Evaluation of the Properties of Particleboard Made Using Oil Palm Starch Modified with Epichlorohydrin

Nurul Syuhada Sulaiman,[a] Rokiah Hashim,[a,*] Mohd Hazim Mohamad Amini,[a] Othman Sulaiman,[a] and Salim Hiziroglu [b]

The objective of this study was to investigate the physical properties, including density, moisture content, thickness swelling, and water absorption, in addition to the mechanical properties such as modulus of rupture, modulus of elasticity, and internal bond strength of experimental panels that were made from rubberwood particles using oil palm starch modified with epichlorohydrin as a binder. The samples were also examined using X-ray diffractometry, scanning electron microscopy, thermogravimetric analysis, and differential scanning calorimetry. The panel properties were compared with the properties of panels manufactured using native oil palm starch. The properties of starch and starch adhesives were also investigated. The panels were produced based on 0.60 g/cm^3 and 0.80 g/cm^3 target densities and two press times of 15 and 20 min. The results showed that the 0.80 g/cm^3 panels manufactured using modified oil palm starch and with 15 min of press time had better properties than did the others. However, a lower thickness swelling value was found for panels with density 0.60 g/cm^3 and with 15 min of press time. Based on the results in this study, it can be concluded that the use of oil palm starch modified with epichlorohydrin as a binder has the potential to be used as a green adhesive in commercial applications.

Keywords: Physical properties; Mechanical properties; Epichlorohydrin; Oil palm starch

*Contact information: a: Division of Bio-resource, Paper and Coatings Technology, School of Industrial Technology, Universiti Sains Malaysia, 11800 Minden, Penang, Malaysia; b: Department of Natural Resource Ecology and Management, Oklahoma State University, Stillwater, Oklahoma 74078-6013, USA; * Corresponding author: hrokiah@usm.my*

INTRODUCTION

The wood composite industry has become a significant industry in many countries. These wood composite products include plywood, oriented strandboard, hardboard, particleboard, and medium density fiberboard (Ashori and Nourbakhsh 2008). The particleboard industry has grown mostly in the furniture industry (H'ng *et al.* 2011). Therefore, substantial efforts have been put forth to reduce some problems related to particleboard manufacture, including reduction of cost, increase in output, optimization of resin content, and control of formaldehyde emissions (Konnerth *et al.* 2009).

The particleboard industry fully depends on the use of formaldehyde-based resins such as urea formaldehyde (UF), due to its low cost and the excellent properties of its panels as binders (H'ng *et al.* 2011). However, the benefit of using UF in terms of cost savings would not last long given the raise of petroleum prices (Konnerth *et al.* 2009). The production of UF, which uses petroleum as the raw material, will increase the cost of

producing this resin (D' Amico *et al.* 2012; Papadopoulou *et al.* 2008). One alternative that could be used to replace or reduce UF utilization in the manufacture of particleboard is a natural adhesive such as starch (Tondi *et al.* 2011; Moubarik *et al.* 2010a). A previous study showed that the properties of starch-based adhesives could be improved by modification (Huber and BeMiller 2010).

Oil palm starch (OPS) is one of the new natural adhesives that has the potential to be used in the particleboard industry. As one of the largest producers and exporters of oil palm, Malaysia consequently has an increased amount of oil palm trunk waste (Malaysian Palm Oil Board 2011). Most of this waste is left to decay or is burned, which results in air pollution, a significant environmental problem. The relatively high starch content in the oil palm trunk is one of the main components that could be utilized for many other purposes, including the manufacture of a possible green adhesive (Noor *et al.* 1999).

Epichlorohydrin is a colorless liquid that reacts with many types of compounds because of the existence of an epoxide ring and a chlorine atom within the molecule. This feature allows epichlorohydrin to undergo numerous chemical reactions, such that it can be used in a wide variety of applications. The modification of starch with epichlorohydrin results in the formation of highly resistant starch that could be utilized as a coating material in the paper industry, as a component in water-resistant adhesives, or in the food industry (National Toxicology Program 2011). In the food industry, starch is used for stabilizing, thickening, gelling, fat replacement, and many other functions. The modification of native starch may be required, as the properties needed in the food products are not necessarily fulfilled by the use of native starch. Undesirable properties of native starch could be overcome by crosslinking reaction with crosslinking agents such as epichlorohydrin (Ačkar *et al.* 2010). However, the concentration of epichlorohydrin as a crosslinking agent in the food industry must be below 0.3% (National Toxicology Program 2011). The reaction of starch with epichlorohydrin leads to the formation of distarch glycerols, which are comprised of ether linkages between the cross-links and the hydroxyl groups in the starch (Jyothi *et al.* 2006; Gough 1967).

Much research has been done with the objective of reducing or eliminating the use of UF resin and increasing the utilization of environmentally friendly raw materials in the wood-based composite industry. Konnerth *et al.* (2009) did a study on the possibility of producing particleboard using animal protein glue produced from bone. The results showed that it is possible to use bone glue to replace UF resin as a binder. Another study by Bono *et al.* (2010) showed that addition of soy protein extract in palm kernel cake could increase the strength of plywood. Moubarik *et al.* (2010b) studied the physical and mechanical properties of interior plywood using corn starch-tannin as an adhesive. Their results indicated that the properties of plywood prepared from formaldehyde-free corn starch-tannin adhesives were comparable to phenol-formaldehyde plywood, which has already being commercialized.

The application of OPS modified with epichlorohydrin in particleboard production, has not been investigated yet. Hence, the present study was conducted to investigate the potential of using OPS modified with epichlorohydrin in experimental panel production compared with those made with native OPS. Both mechanical and physical properties as well as characterization of the resulting panels were evaluated in order to determine the feasibility of their commercialization in the particleboard industry.

EXPERIMENTAL

Extraction of Starch

The extraction of the starch from the oil palm trunk was carried out in accordance with the preceding work by Noor *et al.* (1999), with modification in the soaking time. The oil palm trunk slabs were cut into smaller size particles and soaked in 1000 mL aliquots of a 0.5% (w/v) aqueous solution of sodium metabisulphite for 48 hours. The particles were then removed from the solution and the residues were filtered using a sieve with a mesh aperture of 250 µm. The filtered solution was then centrifuged at 4750 rpms for 15 min using a Beckman Coulter Allegra X-15R centrifuge. The precipitated starch was carefully removed from the centrifuge bottle and dried in an oven at 50 ± 2 °C for 3 days. The extracted starch was ground using a blender and stored for further use.

Starch Properties

Moisture content

Approximately 1g of starch sample was taken and dried in an oven at a temperature of 103 °C ± 2 until the sample achieved a constant weight. The moisture content of the adhesive was calculated and expressed in a percentage with an accuracy of 0.01%.

Particle size

The particle size analysis was evaluated using a particle size analyzer, the Mastersizer 2000 (Malvern Instruments), with a Scirocco dry powder feeder. The particle size was measured and expressed in micrometer (µm) units.

Starch content

Determination of starch content was completed according to the previous work done by Nielsen (1943). The graph of absorbance versus amount of starch (gram/50 mL) at a wavelength of 650 nm was plotted. A linear regression equation was used to determine the starch content.

Adhesive Preparation

Modified oil palm starch adhesive (starch modification)

The extracted OPS was modified using epichlorohydrin according to the methodology practiced by Jyothi *et al.* (2006). The oil palm starch was dissolved in distilled water and stirred using an overhead stirrer in a water bath that was set to increase the temperature to 90 °C. Epichlorohydrin was added to the starch solution at a ratio of 1:4 (w/w) once the temperature of the solution had passed 55 °C. Ether linkages form between the cross links and the hydroxyl groups as the epichlorohydrin is introduced to the starch granules (Jyothi *et al.* 2006; Gough 1967). The mixture was continuously stirred until the temperature reached 90 °C. The mixture was then left to cool to room temperature before it was used in the manufacture of the particleboard.

