

DEPENDENCE OF POLYURETHANE CONTENT ON PHYSICAL AND MECHANICAL PROPERTIES OF WOOD FIBER/PALM KERNEL SHELL COMPOSITES

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(RECEIVED AUGUST 2022)

ABSTRACT

Wood-based composites with different ratios of wood fiber (WF)/palm kernel shell (PKS) and polyurethane (PU) content have been prepared using the wet-process method. Samples of WF85/PKS15 and WF75/PKS25 were fabricated where each sample was applied with 20% and 70% of PU contents and its physical and mechanical properties had been studied. The physical results show that the samples with 70% of PU content were denser, had low porosity, low moisture content, and low water absorption. Surface morphology observation shows both series samples with high PU content tend to form tube-like shape with different diameter. In mechanical studies, generally, the sample with high PKS and PU possesses high flexural strength, flexural modulus, tensile strength, tensile modulus, and hardness. However, the increased of PKS content in the composite reduces the tensile strength for both samples with 20% and 70% of PU. The effects of the binder and palm kernel shell in the composite were also explained. Based on the Japanese Industrial Standard (JIS) A 5905 standard, the sample composites meet the requirement under medium density fiberboard (MDF) category and classified as board type 5 which suitable as furniture, house, and automotive interior design and construction materials.

KEYWORDS: MDF, wood fiber, palm kernel shell, polyurethane adhesive, mechanical properties.

INTRODUCTION

Wood-based composites have become important for building materials, furniture components, interior and exterior house panels design, and car panel (Aziz et al. 2015). The applications of wood-based composites are typically depending on its mechanical properties such as strength, hardness, tensile, and fractural (Alabduljabbar 2020, Renner et al. 2021). The factors that influence the mechanical properties of wood composites are dependent on wood species, the type of binder, wood geometry and the density of the panel composite itself (Papadopoulos 2019). Composite production is also one of the steps in addressing the lack of wood. This involves a mixture of agricultural waste materials or from agro-industry residues like peanut husk, hazelnut shell, husk, wheat bran, banana stems, orange peels, cotton, and corn stalks (Akgül and Tozluoğlu 2008, Çöpür et al. 2008, Kagarfard and Latibari 2011).

In oil palm industry, the waste produced from the processing of palm oil like empty fruit bunches, mesocarp fiber, palm kernel cake, and palm kernel shell are also getting attention. Palm kernel shell (PKS) is usually thrown away or used as combustion material in boilers for generating electricity (Hamzah 2019). Study on chemical properties shows PKS contents a large amount of lignin, hemicellulose, and cellulose, while its physical properties have high bulk density ($440 - 740 \text{ kg m}^{-3}$), low moisture content (6 - 13%) and 28% of porosity (Romisuhani et al. 2010, Ikumapayi and Akinlabi 2018, Elham 2001). For this reason, PKS has gained intention to serve some applications like lightweight concrete, filler reinforcement materials as well as filtration materials (Alengaram et al. 2013, John et al. 2015, Ogedengbe 1985, Edmund et al. 2014, Baby et al. 2019).

Medium density fibreboard (MDF) is a wood-based composite mainly formed by wood fibers, combined with wax and a synthetic resin such as urea formaldehyde (UF) or binder such as isocyanate by applying high temperature and pressure to form dry-formed panel product (Khalil et al, 2010, Nasir et al, 2014, Alabduljabbar, 2020). In the production of MDF, formaldehyde-based resin likes phenol-formaldehyde, urea-formaldehyde, melamine-formaldehyde, and isocyanate are usually used as binder (Dukarska et al. 2006, Halvarsson et al. 2008, Nuryawan and Alamsyah 2017). However, these substances have been known to cause health problems (Myers 1984, Gonzalez 2011). These include effects on the respiratory system, nervous system, skin problems, eye irritation and certain type of cancers (Casteel et al. 1987, Ülker and Ulker 2019). Therefore, demand on a non-toxic binder is crucial in MDF manufacturing. Polyurethanes (PU) are a versatile class of polymers which have very good mechanical properties, chemical resistance, and resilience. Although PU is the result of the chemical reaction between diisocyanate and polyol, but it is completely inert and harmless to humans (Dernehl 1966). Among the most important types of polyurethanes is Thermoplastic PU. Thermoplastic PU is a melt-processable thermoplastic elastomer with high durability and flexibility which suitable for a wide range of applications (Qi and Boyce 2005). The questions that arise here is either PU compatible to be used as a binder for wood-based composites and its effect on their mechanical properties. This paper presents the bonding effect through mechanical testing on wood fibre and palm kernel shell composite using polyurethane as binder. Discussion in sample preparation and testing including samples performance and use is also presented.

