Effect of water-soluble chitosan content and target density on the properties of corn husk particleboard

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Abstract
Particleboards are wood-based composite panel products produced from lignocellulosic materials, which can be combined with adhesive and hot-pressed into sheets for various purposes. However, the most common adhesive is based on formaldehyde and has been reported to be detrimental to health and the environment. This indicates that there is a need to develop environmentally friendly alternative adhesive with low toxic effects. Therefore, this study aims to evaluate the feasibility of water-soluble chitosan (WSC) as an adhesive in particleboard fabricate based on corn husk as well as to determine the influence of its content and target density variations on the properties of particleboard. The target density of board was varied at 0.6, 0.8 and 1.0 g/cm^3. The variation of WSC content used was 6, 8 and 10% based on the air-dried particles weight. The manufacture of particleboard was made under a press temperature of 180^o C for 15 minutes under 2.5 MPa press pressure. The results showed that WSC is proven as a natural adhesive for particleboard. The properties of particleboard were shown to be affected by the adhesive content and variation of target density. Particleboard with 8% of WSC content and target density of 0.8 g/cm^3 were resulted best properties compared other compositions, relatively.
Keywords: adhesive, corn husk, particleboard, target density, water-soluble chitosan

1 Introduction

Particleboards are wood-based composite panel products obtained from lignocellulosic materials, which can be combined with adhesive and hot-pressed into sheets [1,2]. Furthermore, these products are widely used for various purposes, including furniture, partitions, insulation, and other non-structural components of buildings.
Broad the use of particleboard can be linked to the economy advantage of the low cost of wood raw materials, inexpensive agent and simple processing [3]. The demand for these products has increased substantially worldwide, representing 57% of the total wood-based panel consumption, that percentage continues to grow at a rate of 2-5% per year [4, 5].
The use of lignocellulosic materials, such as wood particles for the production of particleboards is still dominant, approximately until 95%. However, due to the shortage of wood in the panel industry, increased environmental concerns, and interest in sustainable manufacturing based on renewable resources, several studies explored other alternatives, such as agricultural by-products [6, 7]. In this context, several studies succeeded in developing substitutes for wood particles using non-wood and agricultural by-products, such as rice husk [8], sugarcane bagasse-corn cob [9], oil palm trunk [10] and coconut husk [11].
One of the raw materials from agricultural by-products is an attractive alternative to substitute wood due to the numerous useful product properties of which they are the main components is corn husk. Corn husk biomass appears to be a promising candidate for raw materials in particleboard production due to its abundance as an

agricultural by-product, which has no other utilization except animal food and handycrafts in a limited quantity. The data in 2021/2022, Indonesia has corn production area was 3.9 million hectares with yield of 3.26 metric tons per hectare and corn production until 12.70 million metric tons, it is possible to obtain approximately 1.27 to 5.08 million metric tons of corn husk biomass, which potential to be developed as particleboard raw materials [12]. Pandecha et al. [13] reported a study that corn husk has cellulose (47%), hemicellulose (43.96%), lignin (4.13%) and ash (2.93%) which important as components in particleboard manufacture.

Particleboard production typically involves the use of formaldehyde-based adhesive, such as urea-formaldehyde (UF) and phenol formaldehyde (PF) as the main adhesive. However, the use of these adhesives causes problems for the environment and consumer health [14]. This condition encourage highlighting the need for suitable and non-toxic alternative material of adhesive. An effective alternative material is water-soluble chitosan (WSC). WSC is a derivative of chitosan and has been synthesized in various structures. WSC is generally obtained by adding various functional groups to the chitosan back chain which helps to make chitosan obtain polymers with water-soluble properties. Synthesis of WSC is carried out by modifying the chitosan back chain with the right chemical compounds [15]. Due to the limitation of acetic soluble chitosan (ASC) as an adhesive for wood panels and composite products, there has been interest in developing a chitosan derivative, particularly WSC for particleboard fabricate, but there are no studies on its application.

As a product based on lignocellulosic materials, the particleboards are susceptible to decay fungi, particularly outdoors [16]. However, composite wood product such as particleboard generally tends to have better resistance to decay than natural solid wood [17, 18]. The resistance of particleboard against wood decay fungi, including white rot and brown rot, can vary depending on the specific type of particleboard and the materials used in its construction.

