contact angle was measured according to Martha *et al.* (2024). Samples with a size of 50 mm x 50 mm x 6 mm were evaluated by the sessile drop method, using the Drop Shape Analysis system - DSA100 (Krüss GmbH, Hamburg, Germany). Each sample had three water droplets, and the dynamic contact angle was automatically measured every 3 s for 60 s. Twenty data points were analyzed to create a contact angle *versus* wetting time curve. The equilibrium contact angle was determined through the transition point between contact angle and time using SAS (SAS STAT 9.1, SAS Institute Inc., Cary, NC, USA). The contact angle change rate (K-value) was calculated using the S/G model (Shi and Garnder 2021) with XLSTAT to evaluate wettability.

Mechanical properties

The sample size of modulus of elasticity (MOE) and modulus of rupture (MOR) was 200 x 50 x 6 mm^3 in length, width, and thickness based on JIS A 5908 (2003). Samples were tested using a universal testing machine (UTM, AGS-X series 50 kN, Shimadzu, Tokyo, Japan) with a loading speed of 10 mm/min and an effective span of 150 mm.

Durability testing

Roof tile composite was tested against termite and wood decay fungi to determine their durability. According to SNI 7207-2014 (BSN 2014), sample preparation involves keeping a 20 mm x 20 mm x 6 mm sample size in an oven at 60 °C for 48 h, then weighing it to determine its weight before testing and sterilizing sand at 100 ± 2 °C for 48 h. The sample was placed in a bottle containing moist sand and exposed to 200 subterranean termites (*Coptotermes curvignathus* Holmgren) for 4 weeks in a dark room. Samples were cleaned and kept in the oven at 60 °C for 48 h to measure the weight after testing. The durability class was classified based on weight loss percentage. From the high to low durability class (I to V), the weight loss percentage is as follows: I) less than 3.5% indicates very durable; II) 3.5 to 7.4% indicates durable; III) 7.5 to 10.8% indicates moderately durable; IV) 10.9 to 18.9% indicates not durable, and V) > 18.9% indicates very not durable. In addition, termite mortality was also determined.

The durability of roof tile composite against brown-rot decay fungi (*Fomitopsis palustris*) was tested according to JIS K 1571 (JSA 2004) with sample size of 20 mm x 20 mm x 6 mm. Uncoated samples and rubber wood were also tested as a comparison. Before testing, the sample was kept in an oven at 60 °C for 48 h and weighed to get its mass. To prevent contamination from other decay, the test sample was placed in a test bottle that was tightly sealed and filled with rotting decay for 12 weeks. Following testing, the samples were cleaned and 48 h of oven drying at 60 °C until their weight remained consistent. Their durability was assessed using the weight loss percentage.

Statistical Analysis

The average values and standard deviations were computed after each experiment's results were obtained. Analysis of variance (ANOVA) was the single method used to examine the physical, surface, mechanical properties, and durability data using SPSS 23. Duncan's multiple range test was performed to determine whether the results differed significantly ($p \leq 0.05$). Conditions that do not show statistically significant differences are indicated by the same letters, while different letters represent statistically significant differences, as shown in each figure.

RESULTS AND DISCUSSION

Characterization of Tannin Extract Compared to Commercial Tannin

The tannin extract was analyzed using FTIR, XRD, Py-GCMS, and TGA to determine the quality of the extract and compare it with commercial tannin. The FTIR spectra are shown in Fig. 2a. A broad band at 3200 to 3500 cm^{-1} corresponds to –OH stretching vibrations in phenolic and aliphatic structure (Yingprasert *et al.* 2023), as well as within any moisture that is present. In the present work, such bands were centered at 3292 cm^{-1}. As can be seen, a higher intensity of tannin extract compared to commercial tannin enabled a relatively high phenolic content in tannin extract. In other studies, the OH-stretching band's shape offers early indications of a polymerization process. The spectrum of condensed tannins ranged widely, from 3,700 to 3,000 cm^{-1}, due to their different positions of the substituent OH bands in molecules with varying degrees of polymerization and from several interactions between the tannin molecules and certain substrate (Marques *et al.* 2021). Then, the peaks between 1400 and 2000 cm^{-1} show the aromatic nature of the structure. In addition, peaks at 1600 to 1750 cm^{-1} represented C=C stretching bands in tannins (Shnawa *et al.* 2015; Yingprasert *et al.* 2023). The aromatic rings present in the present study gave rise to a peak at 1632 cm^{-1}. For this peak, the intensity of tannin extract and commercial tannin was comparable, suggesting that their contents were the same. Tannin extract lacks the single high-intensity peak at 1031 cm^{-1} observed in commercial tannins. Various peaks in the 600 to 1300 cm^{-1} correspond to substituted benzene rings (Bharudin *et al.* 2013). According to XRD analysis, the tannin extract in the present study was judged to have a relatively higher degree of crystallinity than commercial tannin based on higher peak intensity at $2\theta = 22°$ of 27.43% and 24.78%, respectively (Fig. 2b).

