

Influence of Chemical Components of Oil Palm on Properties of Binderless Particleboard

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The influence of chemical components of oil palm on properties of binderless particleboard were evaluated through a series of mechanical, physical, and chemical analyses in order to assess the self-bonding mechanism. Binderless particleboards were evaluated by relating the physical and mechanical properties to the chemical components. Results revealed that the addition of glucose and sucrose onto the board with and without extraction increased the modulus of rupture and internal bond strength. Glucose and sucrose also reduced the thickness swelling and water absorption of the board. The addition of starch onto the board enhanced the strength of the board, though sugar addition enhanced the board strength more than starch. Adding the sugar also lowered the xylose/arabinose ratio, indicating that the boards consist of short-chain polymers with a large amount of branching with other monosaccharides. This implies that sugar content present in oil palm trunk plays a major role in the bonding of binderless boards.

Keywords: Binderless board; Additives; Self-bonding; Oil palm trunk

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INTRODUCTION

Oil palm has become well established in commercial plantations in Malaysia, and the planting is still increasing dramatically. The cultivation area covered 5 million hectares in 2011, an increase of 3.0% compared to 4.85 million hectares recorded in 2010 (MPOB 2011). The industry produces a large quantity of biomass waste in the form of empty fruit branches, trunks, kernel shell fronds, and other waste waiting to be utilized judiciously. A promising use of plantation waste is the production of particleboards from oil palm trunks. In the production of particleboard, a problem faced by manufacturers is the high cost due to the use of adhesives. Omitting the use of adhesives in particleboard production has the potential to bring new hope to produce low-cost, environmentally-friendly products.

The self-bonding process occurs when wood or non-wood based fragments are converted into boards by steam or heat treatment without the use of adhesives (Shen 1991). This board, called a binderless particleboard, depends only on the activating chemical components that exist in the board during steam or heat treatment (Okuda and Sato 2004). Binderless particleboard strength depends on the self-bonding that occurs within the particles during the production of the board. Chemical components, namely cellulose, lignin, hemicelluloses, sugar, and starch, have the potential to create bonding between

particles without using any synthetic resin. Various researchers engaged in the topic of binderless boards have postulated that the mechanism of self-bonding involves activation of various chemical components of boards. This can include degradation of hemicelluloses and part of the cellulose to produce simple sugars and other decomposition products (Shen 1986; Rowell *et al.* 2002; Widyorini *et al.* 2005), thermal softening of the cell wall matrix (Tanahashi *et al.* 1989), cross-linking between carbohydrate polymers and lignin (Suzuki *et al.* 1998), and an increase in cellulose crystallinity (Tanahashi 2002).

Okuda and Sato (2004) noted that the manufacturing of binderless board mainly involves the application of heat and pressure to an insoluble and infusible polymeric substance, where the natural sugars and other water soluble material within the lignocellulosic material are chemically transformed through the hydrolysis of hemicelluloses and softening of lignin. The sugars eventually act as bonding agents and strengthen the reconstituted composite products with high mechanical strength and dimensional stability, thus improving the properties of the boards. According to Back (1987), application of heat and pressure treatment can produce auto-cross-linking within the fibers.

Research that aims to enhance the properties of the binderless board so that it can compete with commercial products is still in progress. Previous work (Hashim *et al.* 2010; Hashim *et al.* 2011a, 2011b) only dealt with the factors affecting the properties of binderless boards and having a positive effect on the properties of boards. However, there has been no research providing detailed information regarding the bonding mechanism of the boards. Thus, this study investigates the self-bonding mechanism of binderless board, particularly with respect to the role of sugars and starch. In order to investigate the factors that contribute to the self-bonding of the boards, additives were added onto the boards, and the effect of their addition on the properties of the boards were evaluated through mechanical, physical, and chemical analyses. The findings on this study have potential to benefit future work, as the information will help researchers working to enhance the strength of binderless board.

EXPERIMENTAL

Preparation of Samples

Old and waste oil palm trunks were obtained from a plantation in Kedah, Malaysia and were sawn, chipped, and air-dried. Samples were ground into particles within the range 0 to 1000 μm using a Willey Mill before they were screened. Fine particles with average moisture content of 7% were stored in plastic bags. Oil palm trunk particles were extracted with hot distilled water ($60\text{ }^{\circ}\text{C} \pm 3\text{ }^{\circ}\text{C}$ thermostat controlled) for 6 h and then filtered using a Buchner funnel. For chemical analysis, all board types were finely ground to pass a 0.4 mm (40-mesh) screen to allow a complete reaction of the sample with reagents used in the analysis. The sampling and the preparation of wood for analysis were performed based on TAPPI T264 cm-97.

Methods

Board production

There were eight types of boards with six replications for each treatment: unextracted particle (control board), extracted particles by water extraction, unextracted, and extracted particles by water extraction with additives of glucose, sucrose, and starch. The weight of the additives added was 20% based on the oven dried

weight of the particles for board making. This amount was based on the amount of extractives lost during the water extraction process. All the additives were combined by mixing them evenly together with the oil palm trunk particles with and without extraction. All boards were fabricated at a target density of 0.8 g/cm^3 by hot pressing at 180°C for 20 min with 5 MPa pressure. The board dimensions were 205 mm (W) \times 205 mm (L) \times 48 mm (H).