Particleboard products are typically designed to resist moisture, insects, and decay more than solid wood. The manufacturing process often involves treating the wood fibers or particles with preservatives or chemicals that enhance their resistance to decay and other forms of deterioration. These treatments can help protect the particleboard against fungal growth, including white rot and brown rot.

Therefore, this study aims to evaluate the feasibility of WSC as an adhesive in particleboard fabricate based on corn husk as well as to determine the effect of WSC content and target density variations on the physical, mechanical, thermal and biological (decay resistance) properties of particleboards.

2 Materials and Methods

2.1 Raw materials preparation

Corn husk used in this research was collected from the local industry in Cibinong, West Java, Indonesia. Corn husk were cleaned from impurities materials during direct drying under the sun. After that, the corn husk were cut into particles by a Pallmann ring knife flaker mill then classified using vibrating screens to obtain uniform-sized particles (4 to 14 mesh in size). Water-soluble chitosan (WSC) for particleboard adhesive was supplied from CV. ChiMultiguna, Cirebon, Indonesia. WSC used in this study had a particle size of < 150 mesh, moisture regain of < 9.75%, average molecular weight of 153 kDa, deacetylation degree of > 95%, and viscosity of 52.88 mPas, respectively.

2.2 Particleboard fabricate process

In the particleboards fabricate, the WSC content was variated of 6%, 8% and 10% (w/w). The adhesive was prepared by dissolving chitosan in distilled water with a ratio of 1:10 (w/v). Furthermore, the corn husk particles were mixed with different concentrations of WSC solution using a drum mixer machine until homogenous. The mixtures obtained were placed in a technical oven at 80°C for 12 h to reduce the moisture content for avoid the occurrence of blisters during the pressing process. They were then formed into molds using a forming box to produce particleboards measuring 30 x 30 x 0.9 cm. The production of particleboard was completed using a SHINTO hot-pressing machine, with a press pressure of 2.5 MPa, temperature of 180°C, and target density variations of 0.6 g/cm^3, 0.8 g/cm^3 and 1.0 g/cm^3 for 15 minutes.

2.3 Evaluation of particleboard properties

2.3.1 Determination of morphology

The morphology of raw materials which used in this study are corn husk and water-soluble chitosan, also particleboard that produced were viewed and observed using a 3D Digital Microscope of Keyence VHX 6000.

2.3.2 Physical and mechanical properties

The test samples used for the analysis of physical and mechanical properties were prepared and conditioned for 1 week at room temperature of 22 to 24°C and relative humidity of 60 to 70%, then assessed based on the JIS A 5908:2003 [19] and ASTM D 1037-1999 [20]. The physical properties such as actual density and thickness swelling were evaluated using the gravimetric method. Modulus of rupture (MOR), modulus of elasticity (MOE), internal bonding (IB) and screw holding power (SHP), were evaluated as the mechanical properties using a SHIMADZU universal testing machine (UTM) type AG-IS 50 kN. The assessment of the samples was carried out with 5 repetitions.

2.3.3 Evaluation of thermal conductivity

Potential of particleboard served as a heat insulator was assessed through a thermal conductivity (TC) test. The TC measurement of board samples measuring 120 x 25 x 0.9 cm was carried out with a Conductometer (QTM-500) Probe PD-11 at a room temperature of 22 to 24°C and relative humidity of 60 to 70%.

2.3.4 Decay resistance test

The test samples with the best physical and mechanical properties from assessment 2.3.2 were selected for evaluation of decay resistance. The decay resistance (biological properties) of untreated and treated particleboard sample against white rot (*Trametes versicolor*) was done in accordance with JIS K 1571 standards which was carried out by Kartal et al. [18]. The tests were conducted in Integrated Laboratory, National Research and Innovation Agency, Indonesia. The white rot fungus was maintained in Potato Dextrose Agar (PDA). The decay test was conducted in a bottle jar (300 ml) provided with quartz sand amended with JIS media. The media was sterilized using steam at 121°C and 1.1 A for 20 min before being transferred to the pre-sterilized petri jar. The seven-day-old fungi inoculated on the surface of the media in the jar. The jars were kept at 26°C and 70% relative humidity until the media surfaces were completely colonized by the fungi. Treated and untreated wood samples were sterilized before being placed on the surface of the inoculated test fungi in the bottle jar. Subsequently, samples were kept in contact with the fungus for 12 weeks at 26°C and 70% relative humidity in an incubator chamber. After 12 weeks of exposure, the wood samples were weighed once the surface mycelium on the wood samples was cleaned. The mass loss of all samples was determined by the difference between the dry weight of each specimen before and after the decay test.