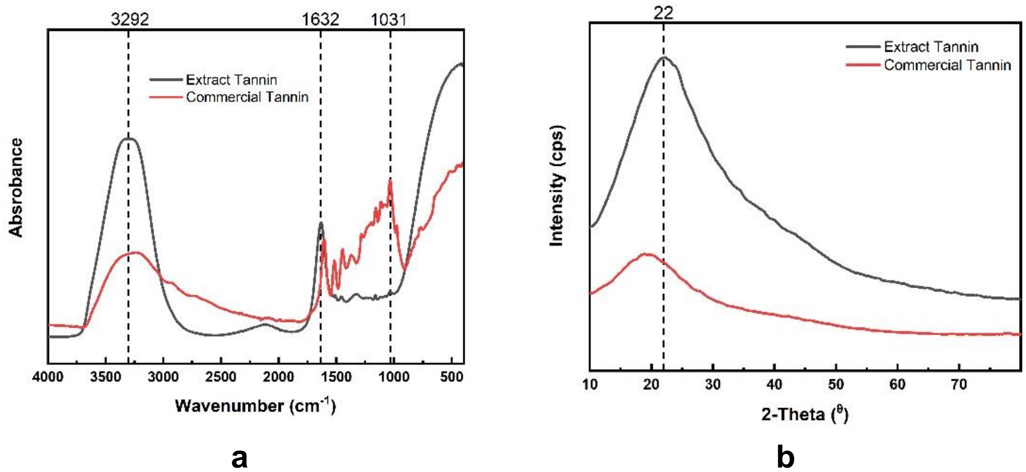
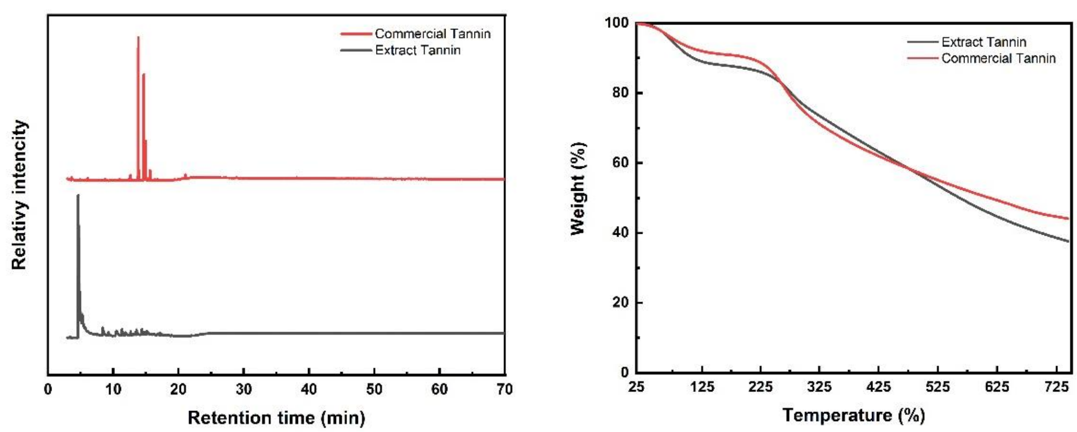