Evaluation on mechanical and physical properties of boards

The modulus of rupture (MOR), internal bond strength (IB), thickness swelling (TS), and water absorption (WA) of the boards were evaluated according to JIS A 508-2003 (Particleboards). A bending test was conducted with a concentrated load of 10 mm/min. Specimen size for bending tests was 50×200 mm with effective span of 150 mm. The sample size for the IB, TS, and WA tests were 50×50 mm. Thickness swelling (TS) and water absorption (WA) tests were performed by immersing the sample in water for 24 h in the conditioning room. Surface roughness of the samples was also conducted by using a portable stylus-type T-500 Hommel tester. Four samples from each panel type with measurements of 5 mm \times 5 mm were used for roughness measurements. Measurements with a tracing span of 15 mm were taken from each side of the samples. Average roughness (R_a) was calculated from acquired digital data (Hiziroglu *et al.* 2004).

Evaluation on chemical components

Preparations of extractive free samples were done according to TAPPI 204 cm-97 with a modification of the solvent ethanol-toluene ratio of 2:1. Holocellulose content was performed by the method of Wise *et al.* (1946), while the alpha cellulose content was measured by the extraction of the holocellulose with 17.5% sodium hydroxide. Lignin content was determined according to TAPPI 222 om-02.

Sugar analysis

The contents of compositional sugars were determined by the two-step H_2SO_4 hydrolysis method by Jeungyil *et al.* (2009). About 100 mg of oil palm trunk particles were hydrolyzed in 1 mL of 72% (w/w) H_2SO_4 at 30°C for 1 h. The mixture was then diluted by adding 7 mL of distilled water, and hydrolyzed at 121°C for 1 h. Then, the mixture was centrifuged at $10\,000 \times g$ for 3 min, and the supernatant was neutralized with 10% (w/v) NaOH. The supernatant was then filtered using a microfilter with the pore size of $0.45 \mu\text{L}$ Nanosep MFGHP and centrifuged for 1 min at 5.0 rpm. Then $100 \mu\text{L}$ of the centrifuged sample was taken and added with $850 \mu\text{L}$ of Double Deionizer (DDI) HPLC grade water and $50 \mu\text{L}$ ribose D-(-) -R7500 from sigma Aldrich as a standard, then stirred evenly. Samples were then placed into the HPLC auto sampler rack for analysis. The sample was also used for measuring total sugar using a colorimetric method devised by Dubois *et al.* (1956).

Microstructure study

The binderless particleboard samples were cut into cross sections of approximately 0.5 mm. All samples were gold-sputtered using sputter coater model Polaron SC 515 ± 20 nm. A LEO Supra 50 Vp field emission scanning electron microscope (FESEM) with ultra-high resolution was used to take the micrographs of the samples.

RESULTS AND DISCUSSION

Evaluation on Mechanical and Physical Properties

Figure 1 presents the modulus of rupture (MOR) values for all the boards. The highest MOR value of 13.6 MPa was determined for the board made with the addition of 20% sucrose (C), and the lowest MOR value, 3.92 MPa (E), was determined for the boards with particles that underwent the extraction process. This result revealed that the MOR increased with the addition of glucose, sucrose, and starch to the board. The additions to both the unextracted and extracted boards improved the strength. Only the board made with the addition of either 20% glucose or 20% sucrose passed the Japanese Industrial Standard (JIS) A-5908 for MOR of 8.0 MPa. Similar trends were also observed for the internal bond strength (IB) values (Fig. 2) when adding the additives to boards, and the MOR and IB results were correlated with each other.

Figure 2 shows that unextracted board made with the addition of 20% sucrose provided the highest IB value of 1.91 MPa (C), followed by samples made with the addition of 20% glucose, which had an IB value of 1.71 MPa (B). All the boards met the requirements of the JIS A-5908 Type-8 for IB values of 0.15 MPa except for the panels that underwent extraction with the addition of starch and had an IB value of 0.12 MPa (H). Comparing the increment in MOR and IB values between the unextracted (control) and extracted boards (A and E) with the boards with the addition of additives indicated that sugar played a major role in improving the properties of boards. Thus, this result suggests that sugar contributed to the bond formation in the boards.

Thickness swelling (TS) and water absorption (WA) values are illustrated in Fig. 3. The TS and WA values of the boards decreased with the addition of 20% glucose and sucrose onto the boards (F, G). The reduction of TS for both unextracted and extracted boards made with the addition of 20% glucose and sucrose was reduced twice when compared to the control and extracted board (A and E). The WA values also decreased from 90.7% to 57.9% and 59.9% for the board made with the addition of either 20% glucose or 20% sucrose. As hot water extraction removed a greater quantity of materials and a portion of the cell wall as well as extracted some inorganic materials (Shebani *et al.* 2008), the TS and WA values for the extraction board increased twice when compared to the control.

