

## Development of Particle Board from Oil Palm Frond Fibers Using Citric Acid and Sucrose as Adhesives

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**Abstract.** Composites made from plant fibers and natural adhesives may call as true green composites. Source of plant fibers can be taken from agricultural wastes such as oil palm wastes. Fronds of oil palm are abundantly available in Indonesia, more than 80 million tons are produced each year. Recently, a new natural adhesive using citric acid and sucrose has been researched for composite. The purpose of this research is to make particleboard from oil palm frond fibers using citric acid and sucrose as adhesives, in particular to obtain optimum adhesive content of citric acid and optimum ratio of citric acid to sucrose. Adhesive solution of citric acid or sucrose was made by mixing with water at 50 % weight. The solution was sprayed to the frond particles, and the particles were dried in an oven at temperature of 80°C for 15 hours. The boards were made by hot pressing at 200°C for 10 minutes with a target density of 0.8 g/cm<sup>3</sup>. Citric acid contents were varied at 0, 10, 20, 30, and 40 wt%. Ratios of citric acid : sucrose were varied at 100:0, 75:25, 50:50, 25:75, and 0:100. Results showed that the optimum citric acid content to make frond particleboard was 20%. While the optimum ratio of citric acid to sucrose was 50:50. Surprisingly, particleboard from oil palm fronds bonded with sucrose only has very good water resistant even after cyclic treatment test, the board properties were met the type 8 particleboard of JIS A5908 standard. This finding opens a possibility to use sucrose only as adhesive for fronds particleboard, as well as combination with citric acid. The Fourier transform infrared spectra (FTIR) analysis was used to confirm the chemical reaction formed between the frond particles and the adhesives.

**Keywords:** Oil palm frond fiber; particleboard; citric acid; sucrose

### Introduction

Oil palm industry grows rapidly in Indonesia. Now Indonesia has the largest area of oil palm plantation in the world. This industry annually generated large amount of wastes such as fronds, empty fruit bunch, trunk, and shell. The wastes are potential source of material for composites industry. More than 80 million tons of fronds are produced each year from 8 million hectares of oil palm plantation in Indonesia [1]. Some research on utilization of oil palm waste for pulp and composites have been done [2-6]. Fronds may be

used for particleboard. Particleboard usually made with formaldehyde based adhesive that harmful to environment and health, therefore natural adhesives are promising as alternative. Citric acid, naturally contains in citrus fruit is an organic polycarboxylic [7], has been developed as adhesive for wood and molding [8-11]. Umemura *et. al* [12] used citric acid to produce particleboard from wood, however utilization of citric acid as adhesive for oil palm frond particleboard has not been done. In this research, particleboard was developed from oil palm frond fibers using citric acid and sucrose adhesives. The purpose was to determine the optimum values of citric acid content, ratio of citric acid to sucrose, and pressing temperature.

## Experimental

### *Materials*

Oil palm frond fibers were obtained from West Java, Indonesia. Citric acid anhydrous and sucrose were obtained from Nacalai Tesque, Inc., Kyoto, Japan.

### *Board Manufacturing*

First experiment was done by making one layer particleboard using citric acid to obtain optimum adhesive content. The citric acid content was varied at 0, 10, 20, 30, and 40 wt%. Frond particles were dried in an oven at 105°C for 24 h, and the particles were sieved to determine the size. Adhesive solution of citric acid was made at 50 wt% by mixing 500 g of citric acid with 500 g of water. Frond particles were mixed with adhesive using spray gun in a drum mixer. After mixing, the particles were dried in an oven at 80°C for 15 h. The moisture content of particles after drying was calculated. Dried particles were matted formed in a forming box and were hot pressed (200°C, 25 kgf/cm<sup>2</sup>) for 10 min. Distance bars with thickness of 0.9 cm were put to control the board thickness. The board target density was 0.8 g/cm<sup>3</sup> and board size was 30 cm x 30 cm x 0.9 cm. The second experiment was conducted to obtain optimum ratio of citric acid to sucrose. The mixture ratios of citric acid/sucrose were 100/0, 25/75, 50/50, 75/25, and 0/100. The resin content was 20 wt.%, and board target density was 0.8 g/cm<sup>3</sup>. The board manufacturing was the same as above. The third experiment was done to search optimum pressing temperature. Pressing temperatures of 140, 160, 180, 200, and 220°C were tried to make boards with ratio of citric acid/sucrose of 50/50, resin content of 20 wt.%, and board target density of 0.8 g/cm<sup>3</sup>.

