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Original Article

Bonding Performance of Maltodextrin and Citric Acid for Particleboard Made From Nipa Fronds[1]

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ABSTRACT

Maltodextrin and citric acid are two types of natural materials with the potential as an eco-friendly binder. Maltodextrin is a natural substance rich in hydroxyl groups and can form hydrogen bonds with lignoselulosic material, while citric acid is a polycarboxylic acid which can form an ester bond with a hydroxyl group at lignoselulosic material. The combination of maltodextrin and citric acid as a natural binder materials supposed to be increase the ester bonds formed within the particleboard. This research determined to investigate the bonding properties of a new adhesive composed of maltodextrin/citric acid for nipa frond particleboard. Maltodextrin and citric acid were dissolved in distillated water at the ratios of 100/0, 87.5/12.5, 75/25 and 0/100, and the concentration of the solution was adjusted to 50% for maltodextrin and 60% citric acid (wt%). This adhesive solution was sprayed onto the particles at 20% resin content based on the weight of oven dried particles. Particleboards with a size of 25 × 25 × 1 cm, a target density 800 kg/m^3 were prepared by hot-pressing at press temperatures of 180℃ or 200℃, a press time of 10 minute and board pressure 3.6 MPa. Physical and mechanical properties of particleboard were tested by a standard method (JIS A 5908). The results showed that added citric acid level in maltodextrin/citric acid composition and hot-pressing temperature had affected to the properties of particleboard. The optimum properties of the board were achieved at a pressing temperature of 180℃ and the addition of only 20% citric acid. The results also indicated that the peak intensity of C=O group increased and OH group decreased with the addition of citric acid and an increase in the pressing temperature, suggesting an interreaction between the hydroxyl groups from the lignocellulosic materials and carboxyl groups from citric acid to form the ester groups.

Keywords : maltodextrin, citric acid, nipa frond, particleboard

1. INTRODUCTION

The synthetic adhesives based on petroleum have been used for along time and had been known to have excellent performance, good working properties and were economically satisfactory. However, amino resins such as urea-formaldehyde (UF) resin or melamine-urea-formaldehyde (MUF) resin are known to release toxic formaldehdye. Formaldehyde

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emitted from these adhesives is a carcinogenic to humans, and can cause irritation of the eyes and throat, respiratory disorders. And the synthetic resins are non-renewable and non-biodegradable materials (Roffael, 1993; IARC, 2012). Many researches have been focusing on reduction of the synthetic adhesives in wood-based materials production. With steadily increasing awareness in environmental and health issue, natural binders from renewable resources regained attraction as alternative bonding agents. Many studies on natural binder using bio-resources have been conducted to develop eco-friendly biocomposites, such as citric acid (Umemura *et al*., 2011), modified lignin (Nasir *et al*., 2013), citric acid with sucrose (Umemura *et al*., 2013; Widyorini *et al*., 2016b) tannin with sucrose (Zhao and Umemura, 2014), glucose, sucrose and starch (Lamaming *et al*., 2013).

Starch is one of the most abundant polysaccharides in the world, and very attractive for the industry due to its low cost and interesting properties for fibre bonding. Starch is found primarily in the seeds, fruits, tubers and pith of stems of plants, most notably in corn, wheat, rice, sago and potatoes. Starch can added as an extender to various types of conventional adhesives in order to enhance their environmental performance by reducing the absolute usage of synthetic resins and the emissions of toxic formaldehyde as well as the production costs (Moubarik *et al*., 2013). Starch also may act as a bonding agent for lignocellulosic materials (Tondi *et al*., 2012; Lamaming *et al*., 2013). One disadvantage of starch was a big size of molecule and insolublity in water. The bigger size of starch molecule is expected to contribute to the lose performance of internal bonding of oil palm trunk particleboard compared to sucrose and glucose (Lamaming *et al*., 2013).

