Eco-friendly Board from Oil Palm Frond and Citric Acid

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Abstract

The utilization of oil palm fronds as composite board raw material has developed due to the declining wood supply. Currently, citric acid is used as natural adhesive on the wood-based moldings to reduce the use formaldehyde-based resin. Citric acid has carboxylic acid functional group that can react with hydroxyl functional group from cellulose by esterification reaction. This paper explain the production of board from oil palm frond and citric acid, then elaborate their physical and mechanical properties. Oil palm fronds were processed with ring flaker to produce particles with 1 ~ 5 cm length. The particles were dried until the moisture content reached 6%. Citric acid solution was obtained by stirring 700 g citric acid in 1000 ml water. Citric acid solution were sprayed onto particles. The weight of citric acid were 10%, 15% and 20% based on oil palm fronds' dry weight. Boards were produced with density target of 0.6 g/cm³ and pressure at 1N/mm² for 10 min. The hot pressing temperatures were varied at 140°C, 160°C, 180°C, and 200°C. Physical and mechanical test were performed according to JIS A 5908-2003. The MOR, MOE, IB and SW values of boards with 15 wt% citric acid content and pressed at 200°C were 5.85 N/mm², 1067.03 N/mm², 0.26 N/mm² and 88.80 N respectively. The optimum condition for board production obtained upon citric acid content and pressing temperature in a range of 15% ~ 20% and 180 °C ~ 200 °C, respectively.

Keywords: oil palm fronds, natural binder, citric acid, esterification, properties.

Introduction

The demand for particleboard products continues to increase. Meanwhile, due to government restriction; wildlife protection, and other environmental concerns, the availability of these raw materials, particularly from wood has been decreasing, leaving an increasing gap between raw materials and product demand. Researchers are finding alternatives to fill this gap. Oil palm fronds, a renewable agricultural by-product from oil palm plantation would be the economical lignocellulosic resources.

Indonesia is the largest producer of oil palm (*Elaeis guineensis*) in the world. In 2009, Indonesia oil palm plantations covered an area of 7,508,023 hectares and increased to 7,824,623 hectares in 2010 (Directorate General of Plantation 2010). During the production period, the fronds of oil palm trees need to be trimmed when harvesting oil palm fruit bunches. In a palm tree age 3 to 4 years, the number of fronds can reach 30 to 40 stalks, and then decreased between 18 to 25 stalks. Within a year, every single hectare of oil palm plantations produce as much as 10.4 tons of oil palm fronds based on dry weight, averagely (Husin 1986). With a total plantation area in 2010 amounted to 7.82 million hectares, there will be oil palm fronds as by product up to 81.32 million tons per year. Such a large amount of oil palm fronds need to be well managed for environmental sustainability.

According to Thole and Hora (2003) referred in Jonoobi *et al.* (2011), more than 90% of the oil palm empty fruit bunches can be converted into fibers, while the part of the oil palm trunk and fronds that can be converted into fibers, respectively, were 25% and 50%. Wan Rosli *et al.* (2004) stated that the oil palm fronds contains 14.81% lignin, 86.53% holocellulose, 62.34% α -cellulose and 1.8%

extractives. Therefore, the oil palm fronds are potential to be processed as source of cellulose.

Commercial particle board production apply urea formaldehyde or phenol formaldehyde as adhesive to bind wood particles. The emission of carcinogenic formaldehyde in the production and during application of particleboard has generated an urgent need for development of a formaldehyde-free wood adhesive for making particleboards.

There has been reports regarding natural adhesives, binderless boards and chemical surface activation of wood, as alternatives for eco-friendly board production. In this research, we paid attention in citric acid which is a natural organic polycarboxylic acid containing three carboxyl groups. Citric acid is widely used in foods, beverages and pharmaceuticals. The utilization of citric acid as a cross-linking agent to improve the physical and mechanical properties of wood has been reported by Vukusic *et al.* (2006). Additionally, Umemura *et al.* (2012) has researched the application of citric acid as the main material of wood adhesive. In this study, citric acid was applied as a natural adhesive for oil palm fronds board production.