### *Board Testing*

The board was cut into specimens and conditioned at 20°C and 65% RH for a week prior to testing. The tests were conducted for bending strength (modulus of rupture/MOR, modulus of elasticity/MOE), internal bond strength, and thickness swelling in accordance with JIS A 5908: Particleboards. Cyclic treatment test was conducted (immersion in water 20°C for 24 hr, dry in an oven 105°C for 10 hr, immersion in water 70°C for 24 h, dry in an oven 105°C for 10 h, boil in water for 4 h, dry in an oven 105°C for 10 h). FTIR (JASCO FT/IR-4200) was used to observe the chemical reaction formed between the particles and adhesives.

## **Results and Discussion**

### *Effect of citric acid content on board properties*

Result of particleboard properties at citric acid content of 0, 10, 20, 30, and 40 wt.% was shown in Table 1. It is shown that the thickness swelling of board without citric acid (binder-less board) was 54%. However after addition of citric acid, the thickness swelling of board decreased with increasing citric acid content. Hydroxyl groups in the cellulose reacts with citric acid to form ester linkages that bound strongly (as shown at FTIR results in Fig. 1) therefore it makes more resistant to water [12]. All thickness swelling values of board with citric acid were met the JIS A5908 standard which requires thickness swelling less than 12%.

The optimum citric acid content was obtained at 20 wt.% which showed the highest MOR and MOE (Table 1). The values are met the JIS A5908 standard where for type 8 board the minimum MOR is 8 MPa and MOE is 2 GPa. At higher citric acid contents (30 and 40wt.%) the values decreased it may caused by excessive of adhesive that weaken the bound between citric acid and frond fibers. Umemura *et. al* [12] using wood particles to produce citric acid particleboard at the same content (20 wt.%) obtained higher MOR (10.7 MPa) but lower MOE (3.3 GPa). Particleboard made from oil palm frond fibers exhibited higher stiffness but lower strength compared to that made of wood particles. The difference in chemical compositions (cellulose, hemicellulose, lignin) between oil palm fronds and wood may have effect on the board properties. Oil palm fronds have higher hemicellulose but lower lignin compare to that of wood. The hemicellulose, cellulose, and lignin contents of oil palm fronds are 35.37%, 47.76%, and 20.15%, respectively [2]; while that of wood are 15-25%, 40-45%, and 23-30%, respectively [13].

It showed that increasing citric acid content decreased the internal bond strength (Table 1). FTIR result of board with citric acid is shown increased of carbonyl groups (Fig. 1). The values of internal bond obtained in this research are all met the JIS A5908 standard which requires minimum value of 0.15 MPa. The internal bond strength values obtained in this experiment using oil palm frond fibers are higher compared to that of using wood particles (0.32 MPa) [12].

Table 1. Effect of citric acid content on the board properties.

Properties	Citric acid content (%)				
	0	10	20	30	40
TS (%)	53.13 (4.32)	6.99 (0.83)	5.10 (1.15)	4.63 (0.64)	3.80 (0.39)
MOR (MPa)	2.87 (0.45)	5.03 (0.75)	9.35 (1.34)	5.66 (0.85)	5.78 (0.85)
MOE (GPa)	0.98 (0.16)	2.01 (0.21)	4.18 (0.08)	2.46 (0.27)	2.37 (0.30)
IB (MPa)	0.08 (0.07)	0.78 (0.14)	0.73 (0.13)	0.66 (0.07)	0.52 (0.14)

Notes: TS=thickness swelling, MOR=modulus of rupture, MOE=modulus of elasticity, IB=internal bond strength. Figures in the brackets are standard deviations.

#### *Effect of ratio of citric acid to sucrose on board properties*

Effect of ratio of citric acid to sucrose on board properties is presented in Table 2. Increasing of sucrose increased of thickness swelling of board. All ratios resulted in less than 6% thickness swelling, which is met the JIS A5908 standard (less than 12%).

The optimum MOR and MOE values were obtained at ratio of 50/50 as shown in Table 2. All ratios resulted in MOR and MOE values met the JIS A5908 standard. It is interesting here that using sucrose only the values of MOR and MOE were met the standard. This new finding opens the possibility to use only sugar-based resin to produce particleboard from oil palm frond.

Table 2. Effect of ratio of citric acid to sucrose on the board properties.

