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Influence of Isocyanate Content and Hot-Pressing Temperatures on the Physical–Mechanical Properties of Particleboard Bonded with a Hybrid Urea–Formaldehyde/Isocyanate Adhesive

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Abstract: Particleboard (PB) is mainly produced using urea–formaldehyde (UF) adhesive. However, the low hydrolytic stability of UF leads to poor water resistance by the PB. This research aimed to analyze the effect of hot-pressing temperatures and the addition of methylene diphenyl diisocyanate (MDI) in UF adhesive on the physical and mechanical properties of PB. The first experiment focused on pressing temperature treatments including 130, 140, 150, and 160 °C. The particles were bonded using a combination of UF and MDI resin at a ratio of 70/30 (%*w/w*). Furthermore, the second experiment focused on UF/MDI ratio treatment, including 100/0, 85/15, 70/30, and 55/45 (%*w/w*), and the particles were pressed at 140°C. All of the single-layer particleboard in this research were produced in 250 × 250 mm, with a target thickness and density of 10 mm and 750 kg/m^3, respectively. This research used 12% resin content based on oven-dry weight wood shaving. The pressing time and pressing pressure were determined to be 10 min and 2.5 N/mm^2, respectively. Before the tests, the board was conditioned for 7 days. When studying the effect of treatment temperature, good physical properties (thickness swelling and water absorption) and mechanical properties (MOR and MOE) were obtained at 140 °C. However, no significant difference was observed in the UF/MDI ratio between 85/15 and 55/45 using the same temperature. The increase in the MDI adhesive ratio improves the MOE and MOR values. However, the internal bond was the contrary. This study suggests that a combination of UF/MDI at a ratio of 85/15 and hot-pressing temperature at 140 °C could produce a PB panel that meets a type 8 particleboard according to the JIS A5908-2003 standard and type P2 according to the EN 312-2010 standard.

Keywords: basic properties; wood shaving; composite; composites materials; adhesive combination

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1. Introduction

Urea–formaldehyde (UF) is an adhesive widely used to manufacture particleboard. An amount greater than 70% of the total UF resin produced is used in the particleboard and medium-density fiberboard industry [1], and its low price is the main reason this adhesive is used. Furthermore, it has disadvantages in terms of low dimensional stability, it is not moisture-resistant, and it has poor durability (biological and weather). Therefore, it is only suitable for interior use. Mansouri et al. [2] and Guru et al. [3] stated that UF adhesive particleboard has low dimensional stability, which has also been supported by other studies [4–10].

The advantages of 4-4 diphenylmethane diisocyanate (MDI)-based resins over UF adhesive include good bonding performance, higher water resistance, aging resistance, and no formaldehyde emission concerns [11]. In addition, the isocyanate group of MDI resins may react with the hydroxyl groups in wood to generate a polyurethane bond, providing direct covalent connections between the adhesive and wood [2]. A number of studies have examined the wood–MDI cure using various techniques, including differential scanning calorimetry (DSC) [12], infrared (IR) spectroscopy [13], and nuclear magnetic resonance (NMR) spectroscopy [14]. In their research, they discovered that wood–MDI cure systems frequently produce biuret, polyuret, and polyurea formations. Only when extremely high doses of MDI were administered, however, were urethane linkages found to form.

Several studies have been conducted on improving composite board quality using UF adhesives. Hybrid resin is one of the studies to improve the performance of UF adhesives. Furthermore, melamine–formaldehyde (MF) has been fortified with melamine urea formaldehyde (MUF) to reduce the weakness of UF adhesives. The UF adhesive was modified by adding isocyanates to improve the thickness swelling properties of the board [11–17]. Mansouri et al. [2] reported that adding a small amount of MDI into the UF improved the performance of the adhesive. This kind of adhesive combination increases the bonding quality of beech (Fagus sylvatica) plywood after immersion in hot (boiling) water. Particleboards made with wheat straw and 4% MDI addition had better mechanical properties and less thickness swelling than resin panels with UF, SPI (soybean protein isolate), and SF (soybean flour). According to [18], three-layer UF-bonded particleboards with 30% waste paper content and pMDI-bonded panels with up to 50% waste paper content in the core layer in terms of mechanical properties meet the requirements of European Standard EN312 for P2-type panels for furniture applications.

Meanwhile, the difference between this study and several others is the application of hybrid adhesive with different ratios during the single-layer board manufacturing process. This is accomplished by spraying the particles individually, with the UF adhesive sprayed first, followed by the MDI adhesive. Iswanto et al. [5] reported that this application technique improved the physical and mechanical properties of particleboards. Therefore, this study aims to analyze the influence of pressing temperature and the ratio of UF and MDI adhesive mixtures on the physical and mechanical properties of particleboards.

