Effect of pre-drying time and citric acid content on *Imperata cylindrica* particleboards properties

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Abstract. The utilization of natural fibers for eco-friendly particleboards production has become very important due to the decreased of wood resources and the human health concern. In this study, *Imperata cylindrica* particles and citric acid were used as the raw material and natural adhesive, respectively, for the particleboards production. Since citric acid solution was sprayed onto *Imperata* particles during particleboards production, the pre-drying treatment was needed to eliminate moisture from particles before hot pressing. The effect of pre-drying time and citric acid content on the physical and mechanical properties of the particleboards was investigated. The boards were produced under pressing conditions of 200°C for 10 min. The citric acid content was varied of 10, 15, 20 wt%. The pre-drying time was for 0, 6, 12 h. The size and target density of boards were 300 x 300 x 9 mm and 0.8 g/cm³, respectively. The physical properties of boards produced using particles after 12 h pre drying time were superior to those of the boards produced using particles after 0 and 6 h pre drying time. The thickness swelling of particleboards with 20 wt% citric acid and pre drying time of 0, 6, 12 h were 5.31%, 3.69%, 2.04%, respectively, fulfilled the requirement of the JIS A 5908:2003. The infrared (IR) spectral analysis showed the presence of ester linkage, representing that the carboxyl groups of citric acid had reacted with the hydroxyl groups of the *Imperata* particles, providing the boards good physical properties.

1. Introduction

The demand for particleboards is continuously increasing accordance to the demand for housing and furniture. In 2015, Indonesia’s particleboard production was 8.31 million m³ and increased to 23.57 million m³ in 2016 [1]. Meanwhile the production of wood log as particleboard raw material was decreasing. Wood log production in 2015 was 43.87 million m³ in 2015 and decreased to 42.25 million m³ in 2016 [1]. The utilization of forest wood log for particleboards production is also being considered as non-environmentally friendly, since it need quite a long time to grow a tree, and will sacrifice the forest ecosystem. For that reason, particleboards mill begun to utilize others lignocellulose sources as particleboards raw materials. One of lignocellulose source is alang-alang (*Imperata cylindrica*). It is an aggressive and perennial grass that is distributed worldwide in the tropical and subtropical region [2]. Due to the high content of cellulose (40.22%) and lignin (31.29%) [3], alang-alang was potentially to be utilized as raw materials for particleboard production. In South East Asia, there was around 35 million ha area covered by alang-alang and 8.5 million ha in Indonesia, especially outside Java island [4].

Alang-alang has been blended with natural rubber [5] or recycled polypropylene [6] for biocomposite production. The physical properties of particleboards from alang-alang and polyvinyl acetate (PVAc) with ratio alang-alang:PVAc = 400:130 (w/w) has fulfilled Indonesian Standard for particleboards but
the mechanical properties has not analyzed yet [7]. Using isocyanate as adhesive with adhesive content of 11%, the alang-alang particleboard’s modulus of elasticity, modulus of rupture and internal bond were met the Japanese Industrial Standard A 5908:2003 for particleboards type 18 [8].

Commercially, particleboard production using formaldehyde based adhesive. Unfortunately, formaldehyde emission cause negative impact to human health. It is known that formaldehyde has a carcinogenic effect on human health [9, 10, 11]. Nowadays, researchers and industries begin to use low formaldehyde emission adhesives or try to find other environmentally friendly adhesives. In 2011, Umemura and his colleagues try to apply citric acid as natural adhesives for wood [12]. Since then, citric acid had been used in particleboards of Agave sisalana fibers [13], bamboo [14], oil palm frond [15], sugar cane bagasse [16], sweet sorghum bagasse [17], corn husk [18].

In this study, Imperata cylindrica particles and citric acid were used as the raw material and natural adhesive, respectively, for the particleboards production. Since citric acid solution was sprayed onto Imperata particles during particleboards production, the pre-drying treatment was needed to eliminate moisture from particles before hot pressing. The effect of pre-drying time and citric acid content on the physical and mechanical properties of the particleboards was investigated.

2. Materials and Method
The raw materials for production of particleboards in this study were alang-alang (Imperata cylindrica), collected from Cibinong, West Java, Indonesia.

2.1. Preparation of materials
Only the above ground part of alang-alang was used in particleboard production. Then, a chipper and a knife-ring flaker machine were used to produce alang-alang particles. The particles were screened using a sieving machine to obtain particles of uniform sizes, and the particles remaining between aperture sizes of 4.76 mm (4 mesh) and 1.41 mm (14 mesh) were used as the raw material. The particles were dried in an oven at 60°C for 16 h to obtain a moisture content of less than 4%.