Maltodextrin was a starch-based polysaccharide produced by hydrolysis of starch down to glucose polymers with an average chain length of 5-10 glucose units. Maltodextrin is soluble in water, what makes it more attractive for industrial applications compared to insoluble starch. Maltodextrin has potential as a natural binder for the replacement of synthetic adhesives based on petroleum and has better adhesion properties than starch (Clare *et al*., 2002; Castro-cabado *et al*., 2016). Citric acid has been known to act as a natural binder that can provide excellent strength at the bonded material (Umemura *et al*., 2011; Widyorini *et al*., 2012; 2014; 2016a). In addition, citric acid has also been studied as a cross-linking agent for wood (Hasan *et al*., 2007) and starch (Reddy and Yang, 2010). Castro-cabado *et al*. (2016), reported the use of crosslinking systems based on maltodextrin (polysaccharides in general) and polycarboxylic acids as a fairly good eco-friendly alternative for binding glass or wood fibers applicable for furniture and insulation industry. Other studies have shown that when citric acid was used as a cross-linking agent, the carboxyl groups from citric acid had reacted with the hydroxyl groups from wood and reduced the hygroscopicity of wood as well as the tendency of wood to swell or shrink

(Rowell, 1991). Umemura *et al*. (2011) pointed out that ester linkages could be detected by fourier transform infrared spectroscopy (FTIR), which indicated that the carboxyl groups from citric acid could had been reacted with hydroxyl groups from wood, improving the performance of boards.

Research on maltodextrin and citric acid and their combination have been initiated in an attempt to increase the quality of the bonded particleboards. In the mixture of maltodextrin/citric acid, the researches on application as natural binders are still very limited. In the non-wood materials, i.e. nipa fronds, has not been studied about the mixture of maltodextrin/citric acid as a binder. Therefore, this paper was designed to investigate the characteristics of particleboard bonded using maltodextrin, and citric acid and their combination with various compositions and resin contents. Since the melting temperatures of maltodextrin and citric acid were different, the effect of pressing temperature was also studied in this research.

2. MATERIALS and METHODS

2.1. Board Materials

Nipa fronds particles were collected from the Jatimalang Beach Purworejo Middle Java Province and were used as the board material for these experiments. The particles were screened and particles which passed through aperture sizes of 10 mesh were used as materials in this research. According to the mesh analysis of particle size, about 69% of the particles used between 10-40 mesh, 6% between 40-60 mesh, 4% between 60-100 mesh and 3% passed through 100 mesh. All particles were air-dried to a moisture content of around 12%.

2.2. Preparation of Adhesive Solution

Maltodextrin DE 10-15 from Zhuceng Dongxiao Biotechnology Co. Ltd. (China) and citric acid anhydrous from PT. Budi Starch & Sweetener (Indonesia), were used without further purification. Maltodextrin and citric acid were dissolved in water with the solution concentration was adjusted variated to 50% for maltodextrin and 60% for citric acid (wt%). The mixture ratios of maltodextrin/citric acid were 100/0, 87.5/12.5, 75/25, and 0/100. The solutions were then used as an adhesive.

2.3. Board Manufacturing

The solution was used as an adhesive and sprayed onto the particles at 20% resin content based on the weight of oven dried particles. The sprayed particles were then oven-dried for 20 hours to 48 hours at 80℃ to reduce the moisture content. The moisture content of the mat was 2% to 4%. The particles were hand-formed into a mat using a forming box followed by hot pressing into particleboards with a distance bar of 1 cm to control the board thickness. The boards were pressed at 180℃ or 200℃ for 10 min under a pressure of 3.6 MPa. The target density was 800 kg/m^3

with board size of 25 × 25 × 1 cm and three particleboard panels were prepared for each experimental unit. Prior to the evaluation of the physical and mechanical properties, all boards were conditioned at ambient conditions for approximately one week.

2.4. Board Evaluation

Particleboards were evaluated according to the Japanese Industrial Standard for Particleboards (JIS A 5908, 2003). Tests were carried out to determine density, moisture content, thickness swelling, water absorption, surface roughness, modulus of rupture (MOR), modulus of elasticity (MOE) and internal bond (IB) strength. The density, moisture content, thickness swelling, water absorption and internal bond strength test was performed on a 5 × 5 × 1 cm specimen cut from each board. Both thickness swelling and water absorption tests were done after water immersion for 24 h at 20℃. The surface roughness was measured using portable surface roughness tester (SRG-4000), where average roughness (Ra) was used to evaluate roughness characteristics of the particleboards and eight measurements were randomly taken from both surfaces of the 20 × 5 × 1 cm specimen. Static three-point bending tests were conducted on a 20 × 5 × 1 cm specimen. The effective span length and cross-head speed were 15 cm and 1 cm/min, respectively. Each experiment was performed in triplicates, and the average value and standard deviation were presented.