2. Material and Methods

2.1. Materials

Sengon (*Paraserianthes falcataria*) wood shavings were obtained from the wood industry in Medan, Indonesia. Furthermore, the commercial UF adhesive (solids content 65.7% and viscosity 210.5 mPa · s) was obtained from PT. Pamolite Adhesive Industry, Probolinggo, Indonesia. The commercial MDI adhesive (solids content 99.5% and viscosity 212.4 mPa · s) was obtained from PT. Polychemie Asia Pacific Permai, Jakarta, Indonesia.

2.2. Characterization of the Hybrid Adhesive Properties

The properties of the hybrid UF/MDI adhesives were determined according to the published methods [19,20]. The viscosity of hybrid UF/MDI adhesives was analyzed using a rotational rheometer (RheolabQC, AntonPaar, Graz, Austria) with a No. 27 spindle at 100 RPM and 25 °C. The gel time of neat UF resins and UF/MDI adhesives was measured at 100 °C using a gel time meter (Techne GT6, Colepalmer, Vernon Hills, IL, USA). The non-volatile solids content of the hybrid UF/MDI adhesives was determined by drying 2 g of the sample in an oven at 105 °C for three hours and dividing the oven-dried weight by the initial weight. Each experiment was repeated in triplicate.

The curing temperature (Tp) of the hybrid UF/MDI adhesives was scanned using differential scanning calorimetry (DSC4000, Perkin Elmer, Hopkinton, MA, USA) from 30 °C to 200 °C with a heating rate of 10 °C/min under 40 mL/min of nitrogen gas. The spectra of hybrid UF/MDI adhesives were also recorded using Fourier transform infrared (FTIR) spectroscopy (SpectrumTwo, PerkinElmer Inc., Hopkinton, MA, USA) with the universal attenuated total reflectance (UATR) method in the range of 400–4000 cm^{-1} at room temperature to detect any changes in the adhesives' functional groups.

2.3. Determination of Slenderness Ratio and Aspect Ratio

One hundred samples of wood shavings were obtained, and the length, width, and thickness were randomly measured. The slenderness ratio (SR) value was determined based on the length and thickness of the particles, while the aspect ratio (AR) value was determined based on the ratio of the width and thickness.

2.4. Particleboard Manufacturing and Testing

The particles in the form of wood shavings were oven-dried at a temperature of 103 ± 2 °C to reach a moisture content of 5%. Furthermore, the adhesive content determined was 12% based on the dry weight of the particles. Single-layer particleboards were produced with a nominal density of 750 kg/m^3 and dimensions of 250 × 250 × 10 mm. The boards were made using a specific pressure of 2.5 N/mm^2 and a pressing time of 10 min. The treatment in the manufacture of the boards was divided into two stages, namely: the influence of pressing temperature on board properties and the influence of the UF/MDI ratio on board properties. After the pressing process, the boards were conditioned for 7 days at ambient temperature. The test sample was cut before testing after the board conditioning process, and it was consistent with the JIS A5908-2003 and EN 312-2011 standards. The samples were air-conditioned under standard defined humidity and temperature conditions until a constant weight was achieved. The test parameters included the physical and mechanical properties of the board. The physical properties include the density, moisture content, thickness swelling, and water absorption. Meanwhile, the mechanical properties include the modulus of rupture and the modulus of elasticity in bending regarding EN 310 and internal bond regarding EN 319.

2.4.1. Influence of Pressing Temperature on Board Properties

In the first stage of the study, the PB was manufactured at different hot-pressing temperatures including 130, 140, 150, and 160 °C. The adhesive used was a mixture of UF and MDI with a ratio of 70/30 UF/MDI (%*w*/*w*) based on the determined content of 12%. Furthermore, the use of adhesive content of 12% refers to Iswanto et al. [5]. The application was conducted by spraying UF and MDI adhesive separately.

2.4.2. Influence of UF/MDI Ratio on Board Properties

At this stage, the board manufacturing process was treated as a mixture ratio of UF and MDI adhesives consisting of 100/0, 85/15, 70/30, and 55/45 (%*w*/*w*) based on the determined adhesive content of 12%. The pressing temperature used was the best in the first stage of the study, which was 140 °C with the same pressing pressure and time as the previous stage. The application was conducted by spraying UF and MDI adhesive separately.

2.5. Scanning Electron Microscopy (SEM) Analysis

The morphological observation of the particleboard surface area was conducted by scanning electron microscopy (SEM) JSM-6360 (JEOL Ltd., Tokyo, Japan). Previous imaging demonstrated that the sample surface was coated with an 80 nm gold layer using a sputter coater and then it operated at an accelerating voltage of 15 kV with a magnification of 500×.