Materials and Methods

Materials

Oil palm fronds were obtained from oil palm plantation in West Java, Indonesia. The leaves were cut off from oil palm fronds, then the fronds were processed with ring flaker to produce particles with 1 ~ 5 cm length. The particles were washed and air dried until the moisture content reached 6%. Citric acid was produced by PT. Budi Acid Jaya, Lampung. Citric acid solution was obtained by stirring 700 g citric acid in 1000 ml water.

Board Production

Oil palm fronds particles were sprayed with citric acid solution. The weight of citric acid were 10%, 15% and 20% based on oil palm fronds' dry weight. Details regarding the formulations are shown in Table 1. Boards were produced with dimensions of 25 cm x 25 cm x 0.8 cm with density target of 0.6 g/cm³. The hot pressing temperatures were varied at 140 °C, 160 °C, 180 °C, and 200 °C. The pressing condition was pressure at 1N/mm² for 10 min. Boards were produced in randomly particles arrangement.

Table 1. Formulation of boards

Citric acid content (wt%)	Weight ratios of OPF to citric acid	Oven-dried Oil palm fronds (g)	Citric acid (g)
10	5 : 1	272.7	27.3
15	7 : 1	260.9	39.1
20	10 : 1	250.0	50.0

Evaluations of Boards

The specimens of 200 x 50 x 8 mm were prepared for static bending test. The static three-point bending test was carried out under air-dry condition according to JIS A5908-2003. The effective span and loading speed were 120 mm and 5 mm/min, respectively. The modulus of rupture (MOR) and modulus of elasticity (MOE) of composites were calculated. Internal bond and screw withdrawal test were also performed according to JIS A 5908-2003. For thickness swelling property testing, water immersion treatment for 24 h was performed using a square specimens (50 x 50 mm). After the treatment, thickness changes were measured under wet condition.

Results and Discussion

Natural oil palm fronds particles' color were light brown. Due to hot pressing at 140 ~ 200 °C, boards of oil palm fronds' color change into dark brown (Figure 1). The obtained board densities ranged from 0.44 to 0.67 g/cm³.

Figure 1. Oil palm frond board.

Effects of Citric Acid Content on Composite Physical Properties.

The thickness swelling of oil palm frond board bound with citric acid is shown in Figure 2. After 2 hr immersion, the board thickness swelling with 10 wt% citric acid and 140

°C pressing temperature showed the highest value which was 46.03% while board with 20 wt% citric acid and 200 °C pressing temperature had the lowest value (0.56%). After 24 hr immersion, the board thickness swelling with 10 wt% citric acid and 140 °C pressing temperature showed the highest value which was 57.34% while board with 20 wt% citric acid and 200 °C pressing temperature had the lowest value (2.53%). The increasing citric acid content and pressing temperature lead to the better performance of boards' thickness swelling properties.

Figure 2. Oil palm fronds board's thickness swelling after (a) 2 hr immersion, (b) 24 hr immersion.

The chemical composition of oil palm frond strands was 14.81% lignin, 86.53% holocellulose, 62.34% α-cellulose, and 1.8% extractives, on an oven-dry weight basis (Wan Rosli *et al.* 2004). The polycarboxylic acids react with hydroxyl groups of cellulose after high-temperature drying or curing, and drive the formation of ester cross-links in cellulosic or lignocellulosic. A covalent bonding is expected in the adhesion mechanism, as the boards have some resistance to swelling. The reaction between carboxyl groups from citric acid and hydroxyl groups from cellulose is given in Figure 3.

Figure 3. Esterification reaction of carboxyl groups from citric acid and hydroxyl groups from cellulose.

Effects of Citric Acid Content on Composite Mechanical Properties.

Effects of citric acid content on the boards' modulus of rupture (MOR) is presented in Figure 4. The maximum MOR value of oil palm fronds boards in this study was 5.85 N/mm², produced from 15 wt% citric acid and 200 °C pressing temperature. Higher pressing temperature was applied during boards production, improved the boards' modulus of rupture properties.

Figure 4. Oil palm fronds boards' modulus of rupture.

