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IMPORTANCE OF LIGNIN ON THE PROPERTIES OF BINDERLESS PARTICLEBOARD MADE FROM OIL PALM TRUNK

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ABSTRACT

This study investigated the influence of soda lignin extracts from oil palm on the binderless particleboard properties especially in against moisture. Characterization analysis of oil palm soda lignin, including carbon, hydrogen, nitrogen, sulphur elemental analysis, ash content, infrared spectra via fourier transform spectrometer, glass transition point, and thermal decomposition were conducted. Moisture resistant properties including thickness swelling, and dimensional changes with changes of relative humidity of binderless board with lignin add-on were investigated. Mechanical strength properties including modulus of rupture and internal bond strength were evaluated. Scanning electron microscopy image of specimens was inspected. Lignin was statistically significant influencing the properties of oil palm binderless particleboard. All specimens with lignin added showed better performances than control specimens that without lignin add-on. Binderless board with 5% lignin add-on showed the best result in moisture resistant properties and mechanical properties.

Keywords: oil palm trunk, Soda lignin, binderless particleboard.

INTRODUCTION

Composite panels including particleboards and fibreboards are manufactured using formaldehyde based adhesives in many countries including Malaysia (Hashim *et al*. 2009). One of the disadvantages of such adhesives is its formaldehyde emission during the service life of final products. It is a well known fact that formaldehyde is not only hazardous to human health but also causes significant environmental pollution if it is above certain limits. The requirement of formaldehyde release from the composite panels has become very strict in many countries. Furthermore, the increasing price of solid wood has initiated the study on non-wood agricultural crop waste as an alternative material (Hashim *et al*. 2011). Therefore, the studies on experimentally manufacture of binderless boards from non-wood material and evaluating their properties have increased in last 10 years or so (Hashim *et al*. 2011, Halvarsson *et al*. 2009, van Dam *et al*. 2004, Anglès *et al.* 2001).

Oil palm (*Elaeis guineensis)* has the largest plantation land among other agricultural crops in Malaysia. Oil that can be produced from the mesocarp of the fruit has important commercial value. However, substantial amount of waste material during the harvesting of such resource is created in the form of biomass. This byproduct is approximately 51.2 million tons per year (Sumathi *et al.* 2008). Unfortunately, biomass from oil palm plantation is not used as efficiently as it should be. Burning and land-filling are common practices, which put an adverse impact on the environment. The intensive use of such agricultural waste should be promoted (Ibrahim *et al.* 2012). Based on the results of some of the previous studies, oil palm biomass would be used as raw material to produce value-added composite panels.

In one of these attempts, binderless composite samples made from oil palm strands met minimum modulus of rupture requirements stated in Japanese Industrial Standard (JIS) (Hashim *et al.* 2010).Binderless fibreboard panels were also made from steam exploded pulp of oil palm resulting satisfactory physical and mechanical strength characteristics (Suzuki *et al* 1998).Previous works showed that different parts of oil palm particularly trunks had good potential as raw material for binderless board manufacture. According to Hashim *et al.* (2010, 2011a), binderless particleboards made from trunk had the highest physical and mechanical properties as compared to those made from other parts of oil palm. This could be due to rich of sugar, carbohydrate, and saccharide compounds of the trunk that contribute to particles bonding each other well.

It seems that using oil palm trunks to manufacture particleboard panel without any adhesives addition would be usable approach to add value such waste raw material efficiently. However, moisture resistant of binderless oil palm composites has always been an important problem. Based on some previous work, the authors found that oil palm binderless board had poor moisture resistant (Hashim *et al*. 2010, Hashim *et al.* 2011a) Similar problem has also been observed in composite panel made from other agricultural crops wastes. Most of agricultural crops including oil palm biomass is a naturally hygroscopic material (Hashim *et al*. 2011b). The poor moisture resistant affects the durability and strength performance of the board when changes of

moisture and humidity condition take place. Especially in high moisture condition and high relative humidity, these conditions usually decrease the bonding of biomass composite (Halligan, 1970).In order to improve the moisture resistant of composite panels, hydrophobic compound is one of the important elements.

