Vol. 11(2): 284-297, May 2023

DOI: https://doi.org/10.23960/jsl.v11i2.704

Jurnal Sylva Lestari

P-ISSN: 2339-0913
E-ISSN: 2549-5747

Journal homepage: https://sylvalestari.fp.unila.ac.id

Full-Length Research Article

Resistance of Chip Block Pallet from Teak Wood Particle against Decay Fungi and Subterranean Termites

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ARTICLE HISTORY:

Received: 13 January 2023
Peer review completed: 5 June 2023
Received in revised form: 30 June 2023
Accepted: 5 July 2023

KEYWORDS:

Biodeterioration
Chip block pallet
Polyurethane
Teak wood particles

ABSTRACT

Wood biomass such as sawdust, particles, and chip obtained wood industry can be used as raw material for chip block pallets (CBP). This study evaluated the resistance of chip block pallets made from teak-sawn waste and polyurethane adhesive against decay fungi (white-rot and brown-rot fungi) and subterranean termites. The CBPs with dimensions of 9 × 9 × 9 cm and a target density of 0.6 g/cm^3 were cold pressed at 9.8 MPa for 4 h using several polyurethane concentrations and composition of particle sizes. The termite and decay resistance tests were conducted following JIS K 1571 2004. The results showed that CBP treated with different polyurethane concentrations and composition of particle sizes was resistant to subterranean termite attacks. However, the CBP manufactured were not resistant to white-rot and brown-rot fungi attacks, showing more than 3% mass loss percentage. Considering the efficiency of polyurethane use, the study suggested the best content for using polyurethane in the manufacture of CBP is 4.5% with a particle size composition of 50 (4-14 mesh): 50 (> 60 mesh).

1. Introduction

The global pallet market reached 6.87 billion units in 2018 (Khan et al. 2021). Over 600 million pallets, approved by the European Pallet Association (EPAL), are available for the global logistics industry. In 2019, 123 million EPAL wooden pallets and other carriers were produced, which increased by 1.2 million compared to 2018 (Khan et al. 2021). A bearing block is one of the most important components in the pallet structure. Block pallets are generally developed using solid wood, but the availability of solid wood is increasingly limited due to deforestation. Hence it is necessary to use alternative raw materials. Rigg-Aguilar et al. (2019) have successfully developed a chip block pallet using *Vitellariopsis ferruginea*, *Cordia alliodora*, and *Gmelina* *arborea* sawdust. CBP's biodeterioration against wood-destroying organisms such as termites and fungi has not been evaluated. The attack of wood-destroying organisms is quite dangerous. The quality of CBP for packaging purposes is determined by its strength and durability.

Indonesia, particularly Jepara, is one of the districts famous for its furniture industry using teak wood. The use of teak wood in Jepara reaches 2500 m^3/month. Every year, Jepara produces 17,355 m^3 of by-products consisting of flakes (22.32%), particles (9.39%), and sawdusts (8.77%), and most of these by-products are teak sawdusts (Sambe et al. 2021). Generally, wood has natural resistance against termites and fungi if it contains extractive substances toxic to both organisms (Luth 2019). Teak extractive compounds such as anthraquinone and 2-methyl anthraquinone (MAQ) are important agents against deterioration organisms such as fungi and termites (Ismayati et al. 2023). Niamké et al. (2012) also proved that teak extractives such as 4',5'-Dihydroxy-epiisocatalponol provide antifungal protection to teak wood. Anthraquinones and naphthoquinone play a role in the resistance of teak wood to insects and fungi. Naphthoquinones are believed to be more toxic to fungi than anthraquinones (Sumthong et al. 2006). The toxic content from teak wood, a by-product of teak particles, can be used as a raw material for chip block pallets because of its resistance to these organisms.