3. Results and discussion

3.1 Morphological characteristics

The micrograph of corn husk (A), WSC (B), and particleboard (C and D) is presented in Figure 1. The results showed that the surface of the corn husk appeared as a rectangular epidermal cell with squiggly anticline walls. Small cells were also observed on the surface of the sample, and they were filled with silica and cork substances. Meanwhile, the micrograph of the inner part of the particleboard showed that WSC covered the corn husk particles, and they appeared shiny brown (C and D). WSC had a relatively variable molecular weight, which increased its compatibility with incorporated materials, such as corn husk. Larnoy et al. and Singh et al. [21, 22] stated that chitosan and its derivatives, such as WSC, with low to medium molecular weight penetrated the wood cell wall, lumen, and intercellular capillary space.

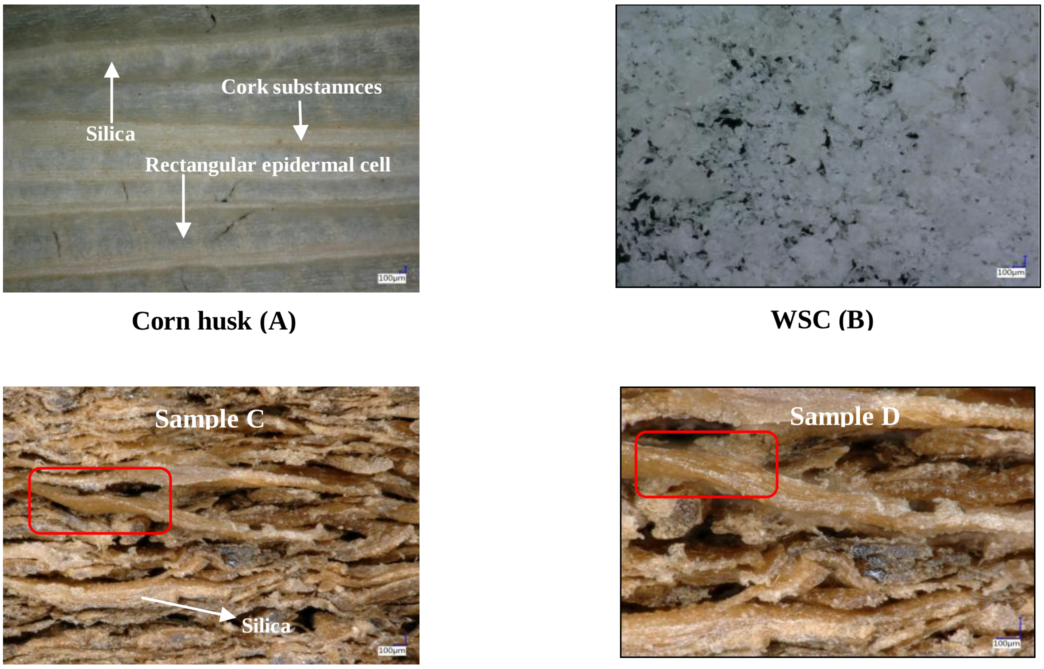

Fig. 1 The surface appearance of corn husk particles (A) (magnification 50x), WSC (B) (magnification 100x), and the inner part of particleboard (sample C with magnification 100x and sample D with magnification 200x)

3.2 Physical and mechanical properties

The actual density of particleboards are presented in Figure 2, which ranged from 0.53 to 0.99 g/cm3. The target density was arranged at 0.6, 0.8 and 1.0 g/cm^3, but in the fact some of the samples did not achieve this target density. The results showed that the particleboard with target density of 0.8 g/cm^3, gave the best results and reached the target compared to other combinations. According to the results from actual density of particleboards which obtained, no significant differences among treatments were found.