Lignin is a hydrophobic compound by its nature (Lu *et al.* 1998). Lignin can be defined as three dimensional amorphous polymers with high-polymer degrees. It is a complex organic polymer derived from plant. It is one of the abundant and renewable chemical compounds found in any lignocellulosic materials. In many plants, the yield of lignin is exceeded only by cellulose (Mansouri and Salvado, 2006). It is the crucial compounds in cell wall for strength properties of woody tissue. However, the lignin is an obstacle to prevent water absorption into the cell wall due to its hydrophilic polysaccharide compound structure. Currently, there is no information or any attempts to evaluate the effect of lignin on properties of binderless oil palm particleboard. Therefore, the main objective of this work was to manufacture such panels using soda lignin extracted from oil palm and use it in particleboard manufacture to investigate its influence on some properties of the panels.

MATERIALS AND METHODS

Material preparation

Oil palm trunks were obtained from FELCRA Kampung Gajah Perak Malaysia. Trunks were left to air dry before they were chipped and grounded into particles with 2 mm aperture mesh size. Lignin was also extracted from particle samples to be used in experimental binderless panels.

Lignin extraction and chemical analysis

Soda lignin was extracted by using 4% of sodium hydroxide at the ratio of 10:1 to samples at the temperature of 100 °C. Black liquor was precipitated using 50% sulfuric acid to pH level of 3.5 and later it was filtered (Khan 2004, Pouteau 2003). In the next step, the crude lignin was washed with hot water and dried with freeze dryer unit. Following the washing of the material with the mixture of cyclohexane and ethanol (1:1, v/v), the solvent was removed via rotary evaporator. Lignin extracted particles were not used to make experimental binderless panels. Carbon, hydrogen, sulphur, and nitrogen contents of the lignin were determined using Perkin Elmer 2400 Series II CHN elemental Analyzer. The percentage of oxygen was predicted by subtracting amount of carbon, hydrogen, sulphur and nitrogen from 100% (Mansouri and Salvado, 2006). The ash content of lignin was also determined gravimetrically after approximately 1g lignin sample calcined in a furnace at the temperature of 575 °C for 3h (Mansouri and Salvado, 2006). The fourier transform infrared spectra of lignin were performed in Thermo Scientific Nicolet IS 10. The spectrums of lignin in the range of 500 cm^{-1} – 4000 cm^{-1} were recorded. The differential scanning analyse was carried out in nitrogenous atmosphere from 50°C to 200°C at the heating rate of 10 °C/min. The thermal gravimetric analyse was also performed in nitrogenous atmosphere from 50 °C to 800 °C at the heating rate of 20 °C/min.

Panel manufacture

A total of 12 single layer binderless boards at a target density of 0.60 g/cm^3 were produced. The purified soda lignin was mixed with oil palm trunk particles at 1%, 3% and 5% (w/w) with three replications, respectively. Three binderless boards without addition of lignin were also made as control samples. The mat layout of each binderless board was performed manually into a mould and compressed under digital controlled hot press at pressure of 10 MPa with temperature of 200 °C for 20 min (Boon *et al*. 2013). Panels with 10% urea formaldehyde resins were made as comparison. These test specimens were conditioned in a climate chamber having the temperature of 20 $\pm$ 2 °C and 65 $\pm$ 5% relative humidity in conditioning room. Density of each panel determined according to EN 323 (1993). Thickness of the board was determined according to EN 324 (1993). Binderless boards that fulfilled the density and thickness tolerance assigned by EN 312 (2010) were cut into test specimens.

Moisture resistant and mechanical strength tests of the samples

Moisture resistant properties and mechanical properties of test specimens were performed according to British Standard and European Standard. Thickness swelling, dimensional changes with changes of relative humidity, bending strength and internal bond strength were performed according to EN 317 (1993), EN 318 (2002), EN 310 (1993) and EN 319(1993), respectively.

Scanning electron microscopy

The binderless particleboard specimens were cut into about 0.5 cm by 0.5 cm cross sections. All samples were gold sputtered using sputter coater model Polaron SC 515 $\pm$20 nm. The specimens micrograph image were taken with LEO Supra 50 Vp field emission scanning electron microscope (FESEM)

Statistical analysis - Tukey Test (Significant Difference Test)

The comparison of the means was performed with Tukey HSD at alpha value < 0.05. Results of evaluation with replication were expressed as mean $\pm$ standard error.

RESULTS AND DISCUSSIONS

Lignin content, elemental analysis and ash content of the oil palm soda lignin

The yield of soda lignin extraction from particles of oil palm trunk was found as 9%. The specimens had 41.22% carbon, 5.32% hydrogen, 0.23% Nitrogen, 0% sulphur. Oxygen content of the lignin was estimated as 53.23% based on the method used by Mansouri et al (Mansouri and Salvado, 2006). Based on the results in the work, the percentage of nitrogen showed that the

contamination due to protein content and nitrogeneted compounds was low. Soda lignin was free from any sulphur existence. The amount of mineral contains in the test sample was 0.30%, suggesting that the sample had only little contamination.