Properties	Ratio of citric acid to sucrose (%)				
	100/0	25/75	50/50	75/25	0/100
TS (%)	3.93 (0.36)	4.43 (0.39)	5.10 (1.15)	4.84 (1.03)	6.02 (1.35)
MOR (MPa)	9.35 (1.34)	9.54 (0.60)	10.75 (0.41)	9.45 (1.05)	8.65 (1.37)
MOE (GPa)	4.18 (0.08)	4.33 (0.29)	4.41 (0.08)	4.12 (0.17)	3.90 (0.13)
IB (MPa)	0.73 (0.13)	0.99 (0.09)	1.01 (0.24)	1.04 (0.05)	1.19 (0.10)

Increased of sucrose content of internal bond strength of board properties is shown in Table 2. It may caused by the formation of caramel when sucrose is heated at high temperature, however this effect needs to be clarified in further research. Similar trend was found when using wood particles as raw material [12]. All internal bond strength values at all ratios were met the JIS A5908 standard (more than 0.15 MPa). Board that used sucrose only exhibited the highest internal bond strength values (1.2 MPa). This result is different with that of obtained by Umemura *et. al* [12] using wood particles they obtained a low value of 0.2 MPa.

Result of cyclic treatment test on the thickness swelling is presented in Fig. 1. It is shown that all of the board types exhibited excellent properties that are very small changed in thickness after severe treatment. The board bonded with sucrose only resulted in the highest thickness change, however the change is still less than 10%. FTIR result (Fig. 2) shown that there is an increase in the absorption peak at  $1734\text{ cm}^{-1}$  due to the addition of citric acid (middle graph) compared to frond fibers (bottom graph). After cyclic treatment the peak did not change (upper graph), it is confirmed that the reaction between citric acid and frond fibers is strong.

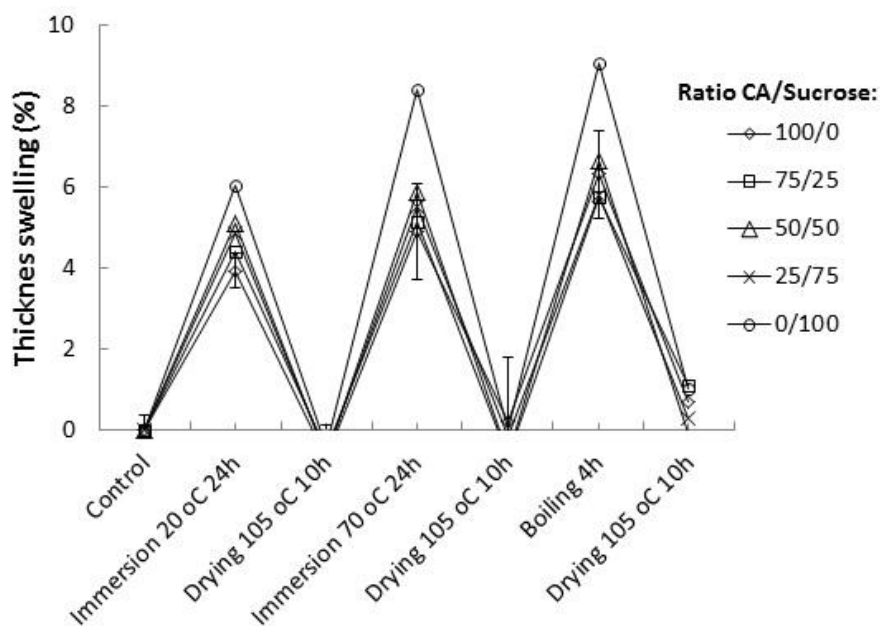


Figure 1. Thickness swelling of board after cyclic treatment test.