When subjected to soaking, the hydrophilic properties of lignocellulosic materials and the capillary action induced an uptake of water and thus increased the TS and WA values (Norul Izani *et al.* 2012). The value decreased with the addition of glucose or sucrose to the extracted particles and improved the physical test results of the boards. This result suggested that the improvement was due to the sugars themselves, similar to the case of MOR and IB. Even though the addition of starch to the board also improved the physical properties to some extent, the improvement was not as great as with the addition of sugars. Despite the improvement with the addition of additive on the properties of boards, none of the panel types of boards were able to meet the JIS A-5908 Type-8 specification, which is 12% for TS.

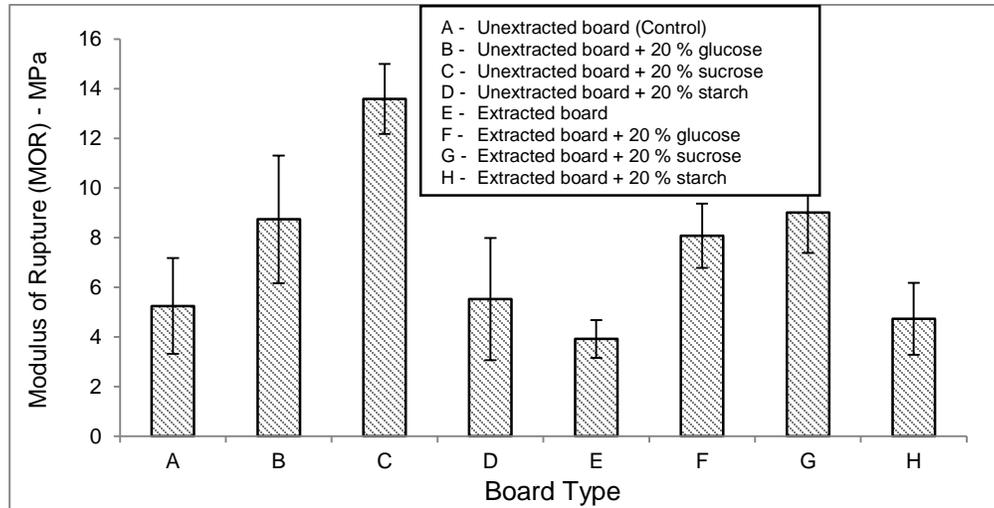


Fig. 1. Modulus of rupture of different panel boards types

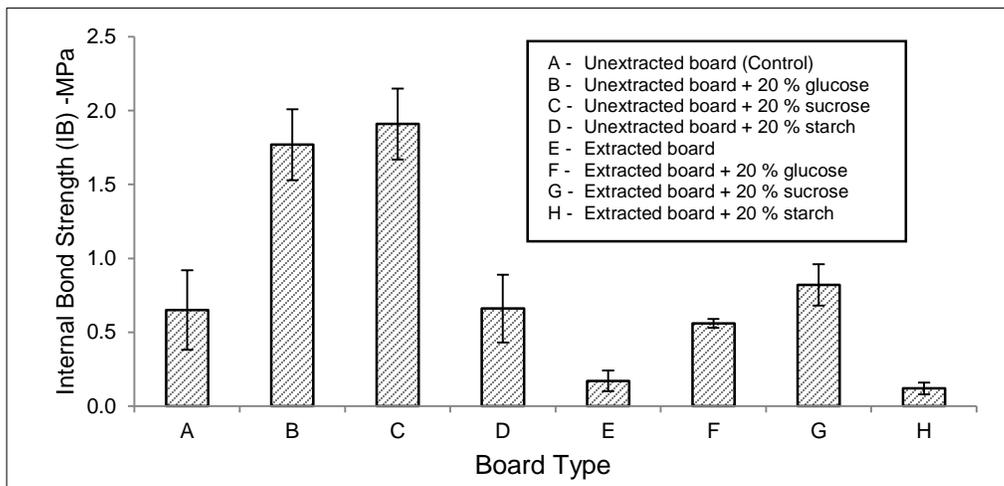


Fig. 2. Internal bond strength of different board types

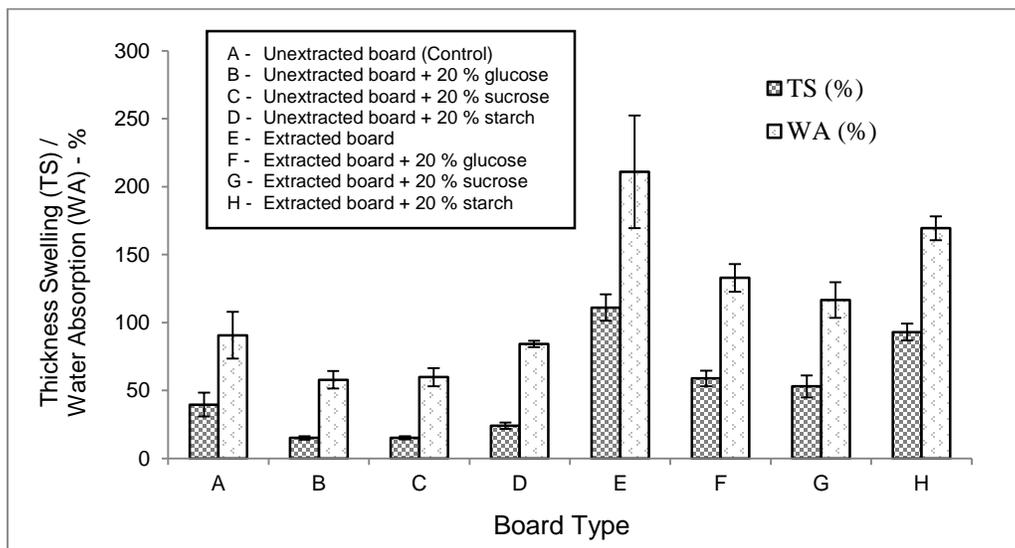


Fig. 3. Thickness swelling and water absorption of different board types

Figure 4 displays average R_a values for six types of manufactured boards. Since typical particleboard has R_a values that range from 3 μm to 10 μm depending on the raw material, particle size, and surface densification (Hiziroglu *et al.* 2004), the roughness values for the manufactured boards in this study falls within the range.

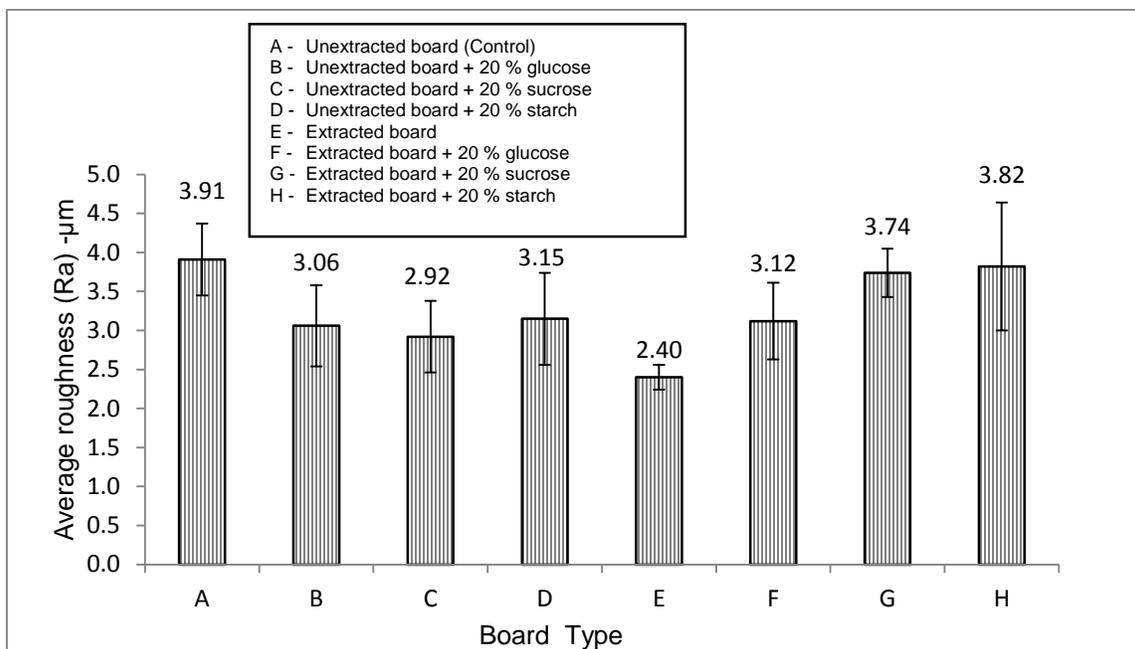


Fig. 4. Average surface roughness (R_a) of all type of boards

Evaluation on Chemical Components