MATERIAL AND METHODS

Sample preparation

The fiber board composites were fabricated using the wet-process method. In this study, two series of samples were prepared. The wood fibre (WF) and palm kernel shell (PKS) were supplied from a local MDF mill and palm oil mill, respectively. The commercial polyurethane (PU) adhesive no. 705-9046 (Geocel limited, United Kingdom) was used. The first series of composite fibreboard samples consist of 85% of WF and 15% of PKS (WF85/PKS15), whilst the second series content of 75% of WF and 25% of PKS(WF75/PKS25). Each series samples were added with 20% and 70% of PU adhesive.

Here, the WF was sieved with a mesh opening of 2.0 mm for homogeneity while the PKS was washed with detergent to removed oil residue and dried in the oven at 140°C for 30 min. After dried, the PKS was grounded using kernel grinder machine model IKA MF 10 Basic and sieved to obtain 600 µm particle sizes. Both samples were added with 20% and 70% of PU resin from the total mass of WF/PKS before being mixed homogenously using electric mixer for 30 min. The mixed compound was placed into the mould with inner dimension 10.5 cm width by 130 mm length and undergone a pre-heated process in the oven at temperature 140°C for 30 min before pressed at 15 MPa using mechanical press and further heated at 140°C for 1 hour. Then, the sample was left overnight at room temperature for curing process. Finally, the board sample was taken out from the mould and again left at room temperature for 2 days for conditioning. The prepared samples thickness is approximately 3 mm.

Sample measurement

Measurement on density, porosity, moisture content, water absorption, surface morphology, hardness, flexural and tensile test of the composite samples have had been conducted. The particle density and surface morphology of the fibreboard samples has been carried out using helium psychometry test (AccuPcy II 1340, Micromeritics Equipment) and scanning electron microscope (SEM-CARL ZEISS MA10).

The tensile and flexural tests were conducted using Universal Testing Machine from HAIDA equipment and TM2101 software. Three tests were carried out for each sample for both measurements. The composite sample under tensile test gives tensile strength (TS) and tensile modulus (TM). TS or ultimate strength is defined as the capacity of a material to break under tension whilst the TM also known as Young's modulus or elastic modulus, is a measure of the stiffness of an elastic material. For three-point flexural test, where P is load applied at the fixed rod to the sample, b and h are the sample's width and thickness, respectively and L is the distance between the two adjustable rods (Fig. 1a). As the load, P is applied, the sample's deflection will be measured to determine the sample bending properties.