Adhesive Properties

Solids content

About 1 to 2 g of adhesive was taken and dried in an oven at 105 °C for 3 h. The adhesive was then reweighed. The solid content of adhesive was calculated and expressed in percentage (Chew *et al.* 1988).

Viscosity

The adhesive viscosity was determined by using a rotary rheometer (AR1000-N) (Jyothi *et al.* 2006). The rheological behaviour measurement was taken at a shear rate of 150 s^{-1} at a temperature of 30°C.

Pot life

The adhesive sample was prepared and left inside a beaker until the adhesive became too thick to be spread. The time taken for the adhesive to become non-usable was recorded as the pot life (Cognard 2005).

Swelling power and solubility

The swelling power and solubility of both native and modified starches were carried out according to previous work done by Ačkar *et al.* (2010). A total of five samples of starch dispersion (2% w/v) were heated in a shaking water bath at five different temperatures (50, 60, 70, 80, and 90 °C) for 30 min. The samples were then cooled before they were centrifuged at 4000 rpm for 30 min. The separation of gel and supernatant resulted from the centrifugation process and the weight of the gel was recorded. Both the supernatant and gel was then heated in an oven at 105 °C until they achieved their constant weight. The swelling power and solubility of starch were calculated and expressed in percentage.

Particleboard Manufacture and Evaluations

A total of 40 panels were produced based on two target densities (0.60 g/cm^3 and 0.80 g/cm^3) and two press times (15 min and 20 min) with 5 replicates for each set. Rubberwood (*Hevea brasiliensis*) particles were obtained from HeveaBoard Sdn Bhd located in Seremban, Negeri Sembilan, Malaysia. Before the panels were manufactured, the particles were dried in an oven until they had obtained a moisture content of about 2%. About 15% the oven-dried weight of the wood in native or modified OPS adhesive was mixed manually with the rubberwood particles. The mixture was then put into a mold with dimensions of 20.1 cm x 20.1 cm x 0.5 cm, followed by pre-pressing with a pressing machine to form a mat. The mat was then hot-pressed using a hot press machine with pressure of 5 MPa and at a temperature of 165 °C for 15 or 20 min. The panel was then cooled and conditioned in a climate room at a temperature of 25 ± 2 ºC and a relative humidity of 65 ± 2% for several days before the tests were carried out.

The physical properties of the samples, including density, moisture content (MC), thickness swelling (TS), and water absorption (WA), and the mechanical properties, including modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond (IB) strength, were carried out in accordance with the Japanese Industrial Standard Committee (2003). Thirty samples, size 5 cm x 5 cm, from each type of panel were used for density, TS, and WA tests. The density test was done by taking measurements of the length, width, thickness, and mass. For TS and WA, readings for the thickness and weight of each sample were taken before they were soaked in distilled water. The increases in thickness and weight of each samples were measured after soaking for 2 or 24 h. The moisture content (MC) value was taken from the average reading of 5 samples from each type of panel. The initial masses of the panel samples were taken, and the panels were then oven dried until they achieved a constant weight. The constant weights of the samples were recorded, and the MC value was calculated based on the formula stated in the JIS standard. For the evaluation of the bending properties, twenty samples of

size 5 cm x 20 cm were cut from each set for the MOR and MOE tests, while 30 samples were cut from each set for the internal bond strength tests. The Instron Tensile Machine Model 5582 was employed for both tests, with a crosshead speeds of 10 mm/min and 2 mm/min for the bending and internal bond strength test, respectively.

Particleboard Characterizations

X-ray diffractometry (XRD) analysis

The particleboard sample was ground using an MF 10 basic IKA® WERKE grinder with a mesh size of 0.50 mm. The powdered sample of the particleboard was then analysed with a Kristalloflex D-5000 X-ray diffraction system (Siemens, Germany). The powdered sample was used to fill a sample holder, and an air blow was used to smooth the finished surface of the sample. Step scan measurements were done using X-rays (Cu-Kα) generated at the opening voltage and current of 40 kV and 30 mA, respectively. The scan was made from a diffraction angle of 2θ ranging from 10° to 40°, corresponding to a scanning speed of 0.02° and 2°/min. The crystallinity index (CrI) was calculated using the formula (Eq. 1),

$$\text{CrI}\ (\%) = (I_{200}\text{-}I_{\text{am}})/\ I_{200} \text{ x } 100 \quad (1)$$

where I_{200} is the peak intensity of the crystalline fraction and I_{am} is the peak intensity of the amorphous fraction.

Thermogravimetric analysis (TGA)

The powdered sample of the particleboard was prepared based on the method described previously. A thermal analysis of the sample was carried out using a Mettler Toledo TGA/SDTA851e thermogravimeter (Mettler Toledo Corp., Switzerland) with STARe software (version 9.20). Approximately 10 mg of the powder sample was placed in an aluminium pan before being heated at a rate of 20 °C/min over the temperature range between 30 °C and 800 °C in a nitrogen atmosphere.

Differential scanning calorimetry (DSC) analysis

The melting temperature (T_{m}) of each particleboard was determined using a Perkin Elmer Thermal analysis (Model DSC 8000). Approximately 5 mg of the powdered sample which was prepared based on the method described previously was weighed out into an aluminium pan. The sample was then transferred to the heating pan, with the usage of empty pan as a reference. The sample was heated up at a rate of 10 °C/min within temperature range between -15 °C and 280 °C in a nitrogen atmosphere.

Scanning electron microscopy (SEM) analysis

Small cubes were cut from a cross-cut view of particleboard panels and glued onto a stub using tape. Then, the samples were coated using a Polaron SC515 SEM coating system (Fisons Instrument) with a thin layer of gold to make them conductive. The samples were examined using an acceleration of 15 kV with a Scanning Electron Microscope (Model Supra 50 VP).

RESULTS AND DISCUSSION

Extraction of Starch

The yields of extraction are listed in Table 1. The average yield of starch extraction was 18.99%. This value was higher as compared to what has been reported by Noor *et al.* (1999), which showed a maximum oil palm starch extraction of 7.15%. However, the yield of starch extraction from oil palm trunk acquired in this study is still considered to be low as compared to the yield of starch extraction from the other raw materials such as sago palm trunk (Akmar and Kennedy 2001). A study done by Tomimura (1992) showed that the parenchyma of oil palm trunk contains 55.5% starch content, which is relatively high compared to the vascular bundle of oil palm trunk which contains only 2.4% starch. The parenchyma cells in oil palm trunk comprise a high amount of lignin which makes the starch in this cell difficult to extract (Sulaiman *et al.* 2012; Noor *et al.* 1999).

Table 1. Percentage Yield of Starch Extracted from Oil Palm Trunk (OPT)

Sample	Yield of Starch (%)
Sample 1	16.60
Sample 2	17.19
Sample 3	24.43
Sample 4	18.81
Sample 5	17.93
Average	18.99

Starch Properties

The properties of extracted oil palm starch are shown in Table 2. Moisture content of the oil palm starch was recorded as 10.98%, which is close to the moisture content recorded by Noor *et al.* (1999) at 11.8%. For comparison, other commercial starches also have comparable values of moisture content which were 13% for wheat, corn, and tapioca starch while potato starch showed 19% (Swinkels 1985).