Chemical compositions for all types of boards are presented in Table 1.

Table 1. Chemical Composition and Starch Content of Different Types of Board

Board Type	Extractives (%)	Chemical Compositions (%)			
		Holocellulose	α -cellulose	Hemicellulose**	Lignin
Unextracted Board (Control)	12.2 (0.49)	69.8 (3.59)	59.9 (3.20)	9.9	21.0 (0.29)
Unextracted Board + 20% glucose	22.9 (3.48)	61.1 (1.03)	52.9 (3.23)	8.2	22.7 (3.48)
Unextracted Board + 20% sucrose	27.6 (1.34)	57.6 (1.09)	51.3 (1.25)	5.3	27.6 (1.34)
Unextracted Board + 20% starch	7.5 (0.08)	52.4 (1.26)	48.1 (1.20)	4.3	14.1 (2.46)
Extracted Board	4.5 (0.20)	78.8 (0.43)	51.3 (2.99)	27.5	16.3 (0.14)
Extracted Board + 20% glucose	18.2 (0.53)	72.3 (1.85)	53.2 (2.23)	19.1	18.2 (0.53)
Extracted Board + 20% sucrose	17.9 (1.08)	63.7 (0.53)	55.2 (4.62)	8.5	17.9 (1.08)
Extracted board + 20% starch	3.7 (0.31)	62.9 (6.16)	48.8 (0.30)	14.1	8.19 (4.00)

* Values in parentheses are standard deviation

** Hemicellulose content was calculated based on the deduction of holocellulose to α -cellulose

Oil palm trunk is a lignocellulosic material, which implies that it is a polymeric composite primarily composed of cellulose, hemicelluloses, and lignin. The yield of the extractives was detected as being higher in the board made with the addition of either 20% glucose or 20% sucrose (B and C) compared to the control board (A). The lowest value was determined for the extracted board (E), as the hot water removed water-soluble materials which were 4.5%. Both control and extraction board with sugar addition showed an increase in yield.