Fig. 2. FTIR spectra (a) and XRD (b) of tannin extract and commercial tannin

Different components were identified in a Py-GCMS analysis of tannin extract and commercial tannin. As many as 23 and 20 compounds were found in tannin extract and commercial, respectively. The five highest chemical compounds (± 88%) are presented in Table 2. Tannin extract had the highest peak (Fig. 3a) at a retention time of 1.64 s, which marks it as a carbamic acid compound with a concentration of 68.8% (Table 2). The second largest compound is acetic acid, and the last 3 compounds are only about 2% (2-propanone, furfural, and lactic acid). Commercial tannin is dominated by catechol and resorcinol compounds by 39.21% and 33.50%, respectively. The high content of catechol and resorcinol may contribute to the growth of the pharmaceutical leather, and adhesive sectors (Masendra *et al.* 2021). The third most abundant compound was 2-benzenediol 4-methyl, with a concentration of 10.97%; the rest was phenol at 5.04%. Ismayati *et al.* (2024) reported the tannin extract of *A. mangium* pyrolysis products were catechol, resorcinol, pyrogallol, and benzoic acid. While commercial tannins are predominantly phenolic compounds, the tannin extract was characterized by a higher concentration of organic acids.

Table 2. The Five Largest Compounds of the Identified Pyr-GCMS Product of Tannin Extract and Commercial Tannin

No	Ret. Time (s)	Compounds	Concentration (%)	
			Extract	Commercial
1	1.637	Carbamic acid	68.78	
	13.845	Catechol		39.21
2	2.239	Acetic acid	12.23	
	14.673	Resorcinol		33.50
3	2.611	2-Propanone 1-hydroxy-	2.9	
	14.921	2-Benzenediol 4-methyl-		10.97
4	5.404	Furfural	2.61	
	15.622	Phenol, 6-dimethoxy-		3.17
5	7.513	L-Lactic acid	2.38	
	12.635	Phenol, 2-methoxy-		1.87
Total concentration (%)			88.9	88.72



a b

Fig. 3. Programs of Py-GCMS (a) and TGA (b) of tannin extract and commercial tannin

The TGA analysis of tannin extract compared to commercial tannin is shown in Fig. 3b. The tannin decomposition profile appeared to occur in two steps, at a temperature range of 25 to 100° and above 230°. The first stage can be attributed to removing moisture, volatile organic compounds, and low-temperature-boiling impurities. Furthermore, the degradation began at the second stage with the tannin weight still around 40% at temperatures up to 725°. Similar to the study of Lisperguer *et al.* (2016), the degradation was almost complete at 600 °C, and tannin showed a remaining weight of approximately 44%. The complex condensed aromatic structure of tannin leads to high thermal resistance. The decomposition profile of extract and commercial tannin are similar, but the percentage residue content of tannin extract was relatively lower at the same temperature. This indicates that tannin extract had lower thermal stability than commercial tannin. Commercial tannin had a larger phenolic hydroxyl content (Table 2), which increased the cross-linking density and created a more stable structure acting as a thermal barrier (Xie *et al.* 2021). Moreover, the tannin extract in this study had a relatively higher degree of crystallinity.

Physical and Mechanical Properties

Figure 4 shows the density and moisture content of SB roof tiles composite (SBRTC), which was 0.72 g/cm^3 before increasing approximately 10% to 0.78 to 0.80 g/cm^3 after coating with PU and PU-tannin. The statistical analysis revealed that the uncoated and coated SBRTC differed significantly ($p \leq 0.05$), while the PU coating and PU-tannin coating had the same value. However, a higher tannin content resulted in higher density, which the mass contribution of the coating materials can explain. Both Özlüsoylu (2023) and Iswanto *et al.* (2017) reported higher density after coating, with weight increment greater than dimension change. The moisture content (MC) of uncoated SBRTC compared to coated SBRTC was significantly different at 10.3% and 5.12% to 7.20%, respectively. However, there was no significant difference between PU coating and PU-tannin coating. This implies that coated SBRTC increased hydrophobicity and could resist changes in the environmental condition, as seen by reduced MC. Increasing the tannin content in coating formulations provided lower MC. The performance of wood-based panels is strongly influenced by moisture, while post-treatment conditions affected their water affinity (Copak *et al.* 2021).