This phenomenon may be related with loss of adhesive during application and material loss during the manual formation of the particleboard fabricate, as well as during the pre-pressing and hot-pressing steps. Melo et al. [23] also reported this fact and attributed factors such as loss of adhesive at the application time due to adhesion on the edges of the drum and differences in the specific mass and moisture content of particles. As we know, the type and amount of adhesive used can also affect the quality of particleboard. The quality of particleboard is increased by increasing the amount of adhesive due to better adhesive distribution on the particles and increasing the connection and compatibility points between particles and adhesive [24].

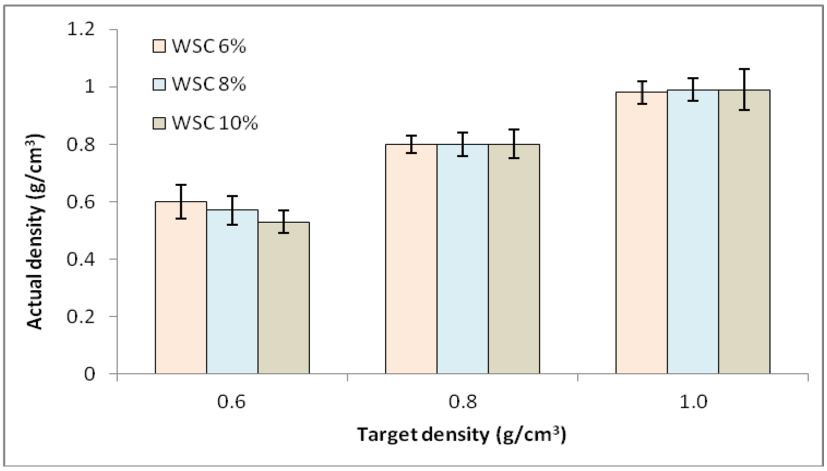

Fig. 2 Actual density from particleboard

The thickness swelling (TS) of particleboard after soaked in water for 24 hours was viewed in Figure 3. The results presented that particleboard pressed with target density of 0.6 g/cm^3 produced the lowest TS (36.31 to 66.8%) compared to others. However, the values obtained from the samples did not meet the JIS A 5908:2003 standard, namely a maximum of 12%. The increase in WSC content from 6 to 8% led to a decrease in the TS value, relatively. A slight increment was observed in samples with 10% WSC, which were pressed at target density of 1.0 g/cm^3. Generally, the graph revealed that the target density with the least TS value was 0.6 g/cm^3 and the increasing TS trend occurred at 1.0 g/cm^3. This result was related to the actual density value, which increased along with the target density. Maraghi et al. [25] reported that the increasing of adhesive content, the dimensional stability of particleboard made of poplar wood mixing citrus branches and beech twigs particles improved partially. However, by increasing the density, water absorption of particleboard decreased but its thickness swelling increased.
Melo et al. [23] also argued that the highest percentages of water absorption from particleboards which related to thickness swelling properties can be attributed to the high content of silica and to the lower content of lignin in these materials. Such components interfere with the particles' adhesion and gluing processes, such as silica, or with the increase on the proportion of celluloses and polyoses chemical constituents with hydrophilic behavior, such as lignin.

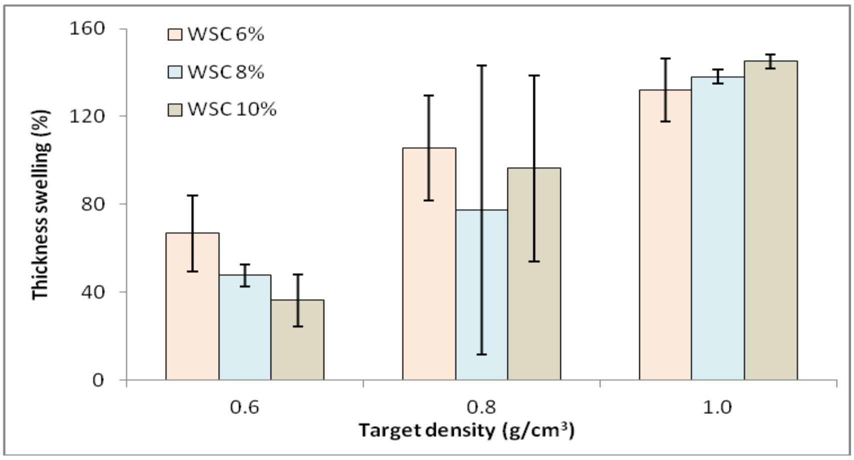

Fig. 3 Thickness swelling of particleboard

In this case, the low bulk density of the corn husk particles needs a greater amount of particles to form the particleboard, causing a higher compression rate in the pressing time and a greater amount of hygroscopic sites, with the particleboard being more vulnerable to water absorption and dimensional instability which can be seen from the thickness swelling value. The small size particle of corn husk can also affect the TS properties. Bazzetto et al. [26] stated that the adhesive content may not fully cover the surface area of the smaller particle, therefore they do not form an effective barrier to the water absorption by the fiber cell wall in relation to the larger particles with greater coating capacity.