Citric acid (CA) can be functionalized as particle adhesive by the present of covalent bonding between carboxyl groups from citric acid and hydroxyl groups from cellulose which was effective for development of good bending properties. Although oil palm fronds board with 15 wt% citric acid content pressed at 200 °C (5.85 N/mm²) did not fulfill the JIS standard for particle board type 8, MOR of oil palm fronds board was higher than sisal fiber board or vetiver root board at the same citric acid content. According to Syamani *et al.* (2010), MOR of sisal fiber board with 15 wt% and 20 wt% citric acid were 1.67 and 1.52 N/mm², respectively while MOR of vetiver root board with 15 wt% and 20 wt% citric acid were 1.96 N/mm² and 2.53 N/mm², respectively (Syamani *et al.* 2012).

Oil palm fronds board's modulus of elasticity (MOE) also showed a similar trend (Figure 5). The maximum MOE value of oil palm fronds board in this study was 1067.03 N/mm², produced from 15 wt% citric acid and 200 °C pressing temperature. The MOE of oil palm frond board was higher than sisal fiber board or vetiver root board at the same citric acid content. According to Syamani *et al.* (2010), MOE of sisal fiber board with 15 wt% and 20 wt% citric acid were 220.74 N/mm² and 308.96 N/mm², respectively while the MOE of vetiver board with 15 wt% and 20 wt% citric acid were 221.6 N/mm² and 260.7 N/mm² (Syamani *et al.* 2012).

Figure 5. Oil palm frond's modulus of elasticity.

The increasing pressing temperature leads to the improvement of bending properties of oil palm frond boards. Citric acid melts at 153°C and dehydrates to give aconitic acid on heating at 175°C. Further heating results in the formation of methyl maleic anhydride (Barbooti and Al Sammerai, 1986). The citric acid began to melt from 157°C temperature and turns into gas form at the temperature of 175°C (Munawar *et al.* 2009). In this phase, the carboxyl group of citric acid will begin to bind the hydroxyl group in acacia bark through esterification process (Umemura *et al.* 2010). Esterification process in this phase causes the bonds between citric acid and fiber becomes stronger than at other temperatures.

Oil palm frond acid boards' internal bond properties which produced from 15 wt% citric acid and 200 °C pressing temperature were 0.26 N/mm² (Figure 6). Generally, internal bond properties of oil palm frond boards were improved with the increasing of citric acid content and pressing temperature. Based on analysis of variance, board's internal bond strength with pressing temperature at 140 °C and 160 °C showed no significant difference, but differ from board's internal bond strength with pressing temperature at 180 °C and 200 °C.

Figure 6. Oil palm fronds' internal bond.

The bonding mechanism between the carboxyl groups from citric acid and hydroxyl group from oil palm frond particles occur in hemicellulose components of oil palm particles. Only the amorphous region of cellulose and low molecular weight hemicellulose fraction are expected to be accessible to chemicals and more accessible to aqueous reagents. As reported by Umemura *et al.* (2010), optimization of bonding occurred through ester linkage between carboxyl group from citric acid and hydroxyl group from wood hemicellulose.

Efforts to improve the internal bond of the board from high cellulose content material using citric acid as a bonding agent has been done by Sugihara *et al.* (2010). The research results showed that with the addition of sucrose into the citric acid from 25% ~ 50 wt% have increased the internal bond of the wood particle boards by 150% ~ 300%.

Figure 7. Oil palm fronds' screw withdrawal.

Oil palm frond acid boards' screw withdrawal properties produced from 15 wt% citric acid and 200 °C pressing temperature were 88.80 N (Figure 7). Based on analysis of variance, the values of boards screw withdrawal showed no significant difference at varied citric acid content and pressing temperature.

According to Umemura (2012), when the plant-derived material is mixed with a powdery polycarboxylic acid and is pressured, it is preferred that a maximum length is controlled to 10 mm or less, although it is possible to sufficiently cure a plant-derived materials which is in form of small pieces having a maximum length of 50 mm. The dimension of oil palm frond particles used in this study were varied between 1 ~ 5 cm length and randomly composed inside the board, resulted in a similar screw withdrawal properties.

Conclusions

Citric acid was used as a natural adhesive to produce oil palm frond boards. The boards obtained had good bending properties. The MOR, MOE, IB and SW values of boards with 15 wt% citric acid content and pressed at 200°C were 5.85 N/mm², 1067.03 N/mm², 0.26 N/mm² and 88.80 N respectively. The optimum condition for board production obtained upon citric acid content and pressing temperature in a range of 15% ~ 20% and 180 °C ~ 200 °C, respectively.

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