The development of chip block pallets is also inseparable from using adhesives. Rigg-Aguilar et al. (2019) developed chip block pallets using formaldehyde-based adhesives with high adhesive consumption. Some adhesives release formaldehyde emissions that affect health disorders (Beane Freeman et al. 2013; Dorieh et al. 2019; Salthammer et al. 2010). Therefore, it is necessary to develop human-friendly adhesives with high reactivity and efficient use. Polyurethane is a polymer material that contains a urethane group (-NH-CO-O-) by the reaction of polyols with isocyanates in the form of liquid (Chattopadhyay and Webster 2009; Triwulandari and Ghozali 2013; Zia et al. 2014). Isocyanate-based adhesives have advantages such as curing, better physical and mechanical properties, and zero-emission of formaldehyde (Zia et al. 2014). Particleboards from sago (*Metroxylon sagu*) with polyurethane resins have a lower mortality and mass loss percentage against deterioration organism attacks than those of the particleboard bonded with phenol-formaldehyde (Zulfiana and Kusumah 2014), and the use of 10% polyurethane content resulted in the physical and mechanical properties that met the standards (Zulfiana and Kusumah 2014). Particle geometry is one of the most important factors in the manufacture of composites. The smaller particle size will cause low bondability between particles. The smaller particles will affect the higher adhesive consumed, hence the adhesive could not cover the surface area of all particles (Santoso et al. 2022). There are many studies on CBP manufacturing, but the high susceptibility of CBP durability has not been evaluated. Therefore, this study aimed to evaluate the resistance of CBP from teak particles with polyurethane adhesive against subterranean termites and fungi attacks.

2. Materials and Methods

2.1. Materials

Teak wood (*Tectona grandis*) materials in the form of chips, particles, and sawdust were obtained from sawmills in Jepara, Central Java, Indonesia. The raw material was sifted to a size of 4-14 mesh and > 60 mesh, then dried at 80ºC until the moisture content reached 5%. Polyurethane adhesive obtained from PT. Anugrah Raya Kencana, with a solid content of 99.47% and viscosity ranging from 712.01–1524.45 mPa.s (Aisyah et al. 2023; Hariz et al. 2023), was used as a matrix in the manufacture of CBP.

2.2. Manufacturing of Chip Block Pallets

The chip block pallets (CBP) were manufactured in two stages (**Table 1**). The first stage evaluated the effects of polyurethane adhesive content of 2.5%, 3.0%, 3.5%, and 4.5% with a particle size composition of 50:50. Particles were mixed with a mixer and sprayed with adhesive using a spray gun (**Fig. 1**). Then, the sprayed particles were mat formed using a forming box with a size of 9 cm × 9 cm × 9 cm and a target density of 0.6 g/cm^3. The mat was then cold pressed at a specific pressure of 9.8 MPa for 4 h. Conditioning was carried out for seven days at room temperature (Kusumah et al. 2016). The second stage evaluated the effects of the composition of coarse particles and powder particles, as described in **Table 1**.

Table 1. Manufacturing formulation of CBP

Manufacture	Formulation	Notes
Raw materials	*Tectona grandis*	
Adhesive	Polyurethane	
Target density	0.6 g/cm^3	
Size	9 cm × 9 cm × 9 cm	
Press conditions	Cold press, 4 h, 9.8 MPa	
Stage one (various polyurethane content)	2.5% 3.0% 3.5% 4.5%	Particle size 4 – 14 mesh and > 60 mesh
Stage two (Various particle size compositions)	Optimal polyurethane Content 4.5%	100% 4-14 mesh 100% > 60 mesh 50% 4-14 mesh: 50% > 60 mesh 70% 4-14 mesh: 30% > 60 mesh 30% 4-14 mesh: 70% > 60 mesh

Fig. 1. The manufacturing of chip block pallets.

2.3. Termite Resistance Test

The bioassay test of the CBP against subterranean termites (*Coptotermes curvignathus*) followed the method from JIS K 1571:2004 for determining the effectiveness of wood preservatives and their performance requirements. 150 worker termites and 15 soldier termites were placed into the acrylic cylinder containing CBP samples (**Fig. 2**). Rubberwood (*Hevea brasiliensis*) sample was used as a control (Ismayati et al. 2011; Meisyara et al. 2021; Tarmadi et al. 2014). The forced-feeding test was conducted for 3 weeks and incubated under dark conditions at 28°C and 70% humidity. The observation was done after 3 weeks. Following the termite inspection, the test samples were cleaned and dried in an oven at 60°C for 48 h. The test samples were then weighed to obtain the mass after the test. The mass ratio after testing (W_2) and before testing (W_1) was calculated to determine the level of attack by subterranean termites using Equation 1, and mortality was calculated using Equation 2.

$$Mass\ loss\ (\%) = \frac{W_1 - W_2}{W_1} \times 100\% \quad (1)$$

$$Mortality\ (\%) = \frac{Amount\ of\ termite\ mortality}{150} \times 100\% \quad (2)$$