2.6. Data Analysis

A non-factorial, completely randomized design was used, and the first stage of treatment was pressing temperatures consisting of 130, 140, 150, and 160 °C. Furthermore, the second stage of treatment was in the form of a comparison of UF/MDI adhesives consisting of 100/0, 85/15, 70/30, and 55/45 (%*w*/*w*), and the number of board repetitions for each treatment was three replications.

3. Result and Discussion

3.1. Properties of the Hybrid UF/MDI Adhesives

The characteristics of UF/MDI adhesives at various ratios are shown in Table 1. The basic characteristics including non-volatile solids content, gel duration, and viscosity influence the performance of the adhesives in wood-based panels. In general, the solids content and viscosity of UF/MDI adhesives increased as a function of MDI content while the gel time decreased. The increase in solids content is related to the addition of the MDI, which has a solids content of 99.5%. The high solids content of MDI indicates a high content of active materials for bonding, resulting in the more excellent adhesion and cohesion strength of the MDI adhesive [11–14]. Furthermore, the presence of MDI in UF resins markedly increased the viscosity of hybrid adhesives. The increase in viscosity is probably because of the reaction between –NCO groups and –CH_2OH groups of the UF [21]. As a result, the gel time of the UF/MDI hybrid adhesive decreased with a higher MDI content, which in practice means that a higher MDI content makes the life of the UF/MDI hybrid adhesive shorter.

Table 1. Basic properties of hybrid UF/MDI adhesives.

Properties	UF/MDI Ratio (%*w*/*w*)			
	100/0	85/15	70/30	55/45
Non-volatile solids content (%)	65.7 ± 0.32	67.4 ± 0.23	69.3 ± 0.24	70.6 ± 0.34
Viscosity (mPa · s)	210.5 ± 6.48	230.4 ± 3.91	275.7 ± 8.68	332.4 ± 6.98
Gel time (s)	202.0 ± 2.88	195.6 ± 2.89	180.2 ± 4.04	165.8 ± 3.21

The value after ± indicates the standard deviation.

The DSC analysis showed the curing temperature (Tp) of the hybrid UF/MDI adhesives as an exothermic reaction (arrow) (Figure 1a). The neat UF adhesive with 0% MDI had a Tp around 136.2 °C. Regardless of the MDI content, incorporating MDI into the UF adhesive decreased the Tp to 96.8 to 123.5 °C (asterisk). The decrease in Tp was probably because of the reaction between the –CH_2OH groups of the UF and the –NCO groups of the MDI to form urethane bonds [21]. The –NCO groups of MDI are known to have greater reactivity than other adhesive functional groups [12,14]. The ATR-FTIR spectra of hybrid UF/MDI adhesives are displayed in Figure 1b. The spectra reveal several specific functional groups, such as free –NCO groups and the C–C aromatic of MDI that were observed at 2250 cm^{-1} and 1525 cm^{-1}, respectively, while the neat UF adhesive had C=O groups at 1650 cm^{-1}. Regardless of the MDI content, adding MDI into the UF adhesive increased the intensity of N–H at 3300 cm^{-1} and C–H at 2948 cm^{-1} and 2880 cm^{-1}. The addition of MDI into the UF adhesive formed urethane linkages at 1730 cm^{-1} due to the reaction between the –CH_2OH groups of the UF and the –NCO groups of the MDI. The hybrid adhesives containing higher MDI content had –NCO groups at wavenumber 2270 cm^{-1} from the excess MDI. The free –NCO groups could further react with the –OH of wood [12–14].

