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THE EFFECT OF NCO/OH EQUIVALENT WEIGHT RATIOS OF POLYURETHANE (PU) BINDER AND PU BINDER CONTENT ON PROPERTIES OF MEDIUM DENSITY FIBREBOARD (MDF)

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ABSTRACT

The potential of using bio-based binders for medium density fibreboard (MDF) has received increased attention. However, no study was done on the potential of palm-based polyurethane (PU) as biobased binder in MDF. Therefore, an investigation on the effect of equivalent weight ratios of isocyanate (NCO) and hydroxyl group (OH) of palm oil-based PU binder and binder content on the properties of MDF was conducted. Palm oil-based PU binders were prepared at different equivalent weight ratios of NCO and OH at 1.5/1 (PU1.5), 2.0/1 (PU2.0) and 2.5/1 (PU2.5) and binder content at 3%, 5%, 7% and 10% were used in the preparation of MDF. The polyol, NCO and palm oil-based PU binders were characterised using Fourier transform infrared spectroscopy and thermogravimetric analysis. As equivalent weight ratios of NCO and OH and binder content increased, the modulus of rupture, modulus of elasticity and internal bonding of MDF were consistently increased. Thickness swelling and moisture content decreased with increased equivalent weight ratios of NCO and OH and binder content. In general, MDF prepared with PU2.0 with 7% binder content fulfills all British Standard (BS EN 622-5, 2009) requirements, indicating its suitability for MDF application.

Keywords: bio-based polyol, mechanical and physical properties of MDF, medium density fibreboard (MDF), palm oil-based PU binder.

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INTRODUCTION

In recent years, many countries have seen a surge in demand for wood-based composite panels, including plywood, particleboard and fibreboard. Medium density fibreboard (MDF) is one of Malaysia's most important and largest wood-based industries. MDF is a wood panel made by mixing wood fibres with a binder and then pressing the mixture at high temperatures and a density of around 750-800 kg m^{-3} (Pizzi *et al*., 2020). MDF's qualities are influenced by several factors, which can be divided into three main groups. The first group is wood species (Ayrilmis *et al*., 2017), density (Cai *et al*., 2006; Hong *et al.*, 2017), fibre length (Benthein *et al.*, 2014), moisture content of fibres, the strength of fibre, wettability of fibre surface and surface free energy (Cai *et al*., 2006). The second group is related to resin binder such as types of resins (Imtiaz *et al*., 2014), resin reactivity, chemical structure and composition of resin (Lu *et al*., 2021), level of resin spread, viscosity and rate of resin curing (Hong and Park, 2017). The third group is related to the preparation of MDF processes such as hot-pressing

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temperature (Wang *et al*., 2019), pressure (Gul *et al*., 2017), mat moisture content (Magalhães *et al*., 2021) and assembly time.

Apart from that, the binder consumption and composition of the binder are the two most important factors that affect the cost and quality of the board. The amount of binder used in the production of MDF panels accounts for at least 25% of the entire cost of production. Cyr *et al*. (2006) reported the efficiency of the binder has a considerable impact on the cost and performance of MDF. There are several studies that have been conducted toward getting a good quality MDF by determining the optimal binder content. Hong *et al.* (2017) and Praveen *et al*. (2019) reported that an increase in binder consumption of MDF led to a corresponding increase in the quality of MDF board mechanical properties but a decrease in thickness swelling (TS) and water absorption (WA). Meanwhile, Khairiah and Khairul (2006) and Mahzan *et al.* (2010), had prepared the boards from the composition ratios between wood fibre and binders. They found that as the composition ratio of the binder increased, the properties of the boards also increased.

Currently, conventional thermosetting wood binders are frequently manufactured with fossil-derived ingredients such as urea-fromaldehyde (UF), phenol formaldehyde, melamine formaldehyde, *etc*. (Kumar and Pizzi, 2019; Mantanis *et al*., 2018; Wibowo *et al*., 2020). However, prolonged exposure to formaldehyde has been reported to lead to cancer in humans, as indicated by the International Agency for Research on Cancer (IARC) (IARC, 1995). IARC officially classified formaldehyde as a "classified human carcinogen" for nasopharyngeal cancer in 2004 and leukemia in 2012 (IARC, 2006; 2014). These classifications have driven manufacturers of MDF to find non-toxic binders as an alternative. In order to eliminate formaldehyde emissions from both formalin-based binder and resin-based formaldehyde-bonded wood products, the use of bio-based binders or isocyanate (NCO) binders has been increased due to reduced health hazard effects (Emelie *et al.*, 2017; Manggar *et al.*, 2021). Thus, many studies were conducted to change the composition and technology of UF resins for composite wood products, especially without the inclusion of formaldehyde. Besides, concern about free formaldehyde emissions from wood composites, particularly for indoor applications, as well as increasing environmental knowledge about the sustainability of raw materials and finished goods, along with new stringent environmental legislation, are the major contributing reasons for scientists and industries to move from traditional formaldehyde-based synthetic resins to new bio-based binders for the development of eco-friendly products (Navarrete *et al*., 2013 and Valyova *et al*., 2017).