2.5. Fourier Transform Infrared (FTIR) Spectroscopy

The FTIR (Shimadzu IR Prestige-21, Japan) analysis was performed. For this purpose, the bending strength test samples were used. To remove excess from free carboxylic acids and unreacted citric acid, the particleboard specimen was previously immersed in boiling water for 2 h and in water at room temperature for 1 h, respectively. The material was then dried at 40℃ overnight and powdered using a mill machine. All infrared spectra were obtained using the KBr disk method and recorded by means of an average of 10 scans at a resolution of 16 cm^{-1}.

2.6. Scanning Electron Microscope (SEM)

The 20% maltodextrin bonded particleboard samples were cut into cross sections and parallel sections approximately 0.05 cm. All samples were platinum-sputtered using sputter coater model Polaron SC 515 ± 20 nm. A LEO Supra 50 Vp scanning electron microscope (SEM) with ultra-high resolution was used to take the micrographs of the samples.

3. RESULTS and DISCUSSION

All of particleboards in this research could be manufactured without any delamination. The boards bonded with maltodextrin were lighter in color and come to darker with addition of citric acid. The different colors of the board with different binders might be attributed of the binder and other modification of chemical components

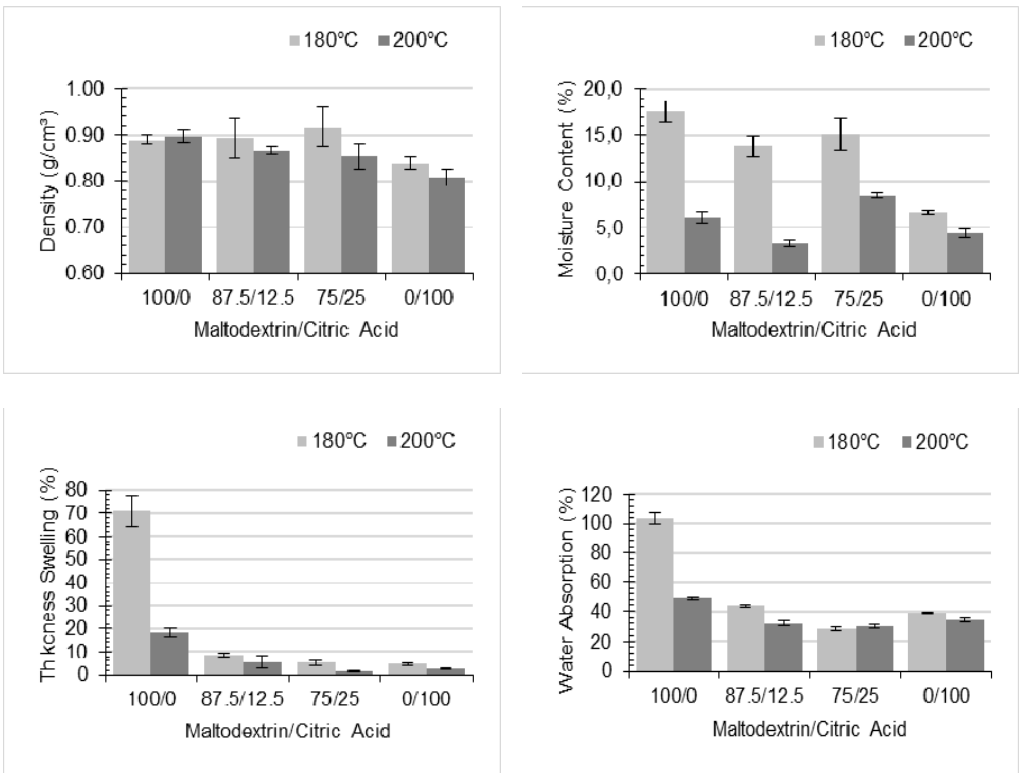

Fig. 1. Density, moisture content, thickness swelling and water absorption of particleboard at various maltodextrin/citric acid compositions and press temperatures. The vertical bar represents standard deviation.

during process. The darker color was also found at boards made from softwood and hardwood particles with sucrose and citric acid and their combination as binder (Umemura *et al*., 2011; Widyorini *et al*., 2016b). The darker color of boards with increasing press temperature indicating a high degree of hydrolysis or other modification of chemical components during treatment (Widyorini *et al*., 2005).