Fourier transform infrared spectroscopy

The FT-IR spectra of oil palm lignin are shown in Figure-1. Aromatic C-H out of plane bending appears at the band of 897.85 cm^{-1}. The bands of 1324.25 cm^{-1} associated with the sinapyl ring breathing with C-O stretching, while the band of 1242.24 cm^{-1} is associated with coniferyl ring breathing with C-O stretching (Sun *et al*. 1998).The band of 1460.16 cm^{-1} corresponds to C-H deformation and aromatic methyl group vibrations. The bands of 1420.71, 1503.25 and 1595.02 $cm^{-1}$are referring to aromatic skeleton vibrations of the lignin (Mohamad Ibrahim *et al.* 2011). The band of 2924.36 cm^{-1} refer to C-H stretching of methyl, methylene and methoxyl groups, and 3650.64 cm^{-1} band is O-H stretching associated with phenolic OH and aliphatic OH compounds (Mohamad Ibrahim *et al*. 2011).

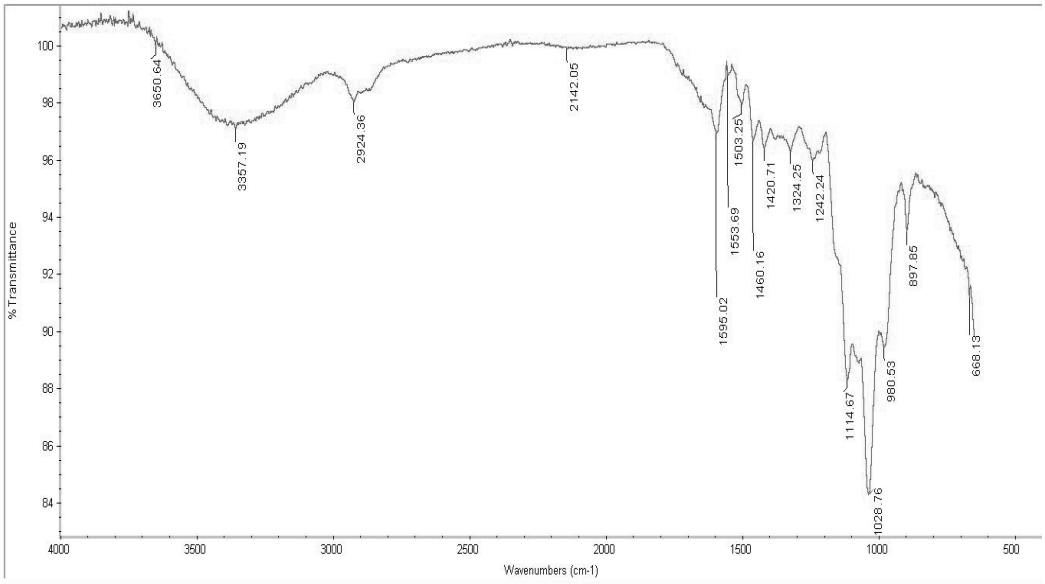

Figure-1. Fourier transform infrared spectrum of oil palm trunk soda lignin.

Differential scanning calorimetry analysis and thermo gravimetric analysis

The curve of DSC of the lignin is shown in Figure-2. The second peak of the heat can be attributed to the glass transition point of oil palm lignin, which is located at around 140 °C. Similar glass transition point of soda lignin has been observed in other studies (Pucciariello, 2004; Lora and Glasser, 2002).