An increment of lignin content was observed after the addition of sugar, probably due to changes in structure of oil palm trunk during heat treatment. An increase in lignin content also can be attributed to the loss of hemicelluloses or fragile pentoses and hexoses during heat treatment (Kamden *et al.* 2002; Salim *et al.* 2008). The polymerization of carbohydrates and polycondensation reactions involved in cross-linking of the lignin network can also contribute to the apparent increase of lignin content (Boonstra and Tjeerdsma 2006). It has been reported that at a temperature around 180 °C, homolytic cleavage of β -ether linkages and formation of radicals, condensation products, and cross links between lignin and polysaccharides have occurred (Tjeerdsma *et al.* 1998; Salim *et al.* 2008). The lignin also covers the hemicelluloses and cellulose and prevents the hydrophilic groups of these two sugar polymers from taking up water. This can explain why the thickness swelling and water absorption properties decreased as the lignin content increased for the board with the addition of additives. A better inter-fiber bonding occurred due to the lignin fluidity on fibers, which contributed to the improvement of lignin distribution of the boards (Quintana *et al.* 2009).

The holocellulose content in the binderless particleboard showed a decrease in value, as depicted in Table 1. The changes in holocellulose content occurred when sugar and starch were added to either the unextracted and extracted boards. The reduction of holocellulose content was explained by the depolymerisation of the hemicelluloses to form soluble sugar and by some degradation of cellulose during hot pressing (Boonstra and Tjeerdsma 2006).

Cellulose gives mechanical strength to plant tissues, while lignin provides rigidity and stiffness. Therefore, it is well known that intensive degradation of celluloses and lignin decrease mechanical quality of the boards (Widyorini *et al.* 2005). Cellulose was found as the main constituent in oil palm trunk binderless boards. From the Table 1, it can be seen that the cellulose content ranged from 48% to 60%. A high cellulose content of 40% to 50% contributes and supplements to the binding property (Pandey and Nema 2004). Generally, high cellulose content will give strength and stability to fiber cell walls. Fibers that contain a high level of cellulose and low micro fibril angle when being pulled will create a higher tensile strength. In other words, a low microfibril angle will give ductility to the fiber. As reported before, cellulose degradation reactions happened at a temperature above 210 to 220 °C (Back 1987). Different process conditions and treatment time applied during the heat treatment may influence degradation rate of cellulose content (Boonstra and Tjeerdsma 2006; Salim *et al.* 2008). However, in this experiment, the temperature applied was only 180 °C for 20 min.

From Table 1, the hemicelluloses content for the control and extracted board samples with and without addition of additives were in the range of 4 to 28 %. During heating, acetic acid is formed from acetylated hemicelluloses by hydrolysis. The released acid serves as a catalyst in the hydrolysis of hemicelluloses to soluble sugars. In addition, the acetic acid that has formed depolymerizes the cellulose micro fibrils in the amorphous area. The acid hydrolyses the bonds joining the units of glucose, breaking the cellulose into shorter chains. However, the breaking of a hemicellulose chain does not reduce the strength of oil palm particles as much as the breaking of cellulose chains would do. In the dry state, the glass transition of hemicellulose has been reported to be around 170 °C (Back 1987). The

hemicelluloses content presented in a low amount may be due to degradation of the hemicelluloses onto soluble sugars. The degradation of the hemicelluloses results in free sugars, which in turn form furan intermediates that can undergo polymerization during hot pressing, resulting in the formation of adhesives that bind the particles together (Rowell *et al.* 2000).

Sugar Analysis

The amount of compositional sugars obtained after hydrolysis, as well as the total sugar determined by colorimetric method are exhibited in Table 2. All samples consisted mainly of glucose, xylose, arabinose, and fructose. Glucose and xylose were predominant sugar constituents in all samples tested using HPLC, and this result matched previous research (Hashim *et al.* 2011a; Tomimura 1992). We did not detect sucrose in our analysis, contrary to the results reported by other researchers (Yamada *et al.* 2010; Kosugi *et al.* 2010), who reported that sucrose is one of the main sugars in the oil palm. This may be due to the degradation of sucrose to glucose and fructose. In this experiment, the sample was in particulate form and not in sap. For the sample without extraction (A-D), addition of starch onto the board had the highest glucose content of 13.4 mg/mL (D) followed by panels added with sucrose and glucose which were 9.7 mg/mL (C) and 9.3 mg/mL (B), respectively. All boards showed an increment in the glucose content when comparing with the control board of 8.9 mg/mL (A).