The particle size distribution graph is shown in Fig. 1. Particle size analysis of the extracted oil palm starch resulted in average particle size of 11.38 μm. This is 3.24 μm smaller as compared to the previous work carried out by Noor *et al.* (1999) in term of its average. Oil palm starch particle size is smaller than potato starch, which has an average diameter of 27 μm, and larger than corn and wheat starch at 10 μm and 8 μm, respectively (Swinkels 1985).

The starch content, which provides a measure of the purity of the starch, yielded 80.43%. The purity of starch really depends on the size of the debris such as cellulose cell wall, as debris with the same or smaller size than the starch granule cannot be removed even with multiple filtering.

Table 2. Extracted Oil Palm Starch Properties

Properties	Oil Palm Starch
Moisture Content	10.98%
Particle Size	11.38 μm
Starch Content	80.43%

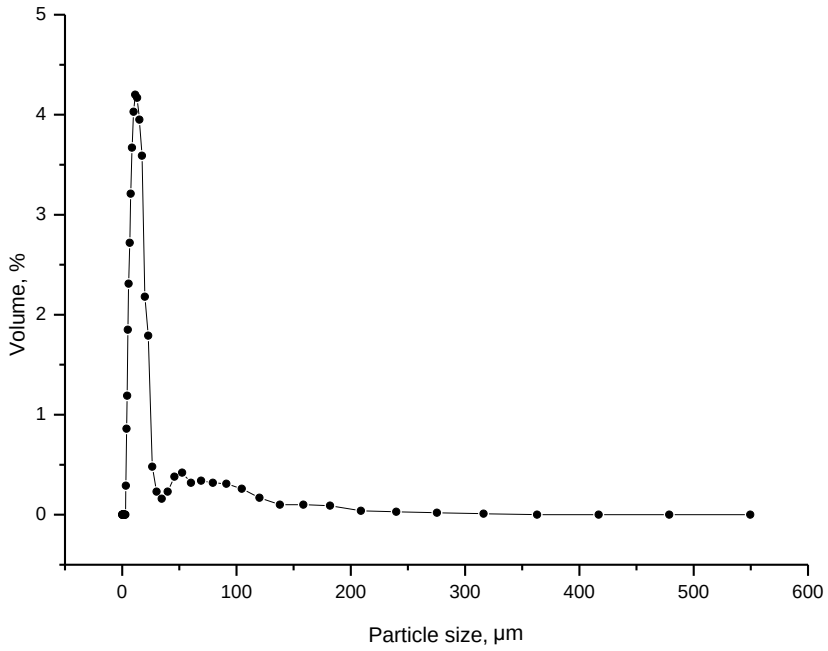


Fig. 1. Particle size distribution of extracted oil palm trunk starch

Adhesive Analysis

Table 3 presents the properties of both native and modified oil palm starch adhesives. The solid content of native starch adhesive was found to be lower than that of modified starch adhesives. The loss of moisture is believed to occur during the heating process to crosslink the starch. Thus, the solid content of modified oil palm starch adhesive was higher than that of the native oil palm starch adhesive.

However, the same trend was found on the rheological properties of those adhesives by having lower viscosity reading on the native starch adhesive compared to the modified starch adhesive. The pure starch pastes can be described as a suspension of swollen particles. The starch granules break and swell after being heated, releasing the entanglement of amylopectin. The presence of the polar groups causes further swelling of the particles, resulting in greater expansion of the coiled starch macromolecules.

Therefore lower viscosity was observed for the native oil palm starch adhesive (Zhu and Chen 2007).

The pot life of native starch adhesive showed a minor difference with the modified starch. Native starch exhibited 5 days pot life, compared to 3 days pot life of the modified starch adhesive. The epichlorohydrin as the crosslinking agent accelerated the process of adhesive hardening of modified starch compared to native oil palm starch, which contains only water as the carrier.

Table 3. Adhesive Properties of both Native and Modified Oil Palm Starch

Analysis	OPS Adhesive	Modified OPS Adhesive
Solid Content (105 °C)	27.43%	29.97%
Viscosity at 30 °C	2.059 Pa.s	2.73 Pa.s
Pot Life	5 days	3 days

[a] *OPS: oil palm starch*

Swelling power and solubility

The swelling power and solubility of native and modified oil palm starches at different temperatures are summarized as in Table 4.

Table 4. Swelling Power and Solubility of both Native and Modified Oil Palm Starch

Temperature, °C	50		60		70		80		90	
	SP (g/g)	SOL (%)	SP (g/g)	SOL (%)	SP (g/g)	SOL (%)	SP (g/g)	SOL (%)	SP (g/g)	SOL (%)
Native OPS	7.20	4.38	11.74	7.99	13.50	16.65	27.63	27.13	33.35	30.69
Modified OPS	4.81	5.80	6.43	6.29	10.31	12.16	17.70	14.11	28.67	20.63

[a] SP: swelling power, SOL: solubility

Table 4 shows that there was an increasing of swelling power and solubility with an increase in temperature for both investigated starches. The modified starch exhibited low swelling power in comparison to the native starch. The percentages of solubility of native and modified starches also presented the same trend as the results obtained in swelling power. The significant decrease of both swelling power and solubility between native and modified starch was observed at a temperature of 80 °C. The crosslinking reaction caused the starch's granular structure to become more compact (Kartha and Srivastava, 1985). Thus, a reduction in swelling power and solubility of modified starch could be seen. This result was consistent with the previous work done by Ačkar *et al.* (2010), who carried out a study on wheat starch crosslinked with epichlorohydrin.

Physical Properties of Particleboard

The physical properties of the panels bonded with native and modified starch at different density levels and with different press times are given in Table 5 (native) and Table 6 (modified).

Table 5. Physical and Mechanical Testing of Particleboard Panels Bonded with Native Oil Palm Starch

Target density (g/cm^3)	Press time (min)	Physical testing						Mechanical testing		
		Density (g/cm^3)	Moisture content (%)	Thickness swelling (%)		Water absorption (%)		MOR (N/mm^2)	MOE (N/mm^2)	IB (N/mm^2)
				2h	24h	2h	24h			
0.6	15	0.57 (0.02)	5.64 (0.19)	58.07 (10.24)	74.45 (11.30)	119.43 (5.25)	140.37 (17.58)	7.80 (1.28)	1695.22 (203.57)	0.19 (0.07)
	20	0.57 (0.03)	5.70 (0.15)	58.12 (12.36)	75.74 (11.59)	133.36 (3.30)	169.96 (6.97)	7.55 (2.67)	1839.42 (335.95)	0.13 (0.06)
0.8	15	0.74 (0.03)	5.75 (0.30)	79.54 (11.91)	99.28 (7.48)	117.4 (5.60)	163.31 (7.72)	12.87 (2.35)	3411.06 (139.63)	0.24 (0.13)
	20	0.73 (0.02)	6.34 (0.62)	76.28 (7.34)	92.56 (10.73)	111.67 (4.57)	163.05 (7.53)	11.86 (2.71)	3137.92 (332.73)	0.21 (0.09)

[a] data is expressed as average
[b] values in parenthesis indicate standard deviation
[c] MOR: modulus of rupture, MOE: modulus of elasticity, IB: internal bond strength

Table 6. Physical and Mechanical Testing of Particleboard Panels Bonded with Oil Palm Starch Modified with Epichlorohydrin