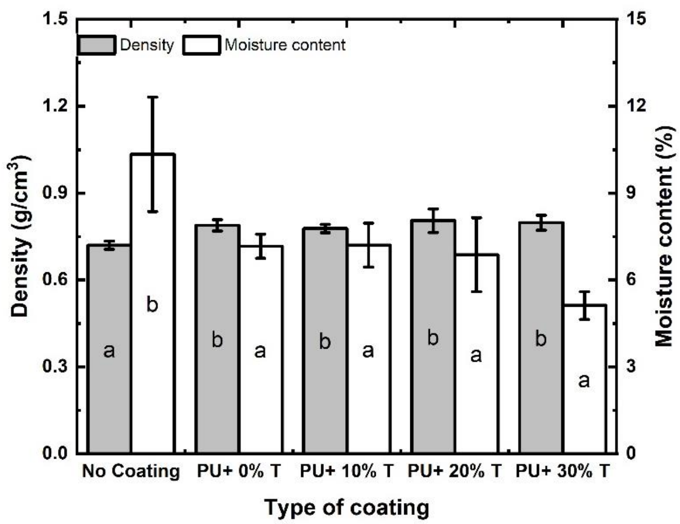


Fig. 4. Density and moisture content of SB roof tiles composite at different compositions of PU-tannin coating compared to PU-coated and uncoated samples

The dimensional stability of the SBRTC is shown by water absorption and thickness swelling (Fig. 5). After 24 h of immersion, water absorption of the uncoated sample was 80.2%, whereas the sample with PU coating was 66.7% and lower in PU-tannin coating (29.8 to 42.9%). This shows that the coating as an outer protector prevented water from being absorbed. A similar result was obtained in the previous study on aluminum alloy coated with a tannin-modified coating. When tannin was present, hydrogen bond numbers increased dramatically, delaying water absorption by more than one order of magnitude (Nardeli *et al.* 2020). Adding tannin up to 30% to the formulation resulted in significantly different water absorption than uncoated SBRTC and PU-coated SBRTC ($p \leq 0.05$).

Thickness swelling (TS) of SBRTC coated with 30% PU-tannin coating had a lower value (5.12%) than the others. Although not significantly different ($p \leq 0.05$), the coating treatment reduced the TS approximately 30 to 50% compared to uncoated SBRTC. The lowest water absorption and thickness swelling were obtained in SBRTC coated with 20% and 30% PU-tannin coating, respectively. According to JIS A 5908 (2003), particleboard should have maximum thickness swelling values of 12%, but Tabarsa *et al.* (2011) recommend 8% and 15% for two and 24 h immersion times, respectively. Even in uncoated SBRTC, the TS of the SBRTC in this study met all the requirements.

Physical properties in this study (density, moisture content, and thickness swelling) of the SBRTC satisfied the JIS A 5908 (2003), both uncoated and coated. The surface coating material affected physical properties, such as density and thickness swelling after 24 h immersion. The result showed that the thickness swelling of particleboard coated with lacquer paint is high compared to overlay with continuous press laminates. They suggest covering the panels' edges with edge bands for final-use applications (Nemli *et al.* 2005a).

The MOE and MOR of SBRTC coated with different tannin contents are shown in Fig. 6. The increased tannin content in the coating formulation contributed to higher MOE and MOR of SBRTC coated with PU-tannin coating. The MOE and MOR of uncoated SBRTC were 1.15 and 9.37 GPa, respectively. Following coating, these values rose roughly 20 to 50% and 20 to 40% for MOE and MOR, respectively. Because density and strength are known to have a positive effect, this rise was in accordance with the density increment in this study. Similar results were obtained by Nemli *et al.* (2005a). The tannin extract in this study has a crystallinity of 27.4%; this high crystallinity may contribute to increased MOR of coated samples. According to Sala *et al.* (2020), moisture content also affects mechanical properties; a slight decrease in MOE and MOR results from increased MC. This study found that coated SBRTC had a lower MC than MOE and MOR increase.