The extraction process removed water-soluble materials from the particles. Therefore, the sugar content was expected to be decreased in the extracted board. With the series of extracted board (E-H), the trend of the increment and higher content of glucose was similar to the control board; the board with the addition of starch exhibited the highest glucose content of 13.8 mg/mL (H), followed by the extracted board with sucrose or glucose added, which exhibited 12.2 mg/mL (G) and 9.9 mg/mL (F), respectively. The higher content and the increase of glucose were due to the sum of glucose derived from cellulose, starch (Xiao *et al.* 2001), and the additives.

Previous studies by several researchers have reported that self-bonding reactions in the board could be due to the degradation of both hemicelluloses and part of the cellulose to produce simple sugar and other decomposed substances (Shen 1986; Rowell *et al.* 2002; Widyorini *et al.* 2005). Hemicelluloses share the basic chemical structure but differ in the manner of substitution of the xylan backbone. The main differences were found in the ratio of xylose to arabinose (xyl/ara). The ratio of xylose to arabinose indicates the degree of linearity or branching of hemicelluloses. A high xylose to arabinose ratio would indicate a higher degree of polymerization with little bonding with other monosaccharide constituents. A low xylose to arabinose ratio suggests a short chain polymer with a large amount of branching with other monosaccharides (Lawther *et al.* 1995; Xu *et al.* 2007).

From Table 2 it can be observed that the board with and without addition of sucrose had the lowest xylose/arabinose ratio, which was 1.29. The board manufactured with the addition of additives also showed the lowest ratio for the extracted board with the addition of sucrose, which was 1.52. Adding the additives to the unextracted and extracted board lowered the xylose/ arabinose ratio. Based on the xylose/arabinose ratio result, it can be concluded that the board made with the addition of sucrose consisted of short chain polymers with a large amount of branching with other monosaccharides. This can explain why the panel made with the addition of sucrose showed the highest MOR value when compared to the unextracted and extracted boards.

The trend of total sugar content in all boards using the phenol sulphuric method showed the same pattern as those obtained by compositional sugar analysis. From Table 2, the board made with the addition of sucrose for both unextracted and extracted showed the highest amounts of sugar, which were 20.8 and 22.5 mg/mL. The lowest total sugar content of 15.86 mg/mL was detected in the extracted board, which was likely because the hot water removed some water-soluble sugar during the extraction process.

Table 2. Compositional Sugars of all Types of Board

Sample	Sugar mg/mL					Xylose/Arabinose ratio	Total sugar (mg/mL)
	Glucose	Xylose	Arabinose	Fructose	Total		
Unextracted board (control) (A)	8.9 (0.45)	4.8 (0.72)	3.0 (0.54)	1.4 (0.03)	18.1 (1.16)	1.63	16.4
Unextracted board + 20% glucose (B)	9.3 (1.18)	5.2 (1.32)	3.3 (0.97)	2.1 (0.10)	20.0 (3.38)	1.58	18.4
Unextracted board + 20% sucrose (C)	9.7 (0.52)	5.3 (1.71)	4.1 (0.94)	2.9 (0.54)	21.9 (0.43)	1.29	20.9
Unextracted board + 20% starch (D)	13.4 (0.52)	3.4 (0.47)	2.0 (0.54)	2.8 (0.24)	21.6 (1.77)	1.66	20.4
Extracted board (E)	6.6 (0.04)	2.8 (0.50)	1.2 (0.13)	4.4 (0.04)	15.1 (0.54)	2.27	15.9
Extracted board + 20% glucose (F)	10.0 (1.69)	7.0 (1.29)	3.9 (1.29)	-	20.9 (0.45)	1.77	21.0
Extracted board + 20% sucrose (G)	12.2 (0.30)	6.6 (1.35)	4.4 (1.01)	-	23.2 (2.23)	1.52	22.5
Extracted board + 20% starch (H)	13.8 (0.30)	5.0 (1.08)	2.7 (0.79)	-	21.4 (2.82)	1.83	22.0

*Values in parentheses are standard deviation

Microstructure Study

The SEM micrographs of all boards after board making are shown in Figs. 5 and 6. Many bell ring-like starch granules can be seen clearly. Most of the granules were of oval shape, although round- and spherical-shaped granules were also found (Hashim *et al.* 2010; Hoover 2001). Compressed fibers and starch granules arranged uniformly can be seen in Figs. 5a and d, whilst in Fig. 6a and b, the starch granules are disorderly arranged. The starch granules can be found between the parenchymatic ground tissues or in the lumen of the cells.

Some of the starch granules and parenchyma cells were ruptured; this may be due to loss of the water-soluble components that hold the fibers during the extraction process, as can be observed in Fig. 6a. The surfaces of the granules appear smooth in the SEM images, with no evidence of any fissures.