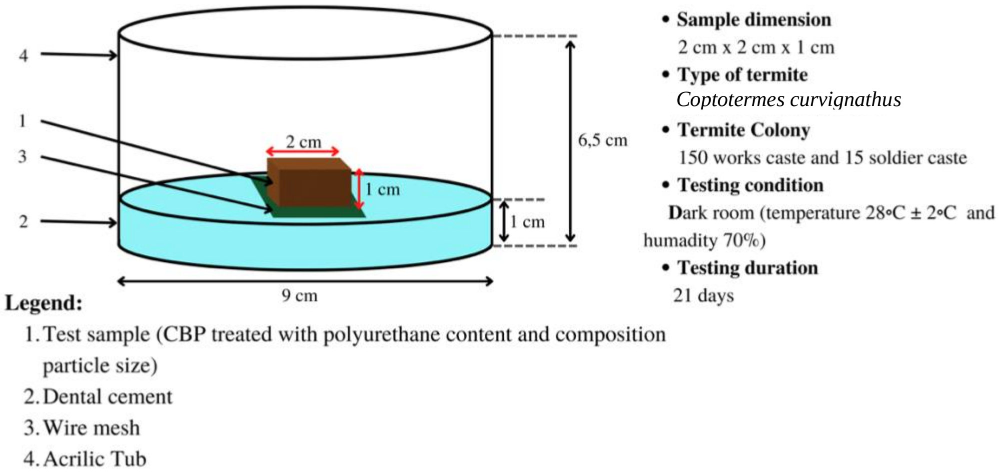

Fig. 2. Non-choice feeding test against subterranean termite.

2.4. Decay Properties Test

The decay test method referred to JIS K 1571, 2004 (**Fig. 3**). The decay test was tested with the white-rot fungi (*Trametes versicolor*) (FFPRI 1030) and brown-rot fungi (*Fomitopsis palustris*) (FFPRI 0507). The fungal cultures were grown on potato dextrose agar for seven days. Then the fungal culture was inoculated into several jars, each jar containing 150 g sterile sand and 45 ml sterile basal medium. All jars were incubated at room temperature for ten days until the mycelium covered the entire surface of the sand medium. The composition of the basal medium per liter was as follows: 3 g of KH_2PO_4, 2 g of $MgSO_4 7\ H_2O$, 25 g of glucose, 5 g of peptone, 10 g of malt extract, and 1000 ml of distilled water. Sterilized CBP specimens were inserted into the jars aseptically and then incubated at room temperature for 12 weeks. Rubberwood block was used as a control (Ismayati et al. 2011; Meisyara et al. 2021; Tarmadi et al. 2014). This test used nine replicates. The mass loss percentage (%) was calculated based on the difference between the initial weight of the specimen (W_1) and the final oven dry weight (W_2). The mass loss percentage (%) was calculated using Equation 1 to indicate the degree of fungal attack.

2.5. Fourier Transform Infrared Spectroscopy (FTIR) Analysis

The Fourier transform infrared spectroscopy (FTIR) was carried out to determine changes in chemical groups after testing CBP's durability. The test samples were ground using a mortar. Spectra were obtained with an FTIR spectrophotometer (FTIR 4000, PerkinElmer Inc., USA).

Spectrums are recorded at wavenumbers ranging from 400 to 4000 cm^1 and 400 cm^{-1} with a resolution of 4 cm^{-1}.

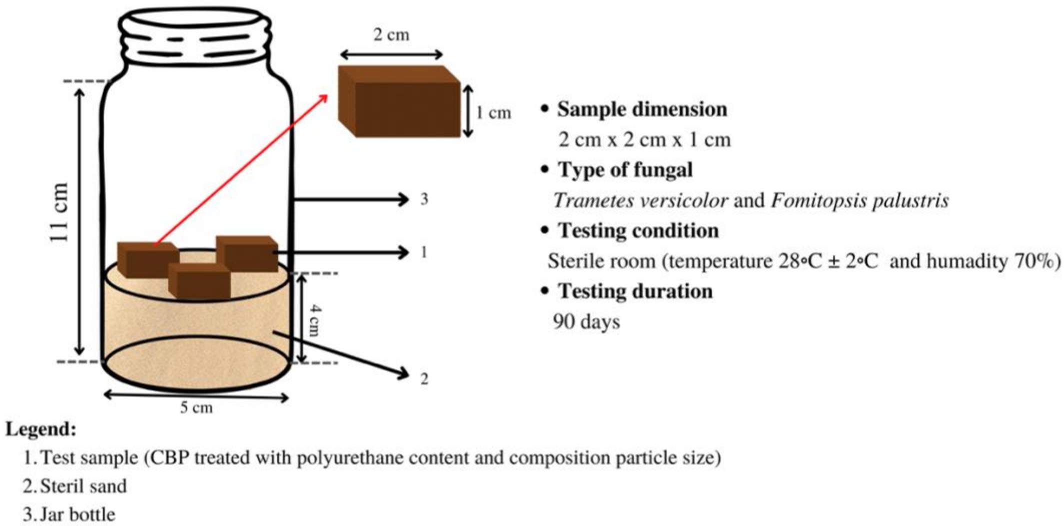

Fig. 3. CBP resistance test to the decay.