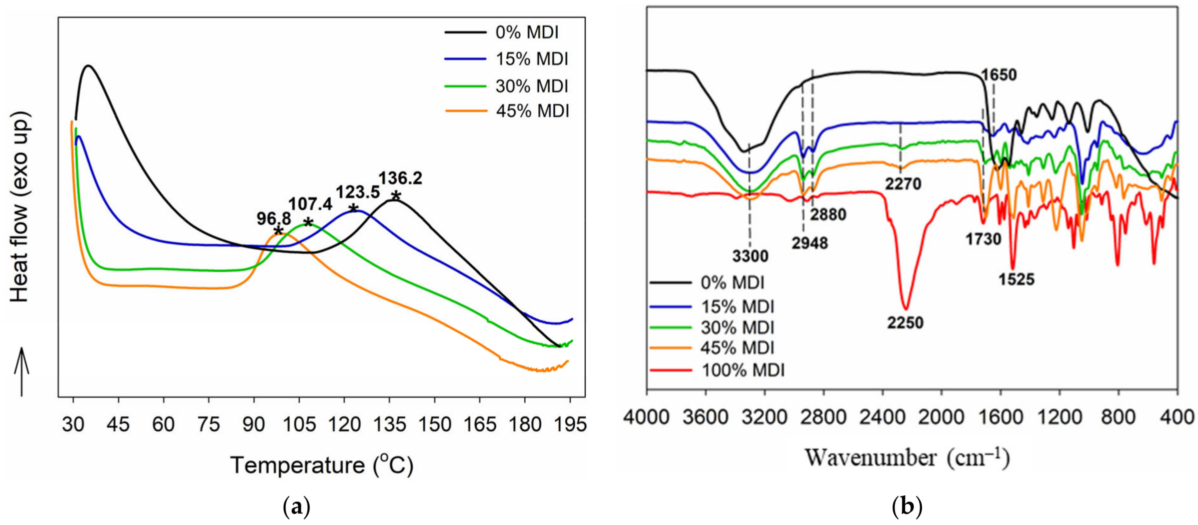


Figure 1. Characteristics of the UF resin and the UF/MDI hybrid adhesives (**a**) DSC thermogram of the hybrid adhesives and (**b**) ATR-FTIR spectra of the hybrid adhesives.

3.2. Slenderness Ratio and Aspect Ratio of Particles

The distribution of SR and AR particles and the calculation results of these two values are displayed in Figure 2. The results indicate that the SR value in wood shavings is dominated in 20–30, while the AR is still below 2. The SR value is included in the low category, and Maloney [21] stated that the ideal SR and AR values are 150 and 3, respectively. The low SR value is one of the causes of the low bending value of the resulting board. According to Maloney [21], particle geometry is one factor that influences the panel modulus of rupture value. This particle geometry deals with size, slenderness, and aspect ratio. The particles with a high slenderness ratio will be more accessible to orient since the resulting board strength will increase and need less adhesive per surface area to bond the particles.

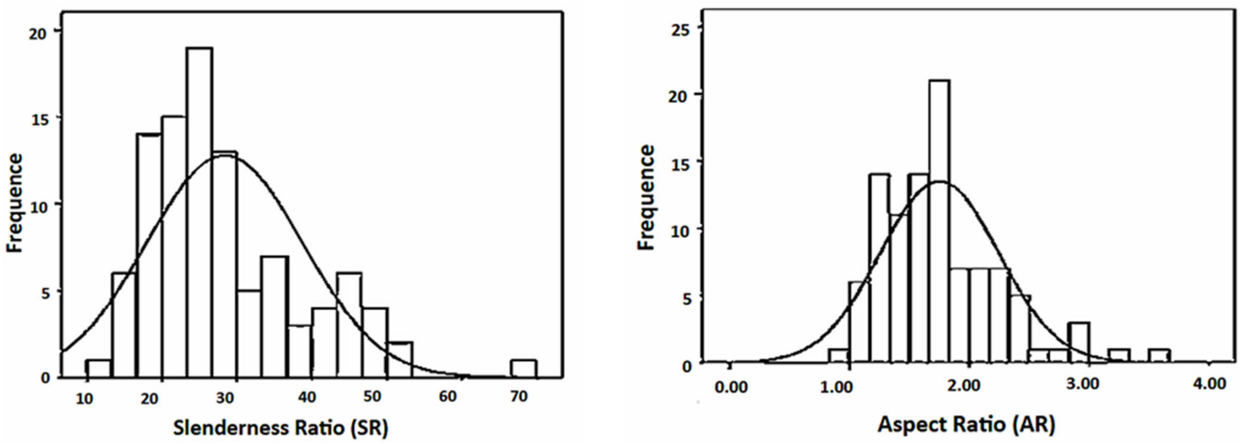


Figure 2. The distribution of the particles' slenderness ratio and aspect ratio.

3.3. Density

Figure 3 shows that the board density value for the pressing temperature treatment ranges from 545–689 kg/m^3. The board produced the lowest and highest density value with a pressing temperature treatment of 140 °C and 150 °C. In addition, the density values ranged from 545–640 kg/m^3 at different UF/MDI ratios. The lowest and highest were obtained at UF/MDI ratios of 70/30 and 100/0. The resulting board density values for the treatments, pressing temperatures, and the overall UF/MDI ratio were still below the target. This is because the particles were removed during the board manufacturing process. Bufalino et al. [22] stated that the low-density value was caused by the loss of particles during the manufacturing process. Similarly, Kelly [23] stated that the factors that influence the board density value include the type of wood (wood density), the amount of pressing pressure, the number of wood particles in the plinth, the adhesive content, and other additives.