The bending properties of the samples, including modulus of rupture (MOR) and modulus of elasticity (MOE) were presented in Figures 4 and 5. The MOR served as an indicator of strength, flexibility, and rigidity, while the MOE described the stiffness of the particleboards. The value of MOR from particleboards ranged from 0.82 to 14.23 MPa which only on board fabricated using WSC of 8 to 10% with pressured at target density of 0.8 g/cm^3 and 1.0 g/cm^3. As for, the MOE value of board ranged from 0.02 to 3.43 GPa. The satisfactory MOR (11.73 MPa) and MOE (3.43 GPa) value and met the JIS A 5908:2003 standard were obtained from particleboard which pressured at target density of 0.8 g/cm^3 and WSC content of 8% than the board which other combinations, respectively. The increased resistance of the particleboards with a higher density is related to the higher compaction rate due to the accommodation of the particles in the pressing process.

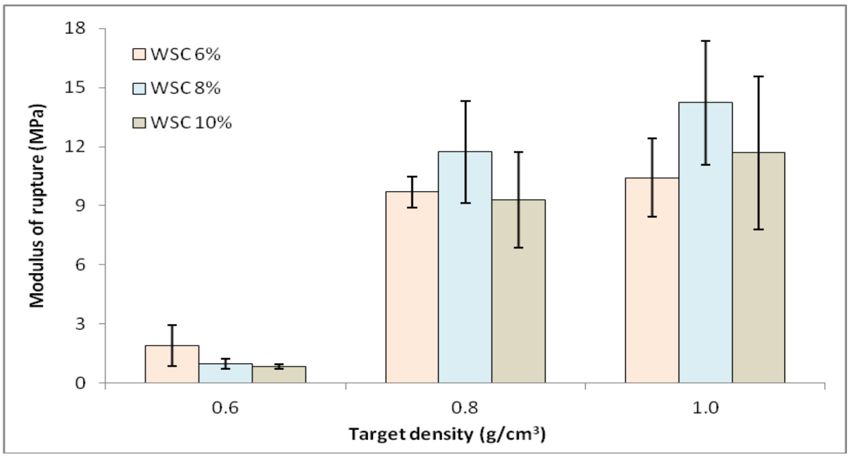

Fig. 4 Modulus of rupture value from particleboard

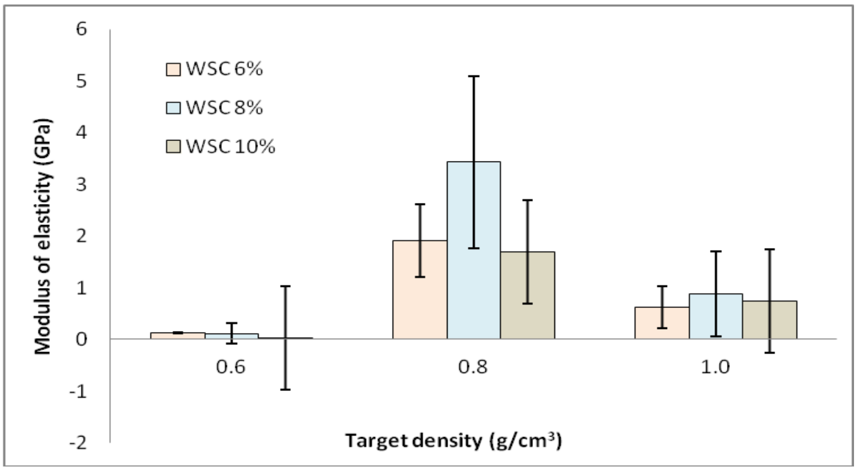

Fig. 5 Modulus of elasticity from particleboard

By analyzing data in Figure 4 and 5, it was possible to observe that the content of WSC increased in particleboards, there was a tendency for the MOR and MOE values to decrease. It is assumed that the adhesive content applied was not enough to cover the larger surface area of smaller-size particles from corn husk, and therefore the increase in the contact area among particles did not result in sufficient number of adhesive bonds. Brito et al. [27] stated that a greater contact area of the particles is promoted with the increase in the compaction rate, as well as a greater slenderness ratio with a reduction in empty spaces. Based on these results, corn husk was feasible and had the potential to be developed as an alternative raw material for particleboard fabricate bonded with WSC as an adhesive, thereby improving the bending properties.