Target density (g/cm^3)	Press time (min)	Physical testing						Mechanical testing		
		Density (g/cm^3)	Moisture content (%)	Thickness swelling (%)		Water absorption (%)		MOR (N/mm^2)	MOE (N/mm^2)	IB (N/mm^2)
				2h	24h	2h	24h			
0.6	15	0.62 (0.02)	5.33 (0.15)	43.15 (5.87)	54.81 (7.94)	114.02 (3.99)	123.84 (11.92)	10.59 (1.51)	1975.96 (370.49)	0.35 (0.06)
	20	0.63 (0.03)	5.35 (0.10)	46.08 (6.51)	64.45 (5.81)	122.63 (6.12)	154.66 (17.06)	9.61 (1.17)	2124.18 (482.52)	0.32 (0.10)
0.8	15	0.78 (0.04)	5.49 (0.12)	54.35 (7.66)	71.50 (10.51)	106.57 (6.78)	126.46 (5.98)	19.09 (1.62)	3471.64 (460.52)	0.49 (0.15)
	20	0.78 (0.02)	5.47 (0.12)	50.07 (14.91)	69.62 (8.46)	104.23 (3.47)	120.32 (7.31)	15.92 (1.90)	3073.16 (474.54)	0.45 (0.06)

[a] data is expressed as average
[b] values in parenthesis indicate standard deviation
[c] MOR: modulus of rupture, MOE: modulus of elasticity, IB: internal bond strength

The average actual densities for the panels bonded with native starch and with target densities of 0.60 g/cm^3 were the same, *i.e.*, 0.57 g/cm^3, for the press times of both 15 and 20 minutes. Meanwhile, for those manufactured with a 0.80 g/cm^3 target density, the average actual densities were 0.74 g/cm^3 and 0.73 g/cm^3 for the 15 and 20 min press times, respectively. For the 0.60 g/cm^3 panels bonded with modified starch, the average actual densities for the 15 and 20 min press time were 0.62 g/cm^3 and 0.63 g/cm^3, respectively, whereas the particleboards with target densities of 0.80 g/cm^3 had the same density level, *i.e.*, 0.78 g/cm^3, for both the 15 and 20 min press times.

Observation of the MC readings resulted in only a slight difference in MC readings between each type of panel at the same density level. Native starch panels with the target density of 0.60 g/cm^3 had MC readings of 5.64% and 5.70% for the 15 and 20 min press times, respectively. For those manufactured with a target density of 0.80 g/cm^3, the MC readings were 5.75% and 6.34% for the 15 and 20 min press times, respectively. The MC readings for modified starch panels manufactured with the target density of 0.60 g/cm^3 were 5.33% and 5.35% for 15 and 20 min press times, respectively. In contrast, panels with a target density of 0.80 g/cm^3 had MC readings of 5.49% and 5.47% for 15 and 20 min press times, respectively. All the MC values obtained were in the range as stated by the Japanese Industrial Standard Committee (2003), *i.e.*, 5 to 13%.

According to the results obtained in this study, panels manufactured using modified starch had lower TS and WA values than did the panels manufactured using native starch. The modification of starch with epichlorohydrin leads to the formation of distarch glycerols which comprise of ether linkages between the crosslinks and the hydroxyl groups in the starch (Jyothi *et al.* 2006). This resulted in the formation of highly water-resistant starch. In addition, the compact structure of modified starch which caused decreasing in swelling power and solubility influenced the decreasing in TS and WA of the panel as well. Thus, this modification improved the dimensional stability of the manufactured panels.

The TS results showed that the panel manufactured with the highest density level had a higher TS value than those made with low density levels. Moreover, panels pressed for 20 min had higher TS values than those pressed for 15 min. However, this trend was only found on the 0.60 g/cm^3 particleboards, whereas with the 0.80 g/cm^3 particleboards, the reverse outcome was observed. These trends were observed for both types of panels, those with native and modified starch. A non-linear relationship was found between the TS and WA properties of the samples. The panels with high density levels had lower WA values than those panels with low density levels. A more simplified description would be that high-density panels had high TS and low WA, while low-density panel had low TS and high WA.

The presence of a large amount of particles in the high-density panels caused a high quantity of water to be absorbed by the particles, and this was reflected in the high TS values of the samples. However, in the high-density panel the particles were compressed and had a greater compaction ratio, which facilated the development of better contacts between the particles and the starch. Thus, the WA decreased because there were only a small number of void spaces available for water storage in the panel despite the fact that the particles in the panel absorb a high amount of water. The high TS value in the high density particleboard was a normal occurrence, as reported by Loh *et al.* (2010).

According to the Japanese Industrial Standard Committee (2003), the maximum acceptable level of TS is 12%. All the samples did not meet the requirements stated in this standard. However, this problem could be solved by using several alternative

methods, such as treating particles with water repellent before panel manufacture or surface-coating the end-product (Yalinkilic *et al.* 1998). Nourbakhsh (2010) also mentioned other methods that could be applied to reduce the percentage of TS in particleboard, such as the use of heat treatments or the application of melamine-impregnated papers or laminates to coat the surfaces of the particleboards. Other alternatives suggested by Nourbakhsh (2010) include reducing the density of the particleboard (if the high density level is not desirable) or adding 0.50 to 1.00% wax to the mixture of resin and particles during the production process.

Mechanical Properties of Particleboard

The mechanical properties (MOR, MOE, and IB) of the panels were tested, and the mean value of each mechanical property is illustrated in Table 5 and Table 6, for panels bonded with native and modified starch, respectively. From these tables, it can be seen that the panels bonded with modified starch demonstrated the best mechanical properties. This phenomenon occurred because the inferior properties of native starch were altered through the crosslinking reaction that occurred when epichlorohydrin was introduced to the starch granules. The crosslinking reaction resulted in the formation of starch that is highly resistant to mechanical shear (Jyothi *et al.* 2006). The crosslinking reaction that occurred with the starch granules also contributed to the better bonding between the starch granules and the rubberwood particles.

The results of the mechanical testing also showed a great influence of density level on panel strength. Panels with a low density level (0.60 g/cm^3) had inferior mechanical properties than did those with a high density level (0.80 g/cm^3). The highest mean values for the MOR and MOE were 19.09 N/mm^2 and 3471.64 N/mm^2, respectively, and this was found for the 0.80 g/cm^3 density panel bonded with modified starch. Both of these panels were produced using 15 min of press time. The lowest values, *i.e.*, the MOR and MOE values of 7.55 N/mm^2 and 1695.22 N/mm^2, respectively, were found for the 0.60 g/cm^3 density panel bonded with native starch. On the other hand, the panel produced with a 0.80 g/cm^3 target density and with 15 min press time also had high mean values of IB strength.

A high amount of particles and starch content in the high density panels led to better mechanical strength due to the increased interaction between the particles and the starch when the panel composite was compressed. The structure of the manufactured panel was more compact and tighter, and the curing of the binder occurred more efficiently (Yimsamerjit *et al.* 2007), which resulted in the high mechanical strength of panel. The high amount of particles also indicated that a large amount of fibrous material was present and was resisting the mechanical loading (Yalinkilic *et al.* 1998).

All the mechanical property values obtained for each type of panel met the minimum requirement as stated in the Japanese Industrial Standard Committee (2003). The minimum requirement for the MOR in JIS standard is 8 N/mm^2, where the mean values of the MOR obtained in this study were in the range of 9.61 N/mm^2 to 19.09 N/mm^2. For IB strength, the minimum requirement is 0.15 N/mm^2, where the mean values for the IB strength of the samples found in this study were in the range of 0.32 N/mm^2 to 0.49 N/mm^2.