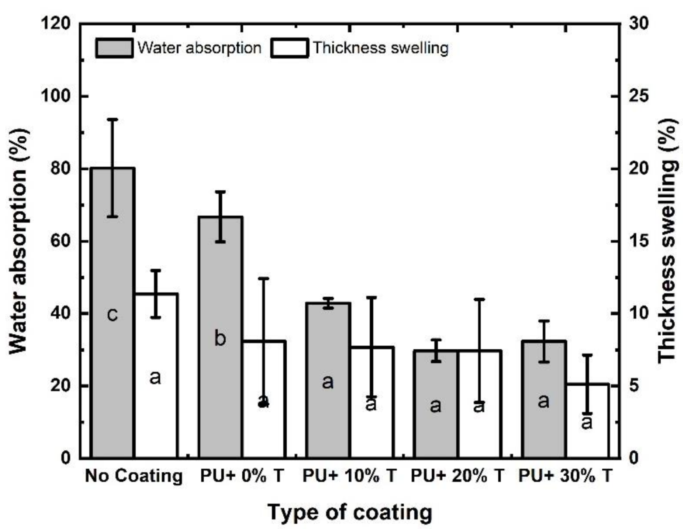


Fig. 5. Water absorption and thickness swelling of SB roof tiles composites at different compositions of PU-tannin coating compared to PU-coated and uncoated samples

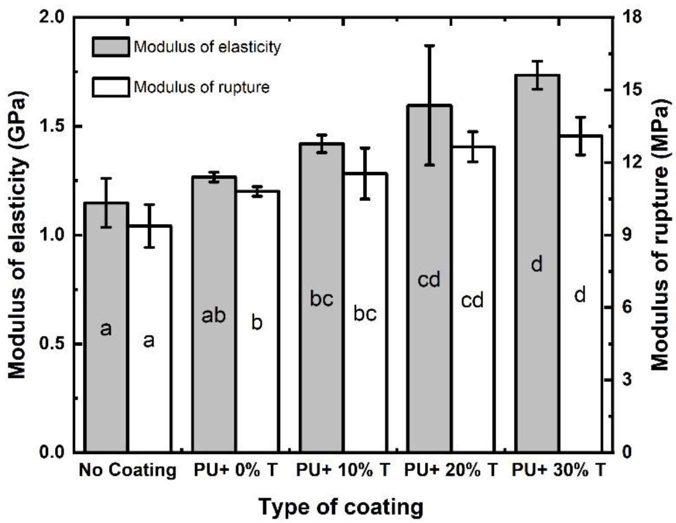


Fig. 6. MOE and MOR of SB roof tiles composite at different compositions of PU-tannin coating compared to PU-coated and uncoated samples

Surface Performance

Surface properties define the finished product's quality (Copak *et al.* 2021). Surface roughness of the uncoated was greater (4.44 µm) than coated SBRTC (1.91, 1.89, 1.87, and 1.82 µm for 0% T, 10% T, 20% T, and 30% T, respectively). These results indicated that the coating application (both PU coating and PU-tannin coating) produced a smoother surface, as shown by a lower average in R_a value (Fig. 7). Applying coating on the surface of SBRTC significantly affected the surface roughness ($p \leq 0.05$). Tomak *et al.* (2018) also found that the surface of Scots pine wood became smoother after applying a tannin containing coating, in which higher tannin content in the coating formulation contributed to a smoother surface. In the present study, low surface roughness could be accounted by total amount of coating substance penetrating the particleboard's porous structure, which strengthens the bonds and contacts between the SB particles. Furthermore, a more compact structure will be obtained. A study by Özlüsoylu (2023) indicated that the roughness values varied depending on the different types and solid content of the varnishes. Smoother and glossier surfaces were achieved on higher-density particleboard (Nemli *et al.* 2005b; Özlüsoylu 2023). In line with the results of this study, the high density of coated SBRTC (Fig. 4) had a low surface roughness value. This can be explained by lower porosity, more compactness, and tighter structure than lower-density particleboard (Nemli *et al.* 2005b). A different result was reported by Yalcin and Ceylan (2017), who showed that samples impregnated with mimosa and quebracho tannin extract showed an increase in the mean surface roughness of varnished wood.