3.1. Physical Properties

Fig. 1 shows the results of densities, moisture content, thickness swelling and water absorption of the particleboards with various of maltodextrin/citric acid compositions and press temperature. The densities of bonded particleboards ranged from 0.81 to 0.92 g/cm^3, irrespective of the condition of manufacture. The moisture content of the all particleboards at press temperature 200℃ met the requirements of JIS A 5908 (5 - 13%). In the press temperature 180℃, only particleboard using 100% citric acid that met the JIS A 5908 moisture content standard. Adding citric acid and press temperature significantly effect to the lower moisture content.

On the maltodextrin bonded particleboards without added citric acid, dimensional stability

of the board was extremely low. After the addition of citric acid, the thickness swelling and water absorption were decreased significantly, and the thickness swelling met the requirement of JIS A 5908 (max 12%), indicating the particleboard had good dimensional stability. This is supposedly due to the ester-linked bonds that were formed. The same results also deduced for particleboard made from softwood and hardwood bonded with sucrose/citric acid (Umemura *et al*., 2013; Widyorini *et al*., 2016b). Increasing press temperature from 180℃ to 200℃ also improved the dimensional stability at the same composition. The pressing temperature was an important component of the reaction between carboxyl groups and hydroxyl groups to form ester groups (McSweeny *et al*., 2006). It was concluded that a press temperature of 200℃ was needed to obtain good dimensional stability of the particleboards, as also found by Umemura *et al*. (2014) and Widyorini *et al*. (2016b).

On the maltodextrin only bonded particleboards, increasing the press temperature greatly reduced the moisture content, thickness swelling and water absorption values of the particleboard. This is supposedly due to the melting point of maltodextrin has been reach on the press temperature 200℃. Research on citric acid found the melting point is one of the critical factors on the particleboard processing. The excellent properties on the citric acid bonded particleboard observed at 180℃ press temperature, the properties were extremely low because this temperature is lower than the melting temperature of citric acid (Umemura, *et al*., 2014).

3.2. Surface Roughness and Mechanical Properties

Fig. 2 shows the surface roughness, internal bonding and bending properties of the particleboards bonded with different ratios of maltodextrin-citric acid and press temperature. The surface roughness values of nipa frons particleboard decreased simultaneous with added the citric acid. On the other side, pressing temperature did not significantly influence to the surface roughness of particleboard. The best surface roughness on this research were 4.19 μm and 4.90 μm taken from citric acid only bonded particleboard and pressing temperature 200℃ and 180℃ respectively, follow by particleboard bonded with maltodextrin/citric acid 75/25 on press temperature 200℃ were 5.19 μm. The average roughness values of commercially manufactured particleboard in Japan ranged from 3.67 to 5.46 μm (Hiziroglu and Suzuki, 2007). Compared to those studies, the average roughness values of nipa frond particleboard bonded with citric acid on press temperature 180℃ and 200℃ and maltodextrin/citric acid 75/25 on press temperature 200℃ were found within the above range. Widyorini *et al*. (2016a) found the addition of citric acid on bamboo particleboard resulted in better contact among particles, hence better adhesion occurred and finally producing smooth surface.