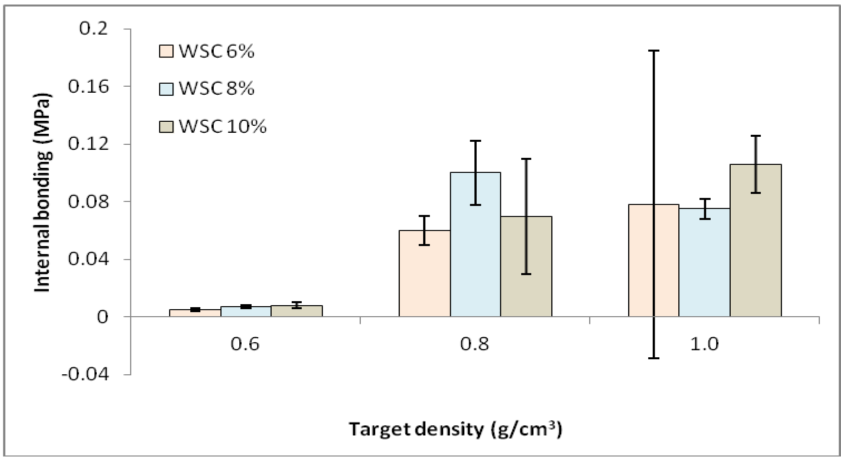

Fig. 6 Internal bonding average value of particleboard

Internal bonding (IB) is one of the important for qualitative characterization of particleboard. This parameter indicates how significant the interaction is between the particles and the adhesive used, and directly related to all other physical-mechanical properties determined in this research. The internal bonding (IB) values of the particleboard ranged from 0.005 to 0.10 MPa presented in Figure 6. Overall, its TS value did not meet JIS A 5908:2003 standard with require minimum value of 0.15 MPa. However, the IB value of particleboard pressured at target density 0.8 g/cm3 and glued with WSC of 8% was slightly higher compared to other combinations. The low values of IB may be related to the form of corn husk particles. Youngquist et al. [28] mentioned that the low permeability of particles affected on the particles' gluing.

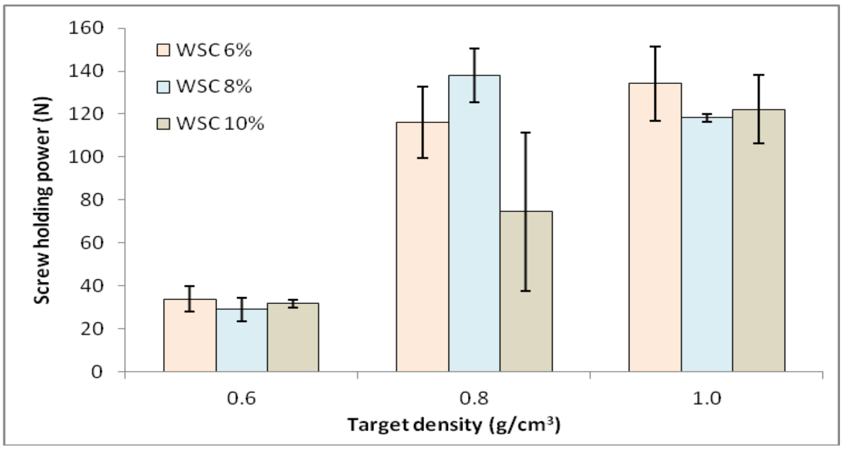

Fig. 7 The value of screw holding power from particleboard

The screw-holding power (SHP) of particleboards indicate a tendency to increase the surface screw withdrawal. Figure 7 showed that the SHP value of particleboard ranged from 28.91 to 138.02 N. Overall, the values was obtained did not met the JIS A 5908:2003 which mentioned the minimum standard value is 300 N. These results may have been affected by the lower density of corn husk particles and the low availability of adhesive per particle due to its greater specific surface area. Particleboards with target density of 0.8 g/cm^3 and WSC content of 8% produced the better value of SHP (138.02 N) compared to other combinations, respectively. Similar study results were also found by Calegari et al. [29] for particleboards made from bamboo-wood and by Melo et al. [30] for wood-rice husk particleboards.