Alternative binder such as NCO is an excellent binder known for total lack of formaldehyde emissions and can be used to bond wood composites at significantly lower levels than UF, MUF and PF binders (Pizzi *et al*., 2020). According to Rowell and Ellis (1981), isocyanate binder has the advantage such as high strength, resistance and is also stable although it is applied on treated wood. NCO binder is very low in molecular weight, low in viscosity and low in surface tension. This binder wets readily and penetrates deeply into wood (Shi and Gardner, 2001). NCO binder reacts with the moisture in the wood to form a polyurea/polybiuret network (Ni and Frazier, 1998). Additionally, the NCO binder has the ability to produce covalent urethane bonding with wood (Zhou and Frazier, 2001), which improves bondline durability. NCOs have traditionally been used in two methods as wood binder. Firstly, NCO is currently widely used in the particle board industry (Arif and Eka, 2017; Charles, 2003). Secondly, through PU prepolymers process with an excess NCO content from the reaction between NCO and polyol (Daneshvar *et al*., 2019). For this study, the second method was used and it seemed important to optimise the ratios between NCO and palm oil-based polyol to achieve better properties of MDF.

The compositions of PU binder, such as NCO and hydroxyl groups (OH), have the greatest effect on binder strength characteristics. When the NCO/OH equivalent weight ratio in the PU binder is higher than 1, binder bonding strength increases due to the perfect reaction between OH group in wood material and NCO group which forms a urethane bond (Khoon Poh *et al*., 2014; Nacas *et al*., 2017). Daneshvar *et al.* (2019) prepared PU wood adhesives from liquefied beech sawdust and pMDI or MDI with different NCO/OH molar ratios from 0.7 to 1.7. They reported that the highest lap shear strength was achieved at an NCO/OH ratio of 1.7 for pMDI and MDI, which is related to excess-free NCO groups. Besides, the lap shear strength of PU based on pMDI was higher than that of PU based on MDI.

Georgieva *et al.* (2008) prepared PU binders with various ratios of OH and NCO groups from 1.0 : 2.2 to 1.0 : 3.0. The PU binders were solid at room temperature with a melting point between 40°C and 60°C. The results showed that the wood composites prepared have a tensile strength of up to 47 N/mm^2 and flexural strength of up to 58 N/mm^2. Recently, Praveen *et al*. (2019) studied the effect of binder content in the production of MDF. The boards were made using 6%, 8% and 10% PF resin and hot pressed at 17.5 kg/cm^2 specific pressures and a temperature of 150°C for 12 min. The results showed that the boards prepared with 10% resin content met the minimum requirement specification of Indian standard for medium density fibreboard for general purpose (ISO:12406, 2003), except for some moisture-related properties, *i.e.*, surface absorption, water absorption, thickness swelling due to general absorption *etc.*, which can be controlled by suitable treatment. Wan Mohd Nazri *et al*. (2019) reported that an increase in the resin content from 8% to 12% led to a corresponding increase in the modulus of rupture, modulus of elasticity and internal bonding of MDF.

In the PU industry, polyols have always played an important role as the main raw material primarily produced from petrochemical. The process of producing polyols from petroleum-based involves high energy consumption, is expensive and has an adverse effect on the environment. Due to the environmental issues and economic point of view, it is significant to replenish petroleum-based polyols with more environmental-friendly vegetable oil-based polyols as alternative raw materials that are renewable, more versatile, and less costly (Chain and Gan, 1998). Several studies have reported on the application of renewable resources such as natural oil-based polyols in the production of PU binders for MDF applications. Fiorelli *et al.* (2011) developed a board using sugarcane bagasse bonded with PU derived from castor oil and determined the board's properties. They managed to get a high-density board which was suitable for industrial use. Campos *et al.* (2014) evaluated the mechanical and physical properties of the MDF board bonded with PU derived from castor oil. They found that the MDF boards showed higher values than those recommended by the European MDF standards. Pereira *et al.* (2016), evaluated the performance of MDF boards bonded with castor oil-based PU and found that the shear strength of MDF with PU samples showed better results compare to PVA and contact adhesives which were, mostly commercial. These natural oil-based polyols have the potentials to be used as PU binders in wood-based composites.

In Malaysia, with the abundant supply of palm oil (Parveez *et al*., 2021), efforts to develop palm oil-based PU through the chemical modification of palm oil started in the early 1990s. This creates an opportunity to use palm-based PU resin as a bio-binder in the production of MDF. Moreover, the utilisation of palm-based PU binder in MDF and its influence on the physical and mechanical properties of MDF is not well reported. Therefore, understanding the above correlation is necessary to produce MDF with higher or comparable physical and mechanical properties to those using petroleum-based commercial binders. The outcome of this study may indirectly increase the demand for sustainable palm oil for the PU binder industry in MDF production.

Most of the binders rely on petroleum as the feedstock. As an alternative, the preparation of green binders utilising green palm oil-based polyols with zero formaldehyde emission need to be studied. Besides, it also increases the amount of green carbon content in the PU materials, a major advantage of using bio-based polyols instead of petroleum-based polyols. There have been no studies on the interaction of MDF with polyurethane binder made from palm oil-based polyol, thus its properties must be studied. Therefore, the potential application of palm oil-based polyol as one of the primary raw materials for the preparation of palm oil-based PU binder in the production of MDF and the effect of equivalent weight ratio of NCO/OH and binder contents on properties of MDF are reported in this paper.

MATERIALS AND METHODS

Materials

The palm oil-based polyol was prepared using epoxidation and alcoholysis processes established by Tuan Noor Maznee *et al*. (2018). The properties of palm oil-based polyol are shown in *Table 1*.

TABLE 1. PROPERTIES OF PALM OIL-BASED POLYOL

Properties	Value
Hydroxyl value (mg KOH/g)	147.20
pH	6-7
Moisture content (%)	0.1907
Viscosity at 25°C, cP	6 000-8 000
Acid value (mg KOH/g)	0.8095

Source: Tuan Noor Maznee *et al*. (2018).