Starch and Particleboard Characterizations

X-ray diffractometry (XRD) analysis

The X-ray diffractometry (XRD) analysis was run to determine the crystallinity index of starch and manufactured panel. The X-ray patterns of the starch and panels bonded with native and modified oil palm starch are shown in Figs. 2(a) and 2(b), respectively.

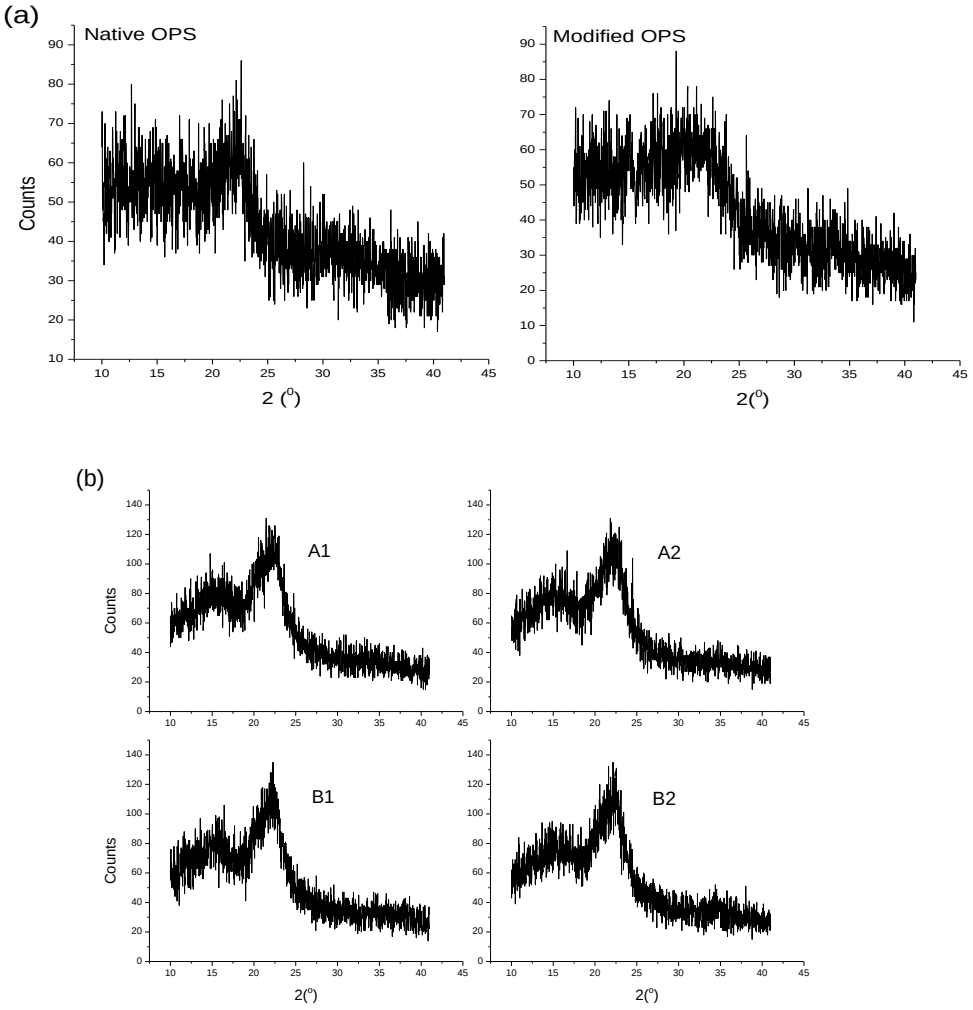


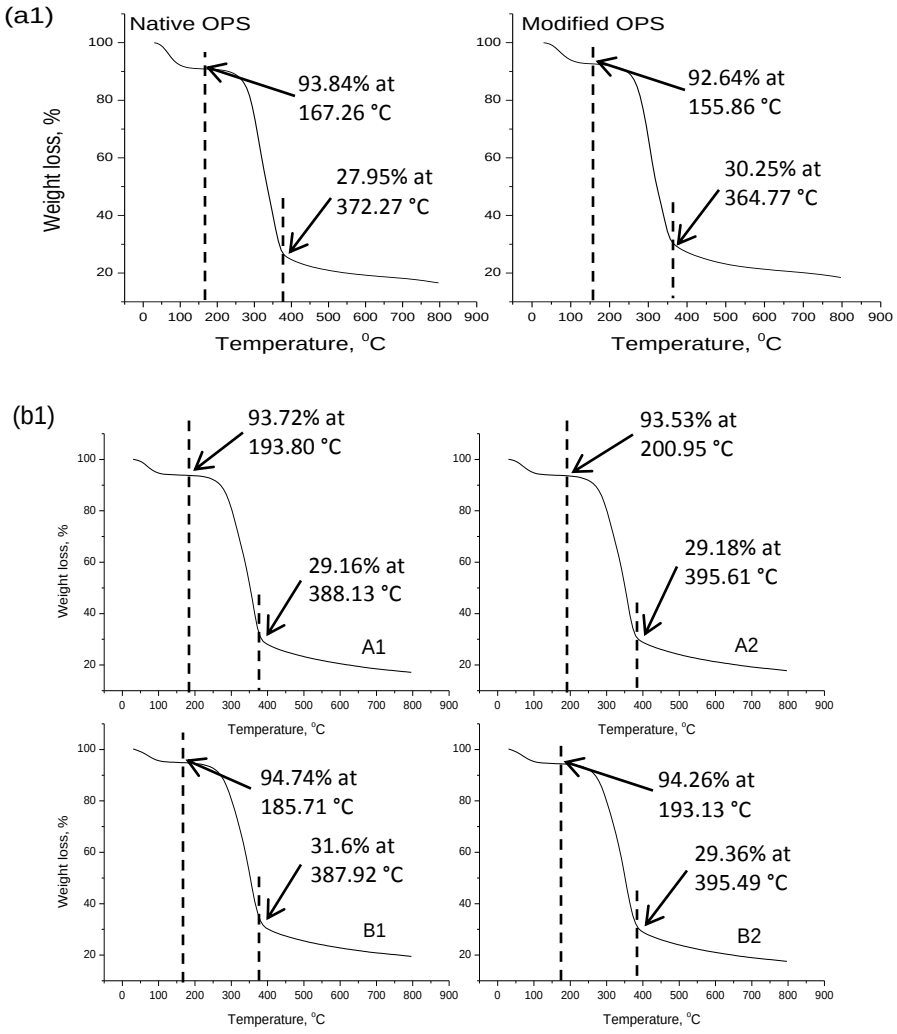
Fig. 2. (a) X-ray diffractometry patterns of native and modified oil palm starch (OPS) adhesive; (b) X-ray diffractometry patterns of 0.80 g/cm^3 density panels bonded with native oil palm starch with 15 min (A1) and 20 min (A2) press times; panels bonded with oil palm starch modified with epichlorohydrin with 15 min (B1) and 20 min (B2) press times

The X-ray patterns revealed that a major intensity line was close to 22 degrees of the 2θ angle. The crystallinity index was computed based on Eq. 1. Based on the calculation, the crystallinity index for native OPS and modified OPS were 31.82% and 29.07%, respectively. These results showed that the crystallinity index of starch was reduced after the modification process had been done. The crystallinity index for 0.80 g/cm^3 density panel bonded with native starch was 48.09% and 48.85% for 15 and 20

min press times, respectively. Meanwhile, a panel manufactured using modified starch had a crystallinity index of 47.33% and 47.41% for 15 and 20 min press times, respectively. The panels bonded with modified starch had the lowest crystallinity index compared to those bonded with native ops. However, whole panels manufactured in this study showed a slight difference in crystallinity percentage. In addition, it was observed that the panel that had the lowest crystallinity index had better physical and mechanical strength. Low crystallinity index indicates that more amorphous material existed on the manufactured panel. The amorphous material creates more bonding than the crystalline material. Thus, the manufactured panel with low crystallinity index will result in higher physical and mechanical strength.