All of the internal bonding of particleboards pressed at 200℃ met the requirements of JIS A

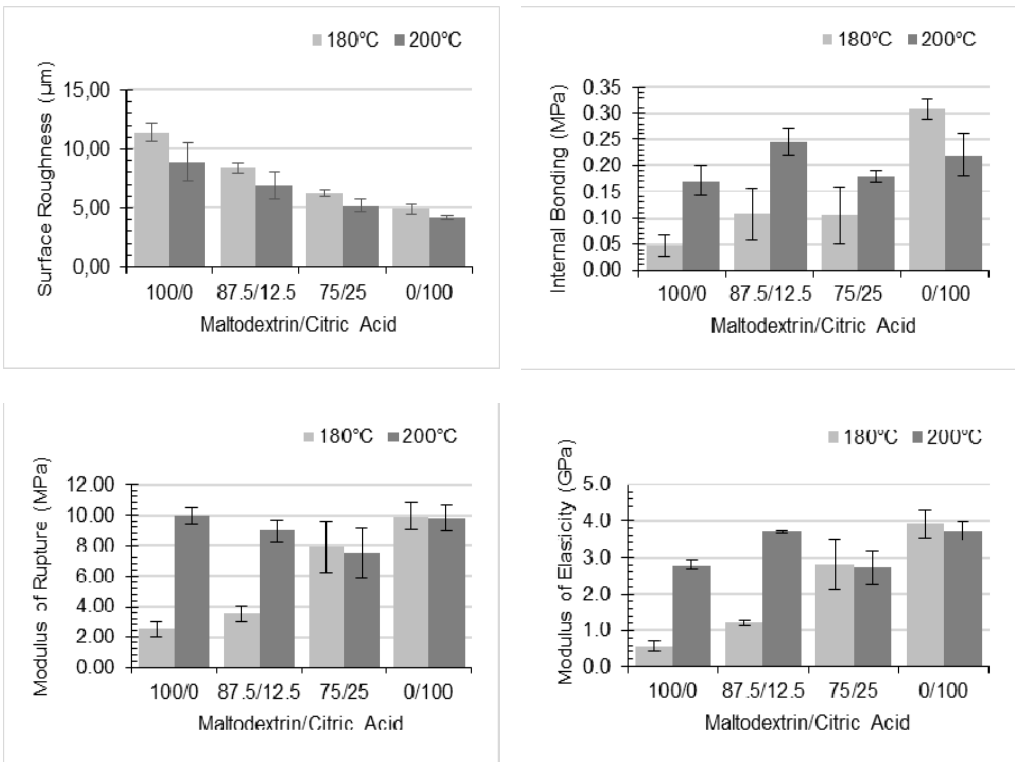

Fig. 2. Surface roughness and mechanical properties of particleboard at various maltodextrin/citric acid compositions and pressing temperature. The vertical bar represents standard deviation.

5908 (2003) type 8 (greater than 0.15 MPa). At the press temperature 180℃, only the particleboards bonded using 100% citric acid that met the JIS A 5908. The highest internal bonding was achieved at press temperature 180℃ bonded using 100% citric acid with an internal bonding of 0.31 MPa, which met the requirement of JIS A 5908 (2003) type 18. Very interesting, on the maltodextrin and its combination with citric acid, the value of internal bonding improves significantly with increasing the press temperature from 180℃ to 200℃. In the ratio of maltodextrin/citric acid 100/0, the internal bonding increased up to three time and in ratio maltodextrin/citric acid 87.5/12.5 increased up to two and half time. The melting temperature of maltodextrin could be playing as a major role here. The same trend also found on another natural binder research i.e. combination of sucrose/citric acid on softwood and hardwood. On softwood particleboard, Umemura *et al*. (2013) reported that the optimum condition was achieved at press temperature 200℃ using citric acid/sucrose ratio of 75/25. On hardwood particleboard (teakwood particle), Widyorini *et al*. (2016b) report the optimum condition was achieved at a press temperature 200℃ using citric acid/sucrose ratio of 100/0.