The NCO component of the palm oil-based PU binder system was purchased from Huntsman Polyurethane (Selangor, Malaysia), a polymeric diphenylmethane diisocyanate (pMDI), Suprasec 5005 with NCO content of 31.0%. Rubberwood fibre was supplied by Donghwa Malaysia Sdn. Bhd. (Negeri Sembilan, Malaysia).

Preparation of Palm Oil-based PU Binder

The palm-based polyol used in the preparation of palm oil-based PU binder was dried at 70°C-80°C for 24 hr under a vacuum at 1-3 mm Hg with continuous mixing using a magnetic stirrer. The water content of the polyol after drying was determined using a Karl Fischer Titrator (Mettler Toledo LLC, Ohio, USA). The palm oil-based PU binders were prepared according to the formulation shown in *Table 2*. The dried palm-based polyol and NCO were weighed in a speed mixer cup and mixed at 3000 rpm using a speed mixer (Flak Tek Inc., Colorado, USA) for 1 min at room temperature.

The NCO/OH equivalent weight ratios of the palm oil-based PU binder were varied at 1.5, 2.0 and 2.5. After mixing, a small amount of palm oil-based PU binder was taken out to determine free NCO content according to ASTM D-2572 (American Standard Test Method, 1997), and *Table 2* displays the results of measured free NCO content. The palm oil-based PU binders were diluted in a fixed weight ratio of 70:30 between the palm-based PU binder and ethyl acetate. The mixtures were homogenised at 2200 rpm for 30 min at room temperature (28ºC) using a high-speed mixer. Herein, the ethyl acetate was used as a solvent to reduce the viscosity of the palm oil-based PU binder for optimal distribution onto rubberwood fibres and used as binders in the preparation of MDF.

Preparation of Medium Density Fibreboard (MDF)

An industrial dry process was adopted to prepare MDF in the laboratory using rubberwood fibres. In the preparation of the palm oil-based PU binder, the equivalent weight ratio of NCO/OH was varied from 1.5 to 2.5, in which the obtained palm oil-based PU binder was designated as PU1.5, PU2.0 and PU2.5, respectively. The amount of palm oil-based PU binder was fixed at 10% based on the oven-dry mass of fibres. The palm oil-based PU binder was sprayed onto rubberwood fibres in a rotary drum blender. The fibres covered with binder were formed into mats. The fibre mats were subjected to a process of cold pressing with a pressure of 49 bar to tack the fibres together and followed by a hot press at 180ºC and a pressure of 172 bar for 5 min to obtain the final board. The final thickness of the board was adjusted to 12 mm using two stop bars during the hot press. After the pressure was released, the board was cooled to room temperature at 25ºC and cut according to the predetermined dimensions. After that, the boards were conditioned for 7 days in a humidity chamber at 65% relative humidity and a temperature of 20ºC to obtain a moisture content of about 10%. The targeted density of the boards was 720 kg/m^3 with board dimensions of 300 x 300 x 12 mm. The boards were designated as MDF 1.5-10, MDF 2.0-10 and MDF 2.5-10, respectively. The same methods were used to produce MDF at different binder contents of 3%, 5%, 7% and 10% based on the oven-dry mass of rubberwood fibres. The equivalent weight ratio of NCO/OH of the PU binder used was 2.0. The designation of the prepared MDF with different equivalent weight ratios of NCO/OH palm-based PU binders and PU binder contents is shown in *Table 3*.

Characterisation of Palm Oil-based PU Binder and MDF

Pot-life analysis of palm oil-based binder. From the perspective of PU binders system's application, the pot life of PU binders is important. The pot life is the maximum amount of time that the system can be applied to a substrate while still being suitably fluid. The palm oil-based PU binders were prepared according to the formulation shown in *Table 2*. The dried palm oil-based polyol and NCO was weighed in a speed mixer cup and mixed at 3000 rpm using a speed mixer (Flak Tek Inc., Colorado, USA) for 1 min at room temperature. The gelation time was recorded as the pot life of PU samples.

ATR-FTIR Analysis of Palm Oil-based PU Resin and MDF

The ATR-FTIR analysis was carried out using the Perkin Elmer Spectrophotometer model Spectrum 100 equipped with *Diamond Attenuated Total Reflectance* (DATR) (Perkin Elmer, Massachusetts, USA). The FTIR spectra were recorded between 4000-650 cm^{-1} wavenumbers with a 20 kHz/scan frequency and an average of 5 scans for each sample at 4 cm^{-1} resolutions. Before evaluating the sample with similar scan details, the background spectrum was first acquired. The resultant spectrum was analysed for functional groups of -OH, -C=O, C=N, N=C=O, N-H and C-O-C.

TABLE 2. DESIGNATION AND COMPOSITION OF PALM OIL-BASED PU BINDER AND MEASURED FREE ISOCYANATE (NCO) CONTENT

Designation of palm oil-based PU binder	NCO/OH equivalent weight ratios to prepared palm oil-based PU binder	Isocyanate (NCO) (g)	Palm-based polyol (g)	Measured free NCO content (%)
PU1.5	1.5	34.42	65.58	5.53
PU2.0	2.0	41.17	58.83	8.48
PU2.5	2.5	46.66	53.34	13.52

TABLE 3. DESIGNATION OF THE PREPARED MDF WITH DIFFERENT EQUIVALENT WEIGHT RATIOS NCO/OH AND BINDER CONTENT OF PALM OIL-BASED PU BINDER

Designation of MDF	Palm oil-based PU binder	Binder content (%)
MDF 1.5-10	PU1.5	10
MDF 2.0-10	PU2.0	10
MDF 2.5-10	PU2.5	10
MDF 2.0-3	PU2.0	3
MDF 2.0-5	PU2.0	5
MDF 2.0-7	PU2.0	7
MDF 2.0-10	PU2.0	10

Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) is a test for determining a change in the sample weight in correlation with increasing temperature. The test was carried out using a TGA Perkin Elmer model STA 6000 (Perkin Elmer, Massachusetts, USA). The carrier gas was nitrogen. The sample weight was about 10 mg. The temperature was set from 50°C to 700°C at a heating rate of 10°C/min. The sample was held in an isothermal at 50°C for 1 min and heated up to 750°C. An isothermal process at 750°C was held for 1 min before the experiment ended.