2.6. Field Emission Scanning Electron Microscopy (FESEM) Analysis

FESEM (Thermo Scientific – Quattro S) was used to study the surface morphology of the samples after testing the durability of CBP. Samples were prepared with a size of 50 mm × 50 mm × 6 mm and cut laterally about 0.05 cm. Then the samples were tested with magnifications of 1500x, 2000x, and 8000x.

2.7. Statistical Analysis

Each test's data were examined, such as polyurethane content (2.5%, 3.0%, 3.5%, and 4.5%) and particle size composition (4-14 mesh, > 60 mesh, 50 (4-14 mesh): 50 (> 60 mesh), 70 (4-14 mesh): 30 (> 60 mesh) and 30 (4-14 mesh): 70 (> 60 mesh). Analysis of variance (ANOVA) was used to assess the importance of the variation between the variables and levels to determine whether groups had substantially different means at the 95% confidence level. The Duncan's multiple range test (DMRT) was used to compare the means. IBM Statistical Product and Service Solution Version 23 was used for statistical analysis.

3. Results and Discussion

3.1. The Resistance of CBP against Subterranean Termite Test

The resistance of CBP to subterranean termite infestation (*C. curvignathus*) was observed in terms of mass loss and mortality (**Fig. 4a**). CBP with polyurethane content of 4.5% and particle size composition of 50:50 resulted in the lowest mass loss of 1.52%. In comparison, the highest was 2.92%, with a particle size composition of 30:70. The statistical test results showed that polyurethane content and particle size composition did not significantly affect the mass loss due to termite attacks. Based on JIS K 1571 2004, the CBPs produced with various polyurethane contents and particle size composition were resistant to subterranean termite attacks. This behavior showed the resistance of the teak wood due to the extractive content. Extractive substances in teak wood contain lapachol, 1-hydroxy-2-methyl-anthraquinone, 2-(hydroxymethyl) anthraquinone, tectoquinone, anthraquinone, and naphthoquinone compounds that are involved in teak wood resistance to insects and fungi (Niamké et al. 2012; Sumthong et al. 2006). It is supported by the mass loss of rubberwood samples (control) that was much greater than that of the CBP samples, indicating the presence of termite-repellent extractive components in teak wood. Lukmandaru et al. (2018) stated that teak wood fed to subterranean termites had a mass loss of 8.70%. Compared to CBP, the mass loss due to subterranean termite attacks was lower.

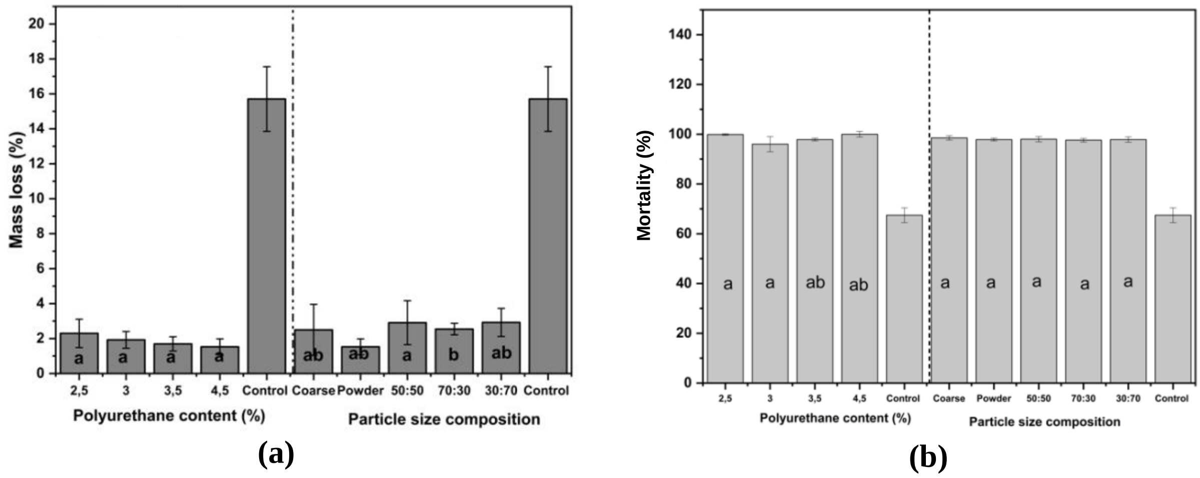

Fig. 4. CBP after subterranean termite tests: (a) mass loss and (b) mortality (the results of statistical tests denoted by the letters a, b, c, and d of the same letter indicate the absence of noticeable differences).