Thermogravimetric analysis (TGA)

Thermogravimetric analysis (TGA) was done to measure the weight loss of the starch and particleboard samples in powder form that were subjected to a temperature program. The TGA analysis results were illustrated by a weight loss curve (TG) and derivative thermogravimetric (DTG) diagram. The TG and DTG curves for starch sample were shown in Figs. 3(a1) and 3(a2), respectively. The TG and DTG curves for particleboard sample were shown in Figs. 3(b1) and 3(b2), respectively.



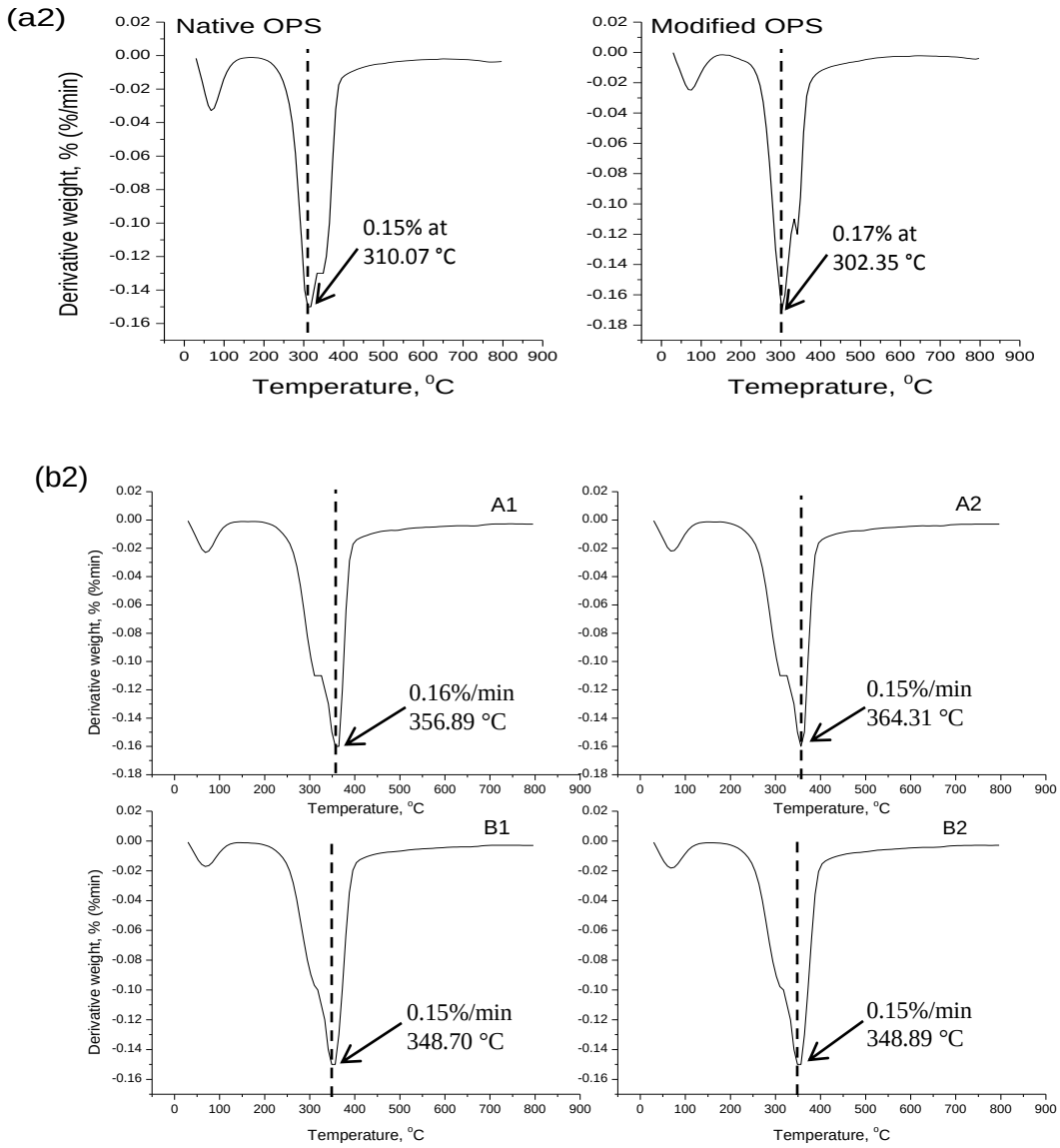


Fig. 3. (a1) TG and (a2) DTG curves of native and modified oil palm starch (OPS) adhesive; (b1) TG and (b2) DTG curves of 0.80 g/cm^3 density panels bonded with native oil palm starch with 15 min (A1) and 20 min (A2) press times; panels bonded with oil palm starch modified with epichlorohydrin with 15 min (B1) and 20 min (B2) press times.

All samples experienced weight loss in three steps, as shown in Figs. 3(a1) and 3(b1). A minor decrease in weight, with initial temperature around 29 °C, indicates the first stage, which involves the evaporation of water and easily volatile materials (El-Wakil *et al.* 2007). The second and third stages are comprised of the dehydration of polymer chains and a complete decomposition of samples residues, respectively (Wang *et al.* 2011). The observation on TG curve of starch sample revealed that native OPS had the highest starting temperature for the second and third stages. The residue left for native OPS is 18.44% which was higher compared to the modified OPS that have 16.59% residue left. According to the DTG curve, native OPS showed the fastest rate of

decomposition at a higher temperature. These results indicated that native OPS has the higher thermal stability as compared to the modified OPS. This can be related to their crystallinity index in which sample that has high crystallinity index possess high thermal stability.

The highest starting temperatures for weight loss at the second and third stages were found on the panels manufactured with native starch with a 20 min press time, and the lowest was found on panel manufactured using modified starch and with a 15 min press time. The starting temperature and percentage of weight loss for each sample is shown in Fig. 3(b1). At the end of the analysis, the residue left for panels manufactured using native starch were 17.81 and 19.50% for 15 and 20 min press times, respectively. On the other hand, the residue left for panels manufactured using modified starch were 17.12 and 17.57% for 15 and 20 min press times, respectively. The DTG diagrams also revealed that the fastest rate of decomposition at a higher temperature was found on the panel manufactured using native starch and with a 20 min press time. In conclusion, the panel manufactured using native starch and with a 20 min press time had a high initial degradation temperature and a lower weight loss compared to the other panels. This revealed that panel manufactured using native starch and with a 20 min press time is more resistant to heat.

Differential scanning calorimetry (DSC) analysis

The DSC thermograms of starch and 0.80 g/cm^3 density particleboard panels are shown in Figs. 4(a) and 4(b), respectively.