Citric acid (anhydrous) of extra purity grade was purchased from MERCK and was used without further purification. Citric acid was dissolved in water at a concentration of 59 wt%, and this solution was used as the adhesive. No other chemical compounds were used.

2.2. Particleboards production
Citric acid solution was sprayed onto dried particles to achieve various citric acid contents, i.e., 10, 15 and 20 wt%. Particles that had been sprayed with citric acid solution were prepared under the following conditions: (a) wet particles that had been sprayed with citric acid solution were used directly to form mats and were called non-dried particles or type (0) particles; (b) wet particles that had been sprayed with citric acid solution were dried at 80°C for 6, 12 h and were called pre-dried particles or type (6, 12) particles. Subsequently, these particles were formed into mats using a forming box with the size of 300 x 300 mm.

The particle mat was hot-pressed at 200°C for 10 min [19] to produce particleboard size and target density of 300 x 300 x 9 mm and 0.8 g/cm³, respectively. A 9-mm steel-thick bar was used to control the board thickness during the hot-pressing process. The maximum pressing pressure was 6.5 MPa when the upper side of pressing plate reached the steel-thick bar.

2.3. Evaluation of the particle boards properties
2.3.1 Physical properties: Thickness swelling (TS) and water absorption (WA) analysis. After conditioning for 1 week at a room temperature, the boards were tested according to the Japanese
Industrial Standards for particleboards (JIS A 5908:2003). The specimen size for TS and WA evaluation was 50 x 50 x 9 mm. Specimens were immersed in water for 24 h. The thickness and weight difference before and after immersion were calculated.

To observe particle boards thickness swelling in severe condition, specimens were subjected to a cyclic 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, immersion in boiling water for 4 h, and drying at 105°C for 10 h). The thickness and weight changes of the specimens that occurred throughout the treatment were determined. Each experiment was performed in five replications, and the average values and standard deviations were calculated. The thickness swelling (TS) and water absorption (WA) values of each board after water immersion at room temperature for 24 h were measured.

2.3.2 Mechanical properties: Modulus of elasticity, modulus of rupture analysis, internal bond. The bending properties of the boards, i.e., the modulus of rupture (MOR) and the modulus of elasticity (MOE), were evaluated by conducting a three-point bending test on a 200 x 50 x 9 mm specimen of each board under dry conditions using Universal Testing Machine (UTM, Shimadzu). The loading speed and effective span were 10 mm/min and 150 mm, respectively. The internal bonding (IB) strength was investigated using a 50 x 50 x 9 mm specimen of each board.

2.3.3 Impact strength analysis. The charpy impact test was completed using a digital impact tester DG-CD (Toyo Seiki Seisaku-sho, Ltd. Tokyo, Japan). A rectangular specimen of 80 mm x 10 mm x 9 mm was prepared, and the impact strength in the flatwise direction was measured with an unnotched sample.

2.4. Evaluations of functional groups in alang-citric acid particles
The edge of an alang-citric acid particleboard was scratched to obtain particles. The particles were ground into a powder, and the powder obtained was dried in a drying oven at 60°C for 16 h. Infrared (IR) spectral data were obtained with a FTIR spectrophotometer (Spectrum Two, Perkin Elmer) using the Universal Attenuated Total Reflectance (UATR) method and were recorded with an average of 16 scans at a resolution of 4 cm⁻¹.

2.5 Evaluations of alang-citric acid thermal properties
The edge of alang-citric acid particleboard was scratched to obtain particles. The particles were ground into a powder. As many as 5 mg of alang-citric acid powder was placed in aluminium pan. Differential Scanning Calorymetry (DSC, Perkin Elmer) was used to analyze thermal characterization of alang-citric acid particle boards powder. Heating scan was performed from 0°C to 200°C with heating rate 10°C/min.

3. Results and Discussion

3.1. Effect of pre drying time
The concentration of citric acid solution used in particleboards production in this study was 59 wt%, thus it was contain plenty of moisture. Whereas, according to Nadzi et al. [20] particles with high moisture content caused inferior mechanical properties particleboards due to the poor adhesion between adhesive and particles. Therefore pre-drying treatment of particles after citric acid spraying was needed to eliminate moisture in particles before hot pressing.