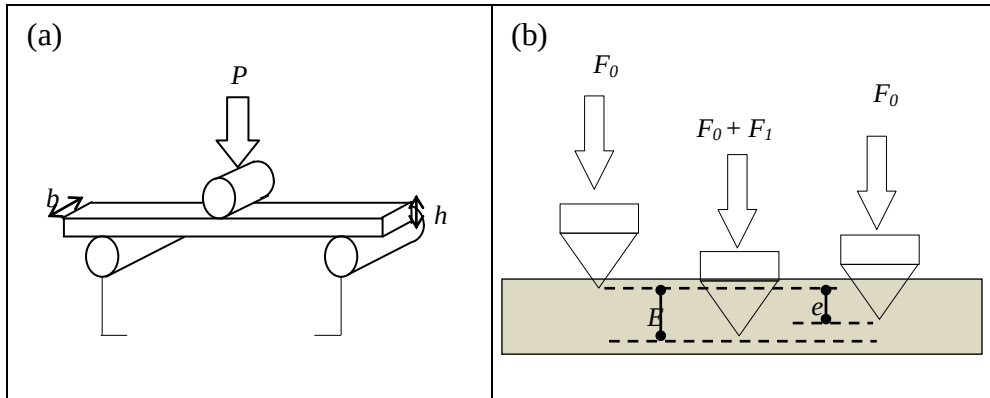


Fig. 1: a) Three-point flexural test, b) Rockwell hardness test.

The flexural stress, F_p for a rectangular cross section sample is given by the Eq. 1:

$$F_p = \frac{3LP}{2bh^2} \text{ (Pa)} \quad (1)$$

where: L is the distance between the two supporting pins, P is for force load given by a pin on the top, b is the sample width and h is for thickness. The flexural strain, E_f , is determined according to the Eq. 2:

$$E_f = \frac{6Dh}{L^2} \quad (2)$$

where: D is the maximum deflection at the center of the specimen. The elasticity or bending modulus, E_b which is equivalent to FM is expressed with the Eq. 3 as follows:

$$E_b = \frac{L^3 m}{4bh^3} \quad (3)$$

where: m is the tangent of the initial straight portion of the stress-strain curve.

The TS is given by the Eq. 4 below;

$$TS = \frac{F}{A} \text{ (Pa)} \quad (4)$$

where: F is the linear force applied and A is the original cross-sectional area of the material. The TM is given by Eq. 5:

$$TM = \frac{Fl_0}{A\Delta l} \text{ (Pa)} \quad (5)$$

where: l_0 is the initial length and Δl is the changes in length.

The bending test is important in classification of MDF follows the JIS A 5905. The sample dimension used for flexural test was 50 x 130 mm and 20 x 130 mm (width x length) for tensile test. The gauge length of the test piece is 80 mm. The crosshead speed was set up to 10 mm·min⁻¹ to follow the requirement of JIS A5905: 2003 fiberboard standard for MDF or insulating board. Hardness measurement was carried out using INOVA Rockwell Hardness tester from CV. The indenter type used was C Rockwell intender with radius 0.2 mm and force applied was 147.1 N (Fig. 1b). Rockwell hardness values were obtained from the average of four readings. The sample is placed on the table prior testing. Initially, the preliminary load, F_0 is applied on the sample surface where depth of penetration by indenter is set to zero when the equilibrium is reached. Then, the major load, F_1 is applied resulting increase of penetration. After equilibrium reached again, the major load is removed causing reduced in depth of penetration. Since, the application and removal of the major load causing permanent increase in depth of penetration, therefore, the Rockwell hardness number, HR is calculated using Eq. 6:

$$HR = E - e \quad (6)$$

where: e is a permanent increase in depth of penetration due to major load, F_1 is measured in units if 0.002 mm, E is a constant depending on form of indenter such as 100 units, 130 units is for diamond and steel ball indenter, resp. (Riggio and Piazza 2010, Webo et al. 2018).

RESULTS AND DISCUSSION

The effect of WF/PKS ratio and PU content to the physical properties of the composite sample; density, porosity, moisture content (MC) and water absorption (WA) of composite samples were given in Tab. 1. The density results show the PU really affecting the sample's density. The density of WF85/PKS15 sample increases from 500 kg·m⁻³ at 20% PU to 720 kg·m⁻³ when the PU 70%. For sample with 25% PKS, similar pattern is observed. The sample initially has 570 kg·m⁻³ with 20% PU and increase to 690 kg·m⁻³ when PU was added to 70%. Comparison between samples with different PKS content shows at 20% PU, the sample's density was slightly increasing from 500 kg·m⁻³ to 570 kg·m⁻³ for PKS15 to PKS25, resp. However, for sample with 70% of PU, the addition of PKS slightly decreases the sample's density. According to on the JIS A5905: 2003 the sample's density falls under medium density fibreboard category and classified as board type 5 which is suitable for furniture, house and automotive interior design and construction materials.