In addition, the use of polyurethane adhesive affected the termite mortality (**Fig. 4b**). The results showed that the termite mortality from all the samples treatment was not significantly different compared to the control. This study also indicated that the termite mortality of CBP treated with polyurethane adhesive increased as increasing the adhesive content. It is in accordance with the research of particleboards made from sago with polyurethane adhesive (Zulfiana and Kusumah 2014). When compared to CBP, CBP's mortality percentage was better. Polyurethane adhesive has toxic properties that cause termite mortality (Bagheri et al. 2022). Aisyah et al. (2023) mentioned that polyurethane has a benzoic acid compound. Benzoic acid and its derivatives are of great interest because of their pharmacological properties, such as antioxidant, radical regulator, antiviral, antitumor, anti-inflammatory, antimicrobial, and antifungal (Cozzolino et al. 2023).

Polyurethane adhesive has good insecticidal properties compared to other adhesives used in composite products, as seen from the termite mortality rate. Compared to phenol-formaldehyde adhesives, polyurethane adhesives are better at protecting the level of termite attacks (Zulfiana and Kusumah 2014). Teak wood also has a good natural resistance against termite attacks, showing termite mortality of more than 90% (Lukmandaru et al. 2018). In most cases, a decrease in wood mass loss is associated with termite mortality (Hassan et al. 2017). However, in some cases, the effect is caused by repellency or antifeedant effects (Alavijeh et al. 2014). There is an effect of mixing teak wood particles and polyurethane adhesive on the termite resistance of particleboard specimens. Gauss et al. (2019) stated that the difference in particle size used could affect the quality of the CBP produced. Smaller particle sizes make the adhesive dissolve imperfectly, making bonding between particles easy. Particle sizes that are too small or fine require a sufficient amount of adhesive because the surface area of the particles is bigger (Desiasni et al. 2022). The adhesive surface of large particles requires a higher amount of adhesive than smaller particles adhesive surface (Ulfah et al. 2015). Previous research explained that a 4-20 mesh particle size has good physical and mechanical properties (Syahfitri et al. 2022). CBP with a composition of 4-14 mesh has better susceptibility to termite attack because the polyurethane adhesive dissolves better than using a composition > 60 mesh. Teak wood particles contribute to the specimen's resistance to termites. It is known that the diet of subterranean termites consists of anything made of or containing cellulose. Termites feed on dead wood and wood by-products; however, they may excavate and damage various non-cellulosic materials in their search for food (Terzi et al. 2009).

3.2. The Resistance of CBP against Fungi Attacks

The resistance of CBP with various polyurethane contents against white-rot and brown-rot fungi is presented in **Fig. 5**. Based on the test results, the mass loss of samples due to white-rot fungi attacks were 6.41–10.87%, while those of the brown-rot fungi attacks were 5.82–7.82%. Furthermore, the mass loss of CBP treated with composition particle size due to white-rot fungi attack was 6.41–8.04%, and those of the sample tested using brown-rot fungi had a mass loss range of 5.82–8.18%. The mass loss of all CBP samples after white-root and brown-rot fungi attacks was lower than that of the rubberwood samples (control). However, the mass loss value of all CBP did not meet the standard JIS K 1571 2004 (≤ 3%).