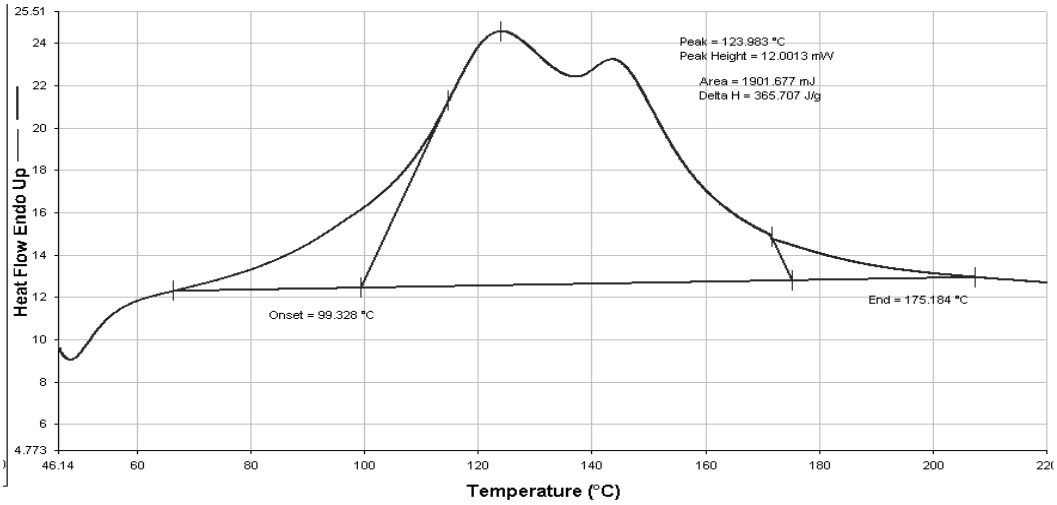

Figure-2. Differential scanning calorimetry analysis of oil palm trunk soda lignin.

Based on TGA curve shown in Figure-3, degradation point of oil palm lignin is at around 240 °C. The first slope in the curve can be attributed to the loss of moisture in the samples. The second slope is belonged to the beginning of the thermal degrades of the lignin. Therefore, the data from DSC and TGA, the temperature of 200 °C used during hot press is suitable for the lignin. This temperature is above the glass transition point and below the thermal degradation point of lignin specimens, which may result in increase flexibility of lignin and preserve it from thermal degradation during hot press.

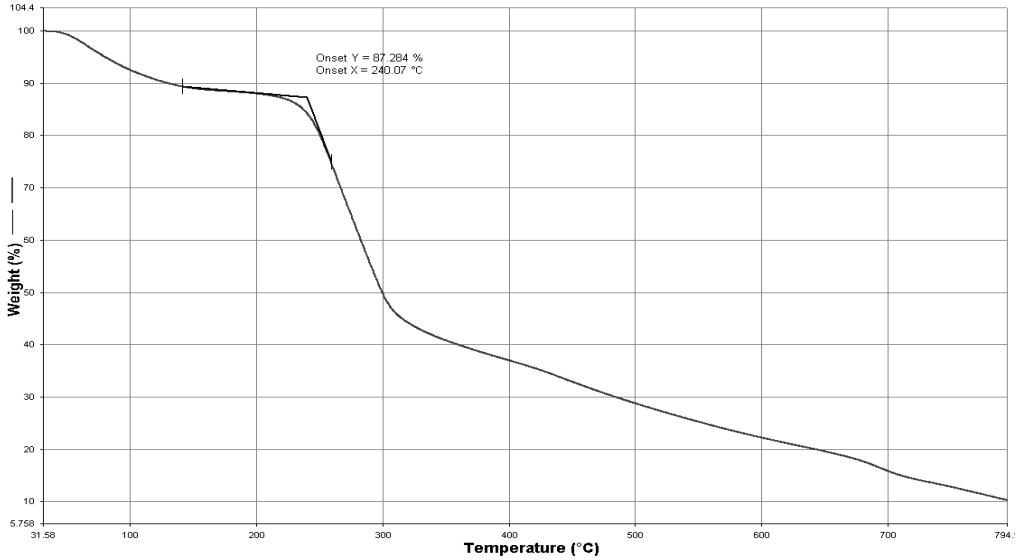

Figure-3. Thermo gravimetric analysis of oil palm trunk soda lignin.

Moisture resistant of the specimens

The result of the dimensional and mass changes with changes of relative humidity and thickness swelling of the samples showed in Table-1,Table-2 and Table-3respectively. Specimens with soda lignin add-on showed better moisture resistant than those of made without lignin. From the result, it showed that the increment of soda lignin add-on amount in the specimens was able to statistically significant improving the moisture resistant of the board. Based on the findings in this study, samples manufactured having 5% lignin add-on resulted in the best dimensional characteristic and thickness swelling properties. Specimens with 5% lignin add-on had an average thickness swelling value of 18.88% and least changes of dimensional when across changes of humidity among these lignin add-on specimens.

Table-1. Dimensional changes of binderless particleboard with changes of relative humidity from 65% to 85%.