A similar trend occurred on physical and mechanical properties of corn husk particleboards was also resulted in some studies which used agricultural by-products and non-wood particles as raw materials for particleboard manufactured such as rice husk [31], sugarcane-bamboo [32] and bamboo [26]. A higher proportion of coarse fibers resulted in interlacing of individual fibers, in the formation of air gaps, and in a lower bulk density [33].

The bulk density of the raw material affects the compression ratio (board density divided by bulk density of the raw material) in panel fabricate. Eventually, this condition affected the properties of particleboard that has been manufactured.

3.3 Thermal conductivity

Thermal conductivity (TC) is a parameter that is needed in the implementation material related to conductors, electric wires, heat exchangers and so on. TC is also required for classify a material in the type of conductor or insulator. The value of TC from particleboards with WSC content and pressing target density variations was presented in Figure 8 that ranged from 0.166 to 0.228 W/m K. From Figure 8, it can be assumed that when WSC content and target density increases, TC increases slightly as well. However, as density corresponds to porosity, increased porosity can transfer heat easier, such as at density of 0.6 g/cm3. While at density of 0.8 g/cm3, porosity of material is very little, resulting in higher TC. Overall, as these values are less than 0.25 W/m K, so it potential to use and safely as insulation material [34].

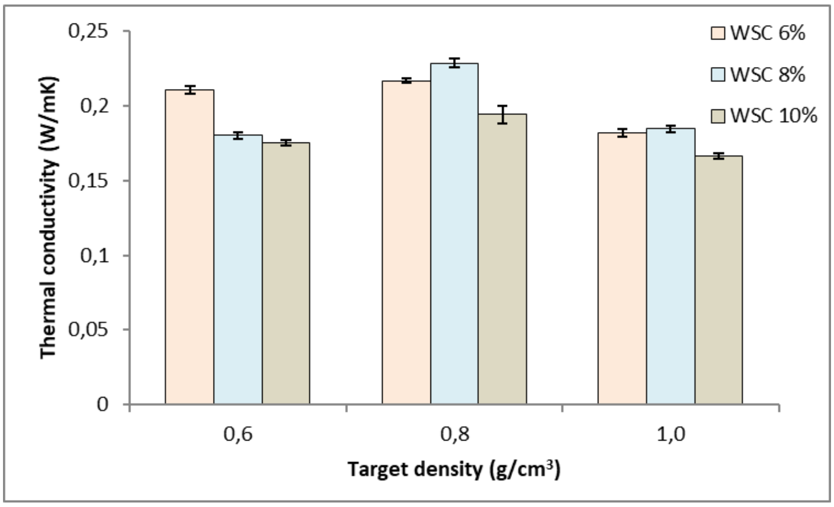

Fig. 8 Thermal conductivity value of particleboard

The TC of particleboards depending on density and were not comparable to natural insulation materials such as wood fibers, flax and synthetic materials. The study conducted by Pandecha et al. [13] presented that insulation board made of corn husk with lower density (0.1 to 0.4 g/cm3) had TC ranges from 0.0287 to 0.0316 W/m K.

3.4 Decay resistance of particleboard

According to Figure 9, the average weight loss of particleboards of both A and B (34.09 and 34.12%, respectively) are lower than that of natural solid wood as control sample (37,88%) which means both samples were more resistant to wood decay fungi, a white rot, than the sample C and natural solid wood (control). Meanwhile, particleboard C showed the highest weight loss (47.53%) than that other samples and natural solid wood. The particleboard A and B showed a similar performance against the white rot fungi, *T. versicolor*. In other words, treatment on both samples (WSC 6% and 8%) improves the toughness of both samples, indicating properties enhancement of A and B compared to control solid wood.