Physical and Mechanical Test of MDF Boards

The MDF was cut into precise test sample sizes in accordance with British Standard (EN 622-5, 2009). The parameters evaluated were the bending strength such as modulus of rupture (MOR) and modulus of elasticity (MOE), internal bonding (IB) strength, thickness swelling (TS) and moisture content (MC). For the IB strength test, samples were bonded to metal blocks with a surface area of 50x50 mm and applied with a hot-melt resin adhesive. The TS test was obtained after immersion in water for 24 hr, and the temperature was maintained at 21°C ± 2°C. For the MC test, the samples were placed in the drying oven for 48 hr at a constant temperature of 103°C ± 2°C. The density of MDF was measured by the procedure of BS EN 323 (British standard, 1993a). *Table 4* below shows MDF's minimum and maximum requirements with a thickness of 12 to 19 mm in dry conditions.

Statistical Analysis

One-way ANOVA and Turkey's significant difference test were performed using an SPSS version 26.0 statistical software package (IBM Corp., New York, USA) on the modulus of elasticity, modulus of rupture, internal bonding strength, thickness swelling and moisture content results for the analysis of variance at a 95% confidence interval ($p \leq 0.05$).

TABLE 4. REQUIREMENTS FOR GENERAL PURPOSE BOARDS FOR USE IN DRY CONDITIONS (TYPE MDF)*

Parameters	Unit	Test method	Requirement
Modulus of elasticity	N/mm^2	BS EN 310 (British Standard, 1993b)	2 200 (minimum)
Modulus of rupture	N/mm^2	BS EN 310 (British Standard, 1993b)	20 (minimum)
Internal bonding	N/mm^2	BS EN 319 (British Standard, 1993c)	0.55 (minimum)
Thickness swelling 24 hr	%	BS EN 317 (British Standard, 1993d)	12 (maximum)
Moisture content	%	BS EN 322 (British Standard, 1993e)	5-8**

Source: * - British Standard (2009); ** - Dongwha Malaysia (2023).

RESULTS AND DISCUSSION

Palm oil-based PU binders were characterised via ATR-FTIR and TGA techniques. The effect of different equivalent weight ratios of palm oil-based PU resins and resin contents as a binder were evaluated based on British Standard (BS EN 622-5, 2009).

Characterisations of Palm Oil-based PU Binders

Pot-life analysis of palm oil-based binder. The palm oil-based polyol (Pioneer E-135) was reacted with NCO at 1.02 NCO index to form a PU binder. The results in *Table 5* clearly indicate that the pot life decreased as the equivalent weight of NCO/OH ratios increased. The palm oil-based PU binders (PU2.5) cured faster compared to PU1.5 and PU2.0 in this study.

TABLE 5. POT LIFE OF PALM OIL-BASED PU BINDER

Designation of palm oil-based PU binder	Isocyanate (NCO) (g)	Palm-based polyol (g)	Measured pot life (s)
PU1.5	34.42	65.58	570
PU2.0	41.17	58.83	460
PU2.5	46.66	53.34	340

The pot life of the PU1.5 and PU2.0 were recorded at 570 and 460 s, while the pot life of the PU2.5 was just 340 s. This means that, after 340 s the PU2.5 cannot be used as a binder over the substrate anymore because it is already in the solid state.

Changes in Functional Groups of Palm Oil-based PU Binders via FTIR Spectra

The FTIR spectra of the palm oil-based PU binders with different equivalent weight ratios of palm-based polyol and isocyanate (PU1.5, PU2.0 and PU2.5), palm-based polyol and isocyanate, as a comparison, are shown in *Figure 1*. The FTIR spectrum analysis was employed to characterise the raw materials and analyse the changes in functional groups of the palm oil-based PU binders when different equivalent weight ratios of NCO/OH were used.

From *Figure 1a*, it was observed that all the palm oil-based PU binders (PU1.5, PU2.0 and PU2.5) possess the following characteristic absorption bands, urethane NH stretching at 3300 cm^{-1}, bending at 1516 cm^{-1}, methylene or alkyl group at 2974 cm^{-1}, carbonyl group at 1700 cm^{-1} and C-O-C stretching at 1100 cm^{-1}. The strong characteristic peak at 2272-2275 cm^{-1} is associated with NCO stretching and supported by the disappearance of the broad absorption band near 3400 cm^{-1} corresponding to the -OH group of polyols. The NCO stretching band and the bands mentioned above indicate polyurethane formation in PU1.5, PU2.0 and PU2.5 with free isocyanate. With increasing NCO/OH equivalent weight ratio, the absorption band at peak 2275 cm^{-1} was slightly increased.