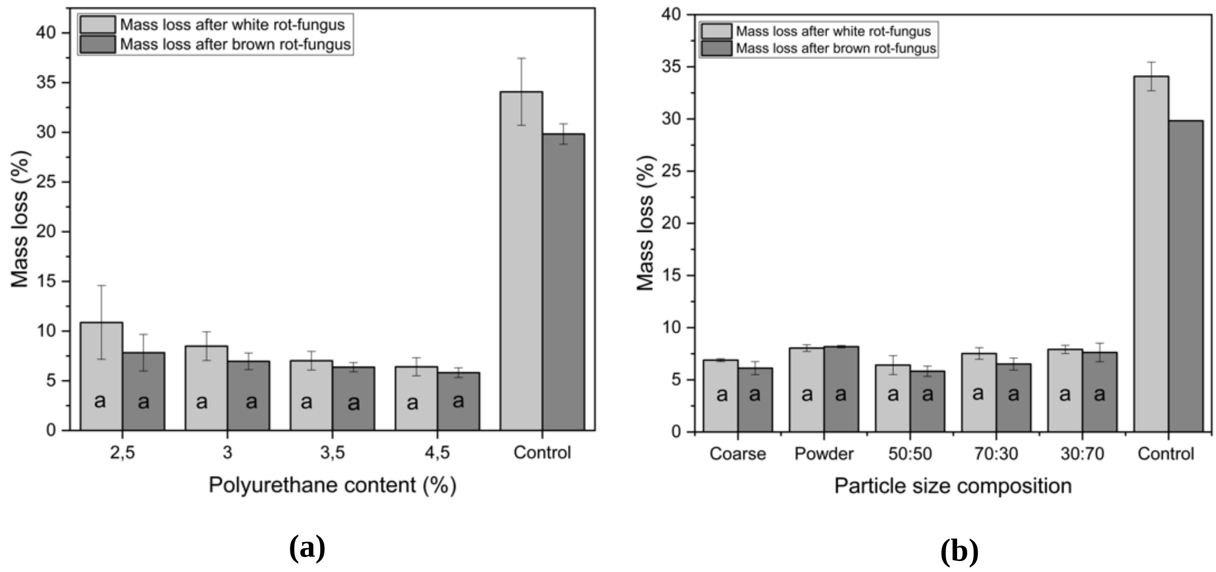

Fig. 5. Mass loss of CBP after white-rot and brown-rot fungi tests: (a) polyurethane content, (b) particle size composition (the results of statistical tests denoted by the letters a, b, c, and d of the same letter indicate the absence of noticeable differences).

The ANOVA results showed that the polyurethane content and particle size composition in the manufacture of CBP did not significantly affect the resistance against fungi attacks. The white-rot fungi can aggressively degrade all components used to manufacture composite (Gonçalves et al. 2021). Polyurethane is one of the adhesives susceptible to degradation. The different degradation patterns of polyurethane are attributed to many polyurethane properties, such as molecular orientation, crystallinity, crosslinking, and chemical groups present in the molecular chain that determine accessibility to degrading enzyme systems (Kay et al. 1991). The regularity in synthetic polymers allows the polymer chains to pack easily, resulting in the formation of crystalline regions. This restricts the accessibility of the polymer chains to degradation, whereas the amorphous area of polyurethane can be more easily degraded (Howard 2002). Bher et al. (2022) and Huang and Roby (1986) observed polyurethane degradation to take place selectively, with amorphous areas degrading before crystalline regions. Polyurethane with long repeating units and hydrolytic groups would be less likely to pack into highly crystalline regions and are more accessible for biodegradation (Howard 2002). A microbial attack on polyurethane could occur through the enzymatic action of hydrolysis, such as ureases, proteases, and esterases (Carr et al. 2020; Skariyachan et al. 2017; Wilkes and Aristilde 2017).

During brown-rot fungi attacks, cellulose and hemicellulose are degraded while lignin remains intact (Polman et al. 2021). The exposed control specimens destroyed the cell wall layer due to cellulose and hemicellulose degradation. The control specimens showed massive penetration of fungal hyphae leading to significant mass loss (**Fig. 5**). White-rot fungi can preferentially or simultaneously degrade lignin, cellulose, and hemicellulose. The degradation rate depends on the wood, fungal species, and the relative content of cellulose, hemicellulose, and lignin in the wood. Mycelial mass increases during degradation. The results showed that after 90 days, the composite underwent natural decay.

3.3. FTIR Analysis

The FTIR spectrum of the control sample and CBP samples after termites and fungi attacks detected an absorption peak of 1721 (**Fig. 6**). This peak is assigned to the C=O stretching derived from the carboxyl group and the C=O ester group (Kusumah et al. 2016, 2017b; a). Polyurethane absorption was seen at the peak of 1536 cm^{-1}-1603 cm^{-1}, according to the C(O)-O ester linkage in the polyurethane polymer (Trovati et al. 2010). The polyurethane absorption peak decreased when CBP was exposed to the decay test. The detected carbonyl and hydroxyl groups prove that there are extractive components that repel termites and fungi, such as the active compounds lapachol, 1-hydroxy-2-methyl-anthraquinone, 2-(hydroxymethyl) anthraquinone, tectoquinone, anthraquinone, and naphthoquinone compounds. Naphthoquinone and its derivatives exhibit antimicrobial, antibacterial, fungicidal, cytotoxic, antiparasitic, insecticidal, and anti-ulcerogenic activities (Babula et al. 2009).

