Modification of rice biomass wastes for eco-friendly particleboard

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Abstract. Rice husk and rice straw are some of abundantly available agricultural wastes that could substitute the utilization of wood as particleboard raw-materials. Nevertheless, recent studies show that rice husk and rice straw instigated low mechanical and physical properties of particleboard. Modification of rice husk and rice straw particle was investigated in order to improve particleboard properties by immersing rice husk and straw in boiling water (100°C) for 0.5, 1, 2 h and in Na₂CO₃ (80°C) for 1 h, and combination of rice husk as face and back layers and rice straw as core layer. The boards were manufactured under pressing conditions of 200°C for 10 min used citric acid and sucrose as adhesive. Pre-treatment of 1 h boiling water of particle resulted the highest quality of all properties and satisfy the JIS 5908 (2003) requirement in Modulus of Rupture (MOR) and Internal Bonding (IB). Combining rice husk as face and back layer and rice straw as core layer resulting in IB improvement. Compared to the other adhesives, citric acid and sucrose adhesives showed better mechanical and physical properties for rice husk and rice straw particleboard manufacturing.

1. Introduction

As an agricultural country with a people that consumes rice as staple food, there will be abundant biomass waste such as rice husk and rice straw in Indonesia. For every ton of rice resulting 0.23 ton of rice husk [1]. Several researches have been conducted in order to utilize rice husk and straw as raw material for manufacturing particleboard. Yang et al. [2] composing particleboard made from wood and rice straw with the optimum condition was 10/90 wood to rice straw. The maximum rice straw content suggested without reducing bending strength was 20%. Boquillon et al. and Li et al. [3,4] proposed high compatibility of wheat straw and oil-based resin adhesive due and MDI for wheat straw and rice straw respectively because urea formaldehyde adhesive exhibit much poorer properties of particleboard. Melo et al. [5] found that rice husk caused in a reduction particleboard quality because of the high percentage of water absorption and thickness swelling that can be related to the high content of silica. Cylindrical and hollow structure of rice husk act as a barrier in resin application, and also the low permeability could instigate low physical and mechanical properties.

As an alternative of UF adhesive, Umemura et al. [6] proposed utilization of citric acid as natural adhesive as wood based panel adhesives. Citric acid is contained in citrus fruits such as lemons and limes and also commercially produced by fermentation of glucose or sucrose-containing materials. Generally citric acid is used in foods, beverages, and pharmaceuticals. Acacia bark powder
and citric acid molding resulted high water resistance and good bending properties. Ester linkage between carboxyl groups derived from citric acid and carboxyl groups of bark component brought adhesiveness causing good physical properties. The addition of sucrose was also implemented by Umemura et al. [7] as an attempt to enhance the bond performance because of the adhesion area in the particleboard is smaller than that in the wood flour molding. Resin content of 20 wt% with 0.8 g/cm³ density and manufactured at 200 °C [8] was chosen as optimal condition to achieve high physical and mechanical properties. Kusumah et al. [9] proposed the addition of pre-dried treatment (12 h at 80 °C) to the sprayed particle to increase physical and mechanical properties of sweet sorghum bagasse particleboard. The optimum condition for manufacturing sweet sorghum bagasse particleboard with citric acid and sucrose adhesive was proposed 10/90 of citric acid/sucrose ratio [10] in 200 °C temperature of pressing for 10 min [11]. For the reason mention above it is important to improve rice husk and straw particleboard properties using alternative natural adhesive by particle pre-treatment and composition arrangement.

2. Materials and Method
2.1. Sample Preparation
Rice (Oryza sativa) husk and straw in this study was obtained from West Java Indonesia. The hammer mill was used to crush the cylindrical and hollow structure of rice husk, meanwhile the rice straw particles were obtained by using chipper and knife-ring flaker machine. The resulted particles were screened and those that remained between aperture size of 1.4 – 4.7 mm were used as a raw material. The rice husk particles were pretreated by soaking for 0 (control), 0.5, 1, and 2 h in boiling water, and 1 h soaking in Na₂CO₃ at 80 °C in order to remove the silica content of the rice husk. The particles were then dried in an oven at 80 °C for 24 h; at that condition, the moisture content was less than 4%. Citric acid and anhydrous sucrose of extra purity grade were purchased from Nacalai Tesque Inc. (Kyoto, Japan) and used without further purification. Citric acid and sucrose were dissolved in water at a concentration of 59 wt%, and that mixture solution was used as adhesives.