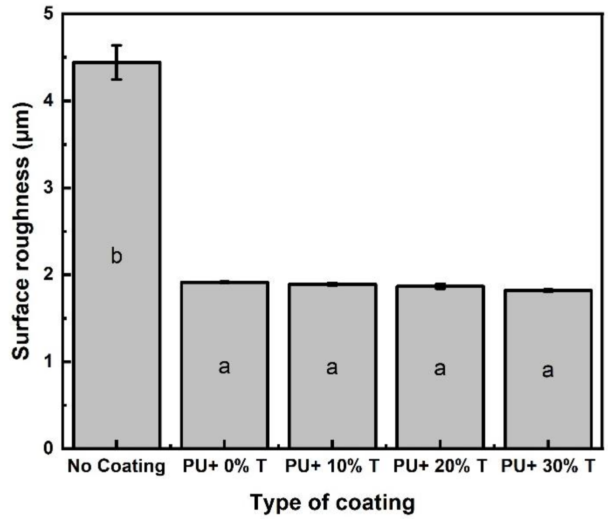


Fig. 7. Surface roughness of SB roof tiles composite after PU-tannin coating compared to PU-coated and uncoated samples

Contact angle and wettability as thermodynamic wetting parameters can be used to measure the interaction between the substrate surface and the coating material (Martha *et al.* 2024). In this study, these values are shown in Table 3. It can be seen that the equilibrium contact angles (θ) increased due to the application of coating. The equilibrium contact angles of SBRTC with PU-tannin coating were higher than those of the uncoated and PU coating. The decrease in surface roughness value led to an increase in the equilibrium contact angle value. Contact angles can also influenced by surface roughness, because a decrease in surface roughness may result in an increase in contact angle (Qin *et al.* 2015). The study's findings confirmed that a coating treatment caused the surface roughness to decrease, which increased the contact angle and decreased the wettability (low K-value). Adding tannin to the PU coating increased the equilibrium contact angle value by approximately 7%. A similar result was obtained by Gholamiyan *et al.* (2022) that particleboards and medium-density fiberboard (MDF) coating with epoxy increase water contact angle value. The high contact angle of water indicated the reduction in hydrophilic properties of coated SBRTC, resulting in a more hydrophobic surface. A contact angle value of < 90° implies that the surface is hydrophilic according to concepts of surface wettability (Barthwal *et al.* 2024; Kartikawati *et al.* 2025).

The wettability, represented by the K-value, was decreased by coating (Table 3). The K-value represents how fast the liquid spreads and penetrates the surface (Shi and Gardner 2001). The K-value of the uncoated sample was 0.027. The value then decreased with an increase of tannin up to 50% for the PU coating. Adding tannin to PU coating materials also reduced the K-value to between 0.003 and 0.009. The high contact angle of the SBRTC was an indication that water did not easily spread and penetrate the surface. The decrease could be seen in the K value. The low K value indicated that the SBRTC surface had less wettability than the untreated surface. The addition of tannin decreased the wettability, indicating that the addition of tannin was able to reduce and prevent water from spreading and penetrating the SBRTC. This result was based on water absorption and thickness swelling (Fig. 5), which was lower in coated samples. Similar results were obtained by Nemli *et al.* (2004), who found that applying Mimosa bark extract by brushing onto particleboard could reduce water absorption and diffusion. Urea-formaldehyde (UF) is used in particleboard manufacturing. Formaldehyde may interact with polyphenolic extracts to produce condensation products with a strong bonding potential. In addition, formaldehyde emission was significantly decreased.

Table 3. Contact Angle Obtained with Water Liquid and K-value of SB Roof Tiles Composite at Different Compositions of PU-tannin Coating Compared to PU-coated and Uncoated Sample

Formula	Contact Angle (θ, °)	K-value
No coating	66.373 ± 5.287	0.027 ± 0.017
PU + 0% T	68.256 ± 2.993	0.014 ± 0.006
PU + 10% T	73.396 ± 0.282	0.009 ± 0.010
PU + 20% T	70.407 ± 1.898	0.003 ± 0.003
PU + 30% T	73.233 ± 1.519	0.006 ± 0.008