The bending properties data show that the composition of maltodextrin/citric acid and pressing temperature affects to the modulus of rupture and modulus of elasticity. At the 180℃ press temperature, the bending properties increased gradually as the citric acid ratio increased. But only maltodextrin/citric acid 75/25 and 100% citric acid that met the requirements of JIS A 5908 (min 8 MPa for modulus of rupture and 2 GPa for modulus of elasticity). An increase in the press temperature from 180℃ to 200℃ made all particleboard on this research met the JIS A 5908, but the composition of maltodextrin/citric acid did not greatly affect here. The best modulus of rupture and modulus of elasticity on this research achieved at a pressing temperature of 180℃ and using 100% citric acid, obtaining values of 9.91 MPa and 3.93 GPa, respectively. The different results found on Widyorini *et al*. (2016b) for teakwood particleboard where the highest modulus of rupture and modulus of elasticity achieved at a pressing temperature of 200℃ and the citric acid/sucrose composition did not greatly affect the modulus of rupture and modulus elasticity. Another research from Umemura *et al*. (2013) also achieved different result, where the optimum condition was noted at a sucrose/citric acid ratio of 75/25 for softwood. These different results could be the effect of chemical components of raw materials and binder. This result was consistent with the internal bonding strength of the particleboards. All of particleboards bonded with 100% citric acid at the 180℃ pressing temperature had higher bending strength compared to another particleboard. But, very interesting to discuss, the mechanical properties of maltodextrin bonded particleboard and its combination with citric acid improve significantly with increasing the press temperature from 180℃ to 200℃, these could be the effect of melting point temperature of maltodextrin that had been reach at 200℃.

3.3. FTIR Analysis

To investigate the bonding mechanism of the board, FTIR analysis were performed. A carbonyl (C=O) of the ester and carboxyl is indicated at approximately 1709 to 1740 cm^{-1} (Castro-Cabado *et al*., 2016). The carbonyl ester group was present because of the reaction of the hydroxyl groups from the lignocellulosic materials and carboxyl groups from citric acid (Umemura *et al*. 2011). On the maltodextrin 100% bonded particleboard (signal number 1 on Fig. 3a and 3b), there was detected the carbonyl group (C=O) at approximately 1720 cm^{-1}. According to Widyorini, *et al*. (2005), these groups resulted from the degradation of hemicellulose during pressing.

Fig. 3a shows an FTIR analysis of particleboard at press temperature 180℃. On the maltodextrin 100% bonded particleboard (signal number 1 on Fig. 3a), comparing to the maltodextrin with citric acid addition (signal number 2 and 3 on Fig. 3a) and the citric acid only bonded particleboard (signal number 4 on Fig. 3a), the peak signal at 1720 cm^{-1} was lower than that. The lower intensity of C=O indicat-

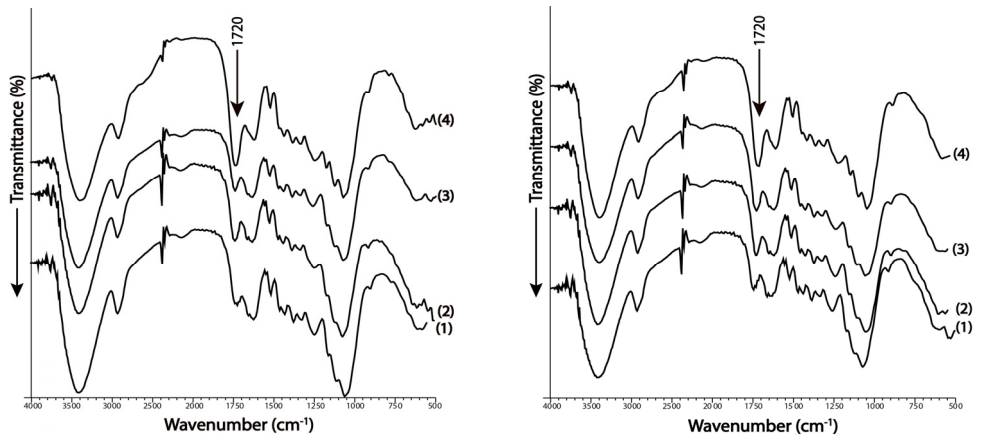

Fig. 3. FTIR analysis of nipa frond particleboard for a) press temperature 180℃ and b) press temperature 200℃: 1. maltodextrin/citric acid 100/0; 2. maltodextrin/citric acid 75/25; 3. maltodextrin/citric acid 87.5/12.5; 4. maltodextrin/citric acid 0/100.

ing the ester linkages is lower too. The specimens after boiling treatment were used as FTIR samples in this research, therefore unreacted citric acid and some water soluble components in the board were expected to be eluted during the treatment. As the result, groups that were present at 1720 cm^{-1} that detected on the FTIR analysis was attributed to the formation of ester linkages in particleboard.