FTIR spectra for rubberwood fibres and MDF 2.0-10 are shown in *Figure 1b*. The absorption band centred at 2270 cm^{-1} for the NCO group was not observed, and the free hydroxyl group (-OH) at 3400 cm^{-1} absorption band decreased in the MDF spectrum. But the presence of carbonyl group (C=O) at the 1740 cm^{-1} absorption band appeared to be strong after the production of MDF, indicating that the diisocyanate group had reacted with the hydroxyl group of palm-based polyol and in lignin as well, thus, generated urethane linkages and amide groups throughout the polymerisation process. Lignin, like cellulose and hemicellulose, is a common natural polymer found in plant cells. Lignin has many functional groups, including aliphatic and phenolic hydroxyls, carboxyl and methoxyl groups (Antonino *et al*., 2021).

Thermal stability of palm oil-based PU binders via TGA. The thermal stability of the synthesised palm oil-based PU binders at different equivalent weight ratios of NCO/OH (PU1.5, PU2.0 and PU2.5), palm-based polyol and NCO was studied by TGA. Thermal stability of PU describes thermal durability as well as heat resistance of PU. According to Javier (2019), polymers with higher thermal stability have higher melting points and thermal decomposition, less softening and less mass loss during high-temperature heating, and a higher heat deflection temperature under load, without losing their basic properties, which determine their functionality.

The TGA and derivative thermagravimetric (DTG) thermograms for palm oil-based PU binder PU1.5, PU2.0 and PU2.5 in *Figure 2* are similar, indicating that the thermal stability was little affected by the equivalent weight ratios of NCO/OH. The pyrolysis process of palm-based polyol and NCO can be divided into one stage, while for palm oil-based PU binder PU1.5, PU2.0 and PU2.5, its pyrolysis can be separated into two stages under different heating rates, are shown in DTG thermogram in *Figure 2*. The first step of temperature degradation (T_{d1}) occurred at 280°C-340°C due to the breakage of the hard segment and urethane linkage, causing low dissociation energies of chemical bonds (Fridrihsone *et al*., 2013). While in the second step, temperature degradation (T_{d2}) was due to the decomposition of the polyol chain's soft segment (Piszczyk *et al*., 2014). The results from TGA confirmed that palm oil-based PU binders are suitable to be used as a binder in MDF production as they will not degrade during hot press temperature at 180°C.

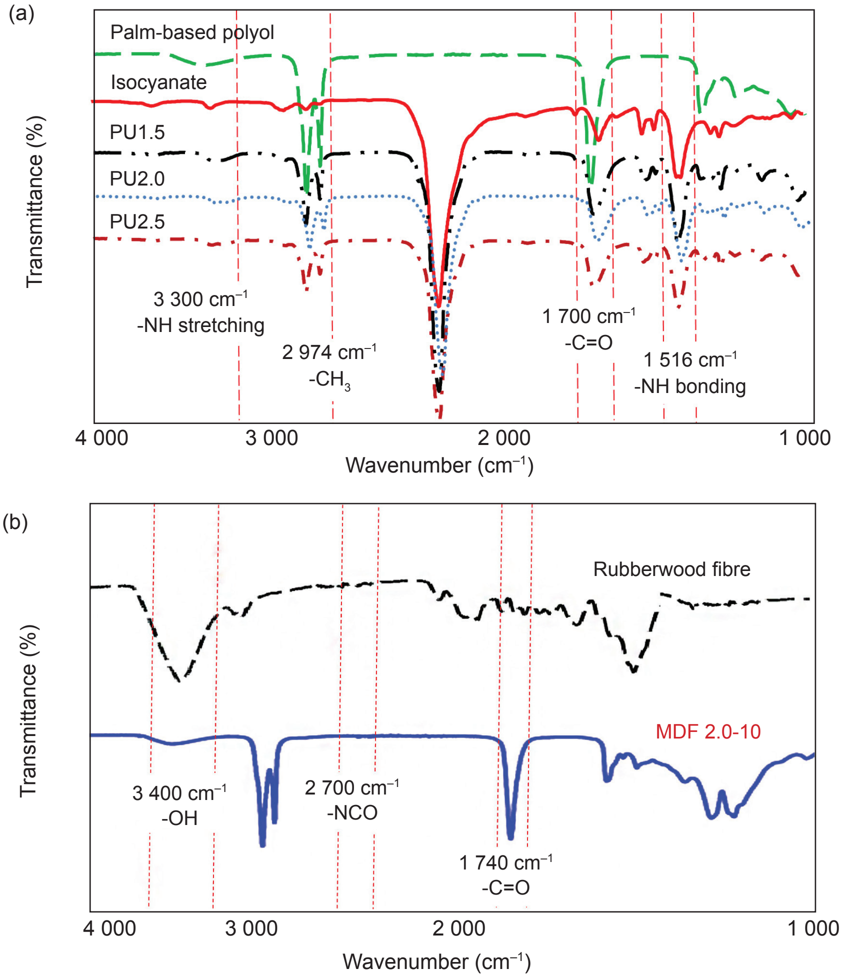

Figure 1. (a) FTIR spectra of NCO, palm-based polyol and palm oil-based PU binders (PU1.5, PU2.0 and PU2.5) and (b) FTIR spectra of rubberwood fibers and MDF 2.0-10.

It has been observed that the temperature degradation (T_d) increases when the equivalent weight ratios of NCO/OH were increased. The thermogram of palm oil-based PU binders PU1.5, PU2.0 and PU2.5 were shifted to the right as compared to polyol and NCO. This is explained by the fact that the polyurethane produced will be NCO terminated as the NCO/OH ratio greater than 1. According to Desai *et al.* (2000), the free NCO groups will react during the crosslinking process resulting in the formation of three dimensional allophanate and biuret linkages and thereby increasing the number of urethane groups. Dutta and Karak (2006) have reported that the urethane linkages formed increased with the increase in NCO/OH ratio and they decomposed considerably at higher temperature. This result shows that PU2.5 has the highest thermal stability, demonstrating that when the equivalent weight ratio of NCO/OH increases, the crosslinking density increases as a result the polymer backbone chain to undergo closer packing, material becoming harder and enhancing its thermostability (Dutta and Karak, 2006).