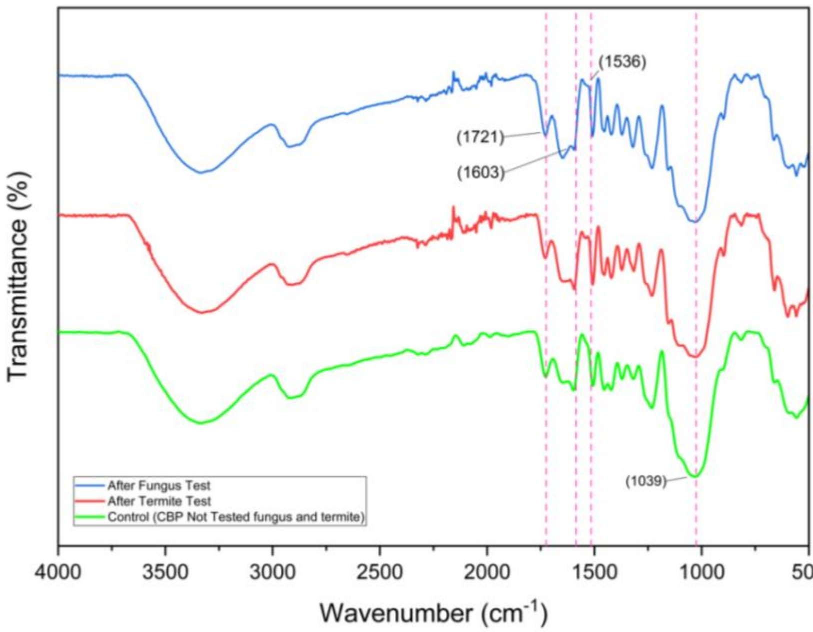

Fig. 6. Fourier Transform Infrared Spectroscopy (FT-IR) of CBP.

Relatively large hemicellulose and lignin content are still present in the raw material. The lignin fraction was shown from the C=C aromatic ring strain at the peak of 1500-1603 cm^{-1}, while the hemicellulose fraction was shown by the acetyl and uronic ester groups at the peak of 1735 cm^{-1} (Bari et al. 2015). The lignocellulosic component degraded as indicated by absorption from 1039 cm^{-1} - 1847 cm^{-1}. The fungus has been shown to degrade cellulose and cause mass loss in CBP. The results of CBP functional groups made from teak wood and polyurethane using FTIR are the presence of alcohols, hydrocarbons, and double bonds or aromatic rings (Syafri et al. 2022).

3.4. FESEM Analysis

An illustration of CBP morphology after being tested for fungal and termite weathering using FESEM is shown in **Fig. 7**. The bioassay test results of the CBP against subterranean termites (*C. curvignathus*) indicated termite infestation occurred in the test samples (**Fig. 7a**). The lignocellulosic content in the test samples are preferred by termites, causing damage to the test samples (Xu et al. 2015). **Fig 7a** shows that after attacking CBP, the termites die. This indicated that polyurethane is an effective barrier agent in the CBP against termite attacks, as mentioned in the previous studies (Awad et al. 2023; Rajendran et al. 2010). Teak wood contains natural preservatives such as tectoquinone, anthraquinone, and naphthoquinone compounds (Babula et al. 2009), which act as a barrier agent, making CBP resistant to subterranean termite attacks. Polyurethane and natural preservatives maintain the moisture of CBP by binding to hydroxyl groups, preventing CBP from being wet and not preferred by termites.