For porosity, the results obtained are in opposite to the density result. The porosity results for WF85/PKS15 and WF75/PKS25 samples show increase PU content from 20% to 70% had slightly decrease the porosity. Increase amount of PKS has decrease the porosity for samples with low PU, however at high PU content, increases of PKS also increase the porosity. The MC for both samples decreases with PU increment but increase with the increases of PKS content. For WA results, the water absorption percentage decrease with increase PU for both samples. However, for low PU content, the increase of PKS had reduced the sample WA while at high PU, the WA increase with PKS increases.

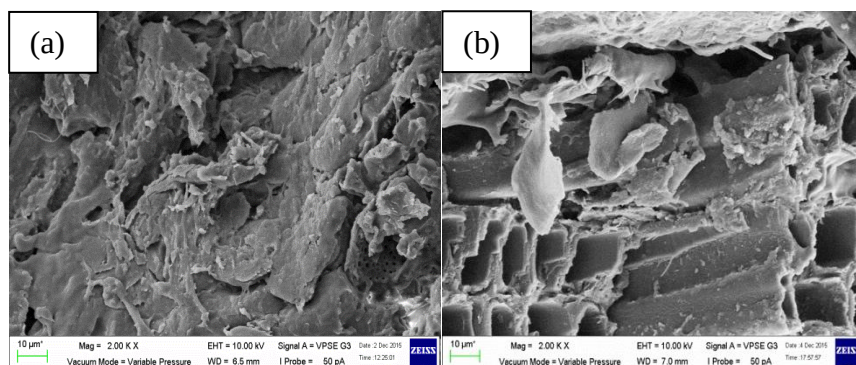
Tab. 1: Effect of PU on the (a) density, (b) porosity, (c) moisture content and (d) water absorption of WF/PKS composites.

Physical properties	Sample WF85/PKS15		Sample WF75/PKS25	
	20% PU	70% PU	20% PU	70% PU
Density (kg m^{-3})	500	720	570	690
Porosity (%)	65.1	45.46	59.7	48.1
Moisture content (%)	9.0	6.5	9.5	8.4
Water absorption (%)	125.7	44.4	108	53.5

Fig. 2 shows the surface morphology of WF composite samples with different PKS and PU content. Figs. 2a,b are the surface morphology for WF85/PKS15 sample with 20% and 70% PU, respectively. While the surface morphologies for WF75/PKS25 sample are shown in Figs. 2c,d with respectively, 20% and 70% PU content. In these figures, both samples composite with 20% PU (Figs. 2a,c) shows very pack structure while for samples with 70% PU (Figs. 2b,d), tube-like formation with different holes diameter were formed. This formation might be due to low wood fiber and the presence of high PU percentage in the composite. As higher percentage of PU added, a more significant tube-like formation is observed (Zamri et al. 2019). The linkage between PU and WF/PKS shows there is strong bonding between them. Xue. et. al. (2014) relates lignin and PU were bonded by covalent bonding under the pre-polymer reaction.

In addition, Ping et. al. (1998) reported that PU pre-polymer reacted with cellulose hydroxyl groups, and it suggested there is hydrogen bonding exist between the molecule of cellulose and PU. Some of PU pre-polymer molecule simultaneously penetrated cellulose gives results to strong interfacial bonding which resulting the mechanical properties, water resistivity, and optical transmittance of the coated films improve significantly. In Fig. 2c, it can be seen clearly that the PKS particles visible as small sphere spots at the surface of the sample. This is because of high amount of PKS and low PU.