Durability

The durability of SBRTC against termite and brown-rot fungi can be seen in Fig. 8. Weight loss of uncoated SBRTC was 12.4% lower than control wood of 17.7%. Furthermore, it decreased significantly around 5.71 to 5.90% in PU-tannin coating with 10 to 30% of tannin addition. Only the PU-coated sample had 9.68% weight loss. The lower weight loss indicates increased coated samples' durability compared to uncoated and control samples. According to SNI 7207 (2014), they were classified as class II (durable) compared to PU-coated at class III (moderately durable), and class IV for uncoated sample and control wood. Gonçalves *et al.* (2021) reported there was no significant difference in the weight loss percentage of particleboard bonded with urea-formaldehyde (UF) and tannin-urea-formaldehyde (TUF). However, the average mortality percentage was higher in particleboard with TUF. This is believed to be caused by the panel's higher density, more ash, and toxic extracts. Secondary metabolite bioactive substances, primarily from tannins and polyphenols, have a protective function (Masendra *et al.* 2021). The presence of phenolic content and carbamic acid with biological activity in tannin extract was verified by FTIR (Fig. 2a) and Py-GCMS analysis (Table 2). According to Yingprasert *et al.* (2023), termites were irritated by the bark extract that covered the outside of the impregnated rubberwood, and they died between the third and fourth weeks of exposure.

Biomass-based panels were found to be susceptible to moisture and attacked by wood decay fungi. Weight loss after exposure to brown-rot decay fungi of uncoated compared to PU-coated SBRTC was 20.4% and 19.8%, and it became lower in PU-tannin coated (around 11.7 to 17.8%). The highest weight loss, 48.5%, was found in control wood. The result of the study showed that coating reduced the level of brown rot attacks, which was indicated by lower weight loss. The coating treatment significantly differed between PU-coated SBRTC and PU-tannin-coated SBRTC and control wood. Tascioglu *et al.* (2013) condensed tannin extract impregnated with wood showed antifungal efficacy following testing against white- and brown-rot fungus. This was due to the high tannin content and fatty acids known to have anti-fungal properties. Furthermore, Yingprasert *et al.* (2023) rubberwood impregnated with bark extract from *Acacia mangium* exhibited much better fungal decay resistance against brown-rot fungi than white-rot fungi.

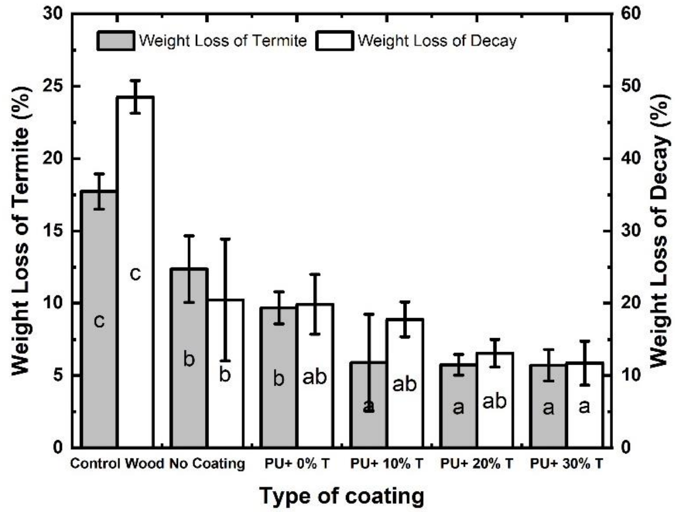


Fig. 8. Weight loss of SB roof tiles composite at different compositions of PU-tannin coating compared to PU-coated and uncoated samples after testing against termite and brown-rot fungi

CONCLUSIONS

1. The polyurethane (PU)-tannin coating performed relatively better with respect to all tested properties in comparison to PU-coated and uncoated sorghum bagasse-based roof tile composites.
2. The presence of tannins in the coating formulation increased the density, followed by an increase in mechanical (MOE and MOR) and durability properties against subterranean termites and brown-rot fungi. In certain parameter tests, while PU-tannin 30% demonstrated lower moisture content, reduced swelling, decreased weight loss, and higher mechanical properties, whereas statistical analysis revealed no significant difference compared to PU-tannin 20%.
3. A concentration of 20% tannin is the optimal addition to the coating formulation for material efficiency. In addition, the lowest wettability was obtained in this concentration.
4. Surface coating on sorghum bagasse-based roof tile composites provided a smoother surface than uncoated samples. As a result, a higher contact angle was obtained, indicating a reduced wettability sign with a low K-value then, resulting in a more hydrophobic surface.
5. However, because the contact angle in this study was less than 90°, the coating treatment can still not achieve a hydrophobic surface. Further study is required to improve coating formulation and achieve a hydrophobic surface by surface treatment.

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