The effect of pre-drying time on thickness swelling and water absorption of alang particleboards bonded with 15% citric acid is presented in Figure 1. Without pre-drying treatment, thickness swelling of alang particleboards reached 26.07%. After pre drying for 6 h and 12 h, the particleboards thickness swelling reduced to 13.32% and 7.66%, respectively. The particleboard made from alang particle after
pre drying for 12 h has fulfilled Japanese Industrial Standard (JIS A5908-2003 for particle board). Similar trend was presented on particleboards water absorption properties. The improvement of particleboard physical properties were attributable to the enhancement of adhesive and alang lignocellulosic materials bondability as will explain in the FTIR analysis section.

A similar trend was presented by particleboards bending properties. Particleboards made from alang particles after 12 h pre drying shows higher modulus of rupture (MOR), which was 7.82 MPa, compare to particleboards made from alang particle without pre drying treatment (4.90 MPa) or after 6 h pre drying (6.19 MPa), as presented in Figure 2. The particleboard modulus of elasticity (MOE) made from alang particle after 12 h pre drying was also higher (1646 MPa) than that of particleboards made from alang particle without pre drying (839 MPa) or after 6 h pre drying (1220 MPa).

Particleboards internal bond (IB) properties (Figure 2) was also confirmed that pre drying treatment could enhanced bondability between alang lignocellulosic materials and citric acid significantly, as particleboards IB made from alang particle after 12 h pre drying was 0.08 MPa, higher than that of particleboards IB made from alang particles without pre-drying treatment (0.01 MPa) and after 6 h pre drying (0.03 MPa).

Figure 1. Effect of pre drying time on particleboards physical properties (thickness swelling and water absorption).

Figure 2. Effect of pre drying time on particleboards mechanical properties (modulus of rupture,
modulus of elasticity properties, internal bond).

All the alang particleboards bonded with 15% citric acid were not fulfilled the requirement of JIS A 5908 yet. Nevertheless, particleboards made from alang particles after pre drying for 12 h showed better physical properties and higher mechanical properties compare to particleboards made from alang particles without pre drying treatment or with pre drying treatment for 6 h. Therefore, in next stage, the effect of citric acid content on particleboards properties was analyzed on particleboards made from alang particles after pre drying treatment for 12 h.

### 3.2. Effect of citric acid content

To evaluate effect of citric acid content of particleboard properties, after spraying citric acid, alang particles were pre drying for 12 h before hot pressing. As presented in Figure 3, particleboard with 20% citric acid content shows the best physical properties compare to particleboard with 10% or 15% citric acid. Thickness swelling (TS) of particleboards with 10% or 15% citric acid were 20.92% and 7.66%, respectively. The particleboards TS reduced significantly, which was 2.04% when alang particles were bonded with 20% citric acid.

**Figure 3.** Effect of citric acid content on particleboards physical properties (thickness swelling and water absorption).

The MOR of particleboard bonded with 15% citric acid (7.82 MPa) was higher than that of MOR of particleboard bonded with 10% citric acid (5.96 MPa). However, MOR of particleboards bonded with 20% citric acid was slightly lower (6.54 MPa). Meanwhile, MOE of alang particleboard bonded with 20% citric acid was 3924 MPa, much more higher than that of MOE of alang particleboard bonded with 10% citric acid (977 MPa) or 15% citric acid (1646 MPa). In the previous study, Azeredo et al. [21] stated that the tensile strength of wheat straw hemicellulose film decreased, but its MOE increased as the citric acid content increased up to 20 wt%, because citric acid acting as a flexible cross linker. The result of this study was similar to Azeredo report, so that, it was assumed that citric acid also acting as cross linking agent in alang particleboards. The acid causes the material to stiff and brittle, therefore the particleboards elasticity was improved but the particleboards tensile strength was decreased.
The internal bond (IB) strengths of alang particleboards with varied citric acid content were not significantly different (Figure 4). Particleboards IB values were 0.077 MPa (CA 10%), 0.076 MPa (CA 15%), 0.074 MPa (CA 20%). The increment of citric acid amount introduced into alang particles, could not enhanced the particleboards internal bond strength. Although chemical reaction between alang lignocellulosic material and citric acid was occurred as presented in Figure 6, it seems some substance in alang materials could hinder bondability between citric acid and alang lignocellulosic materials. Alang contain 3.67% silica [22] which assumed act as blockade in reaction between citric acid and alang lignocellulosic.

![Figure 4](image_url). Effect of citric acid content on particle boards mechanical properties (modulus of rupture, modulus of elasticity properties, internal bond).

Figure 5 shows the Charpy impact strength of alang particleboards made from varied citric acid content and particles pre drying time. The Charpy impact strength of alang particleboards increased gradually with an increasing citric acid content. Generally, a high Charpy impact strength value indicates low brittleness [23]. Consequently, the brittleness of alang particleboards was reduced by adding citric acid. This result was accordance with alang particleboards MOE, which was increased with an increasing citric acid content.