Flexural test is a measure of force required to bend a material and determines the resistance to flexing or stiffness of a material. The fractural strength (FS) is the point of strain where the material physically ruptures whereas the flexural modulus (FM) is a measure of the tendency for a material to resist bending. The results of load-deflection curve for all composite samples are on Fig. 3. Both samples with 20% PU content (Figs. 3a, c) possess low stiffness. However, for both sample with 70% PU content improve the composite stiffness three time. These results suggested that high content of PKS and PU could increase the composite stiffness.



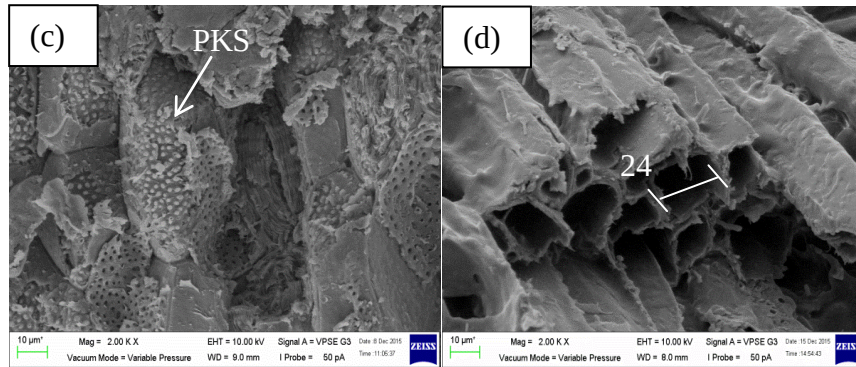


Fig. 2: Surface morphology of WF/PKS samples with different PU content. The WF85/PKS15 sample with PU; (a) 20% and (b) 70%, and WF75/PKS25 sample with PU; (c) 20% and (d) 70%.