2.2. Manufacturing of Particleboard
Citric acid and sucrose content, resin content, pressing temperature, and time were decided upon based on the result from Kusumah et al. [10], where the mixture ratio of citric acid to sucrose was 10:90, resin content was 20 wt%, and the hot press condition was 200 °C for 10 min with specific pressing pressure was 6.5 MPa. The rice husk and straw particles were sprayed with adhesive and then the particles were dried at 80 °C for 24 h to reach moisture content of approximately 10%. The particles were mat-formed using a forming box then was hot-pressed. The target size of manufactured particleboard was 300 mm x 300 mm x 9 mm, and the target density was 0.8 g/cm³. The composition of particleboard manufacturing in this study are present in Table 1.
Table 1. Particle composition and pre-treatment condition of particle board.

| Sample group | Particle composition | Pretreatment | Duration of pre-treatment |
|--------------|----------------------|--------------|----------------------------|
| HB0          | Rice husk            | -            | -                          |
| HB0.5        | Rice husk            | Boiling water 100 °C | 0.5 h               |
| HB1          | Rice husk            | Boiling water 100 °C | 1 h                    |
| HB2          | Rice husk            | Boiling water 100 °C | 2 h                    |
| HN1          | Rice husk            | Na₂CO₃ 80 °C | 1 h                          |
| S0           | Rice Straw           | -            | -                          |
| HSH0         | Face: Rice husk      | Boiling water 100 °C | 1 h               |
|              | Core: Rice straw     | -            | -                          |
|              | Back: Rice husk      | Boiling water 100 °C | 1 h               |
| HSH1         | Face: Rice husk      | Boiling water 100 °C | 1 h               |
|              | Core: Rice straw     | Boiling water 100 °C | 1 h               |
|              | Back: Rice husk      | Boiling water 100 °C | 1 h               |

2.3. Evaluation of particleboard properties

After being conditioned at room temperature and approximately 60% relative humidity for 7 days, the particle board were evaluated its physical and mechanical properties according to Japanese industrial standards for particleboard [12] by 5 replications for each properties. Physical properties of the particleboard such as thickness swelling (TS) and water absorption (WA) was measured of each board after immersion at 20 °C for 24 h using a 50 mm x 50 mm x 9 mm specimen. Following the TS test, thickness changes were measured under a cyclic-accelerated aging treatment (drying at 105 °C for 10 h, warm water immersion at 70 °C for 24 h, drying at 105 °C for 10 h, boiling water immersion for 4 h, and drying at 105°C for 10 h). Mechanical properties of the particleboard, i.e., the modulus of rupture (MOR) and the modulus of elasticity (MOE), were investigated by conducting a static 3-point bending test on a 200 mm x 30 mm x 9 mm specimen of each board in dry condition used 150 mm effective span and 10 mm/min loading speed. The internal bonding (IB) and screw holding (SH) test was performed on a specimen of the same size as those used in TS and WA test. The data was then analyzed statistically using univariate analysis of variance to evaluate the significance of each treatment. Duncan post hoc multiple comparison of observed means was performed to identify which treatment were significantly different from the other treatment. Statistical analysis allowed classifying result into some categories. Treatment that not connected by the same letter are significantly different at 95% confidence level.

3. Results and Discussion

3.1. Physical properties of particleboard

Characteristic of the eight pre-treatments of particleboard were investigated. Figure 1 and Figure 2 show the moisture content and density of rice husk and straw particleboard. The moisture content of the particleboard ranged from 4.9 to 9.1% meanwhile the density ranged from 0.72 to 0.80 g/cm³. Particleboard containing straw particle showed higher moisture content than the other particleboard. All of the particleboard were satisfied the type 18 JIS 5908 (2003) standard [12] unless for the sample without pretreatment (HB0).
Figure 1. Effect of modification of rice husk and particle composition on moisture content of particleboard

Figure 2. Effect of modification of rice husk and particle composition on density of particleboard

Thickness swelling of particleboard were varied from 17.88 to 37.91%, meanwhile water absorption of particleboard was varied from 43.24 to 74.21% as shown in Figure 3. Rice husk particle with 1 h boiling pre-treatment modification showed the lowest thickness swelling and water absorption or results best dimensional stability (17.88%), even it was still above the required JIS 5908 (2003) standard [12]. Modification of rice husk by boiling for 0.5 and 2 h did not improve the dimensional stability of the particleboard than the unmodified rice husk particle. Utilization of rice straw and combination of rice straw and rice husk resulting in low dimensional stability of particleboard as well. Boquillon et al. [3] reported 17 wt% PTP (polymeric material from triglycerides and polycarbonic acid anhydrides) adhesive rice straw particle board resulted 25% of thickness swelling. Melo et al. and Kwon et al. [5,13] reported urea formaldehyde adhesive of rice husk particleboard resulted 49.57% and 23.70% of thickness swelling and 67.25% and 82.40% of water absorption respectively. Compared to the other adhesives, citric acid and sucrose adhesives seems more suitable for rice husk particleboard.
Figure 3. Effect of modification of rice husk particle and composition on thickness swelling and water absorption of particleboard