Mechanical and Physical Properties of MDF

The effects of equivalent weight NCO/OH ratio of palm oil-based PU binder on the mechanical and physical properties of the MDF were evaluated. The results on the bending strength (MOR and MOE), IB strength, TS at 24 hr and MC at 48 hr of MDF are summarised in *Table 5*. The targeted density of MDF boards is 720 kg/m^3 and the density of the prepared boards was in the range of 723 to 727 kg/m^{3,} which was slightly higher than the target density. Meanwhile, a significant interaction effect was observed when different equivalent weight ratios of NCO:OH of palm oil-based PU binders were used to make MDF, both on mechanical and physical properties of MDF, as indicated by ANOVA statistical analysis ($p \leq 0.05$). This implies that the equivalent weight ratios of NCO/OH of palm oil-based PU binder synergistically affect the board properties.

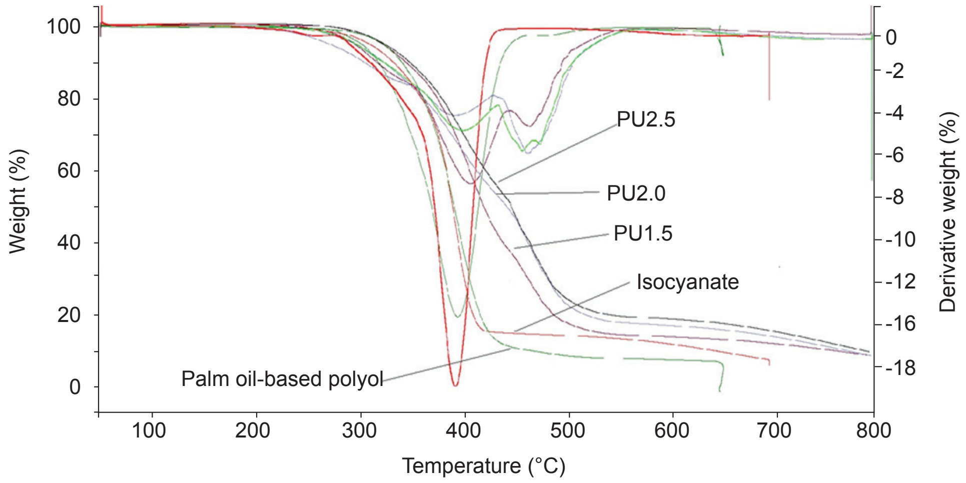

Figure 2. TGA and DTG thermograms of isocyanate, palm-based polyol and palm oil-based PU binders (PU1.5, PU2.0 and PU2.5).

TABLE 6. AVERAGE VALUES OF PROPERTIES OF MDF PRODUCED WITH DIFFERENT EQUIVALENT WEIGHT RATIOS OF NCO/OH OF PALM OIL-BASED PU BINDERS

Designation Of MDF	MOR (N/mm^2)	MOE (N/mm^2)	IB (N/mm^2)	TS at 24 hr (%)	MC at 48 hr (%)
MDF 1.5-10	18.39 ± 1.96^a	2092.33 ± 208.87^a	0.51 ± 0.10^a	14.31 ± 2.51^a	7.01 ± 1.41^a
MDF 2.0-10	21.16 ± 2.10^b	2281.86 ± 188.04^b	0.61 ± 0.09^b	9.81 ± 3.42^b	6.70 ± 1.05^a
MDF 2.5-10	22.44 ± 1.97^c	2391.14 ± 200.07^b	0.70 ± 0.10^c	7.86 ± 2.80^b	5.78 ± 1.07^b

Note: MOR - modulus of rupture; MOE - modulus of elasticity; IB - internal bonding; TS - thickness swelling, and MC - moisture content. Mean followed by the same letters in each column is not significantly different at $p \leq 0.05$ according to the least significant difference Turkey test method.

The MOR measures the maximum bending load that a board can support before it ruptures. It indicates the stress required to cause failure. The MOE measures the boards' stiffness and indicates the ability to resist deflection. MDF boards with palm oil-based PU binder at 1.5, 2.0 and 2.5 of equivalent weight ratios NCO/OH, the MOR and MOE of the boards were 18.39 N/mm^2 and 2092.33 N/mm^2, 21.16 N/mm^2 and 2281.86 N/mm^2 and 22.44 N/mm^2 and 2391.14 N/mm^2, respectively. The MOR and MOE increased proportionately with the equivalent weight ratio of the palm oil-based PU binders (*Table 6*). Thus, MDF 2.5-10 had a relatively higher bending strength value in MOR and MOE than MDF 1.5-10 and MDF 2.0-10. The increment in MOR and MOE is related to the higher percentage of free NCO in the binder (*Table 2*), which allowed for the formation of chemical bonds between free NCO in the palm oil-based PU binder with the hydroxyl group (OH) in rubberwood fibres and water to generate crosslinked polyureas or polyurethane, leading to strong internal bonding strength of the MDF prepared. According to Nacas *et al*. (2017), increasing the NCO/OH ratio increases the free NCO content, leading to higher wood bonding shear strength values.