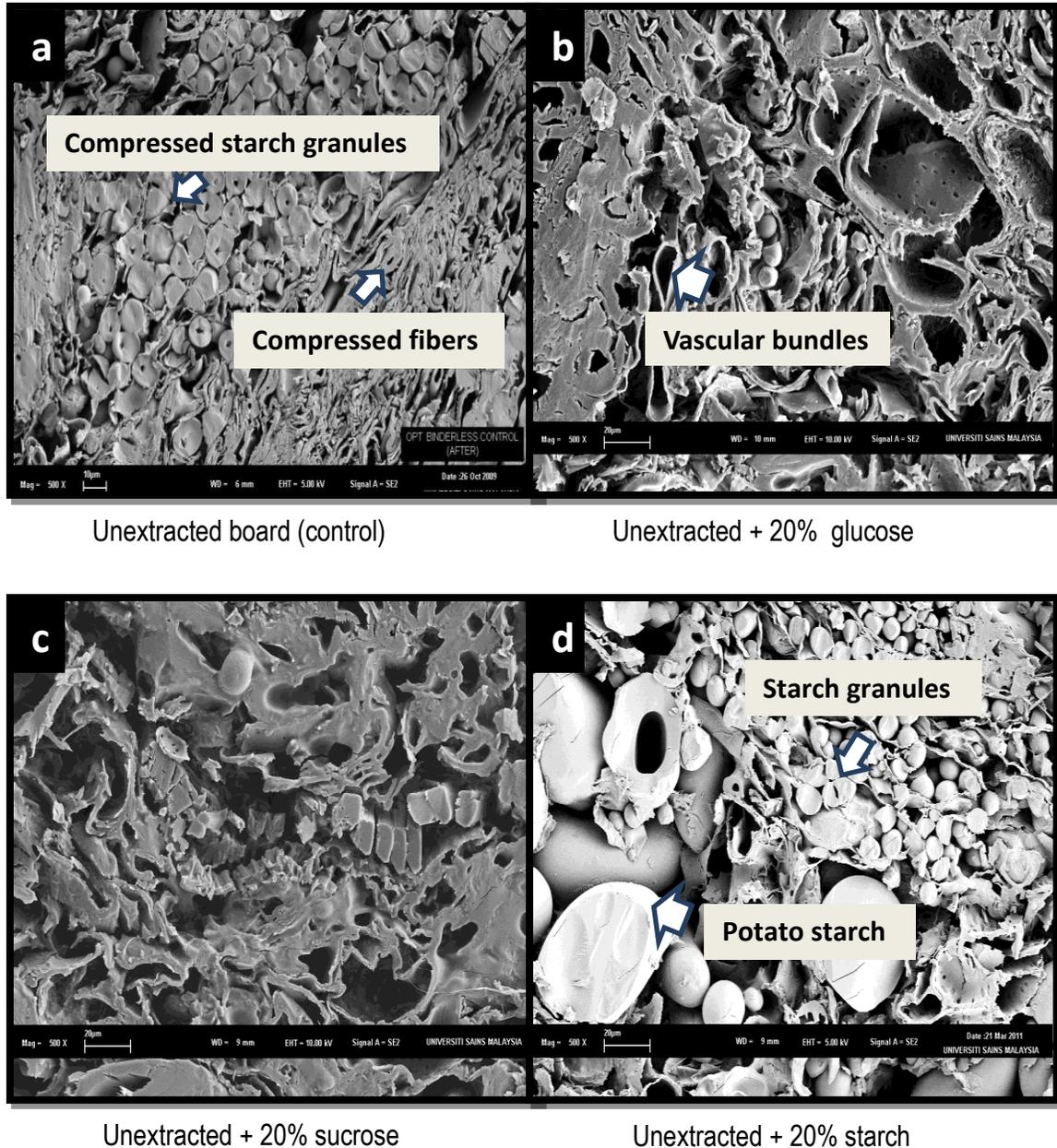


Fig. 5. SEM Micrographs of unextracted board and unextracted board with the addition of sugars and starch

The SEM micrographs revealed the mechanical interlocking between the fibers, as in Figs. 5c and 6c. This may contribute to the higher IB value for the unextracted and extracted board with the addition of glucose and sucrose. During hot pressing, the sugars melt, penetrate into cells, and fill the lumen void area. It also can be stated that sugar can flow, melt, and cover the lignocellulosic fiber and it thus acts as an adhesive to bind the particles together.

Though adding starch to the board improved the properties of the board, the addition of sugars resulted in greater improvement. This is probably due to the size of the potato starch that was bigger than the oil palm starch, and it could not melt together, as can be seen in the Figs. 5d and 6d.