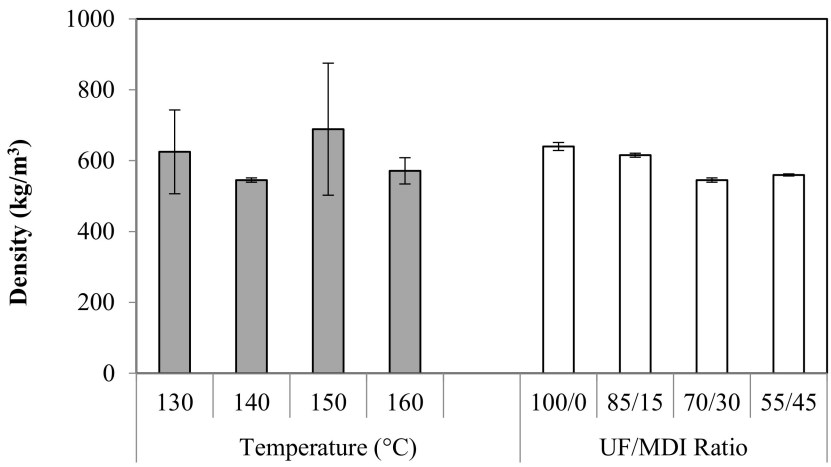


Figure 3. The density of the particleboard. The bars indicate the standard deviation.

The statistical analysis showed that at the 95% confidence interval, the pressing temperature treatment did not significantly affect the density parameter. Meanwhile, the UF/MDI ratio treatment gave a significantly different effect. The final board's density value complies with JIS A 5908-2003 standard requirements, which range from 400 to 900 kg/m^3 [24] and EN 312-2010, whose requirements indicate only ±10% tolerance on the mean density within a board [25].

3.4. Moisture Content

Figure 4 shows that the value of the board's moisture content for the pressing temperature treatment ranges from 5.5 to 6.6%. The particleboard produces the lowest and highest moisture content values with a pressing temperature treatment of 160 °C and 130 °C, respectively. The pressing process at a higher temperature causes a greater decrease in the moisture content of the resulting board. Meanwhile, the temperature of 160 °C showed a drastic decrease in moisture content. This was presumably due to the influence of lignin melting at that temperature. This can lead to a partial closure of the cell cavities in the wood particles and may result in limited water and water vapor accessibility. Ferra et al. [26] stated that the pressing temperature would influence changes in chemical components, such as the liquefaction of lignin. Furthermore, Iswanto et al. [27] reported that pressing at higher temperatures on sorghum bagasse particleboard and jatropha rind produced a lower response to moisture content.

The moisture content value for the treatment of the UF/MDI ratio ranged from 5.8 to 6.5%, where the lowest and highest values were obtained on the boards with a ratio of 100/0 and 55/45, respectively. Figure 3 shows that slightly increasing the MDI adhesive ratio causes an increase in the moisture content value. The MDI proportion and the board density value are inversely related when viewed from the board density value. Furthermore, the low-density value contributed to the increase in the board's moisture content. The statistical analysis showed that at the 95% confidence interval, the pressing temperature treatment and the UF/MDI ratio significantly differed in moisture content parameters. Generally, the moisture content of the resulting boards meets the requirements of JIS A 5908-2003 and EN 312-2010 standards, which specify a moisture content in the range of 5–13% [21,24].

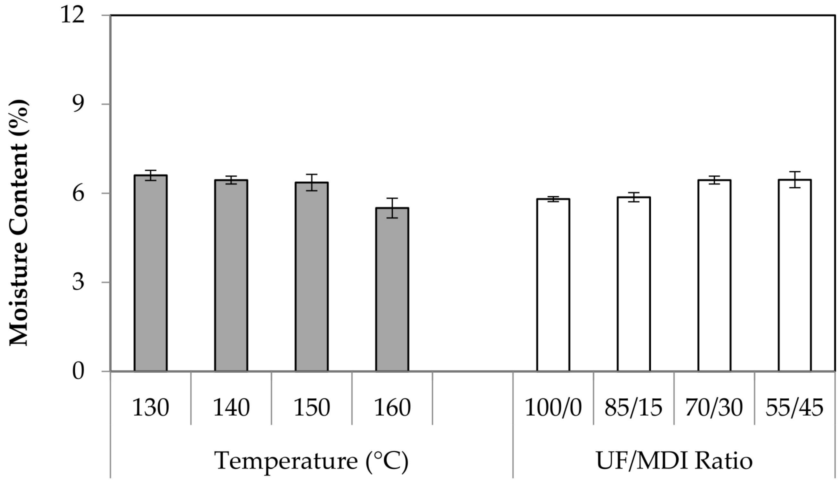


Figure 4. Moisture content of the particleboard. The bars indicate the standard deviation.