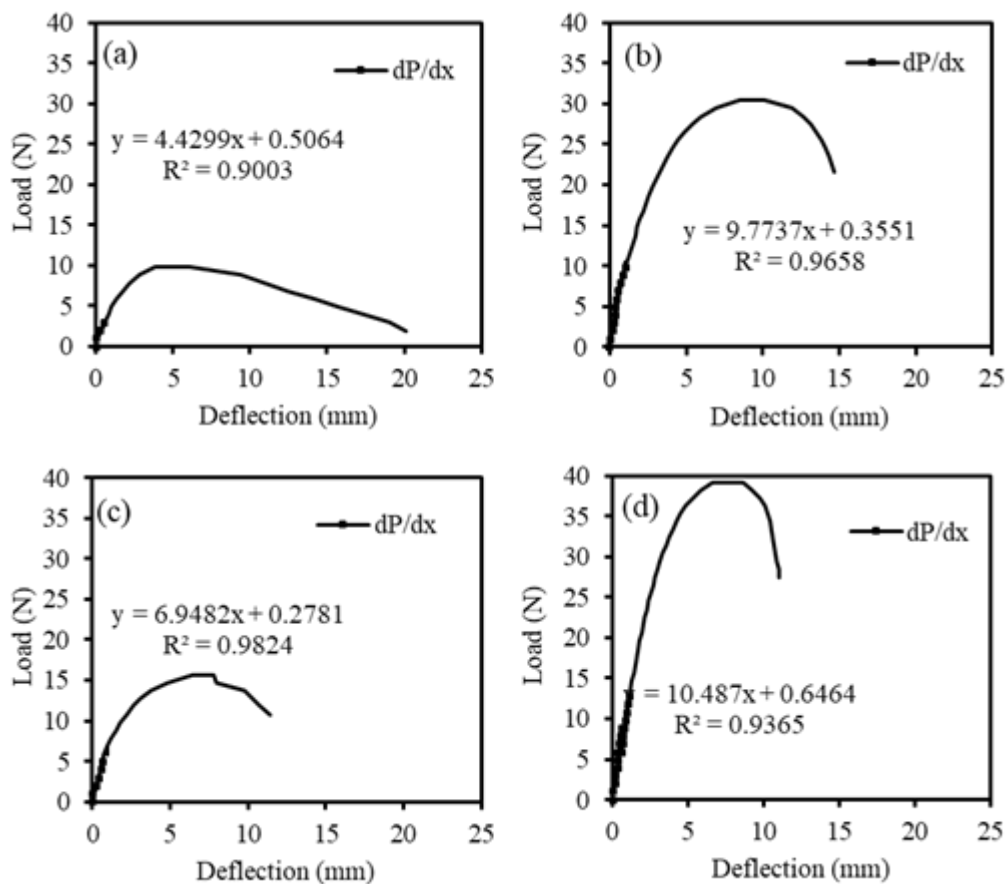


Fig. 3. Load-deflection curve from selected flexural test for MDF. WF85/PKS15 sample with PU (a) 20% and (b) 70%, and WF75/PKS25 sample with PU (c) 20% and (d) 70%.

Fig. 4 shows (a) the FS and (b) FM of WF/PKS composites with different PU contents. Generally, both samples show improvement in FS and FM when the PU increase from 20% to 70% except for sample with PKS 25% and PU 70% content. According to Badri et al. (2005) higher PU levels tends to lower the sample strength due to the decrease wetting of fibre thus contributing to decrease in modulus (Badri et al. 2005). Interestingly, the samples with high PKS

(25%) have higher FS and FM compared to low PKS composite. This might be due to the capability of PKS to absorb stress and better interaction between the WF and PKS filler Ong et al. (2016). Fig. 5 shows (a) the TS and (b) TM of WF/PKS composite with different PU content. The use of PU as the composite binder also affects the TS and TM properties. The TS of PKS15 improves almost 6 times from 1.1 MPa for 20% PU to 5.9 MPa as the PU 70%. While for sample composite with PKS25%, its TS increase almost 4 times from 0.9 MPa to 3.7 MPa when the PU increased. The results also show that the sample with PKS15 has higher TS compared to sample with PKS25. This indicates that the PU has excellent bonding between wood fibers which possibly due to the reaction between the isocyanate groups and hydroxyl groups of the polyurethane and fibres, respectively.

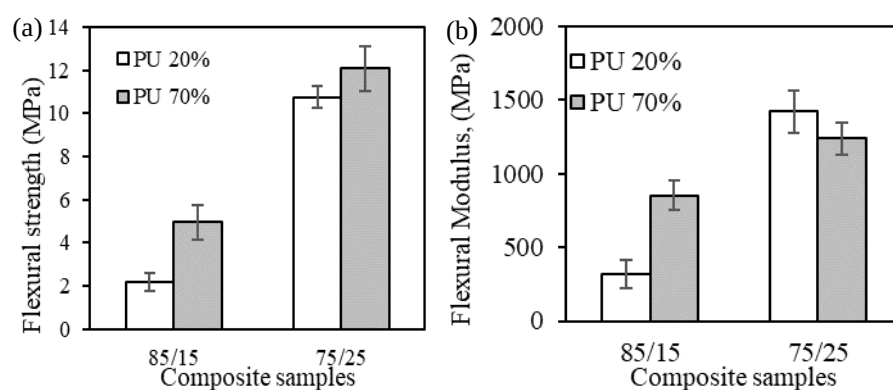


Fig. 4: Flexural strength and flexural modulus of WF/PKS composites with different PU contents.