Amount of added lignin	Dimensional changes with changes in relative humidity from 65% to 85%		
	Length	Thickness	Weight
0%	0.23 ± 0.01a	2.94 ± 0.07a	0.77 ± 0.02a
1%	0.21 ± 0.02a	2.89 ± 0.16a	0.75 ± 0.02a
3%	0.18 ± 0.02ab	2.46 ± 0.11b	0.63 ± 0.02b
5%	0.13 ± 0.02bc	1.99 ± 0.09c	0.49 ± 0.01c
10% UF resin	0.09 ± 0.02c	1.23 ± 0.08d	0.47 ± 0.01c

*Different letter within the same column are statistical significant difference at $\alpha = 0.05$.

Table-2. Dimensional changes of binderless particleboard with changes of relative humidity from 65% to 35%.

Amount of added lignin	Dimensional changes with changes in relative humidity from 65% to 35%		
	Length	Thickness	Weight
0%	-0.07 ± 0.01a	-0.16 ± 0.01a	-0.88 ± 0.02
1%	-0.05 ± 0.02ab	-0.13 ± 0.01b	-0.83 ± 0.01a
3%	-0.04 ± 0.00abc	-0.09 ± 0.01c	-0.75 ± 0.01b
5%	-0.01 ± 0.02bc	-0.07 ± 0.01c	-0.64 ± 0.02c
10% UF resin	0.00 ± 0.00c	-0.04 ± 0.01d	-0.60 ± 0.03c

*Different letter within the same column are statistical significant difference at α = 0.05.

Table-3. Thickness swelling of binderless particleboard.

Amount of added lignin	Thickness swelling (%)
0%	26.00 ± 0.66a
1%	24.05 ± 0.91ab
3%	22.16 ± 1.50b
5%	18.88 ± 1.39c
10% UF resin	15.35 ± 0.76d

*Different letter within the same column are statistical significant difference at α = 0.05.

In this research, 10% glue level of urea formaldehyde particleboard was used as comparison. There is insignificant difference when comparing the performance of 5% lignin add-on specimens with 10% urea formaldehyde resin specimens in the changes of length and weight with changes of relative humidity. The average thickness swelling rate of the urea formaldehyde specimens was 15.35%. Comparing with urea formaldehyde bonded particleboards, the woody material in binderless panel was lacking of coating protection from the synthetic resins that are hydrophobic. Therefore, such synthetic resins are significantly contributes to the moisture resistant of composite panel (Sreekala *et al*. 2002). Consequently, it is not easy to fully compete with performance of synthetic adhesives bonded composite panels. However, the prior objective of this study was to investigate the influence of lignin to the properties of binderless particleboard.

As mentioned in previous chapter, the glass transition temperature of the lignin used in this study was around 140 °C. Enhanced dimensional values of the lignin added specimens could be related to having pressing temperature well above the glass transition temperature of lignin, resulting in its elasticity during pressing, so that lignin could easily fit in and within the particles of the panels to improve their hygroscopicity. It is a fact that lignin is more hydrophobic than solid wood, which could cover the surface area of individual particles making them more resistance against moisture absorption (Rozman *et al*. 2000).

Mechanical properties of the specimens

Table-4 displays mechanical properties of the samples. In this study, the mechanical strength of samples were increased significantly when the amount of added lignin were increased. Similar to dimensional stability of the samples, both MOR and IB strength values were also found the highest for those panels made with 5% lignin add-on. The sample having 5% lignin was shown with modulus of rupture value of 19.65 MPa and 0.81 N/mm^2 of internal bond strength.

Table-4. Mechanical strength properties of binderless particleboard.

Amount of added lignin	Mechanical properties	
	Internal bonding strength (N/mm^2)	Modulus of rupture (MPa)
0%	0.66 ± 0.01a	15.41 ± 0.23a
1%	0.68 ± 0.01a	15.81 ± 0.28a
3%	0.70 ± 0.01a	17.04 ± 0.31b
5%	0.81 ± 0.03b	19.65 ± 0.19c
10% UF resin	0.86 ± 0.01c	20.36 ± 0.43c

*Different letter within the same column are statistical significant difference at α = 0.05.

The improvement of mechanical strength after lignin add-on could be due to the natural adhesion characteristic of lignin. The weak point that created from loose contact of particles in the composite panel could be

one of factors that cause failure when stress is developed on the panel. Lignin that fills in between particles could act as a binder, to improve the contact bonding of particles. Thus, reduction of the ease of breaking increases the mechanical strength of the test specimens.