3.5. Water Absorption

Figure 5 shows that the water absorption value of the board for the pressing temperature treatment ranged from 46.1 to 61.6%. The board produced the lowest and highest water absorption values with a pressing temperature treatment of 140 °C and 160 °C, respectively. The temperature and water absorption capacity of the board is directly proportional. In the water absorption test, the board was immersed in water at a temperature 20 ± 1 °C for 24 h. Therefore, the water absorption ability is largely determined by the adhesives' performance. In the pressing temperature treatment, the particleboard uses a mixture of UF and MDI adhesives with a ratio of 70/30, where the dominance is on the UF adhesive. As previously reported, the ideal UF adhesive works at low temperatures. Therefore, over-curing is impacted when the temperature increases during the same pressing period, which influences the decreased adhesive ability. This is evidenced in the parameters of the internal bonding of the boards produced for the treatment at a temperature of 160 °C, which has the lowest value. Winandy and Krzysik [28] reported that increased pressing time and temperature did not hinder the ability to absorb water from the panels.

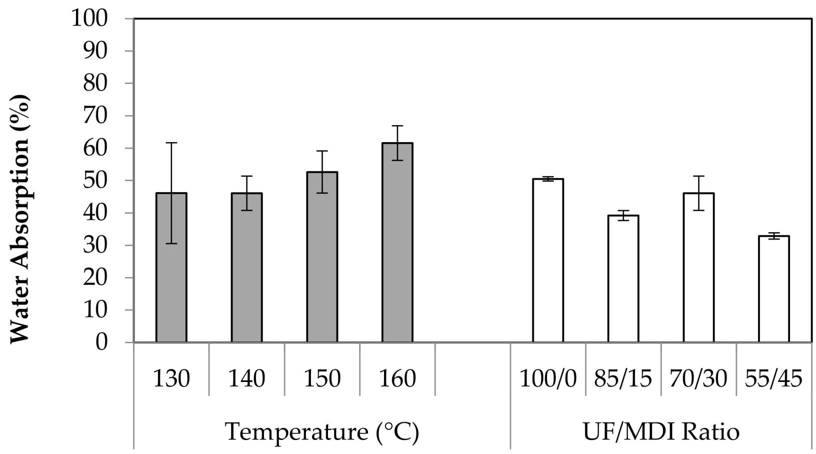


Figure 5. Water absorption of the particleboard. The bars indicate the standard deviation.

Meanwhile, for the treatment of the UF/MDI ratio, the water absorption value ranged from 32.9–50.5%, where the lowest and highest values were obtained on the boards with a ratio of 55/45 and 100/0, respectively. The MDI adhesive ratio and water absorption value of the board are inversely proportional. This is consistent with the trend in the development of board thickness. The statistical analysis showed that at the 95% confidence interval, the pressing temperature treatment did not have a significantly different influence on the water absorption parameters. Meanwhile, the UF/MDI ratio treatment produced a significantly different influence.

3.6. Thickness Swelling

Figure 6 shows that the value of the board thickness swelling for the pressing temperature treatment ranged from 12.1–19.9%. The board produced the lowest and the highest thickness swelling values with a pressing temperature treatment of 140 °C and 160 °C, respectively. An increase in temperature causes a higher thickness swelling value. The UF adhesive underwent over-curing at a higher temperature for the same pressing period. In the hot pressing process of particleboard manufacturing with the UF adhesive, several studies used temperatures varying from 120 °C to 160 °C with time variations between 4–10 min [29–33].

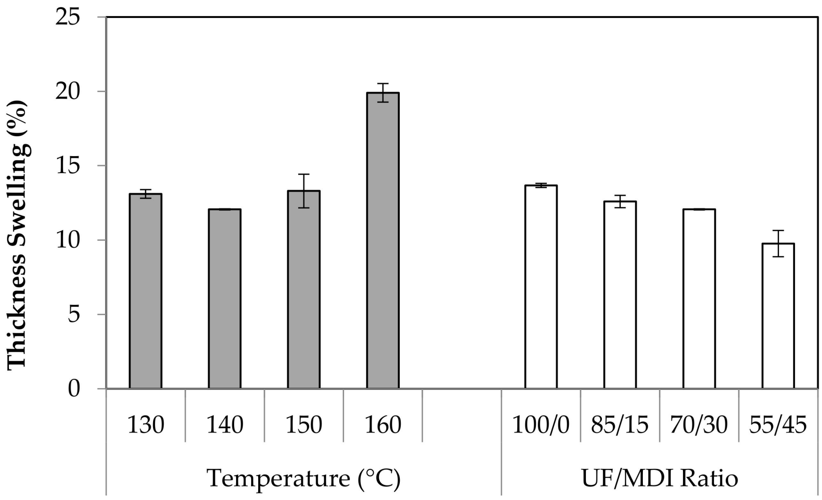


Figure 6. Thickness swelling of the particleboard. The bars indicate the standard deviation.