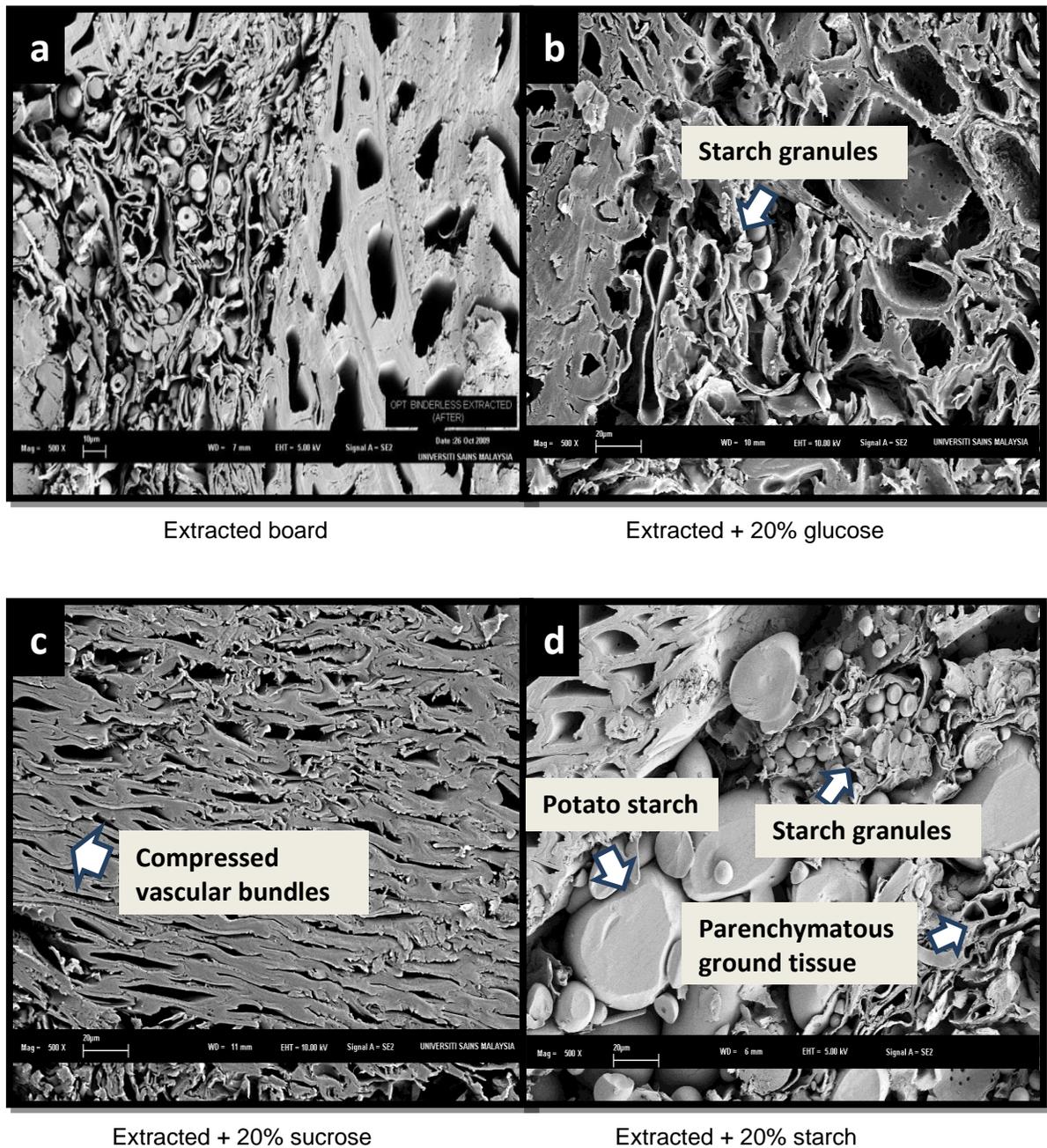


Fig. 6. SEM micrographs of extracted board and extracted board with the addition of sugars and starch

CONCLUSIONS

1. Adding glucose or sucrose during the production of oil palm boards with and without extraction of the particles enhanced the modulus of rupture, internal bond strength, thickness swelling, and water absorption of the resulting panels. The presence of sugar also played a major role in contributing to the self-bonding mechanism of binderless board.

2. Addition of sugar to the board also increased the apparent lignin and cellulose content, indicating that these components react together to form a bond that will bind the particles together.
3. Adding sugar to the control and extracted samples lowered the xylose/arabinose ratio. This indicates that the panels made with the addition of sugar consist of short-chain polymers with a large amount of branching with other monosaccharides compared to the control board.
4. The bonding of binderless board may involve a combination of one or more factors that include the following: degradation of the hygroscopic hemicelluloses to form soluble sugar that may undergo reactions to form less hygroscopic, highly branched polysaccharides; degradation of the hemicelluloses to form free sugar that can undergo polymerization during hot pressing resulting in the formation of an adhesive; thermal softening of the cell wall matrix, mainly lignin, to allow reformation of a new, less stressed matrix after pressing; and crosslinking between carbohydrate polymers and/or between lignin and carbohydrate polymers.

ACKNOWLEDGMENTS

The authors acknowledge Universiti Sains Malaysia for the Fellowship Scheme awarded to Junidah Lamaming and Norafizah Said. The authors would also like to thank the Japan International Research Center for Agricultural Science (JIRCAS) for partially funding this project (304/PTEKIND/650626/J118). Our appreciation also goes to KLK, Kepong, Malaysia of Northern Branch for providing the oil palm samples.

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Article submitted: March 5, 2013; Peer review completed: April 23, 2013; Revised version received: May 3, 2013; Accepted: May 4, 2013; Published: May 9, 2013.