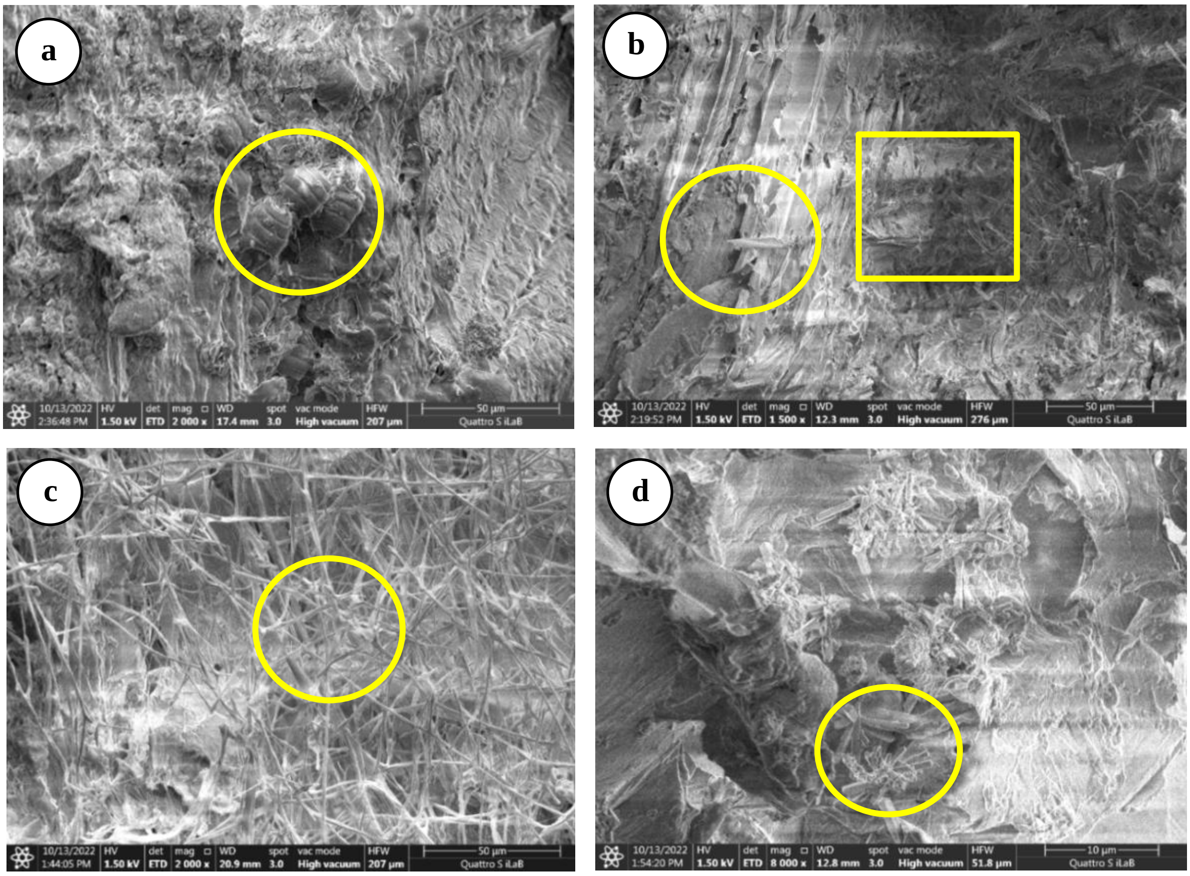

Fig. 7. Morphology of CBP with treatment 50 (4-14 mesh): 50 (> 60 mesh), polyurethane content 4.5%: (a) CBP after subterranean termite attacks, (b) morphology of CBP with visible mold infestation and without infestation, (c) CBP after brown-rot fungi infestation, and (d) CBP after white-rot fungi infestation.

Fig. 7b shows that the fungi attacked some of the CBP surface (yellow square) but there are also parts of the surface that are not affected by fungal attack (yellow circle). The decay of CBP by brown-rot fungi is shown in **Fig 7c**, which shows the fungal attack almost covers all parts of the CBP. In contrast to the brown-rot fungi attacks, the white-rot fungi exhibited non-covering fungal hyphae. It tended to colonize the infected areas **Fig. 7d** (Bari et al. 2015). Wetzstein et al. (1999) reported that the activity in the material attacked by white-rot fungi is allocated to enzymes such as lignin peroxidase, lacquer, and manganese peroxidase which catalyze damage through the diffusion of oxidative agents or specific mediators.

4. Conclusions

The variation in the polyurethane content and particle size composition did not significantly affect the resistance of chip block pallets against white-rot and brown-rot fungi attacks, showing the mass loss percentage of CBP of more than 3%. It did not comply with the JIS K1571 (2004). However, CBP is resistant to subterranean termites and complied with the JIS K 1571 (2004) with a mass loss percentage of less than 3%. The mass loss of CBP decreased with the increase in polyurethane content and variation in particle size. Considering the efficiency of using polyurethane, the best content for using polyurethane in the manufacture of CBP is 4.5% with a particle size composition of 50 (4-14 mesh): 50 (> 60 mesh). Furthermore, a study is needed on the application of polyurethane adhesives as a coating as a protective agent for CBP against deterioration organisms.

Acknowledgments

This research was supported by the Research Center for Biomass and Bioproducts, the National Research and Innovation Agency (BRIN), Indonesia, and the Integrated Laboratory of Bioproducts (iLaB).

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