For the treatment of the UF/MDI ratio, the thickness swelling value ranged from 9.8 to 13.7%, where the lowest and highest values were obtained on boards with a ratio of 55/45 and 100/0, respectively. Figure 6 showed that the MDI adhesive ratio and the thickness swelling value were inversely related. The MDI adhesive can bond chemically. It is stronger than other exterior adhesives, such as PF, which only have mechanical bonding capabilities. Furthermore, Veigel [34] and Mara [35] stated that there is a chemical bond between the MDI adhesive and lignocellulosic material. It produces higher strength and is more stable than mechanical bonds such as PF and UF adhesives. Isocyanates react chemically with hydroxyl groups to form urethane linkages between wood particles [36]. The combination of nonpolar and aromatic compounds from MDI produces resistance to hydrolysis reactions. In addition, isocyanates react physically with the water contained in the wood to form polyurethane. The MDI adhesive usually penetrates the wood surface to a depth of 1 mm [37]. The MDI should penetrate at least 0.3 mm for good adhesion to wood, and the penetration capability results in suitable thickness swelling properties.

The statistical analysis showed that at the 95% confidence interval, the pressing temperature treatment and the UF/MDI ratio produced significantly different effects on the parameters of the thickness of the board. Generally, the resulting thickness swelling value does not meet the JIS A 5908-2003 standard, except for boards with a UF/MDI ratio of 55/45. In contrast, according to EN 312-2010 standard, each board except that obtained by treatment at a pressing temperature of 160 °C meets the requirements (max. 17%) and, due to the value of swelling thickness, is qualified as type P3, i.e., non-load-bearing boards for use in humid conditions [25].

3.7. Modulus of Rupture and Modulus of Elasticity

Figure 7 shows the board's modulus of rupture (MOR) and modulus of elasticity. The MOR value for the pressing temperature treatment ranged from 4.9–8.3 N/mm^2, where the board produced the lowest and highest values with a pressing temperature treatment of 130 °C and 140 °C, respectively. The board's strength decreased at temperatures above 140 °C because the UF adhesive was over-cured at high temperatures. Paridah et al. [38] stated that the adhesive's polymerization rate would increase or decrease depending on the raw materials (wood and adhesive) used. This will directly influence the temperature and pressing time in particleboard manufacturing. The conduction of hot pressing for a long time can affect the over-curing of adhesives, and the strength may be negatively influenced [39].

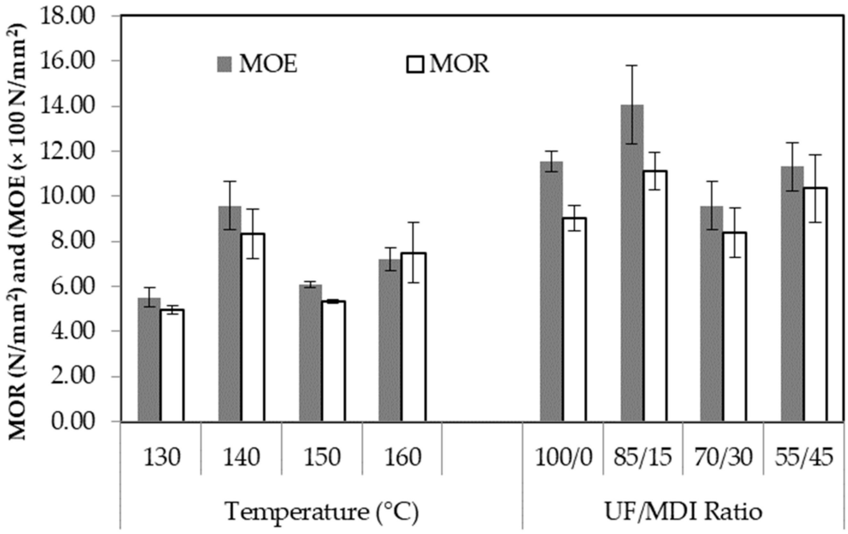


Figure 7. Modulus of rupture of the particleboard. The bars indicate the standard deviation.