However, comparing to commercial adhesives in this study, the adhesions of the lignin is weaker than the commercial adhesives. Only 5% lignin add-on specimens showed insignificant different bending strength comparing with 10% urea formaldehyde specimens. The liquid adhesives are easier to spread evenly on the particles, to have better coverage than the added lignin.

Scanning electron microscopy

The Figure-4 shows the scanning electron microscopy image of specimens with 5% lignin add-on. The figure showed that the oil palm trunks fibres are compressed after hot pressing. Although the binderless specimen was formed without using adhesives, but the contact between particles after hot pressing are relatively close.

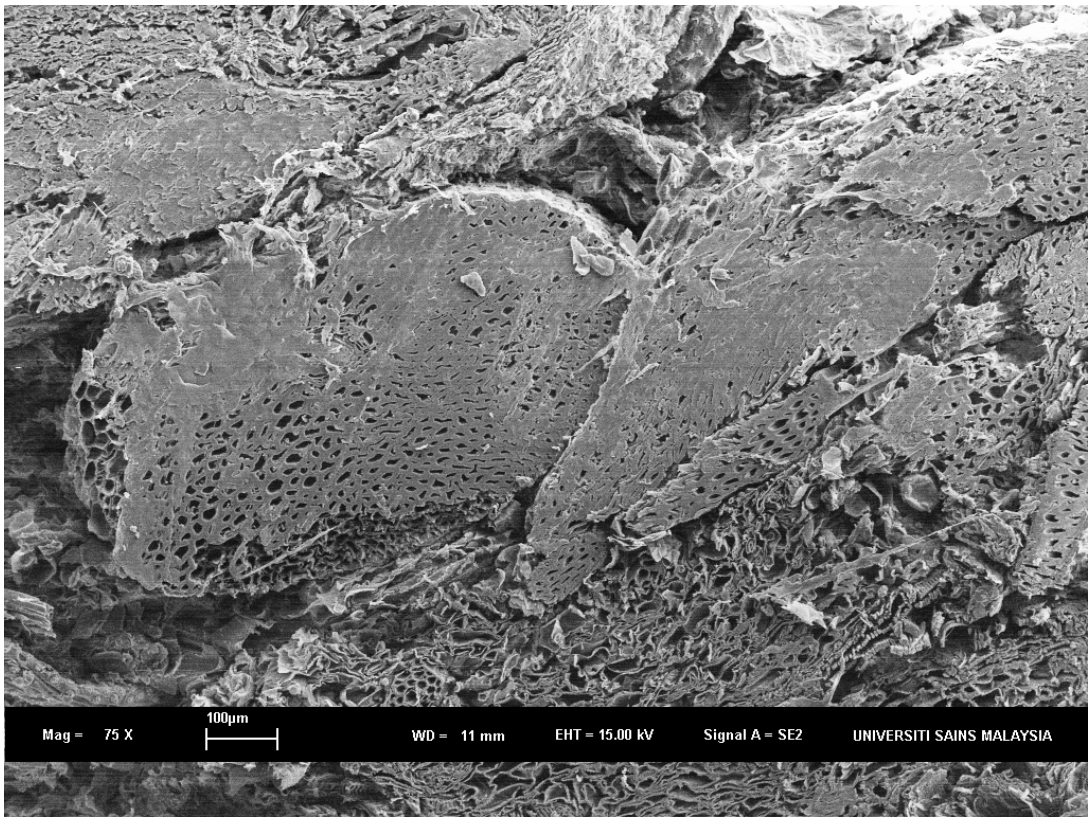

Figure-4. Scanning electron microscopy image of 5% lignin add-on specimens.

CONCLUSIONS

In this study, oil palm soda lignin was significantly influencing binderless particleboards properties positively. The results showed that 5% of lignin addition gave best performance on binderless particleboards in against moisture and its mechanical properties as well. Proposing soda lignin can be work as additive to improve binderless particleboard properties. However, the performances of the treated particleboard still did not meet the most of specifications required for commercial composite panels suggesting that binderless boards still need undergo some improvements to compete with properties of commercially manufactured composite panels. Modification of particles properties using heat or chemical treatment would be considered some of the alternatives to enhance properties of the final product. Consequently, the main advantage of binderless board is their environmental friendly characteristics and lower cost than other panels made with different types of adhesives.

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