Internal bonding (IB) strength is a measurement of the bonding interaction between fibres and correlates to the dimensional stability of the boards, which is important to ensure that the boards will not delaminate during post-processing. *Table 6* shows that the IB strength values of MDF increased with increasing equivalent weight ratios of NCO:OH of palm oil-based PU binder from 1.5 to 2.5, where the IB strength values for MDF 1.5-10, MDF 2.0-10 and MDF 2.5-10 were 0.51 N/mm^2, 0.61 N/mm^2 and 0.70 N/mm^2, respectively, where the highest IB strength was observed in MDF 2.5-10. This correlates well with the highest free NCO content of palm oil-based PU binder (PU2.5) (*Table 2*), which promotes the higher formation of urethane and urea linkages, thus, resulting in higher rigidity and bond strength of MDF 2.5-10.0.

Thickness swelling (TS) is a physical property related to the dimensional stability of the boards when exposed to severe humidity, and it is an important property for exterior-use panel materials. Thickness swelling of fibreboards occurs when the cell walls are bulked with water. As shown in *Figure 3*, the thickness of MDF boards slightly expanded after 24 hr of immersing in water as compared to before immersed in water. According to Norul Izani *et al.* (2012), good bonding strength between fibres resulted in low thickness swelling. In this study, all the MDFs showed a decrease in the TS values when the equivalent weight ratios of NCO/OH of palm oil-based PU binders were increased. The TS of MDF 1.5-10, MDF 2.0-10 and MDF 2.5-10 were 14.3%, 9.8% and 7.8%, respectively. The decrease in the TS values was due to the increase of free NCO groups of NCO when equivalent weight ratio NCO/OH was increased. The addition of excess NCO to the palm oil-based PU binders created more reactions between NCO groups and OH groups, which led to an increase in the cross-linking density due to the formation of urethane bonds to prevent the water absorption and improve the dimensional stability of MDF. Moisture contents (MC) of MDF for four different percentages of palm oil-based PU binder were analysed after being conditioned at a constant temperature of 103°C ± 2°C for 48 hr. MC of the MDF slightly decreased with increasing equivalent weight ratios of NCO:OH of PU binder (*Table 5*). The average MC values for MDF 1.5-10, MDF 2.0-10 and MDF 2.5-10 were 7.01%, 6.70% and 5.78%, respectively.

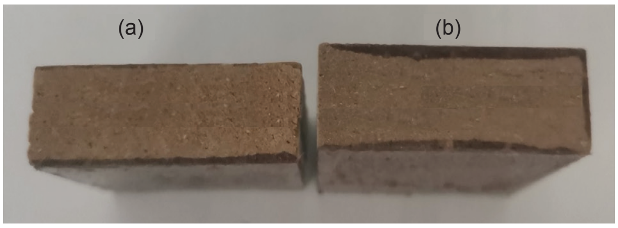

Figure 3. Thickness swelling of MDF 2.0-10 bonded with palm oil-based PU as representation (a) before and (b) after immersed in water for 24 hr.

The results from ANOVA analysis indicated that there was a significant effect between equivalent weight ratios of NCO/OH of palm oil-based PU binder and mechanical and physical properties of MDF at p-value<0.05. This indicated that by increasing the equivalent weight ratios of NCO/OH of palm oil-based PU binder from 1.5:1.0 to 2.5:1.0 in the production of MDF, there was a significant effect on the mechanical and physical properties of MDF. Overall from this study, we can conclude that the optimum equivalent weight ratio of NCO/OH to prepared palm oil-based PU binder for production of MDF that meets all required standard specifications based on BS EN 622-5 (British Standard, 2009) and Dongwha Malaysia (2023) for fibreboard as shown in *Table 4* was 2.0 or MDF 2.0-10.

Effect of Palm Oil-based PU Binder Content on MDF Properties

Other than the NCO/OH equivalent weight ratio of the palm oil-based PU binder, another factor that would greatly influence the MDF qualities is the binder content. Greater binder content is usually associated with higher strength and dimensional stability. However, due to the high cost of binders, the lowest amount of binder is generally employed. Therefore, the effect of palm oil-based PU binder content on MDF properties was studied to optimise the amount of PU binder used. In this study, the palm oil-based PU binder was prepared at a 2.0 equivalent weight ratio of NCO/OH and the palm oil-based PU binder content was varied from 3% to 10% (*Table 3*).

Table 7 displays the mechanical properties such as MOR, MOE and IB strengths values, which positively correlate with palm oil-based PU binder content. The result shows that MOR, MOE and IB values were increased with increasing palm oil-based PU binder content from 3% to 10%. The MOR trend for MDF 2.0-10 with 10% resin content (21.16 N/mm^2) being higher than MDF 2.0-7 with 7% resin content (20.23 N/mm^2), MDF 2.0-5 with 5% resin content (16.47 N/mm^2) and MDF 3 with 3% resin content (14.07 N/mm^2), respectively. This study also indicated that the MDF 2.0-10 exhibited the highest MOE value of 2281.86 N/mm^2 when compared to MDF 2.0-7 (2219.97 N/mm^2), MDF 2.0-5 (1984.87 N/mm^2) and MDF 3 (1618.17 N/mm^2). Meanwhile, the IB strength of MDF 2.0-10 (0.61 N/mm^2) exhibited the highest value as compared to MDF 2.0-7 (0.57 N/mm^2), MDF 2.0-5 (0.46 N/mm^2) and MDF 2.0-3 (0.31 N/mm^2). Generally, by increasing the resin content in MDF from 3% to 10%, the MOR, MOE and IB strength properties were increased significantly. It shows an increment of 50% in MOR and IB strength and 40% in MOE of MDF 2.0-10 in comparison with MDF 2.0-3. The increasing resin content allows for better resin distribution because the interface between the glue and substrate can be improved. This might be due to an increase in the resin content in the board which gives more intimate contact for bonding. According to Phetphaisit *et al.* (2013), free NCO content in PU binder can penetrate the wood and develop urethane linkages by reacting with the wood cellulose hydroxyl group, resulting in higher bonding strength and rigidity. A study by Ashori and Nourbakhsh (2008) found that by increasing the binder content to 9%, 10% and 11%, the MOR, MOE and IB strength of MDF also increased. Similarly, a study by Tabarsa *et al*. (2011) showed that the increase in mechanical properties of the MDF board was influenced by the resin content (10%, 20% and 30%) with higher strength when more resin was added. The MOR, MOE and IB results from this study were expected and consistent with previous studies by Praveen *et al.* (2019) and Wan Mohd Nazri *et al*. (2019).