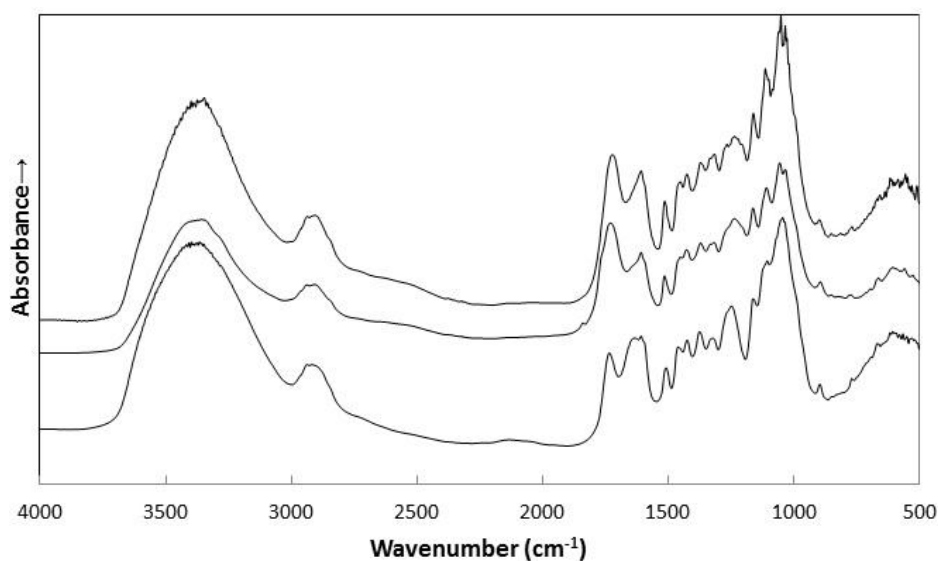


Figure 2. FTIR results of frond particles (bottom), citric acid board (middle), citric acid board after cyclic treatment test (upper).

#### *Effect of pressing temperature on board properties*

Effect of pressing temperature on board properties is presented in Table 3. According to the JIS standard which requires MOR more than 8 MPa, MOE more than 2 GPa, IB more than 0.15 MPa, and TS less than 12%; the board properties using pressing temperatures of 180, 200, and 220 °C are met the standard, while that of 140 and 160°C are not met the

standard. The pressing temperature of 200°C is considered an optimum one. It is stated that thermal decomposition of citric acid is 220°C [14].

Table 2. Effect of pressing temperature on the board properties.

Properties	Pressing temperature (°C)				
	140	160	180	200	220
TS (%)	61.59 (5.52)	23.84 (4.04)	9.83 (2.04)	5.10 (1.15)	3.81 (0.47)
MOR (MPa)	6.92 (0.65)	10.13 (0.60)	10.37 (0.38)	10.75 (0.41)	10.74 (1.08)
MOE (GPa)	1.92 (0.10)	2.71 (0.18)	3.43 (0.08)	4.41 (0.08)	4.35 (0.08)
IB (MPa)	0.71 (0.03)	1.15 (0.03)	1.19 (0.09)	1.01 (0.24)	0.88 (0.06)

Notes: see Table 1

## Conclusion

Oil palm frond fibers with citric acid or sucrose adhesives can be used to make particleboard that met the standard. The optimum citric acid content was 20 wt.%. The optimum ratio of citric acid/sucrose was 50/50. The optimum of pressing temperature was 200°C. A new finding to use sucrose only to produce particleboard from oil palm frond fibers was obtained.

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- [1] A.U. Lubis, P. Guritno, Darnoko, *Prospects of oil palm solid wastes based industries in Indonesia*, Proc. the Third National Seminar on Utilization of Oil Palm Tree and other Palms 1994. Forest Research Institute Malaysia, Kuala Lumpur, Malaysia 1994, p.62-69.
- [2] R. Hashim, M.N.A.W. Nadhari, O. Sulaiman, F. Kawamura, S. Hiziroglu, M. Sato, T. Sugimoto, TG. Seng, R. Tanaka, *Mater. Des.* **2011**, 32, 246.
- [3] N. Laemsak, M. Okuma, *J. Wood. Sci.* **2000**, 46, 322.
- [4] W. D. Wanrosli, Z. Zainuddin, K.N. Law, R. Asro, *Ind. Crop. Prod.* **2007**, 25, 89.
- [5] M.S.M. Rasat, R. Wahab, O. Sulaiman, J. Moktar, A. Mohamed, T.A. Tabet, I. Khalid, *Bioresources* **2011**, 6, 4389.
- [6] C. S. Goh, H.T. Tan, K.T. Lee, *Bioresource Technol.* **2012**, 110, 662.
- [7] A. A. Abou-Zeid, M.A. Ashy, *Agr. Wastes* **1984**, 9, 51.
- [8] K. Umemura, T. Ueda, S. Kawai, *J. Wood. Sci.* **2011**, 58, 38.

- [9] K. Umemura, T. Ueda, S. Kawai, *For. Prod. J.* **2012**, 62, 63.
- [10] K. Umemura K, T. Ueda, S.M. Sasa, S. Kawai, *J. Appl. Polym. Sci.* **2012**, 123, 1991.
- [11] B. Sefc, J. Trajkovic, M. Hasan, D. Katovic, S. B.Vukusic, F. Martina, *Drvna Industrija* **2009**, 60, 23
- [12] K. Umemura, O. Sugihara, S. Kawai, *J. Wood. Sci.* **2013**, 59, 203.
- [13] R.M. Rowell, “*Handbook of Wood Chemistry and Wood Composites*”, CRC Press, Florida 2005, p.35.
- [14] M.M. Barbooti, D.A. Al-Sammerrai DA, *Thermochim. Acta.* **1986**, 98, 119.