Cyclic-acceleration aging treatment was investigated to observe the thickness and weight change. The first step treatment (immersion at 20 °C for 24 h) data was the same as that present in thickness swelling data. Further step instigates high thickness change, generally 1 h boiling pre-treatment show the lowest thickness change in every step. At the end of cycle, particleboard consist of rice straw particle resulted in the lowest thickness change (44.53%) followed by 1 h boiling pre-treatment modification which resulted 59.68% of thickness changed (Figure 4), meanwhile Kusumah et al. [10] reported sweet bagasse particleboard with the same hot press condition and resin content resulted approximately 12% thickness change. High permeability of rice husk could resulted higher thickness swelling [5].
Further step also provokes high weight change in wet condition, and lower weight in dry condition. There was weight reduction in the end of cycle in all treatment. Generally, 1 h boiling pre-treatment showed the lowest weight change in every step. At the end of cycle, 1 h boiling pre-treatment resulted 41.7% of weight change, meanwhile Kusumah et al. [10] reported sweet bagasse particleboard with the same hot press condition and resin content resulted approximately 70% weight change. Weight reduction of particleboard at the end of cycle can be explained by the formation of water-soluble brown pigment called caramelen when sucrose was heated to 200 °C [14].
3.2. Mechanical properties of particleboard

MOE and MOR of particleboard were varied from 1.36 to 1.97 GPa and 6.67 to 9.21 MPa respectively (Figure 6 and Figure 7). Rice husk particle with 1 h boiling pre-treatment showed the highest MOE and MOR even it was still below the required JIS 5908 [12] standard for the MOE. Rice husk pre-treatment of 1 h boiling water increased the MOE and MOR 36.8% and 39.5% respectively than the untreated particleboard and showed significant improvement statistically. Pre-treatment of rice husk particle with 1, 2 h water boiling, Na$_2$CO$_3$, and rice straw particle composition met the required type 8 JIS 5908 [12] standard and improved the mechanical properties of particleboard. Three layered particle board composed of 1 h water boiling of rice husk as face and core layer and untreated rice husk as core layer showed higher mechanical properties than the particleboard composed of untreated rice husk. Meanwhile, utilization of 1 h water boiling as core layer showed lower mechanical properties than the rice husk control particleboard. Li et al. [4] reported rice straw particle board with MDI adhesive resulted approximately 2.5 GPa of MOE and 22 MPa of MOR. Kusumah et al. [10] reported sweet bagasse particleboard with the same hot press condition and resin content resulted approximately 5 GPa of MOE and 30 MPa of MOR. Boquillon et al., Li et al. Charoenwon and Pisuchpen [3,4,15] claimed that increasing size of particle resulting higher MOE.

![Figure 5. Weight change of particleboard on different modification and composition of rice husk and straw](image-url)
Figure 6. Effect of modification of rice husk and composition of particle on MOE of particleboard

Figure 7. Effect of modification of rice husk and composition of particle on MOR of particleboard

Internal bond strength (IB) and screw holding power (SH) of particleboard were varied from 0.14 to 0.49 MPa (Figure 8) and 35.31 to 137.19 N (Figure 9), respectively. Modification of rice husk particle with 1 h boiling pre-treatment and three layered rice husk and straw show significant improvement of IB and met the required type 18 JIS 5908 [12] standard with 167.3-200.5% increase than the untreated rice husk particleboard. All the treatment did not met the SH required type 18 JIS 5908 [12] standard with modification of rice husk particle by boiling in water for 1 h showed the highest SH (128.7% increase), meanwhile application of three layer particleboard showed lower SH than the single layer untreated rice husk particleboard. Previous research of untreated rice husk and rice straw used urea formaldehyde, MDI, and PTP adhesive resulted lower IB, 0.02, 0.45, and 0.3 N respectively [3,4]. Binderless rice straw powder particleboard reported by Kurokochi and Sato [16] also resulted lower IB (lower than 0.2 MPa). Kusumah et al. [10] reported sweet bagasse particleboard with the same hot press condition and resin content resulted approximately 1.1 MPa of IB. There was a tendency that addition
of bigger size particle, i.e. rice straw will increase IB than that used smaller size particle i.e. rice husk. Meanwhile Boquillon et al., Li et al., and Charoenwong and Pisuchpen [3,4,15] claimed that smaller size of particle resulting higher IB.

![Figure 8](image1.png)

**Figure 8.** Effect of modification of rice husk and composition of particle on internal bonding of particleboard

![Figure 9](image2.png)

**Figure 9.** Effect of modification of rice husk and composition of particle on screw holding of particleboard

4. Conclusion
Particleboard composed of modified rice husk with citric acid and sucrose as adhesive showed promising physical and mechanical properties. Pre-treatment of 1 h boiling water of particle resulted the highest quality of all properties and satisfy the JIS 5908 requirement in MOR and IB. It can be suggested that the silica content has been successfully reduced by soaking rice husk in boiling water for 1 h. Further boiling duration and application of Na$_2$CO$_3$ could be expected to lead degradation of rice husk resulting in lower mechanical physical properties. Combining rice husk as face and back layer and rice straw as core layer resulting in IB improvement. Compared to the other adhesives, citric acid and sucrose adhesives showed better mechanical and physical properties for rice husk and rice straw particleboard manufacturing.
5. References

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