According to Fig. 4(a), the higher melting temperature was found on modified OPS with a reading of 88.39 °C. The lowest one was observed on native OPS with a melting temperature of 77.88 °C. The DSC thermograms of starch samples revealed that the melting temperature of starch was increasing as the starch had been modified. This could be contributed by the crosslinking reaction that occurs between the starch granules. The Fig. 4(b) showed that the melting temperature (Tm) of the panel manufactured using modified starch and 15 min press time was higher compared to that of the panel manufactured using native starch and 20 min press time. The melting temperature for the panels manufactured with native starch were 76.62 °C and 76.08 °C for 15 and 20 min press time, respectively. Meanwhile, panels manufactured with modified starch had melting temperatures of 78.14 °C and 77.30 °C for 15 and 20 min press time, respectively. The panel manufactured with modified starch had a higher melting temperature due to the crosslinking reaction with epichlorohydrin that occurred in starch. This leads to better interaction between starch and particles in the panel manufactured. Thus, high temperature was needed to complete the melting process. The higher melting enthalpy was found on the panel manufactured with modified starch, having values of 230.46 and 174.30 J/g for 15 and 20 min press time, respectively. The panel manufactured using native starch had a melting enthalpy of 127.43 and 88.93 J/g for 15 and 20 min press time, respectively. This could be due to the reinforcing of starch granules through crosslinking reaction with epichlorohydrin. It will contribute to the higher strength of panel manufactured and thus cause more heat will be required for melting process to be completed.

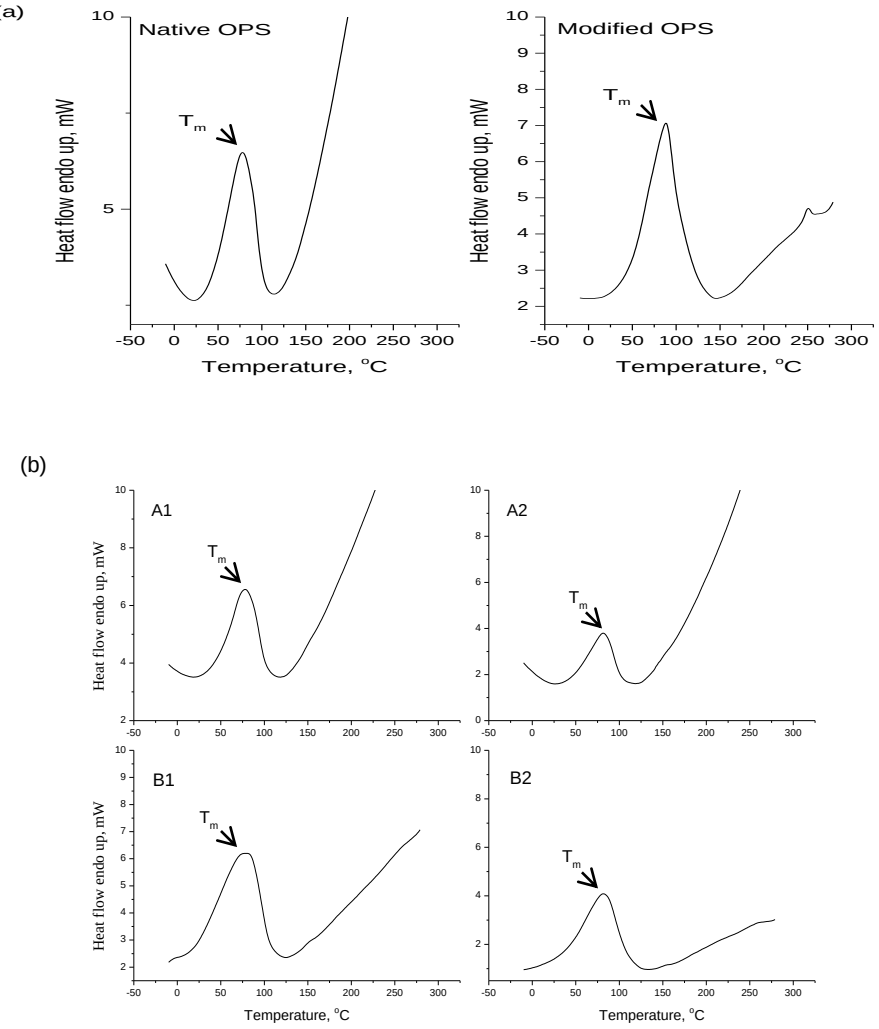


Fig. 4. (a) DSC thermograms of native and modified oil palm starch (OPS) adhesive; (b) DSC thermograms of 0.80 g/cm^3 density panels bonded with native oil palm starch with 15 min (A1) and 20 min (A2) press times; panels bonded with oil palm starch modified with epichlorohydrin with 15 min (B1) and 20 min (B2) press times.

Scanning electron microscopy (SEM) analysis

The compactness and effect of modified oil palm starch as a binder in manufactured particleboard panel was observed by the SEM analysis. The SEM micrographs of the particleboard sample are shown in Figs. 5(a) and 5(b).

From these figures, it can be observed that the compressed cell walls and fibers of the rubberwood particles were compact in structure due to the pressure used during the manufacture of the particleboard panels. By becoming embedded in the rubberwood particles, the starch molecules were dispersed evenly within the particles. These two factors caused more bonding between the rubberwood particles and the modified starch molecules. This was seen more clearly on the panel manufactured with modified starch than on those panels manufactured with native starch. Therefore, the physical and mechanical strengths of the panels will also be enhanced especially for panel manufactured using modified starch.

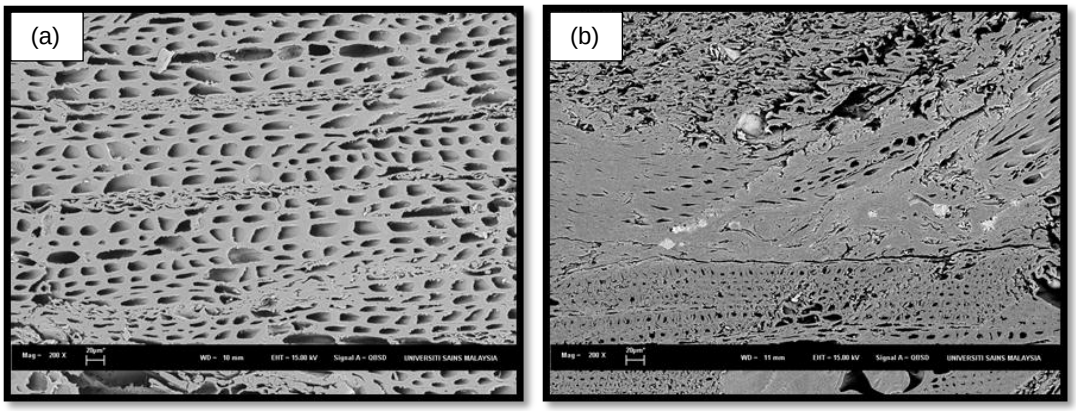


Fig. 5. SEM micrograph showing cross section view of 0.80 g/cm^3 density panel manufactured with 20 min press time, bonded with (a) native oil palm starch and (b) oil palm starch modified with epichlorohydrin, with magnification of 200x

CONCLUSIONS

1. The particleboard panels produced from oil palm starch modified with epichlorohydrin exhibited mechanical strength (modulus of rupture, modulus of elasticity, and internal bond) that satisfied the Japanese Industrial Standards (JIS) as compared to those produced using native oil palm starch.
2. The thickness swelling and water absorption did not meet the minimum requirements as stated in the same standard. This problem could be solved by application of wax on the surface of the panels. The panel with a density of 0.80 g/cm^3 shows better modulus of rupture, modulus of elasticity, internal bond, and water absorption values whereas 0.60 g/cm^3 panels only shows better thickness swelling value. Most of the panels produced with a 15 min press time had better mechanical and physical properties compared to those produced using 20 min press time.
3. The analysis of particleboard through X-ray diffractometry, thermogravimetric analysis, and differential scanning calorimetry showed little difference between panels produced with 15 and 20 min press time, respectively.
4. The scanning electron micrograph showed the modified starch was well dispersed between the compressed rubberwood particles, which contributes to better bonding between them compared to native starch.
5. Thus, modification of starch resulted in better properties of the starch granules, which then contributed to the strength of panels manufactured as proved by this study.