TABLE 7. AVERAGE VALUES OF PROPERTIES OF MDF PRODUCED WITH DIFFERENT PALM OIL-BASED PU BINDER CONTENT

Designation of MDF	MOR (N/mm^2)	MOE (N/mm^2)	IB (N/mm^2)	TS at 24 hr (%)	MC at 48 hr (%)
MDF 2.0-3	14.07 ± 1.39[a]	1 618.17 ± 166.56[a]	0.31 ± 0.08[a]	31.83 ± 5.02[a]	11.27 ± 1.58[a]
MDF 2.0-5	16.47 ± 1.98[b]	1 984.87 ± 183.67[b]	0.46 ± 0.07[b]	19.98 ± 4.32[b]	10.61 ± 1.58[b]
MDF 2.0-7	20.23 ± 1.84[c]	2 219.97 ± 203.67[c]	0.57 ± 0.08[c]	11.62 ± 4.18[c]	7.31 ± 1.32[c]
MDF 2.0-10	21.16 ± 2.10[c]	2 281.86 ± 188.04[c]	0.61 ± 0.09[c]	9.81 ± 3.42[c]	6.70 ± 1.05[c]

Note: MOR - modulus of rupture; MOE - modulus of elasticity; IB - internal bonding; TS - thickness swelling and MC - moisture content. Mean followed by the same letters in each column is not significantly different at $p \leq 0.05$ according to the least significant difference Turkey test method.

The physical properties such as TS and MC values of the boards produced at various percentages of palm oil-based PU binder are shown in *Table 7*. The results show that the MDF's TS and MC values decreased with increasing amounts of resin of palm oil-based PU binder from 3% to 10%. The MDF 2.0-10 gave lower TS (9.81%) and MC (6.70%) followed by MDF 2.0-7 with TS (11.62%) and MC (7.31%), MDF 5 with TS (19.98%) and MC (10.61%), and MDF 2.0-3 with TS (31.83%) and MC (11.27%), respectively. The TS and MC results indicated that the presence of hydrophobic PU on the surface of fibres hindered the expansion of compressed fibres after being soaked in water, and also, the board has improved the moisture resistance and promoted its dimensional stability. Other than that, this improvement in TS and MC properties may be due to the fact that the higher resin content provided more bonding between fibres to prevent water absorption. According to Ali *et al*. (2013), the dimensional stability of the MDF board is enhanced by the bonding between the fibres as the resin cures.

According to the ANOVA analysis, the binder content of palm-oil PU binder and MDF's mechanical and physical properties were significantly correlated, at p-value <0.05. This result indicated that there was a significantly impact on the mechanical and physical properties of MDF when the resin content of palm oil-based PU binder was increased from 3% to 10% during the production of MDF. Thus, based on this study the optimum amount of palm oil-based PU binder that meets all BS EN 622-5 requirements (British Standard, 2009) and Dongwha Malaysia (2023) for fibreboard as shown in *Table 4* was MDF prepared with 7% (MDF 2.0-7).

CONCLUSION

This study demonstrated the potential use of PU derived from palm oil as a PU binder in MDF production. From the above study, it was concluded that palm oil-based PU binders can be utilised for producing MDF. In this investigation varying equivalent weight ratios NCO/OH and amount of palm oil-based PU binder were investigated in order to optimise the amount of palm oil-based PU binders on the mechanical and physical properties of MDF. The mechanical properties of MDF (MOR, MOE and IB strength) and physical properties of MDF (TS and MC) were improved by increasing the NCO/OH equivalent weight ratios of palm oil-based PU binder and palm oil-based PU binders content. Satisfactory MDF boards were prepared by using equivalent weight ratio NCO/OH of 2.0 and the minimum amount of palm oil-based PU binder where 7% palm oil-based PU binder meets the requirement with respect to British Standard, BS EN 622-5 and Dongwha Malaysia for fibreboard. The formulations prepared for use as a binder in MDF boards have shown superior properties and allowed noticeable reduction of isocyanate consumption.

The FTIR spectra have confirmed various important peaks for palm oil-based PU binders, rubber wood fibres and MDF. The disappearance in peak intensity of NCO from palm oil-based PU binder as compared to MDF could probably due to the formation of urethane linkages between the reactive NCO active groups in palm oil-based PU binder and OH groups from cellulose constituent in rubber wood fibres thus forming stronger bonding between the PU matrix and rubber wood fibres. Finally, the TGA diagrams of palm oil-based PU binders show two steps of degradation caused by changes in the soft and hard phases of PU. It has been observed that the temperature degradation increases when the equivalent weight ratios of NCO/OH was increased.

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