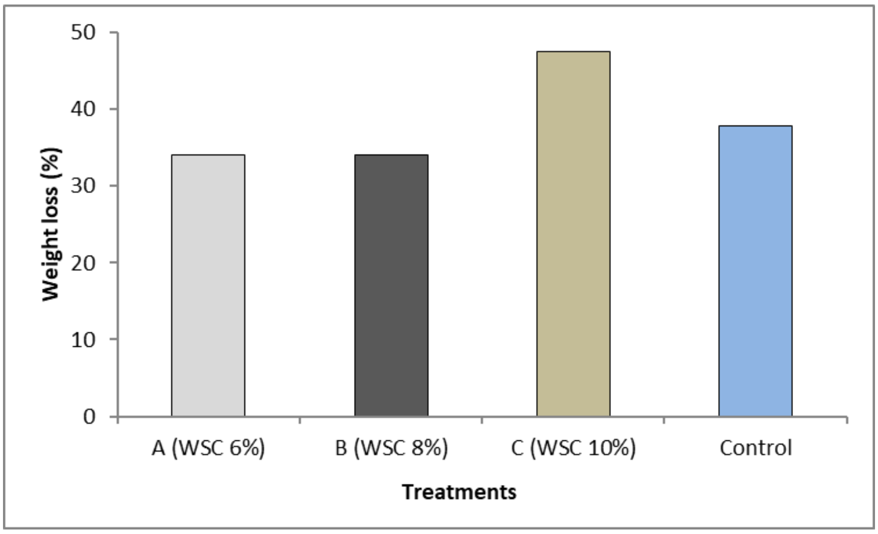

Fig. 9 Weight loss in the specimens subjected to wood rot fungi, *Trametes versicolor,* after 12 weeks of exposure.

The white rot fungi, *T. versicolor,* is a member of Basidiomycetes fungi. The Basidiomycetes are known to use lignin and cellulose as a food source. Hence, *T. versicolor* primarily attacks and decomposes lignin [35]. In general, the fungal attack is always accompanied by high humidity. It was found that the wood-plastic composites indicated 50% of mass loss after intensive action of moisture measured in the culture chamber of about (70 ± 5)%, i.e., for 16 weeks, at (22 ± 2) °C. Then, it was accompanied by changes in bending strength amounting to 30–46% of the final test result [36].

Furthermore, composite wood products such as particleboards are often engineered to have a more consistent density and moisture content, which can reduce the likelihood of fungal attacks. The adhesives or binders used in the construction of particleboard can also provide additional protection against decay by sealing the wood fibers or particles and reducing moisture penetration. It's important to note that the specific resistance of particleboard against wood decay fungi can vary depending on factors such as the type of composite wood, the preservatives or treatments used, and the environmental conditions to which the product is exposed. Therefore, it's recommended to consult the manufacturer or supplier of the specific particleboard product for detailed information on its resistance to wood decay fungi and any necessary maintenance or treatment requirements.

A study stated that the major factors degrading the mechanical properties of the tested composite under detected field exposure conditions were elevated temperature and moisture exposure, whereas UV radiation from the sun had a low impact, if any, on the flexural properties of a WPC board exposed for almost 10 years [37, 36]. Furthermore, study on the decay mechanism of wood composites mentioned that the formation of larger voids present in the material through the connection of microvoids may accelerate the decay process. This condition occurs because of an action of moisture combined with elevated temperature. It would supply the moisture needed for fungal growth and create larger voids, assumably formed by the interconnection of smaller voids, then the decay process was accelerated [38, 37].

4 Conclusion

Particleboards were manufactured using corn husk and water-soluble chitosan as adhesive. Processing combination between WSC of 8% and target density of 0.8 g/cm^3 resulted in the best physical and mechanical properties compared others, respectively. The results also showed that its bending strength (MOR and MOE) values met the JIS A 5908 (2003) standard. Although the samples did not perform as expected in terms of thermal insulation material, they still exhibited good properties. Results indicated that corn husk fiber as particleboard raw material proved to be more suitable insulation material than fiber glass. Treatment by WSC as particleboard adhesive on both samples (A and B) improves the toughness of both samples, indicating properties enhancement of A and B compared to control solid wood. The future proposed research is to improve TS and enhances MOR, MOE, IB, SHP, decay resistance and the compatibility of the materials by increasing WSC concentration and combine it with another additives such as citric acid and lignin.

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