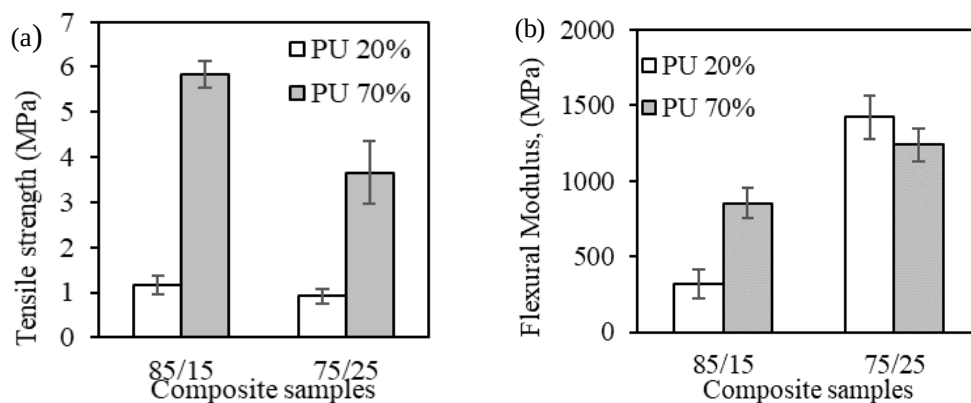


Fig. 5: Tensile strength and tensile modulus of WF/PKS composites with different PU contents.

For TM measurement, the results show sample PKS25 with 70% PU has higher TM as compared to sample PKS15. However, at low PU content (20%), the TM for PKS15 sample has much higher than that PKS25 sample. The results of TS and TM also reveal that sample with lower PU content are influenced by wood fiber amount whereby reducing the amount of wood fiber will also reduce the TS and TM. The prepared high PU content samples were found to have better TS and TM properties as compared to the results of wood-based MDF using phenol

formaldehyde, urea formaldehyde, melamine formaldehyde and standard particle board with respectively, TM; 56.6 MPa, 48.3 MPa, 47.2 MPa, and 35.5 MPa, and TS; 1.8 MPa, 0.92 MPa, 0.90 MPa, and 0.80 MPa (Kannan et al. 2014).

The hardness results of the composites are shown in Fig. 6. Both samples show increases in hardness as the PU increase from 20% to 70%. The increase of hardness due to PU is twice the hardness of low PU content. Comparison between samples with same PU content shows both samples had small increases in hardness property as the PKS increases from 15% to 25%. This result suggest that high PU content promotes good strength and hardness for WF/PKS composites.

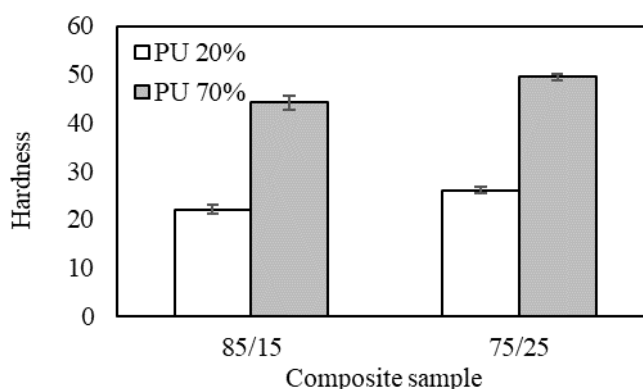


Fig. 6: Effect of PU composition to the hardness of WF/PKS composites.

CONCLUSIONS

The performance of mechanical properties of WF85/PKS15 and WF75/PKS25 samples with different polyurethane content as adhesive has been evaluated. Basically, the use of PU as binder improves the physical and mechanical properties of the two samples. It is also clearly shown that Wood Fiber 75% + PKS 25% samples are better in terms of physical and mechanical properties compared with Wood Fiber 85% + PKS 15% samples. The formation of tube-like structure was found not only responsible in improving the samples mechanical properties, but also minimal materials use and minimal sample weight which in turn lower the materials cost. Both sample composites had met the requirement under medium density fibreboard category and classified as board type 5 based on JIS A 5905 standard.

ACKNOWLEDGMENTS

The authors would like to thank UPM (grant vote no: 9449600) for supporting this research. We also thanked Daiken Sarawak Sdn. Bhd and Human & Eco Energy Resources Development (M) Sdn. Bhd wood fiber and palm kernel shell supply, respectively, and to Sime Darby Austral Sdn. Bhd for allowing us to use palm kernel grinder machine.

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