Fig. 3b shows an FTIR analysis of particleboard at press temperature 200℃. On the maltodextrin only bonded particleboard, the peak around at 1720 cm^{-1} was higher than at press temperature 180℃. This means that a press temperature of 200℃ is likely to be a critical temperature to form carbonyl group on this research. The same trend also found on the bamboo particleboard bonded using 100% citric acid and teakwood particleboard using 100% citric acid and 100% sucrose (Widyorini *et al*., 2014; 2016b). The intensity of peak around at 1720 cm^{-1} increased as the amount of citric acid increased. The peak is generally attributed to C=O stretching derived from the carboxyl group and/or the C=O ester group and the formation of groups ester linkages would contribute to the hydrophobic tendencies and high dimensional stability of particleboard (Umemura *et al*., 2011; Widyorini *et al*., 2016b).

3.4. SEM Analysis

The SEM micrographs of maltodextrin bonded particleboard at 200℃ press temperature is shown in Fig. 4. The SEM micrographs revealed the compressed fiber on cross section area (Fig. 4a).

The maltodextrin granules was not detected (Fig. 4b), suggesting that the maltodextrin during hot pressing, could have been melt, did

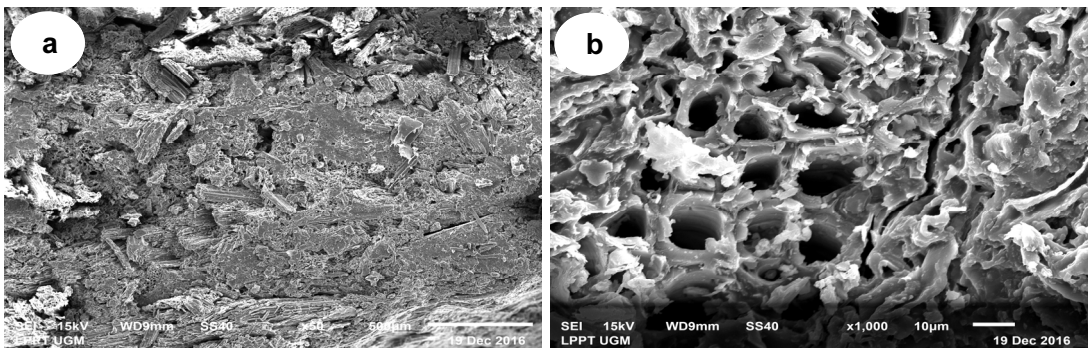

Fig. 4. SEM analysis of nipa frond particleboard 20% resin content and press temperature 200℃ for cross section a) magnified x50 and b) magnified x1000.

flow, penetrated into cells, filled the lumen void area, covered the lignocellulosic fiber and acted as an adhesive to bind the particles together. In the term of molecule size, maltodextrin on this research still better compared to the starch. Lamaming *et al*., (2013) found the size of the potato starch granule was big and it could not melt and flow to penetrate into cells during hot pressing and this phenomenon expected contribute to the lose performance of internal bonding of oil palm trunk particleboard compared to sucrose and glucose.

4. CONCLUSION

Combining maltodextrin with citric acid and increasing the press temperature from 180℃ to 200℃ improves significantly the physical and mechanical properties of nipa frond particleboard. The properties of citric acid-bonded nipa frond particleboard in this research met the requirements of the JIS standard A 5908 for particleboard. The optimum properties in this research taken from citric acid 100% at 180℃ press temperature were internal bond strength 0.31 MPa, modulus of rupture 9.93 MPa, modulus of elasticity 3.93 GPa, average roughness 4.90 µm, thickness swelling 4.58% and moisture content 6.71%. Fourier transform infrared analysis showed that the presence of ester groups was higher with adding citric acid content. The scanning electron microscopy analysis proved the molecule size of maltodextrin extremely small and that make this binder more soluble in water, more easy to applicated and better quality of bending performance compare to starch. Maltodextrin did not act as a good bonding agent when pressed at 180℃; however, its bonding properties could be improved significantly when it was mixed with citric acid and press at 200℃.

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