![Figure 5](image_url). Effect of citric acid content on particle boards impact strength.
3.3. Functional groups in alang-citric acid particle boards

The infrared (IR) spectra of alang particleboards with varied citric acid content is presented in Figure 6. An absorption peak at 1726 cm$^{-1}$ appeared clearly in alang particleboards bonded with 10% citric acid. The peak at 1726 cm$^{-1}$ was typically assigned to C=O stretching due to carbonyl groups and/or the C=O ester groups [24, 25]. The appearance of ester groups in the IR spectra indicates that the carboxyl groups of citric acid reacted with hydroxyl groups of alang to form ester linkages.

![Figure 6. Fourier transform infrared spectra of alang particleboards with varied citric acid content (10%, 15%, 20%).](image)

The lignocellulosic materials contain abundant hydroxyl groups derived from lignocellulosic components such as cellulose, hemicellulose and lignin [26]. The alang particleboards bonded with increased citric acid content showed more ester linkage. The consequent formation of ester linkages would improve the adhesiveness. As a result the physical and mechanical properties of alang particleboards bonded with citric acid were improved.

3.4. Cyclic aging treatment to observe particle boards thickness swelling in severe condition

![Graphs showing moisture content change over time](image)
Figure 7. Thickness change of particle boards with 10%, 15%, 20% citric acid content and 0 h (A), 6 h (B), 12 h (C) pre drying time, during cyclic aging treatment.

The change in the thickness of the particleboards as showing Figure 7. The expanded of thickness change of the particleboard decreased with increasing pre-drying time for each condition of citric acid content. The thickness change after boiling treatment of the particleboard bonded with 20 wt% citric acid and pre-drying times of 6 and 12 h were lower than that of the other conditions of the particleboard. These mean that the pre-drying times treatment of 6 and 12 h with the citric acid content of 20 wt% had given good dimensional stability of particleboard made from alang-alang grass even in the severe conditions.

3.5. Thermal properties of alang-citric acid particleboards

In Figure 8, the DSC curve of alang powder show an endothermic peak at 72.24°C, while alang-citric acid powder show endothermic peaks at 67.47°C; 61.66°C and 66.98°C. Kusumah et al. [23] reported that sweet sorghum bagasse (SSB) powder show endothermic peak at around 90°C while SSB and citric acid show endothermic peak at approximately 75°C. The endothermic peak at approximately 90°C was the consequence of the weakening of the hydrogen bond between carbohydrates [27].

Table 1. Thermal properties of alang particleboards with varied citric acid content

| Sample           | Endothermic peaks |
|------------------|-------------------|
|                  | \( T_1 (^\circ C) \) | \( \Delta H_1 (\text{J.g}^{-1}) \) | \( T_2 (^\circ C) \) | \( \Delta H_2 (\text{J.g}^{-1}) \) |
| Alang Powder (AP)| 72.24             | 146.96            |                |                |
| AP + CA 10%      | 67.47             | 187.53            | 189.83         | 173.04         |
| AP + CA 15%      | 61.66             | 143.95            | 192.84         | 172.66         |
| AP + CA 20%      | 66.98             | 119.11            | 194.49         | 162.34         |

Alang powder shows only one endothermic peak, whereas alang-citric acid powder show two endothermic peaks. The second endothermic peaks at alang-citric acid curves were at 189.83°C; 192.84°C; 194.49°C. Barbooti and Al-Sammerrai [28] mentioned that the endothermic peak at approximately 150°C shows the melting point of citric acid and the endothermic peak at 180°C indicated the decomposition of citric acid to become aconitic acid. The higher the citric acid content in alang particle board, the higher the decomposition temperature of citric acid.
Figure 8. Thermogram of alang powder and varied citric acid content (10%, 15%, 20%)

4. Conclusion

The physical properties of alang-citric acid boards produced using particles after 12 h pre drying time were superior to those of the boards produced using particles after 0 and 6 h pre drying time. The thickness swelling of particleboards with 20 wt% citric acid and pre drying time of 0, 6, 12 h were 5.31%, 3.69%, 2.04%, respectively, fulfilled the requirement of the JIS A 5908:2003. The modulus of elasticity of alang particleboards bonded with 20 wt% citric acid and pre-dried for 12 h, fulfilled The JIS 5908 for particelboards type 8. The infrared (IR) spectral analysis showed the presence of ester linkage, representing that the carboxyl groups of citric acid had reacted with the hydroxyl groups of the alang particles, providing the boards good physical properties.

5. References

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