ACKNOWLEDGMENTS

The authors want to acknowledge Universiti Sains Malaysia for the research university grant (1001/PTEKIND/815066), PRGS Grant (1001/PTEKIND/844104), and Heveaboard Sdn Bhd for providing raw materials to be used in this study.

REFERENCES CITED

Ačkar, Đ., Babić, J., Šubaric, D., Kopjar, M., and Miličević, B. (2010). "Isolation of starch from two wheat varieties and their modification with epichlorohydrin," *Carbohyd. Polym.* 81(1), 76-82.

Akmar, P. F., and Kennedy, J. F. (2001). "The potential of oil and sago palm trunk wastes as carbohydrate resources," *Wood Sci. Technol.* 35, 467-473.

Ashori, A., and Nourbakhsh, A. (2008). "Effect of press cycle time and resin content on physical and mechanical properties of particleboard panels made from the underutilized low-quality raw materials," *Ind. Crop. Prod.* 28, 225-230.

Bono, A., Farm, Y. Y., Krishnaiah, D., Ismail, N. M., Yee, L. Y. K., and Lee, L. M. (2010). "Palm kernel based wood adhesive," *J. of Appl. Sciences* 10(21), 2638-2642.

Chew, L. T., Wong, W. C., Midon, M. S., and Matsumoto, T. (1988). "A study on the properties of synthetic adhesives available in Peninsular Malaysia," *J. Trop. For. Sci.* 3(3), 196-203.

Cognard, P. (2005). *Handbook of Adhesives and Sealants, Elsevier:* United Kingdom: 511.

D' Amico, S., Hrabalova, M., Müller, U., and Berghofer, E. (2012). "Influence of ageing on mechanical properties of wood to wood bonding with wheat flour glue," *Eur. J. Wood Prod.* 70, 679-688.

El-Wakil, N. A., Abou-Zeid, R. E., Fahmy, Y., and Mohamed, A. Y. (2007). "Modified wheat gluten as a binder in particleboard made from reed," *J. Appl. Polym. Sci.* 106, 3592-3599.

Gough, B. M. (1967). "Interaction of wheat starch and epichlorohydrin," *Starch-Starke* 19(8), 240-243.

H'ng, P. S., Lee, S. H., Loh, Y. W., Lum, W. C., and Tan, B. H. (2011). "Production of low formaldehyde emission particleboard by using new formulated formaldehyde based resin," *Asian J. of Scientific Res.* 4(3), 264-270.

Huber, K. C., and BeMiller, J. N. (2010). *Textbook of Modified Starch- Chemistry and Properties: Starches- Characterization, Properties, and Applications*, Bertolini AC, Editor 2010; CRC Press, Taylor & Francis Group: New York, 145-203.

Japanese Industrial Standard Committee (2003). "Standard particleboard (A 5908)," 2003; Japanese Standards Association: Tokyo, Japan.

Jyothi, A. N., Moorthy, S. N., and Rajasekharan, K. N. (2006). "Effect of cross-linking with epichlorohydrin on the properties of cassava (*Manihot esculenta* Crantz) starch," *Starch-Starke* 58, 292-299.

Kartha, K. P. R., and Srivastava H. C. (1985). "Reaction of epichlorohydrin with carbohydrate polymers," *Starch-Starke*. 37, 297-306.

Konnerth, J., Hahn, G., and Gindl, W. (2009). "Feasibility of particle board production using bone glue," *Eur. J. Wood Prod.* 67, 243-245.

Loh, Y. W., H'ng, P. S., Lee, S. H., Lum, W. C., and Tan, C. K. (2010). "Properties of particleboard produced from admixture of rubberwood and mahang species," *Asian J. of Appl. Sciences* 3(5), 310-316.

Malaysian Palm Oil Board (2011). *Overview of the Malaysian Oil Palm Industry*, (http://econ.mpob.gov.my/economy/Overview%202011_update.pdf).

Moubarik, A., Allal, A., Pizzi, A., Charrier, F., and Charrier, B. (2010a). "Preparation and mechanical characterization of particleboard made from maritime pine and glued

with bio-adhesives based on cornstarch and tannins," *Maderas-Cienc. Tecnol.* 12(3), 189-197.

Moubarik, A., Allal, A., Pizzi, A., Charrier, F., and Charrier, B. (2010b). "Characterization of a formaldehyde-free cornstarch-tannin wood adhesive for interior plywood," *Eur. J. Wood Prod.* 68(4), 427-433.

National Toxicology Program (2011). "Report on carcinogens," Research Triangle Park: North Carolina, United States.

Nielsen, J. P. (1943). "Rapid determination of starch content," *Ind. Eng. Chem.* 15(3), 176-179.

Noor, M. A. M., Mohd, A. M. D., Islam, M. N., and Mehat, N. A. (1999). "Physico-chemical properties of oil palm trunk starch," *Starch-Starke* 51, 293-301.

Nourbakhsh, A. (2010). "Mechanical and thickness swelling of particleboard composites made from three-year-old poplar clones," *J. Reinf. Plast. Comp.* 29(4), 481-489.

Papadopoulou, E., Nakos, P., Tsiantzi, S., and Athanassiadou, E. (2008). "The challenges of bio-adhesives for the wood composite industry, in *9th Pacific Rim Bio-based Composites Symposium*; Chimar Hellas SA: Rotorua, New Zealand, 1-10.

Sulaiman, O., Salim, N., Nordin, N. A., Hashim, R., Ibrahim, M., Sato, M. (2012). "The potential of oil palm trunk biomass as an alternative source for compressed wood," *BioResources* 7(2), 2688-2706.

Swinkels, J. J. M. (1985). "Composition and properties of commercial native starches," *Starch - Stärke*, 37(1), 1-5.

Tomimura, Y. (1992). "Chemical characteristics of oil palm trunk," *Bull. Forestry Forest Prod. Res. Inst.* 362, 133-142.

Tondi, G., Wieland, S., Wimmer, T., and Schnabel, T. (2011). "Starch-sugar synergy in wood adhesion science: basic studies and particleboard production," *Eur. J. Wood Prod.* 70, 271-278.

Wang, Z., Gu, Z., Hong, Y., Cheng, L., and Li, Z. (2011). "Bonding strength and water resistance of starch-based wood adhesive improved by silica nanoparticles," *Carbohyd. Polym.* 86, 72-76.

Yalinkilic, M. K., Imamura, Y., Takahashi, M., Kalaycioglu, H., Nemli, G., Demirci, Z., and Ozdemir, T. (1998). "Biological, physical and mechanical properties of particleboard manufactured from waste tea leaves," *Int. Biodeter. Biodegr.* 41, 75-84.

Yimsamerjit, P., Surin, P., and Wong-on, J. (2007). "Mechanical and physical properties of green particle board produce from corncob and starch binder composite," In: *International Conferences on Engineering and Environment- ICEE*: Phuket, Thailand.

Zhu, Z., and Chen, P. (2007). "Carbamoyl ethylation of starch for enhancing the adhesion capacity to fibers," *Journal of Applied Polymer Science, 106*(4), 2763-2768

Article submitted: June 19, 2012; Peer review completed: July 21, 2012; Revised version received: Nov. 13, 2012; Accepted: Nov. 16, 2012; Published: November 27, 2012.