For the treatment of the UF/MDI ratio, the MOR values ranged from 8.3–11.1 N/mm^2, where the lowest and highest values were obtained on boards with a ratio of 70/30 and 85/15, respectively. The MDI adhesive ratio and the MOR value of the board are inversely related. The statistical analysis showed that at the 95% confidence interval, the pressing temperature treatment and the UF/MDI ratio produced significantly different influences on the MOR parameters of the board. The pressing temperature treatment of 140 °C resulted in a MOR value consistent with the JIS A 5908-2003 standard. For the UF/MDI ratio, all of the boards produced were consistent with the standard whereby JIS A 5908-2003 requires a minimum MOR value of 8 N/mm^2 [24]. In terms of the EN 312-2010 standard, the board produced with a UF/MDI ratio of 85/15, whose MOR is 12.6 N/mm^2, meets the minimum requirements (11 N/mm^2) for board type P2, i.e., boards for interior fitments (including furniture) for use in dry conditions, and the board with a ratio of 55/45 for board type 1 (10.5 N/mm^2) [25].

The board's modulus of elasticity (MOE) for pressing temperature treatment ranges from 550–959 N/mm^2. The board produced the lowest and highest MOE values with a pressing temperature treatment of 130 °C and 140 °C, respectively. Meanwhile, for the treatment of the UF/MDI ratio, the MOE values ranged from 959–1406 N/mm^2. The lowest and highest values were obtained on boards with a ratio of 70/30 and 85/15, respectively. According to Maloney [21], various factors influence the MOE value, including the resin type, resin content, adhesive bond, and fiber length. The statistical analysis showed that at the 95% confidence interval, the pressing temperature treatment and the UF/MDI ratio significantly differed from the board's MOE parameters. Furthermore, the treatment did not produce boards that meet both standards as the JIS A 5908-2003 standard requires a minimum board MOE value of 2000 N/mm^2 [24] and the EN 312-2010 standard requires a minimum of 1800 N/mm^2 (type P2) [25].

3.8. Internal Bond

Figure 8 shows that the internal bond value of the board for the pressing temperature treatment ranged from 0.07 to 0.6 N/mm^2. The board produced the lowest and highest internal bond values with a pressing temperature treatment of 160 °C and 140 °C, respectively. Forging at a temperature of 140 °C was the optimal condition in this study and therefore, the bond between the UF/MDI mixed adhesive at a ratio of 70/30 with wood shavings particles. Temperatures below and above 140 °C are suspected of causing pre-curing and over-curing, respectively. Both of these conditions reduce the value of the board's internal ties. Nemli [40] states that increasing pressure, pressing temperature, time, and adhesive content are directly proportional to IB. This is closely related to resin maturation, decreased particle wettability, and increased surface area [40]. Furthermore, Ferra et al. [26] reported that the differences in the characteristics of IB using 10% UF adhesive at five different temperatures were explained by two approaches: (1) the temperature influence the UF bond on wood. It facilitates the movement of fluids in the wood and is accompanied by an accelerated diffusion of resin molecules. The low temperature decreases the resin dispersion in the wood, resulting in a decrease in mechanical interlocking. (2) The pressing temperature influences chemical substrate modifications such as lignin fusion and hydrogen bonds associated with increasing the strength values. Low temperatures can inhibit the mobility of the hydroxyl groups of the polymer. This will prevent the conversion of the methyl ether bridge into a methylene bridge and decrease the bond strength's value.

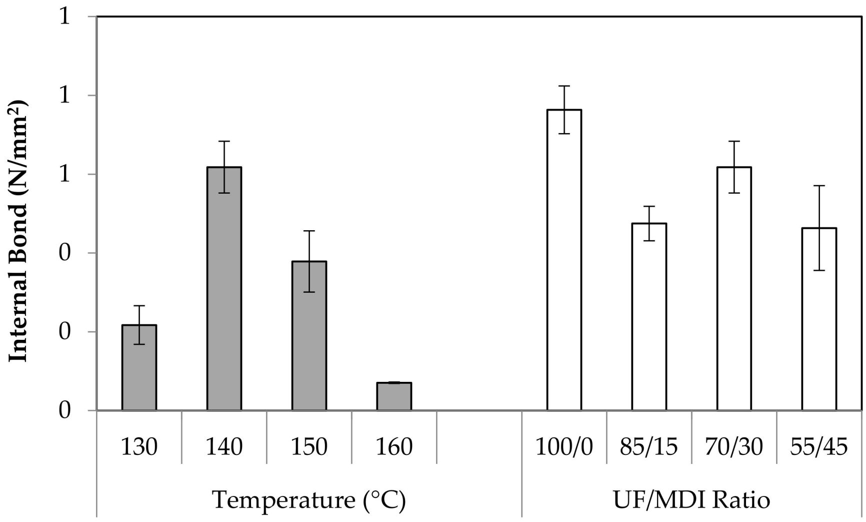


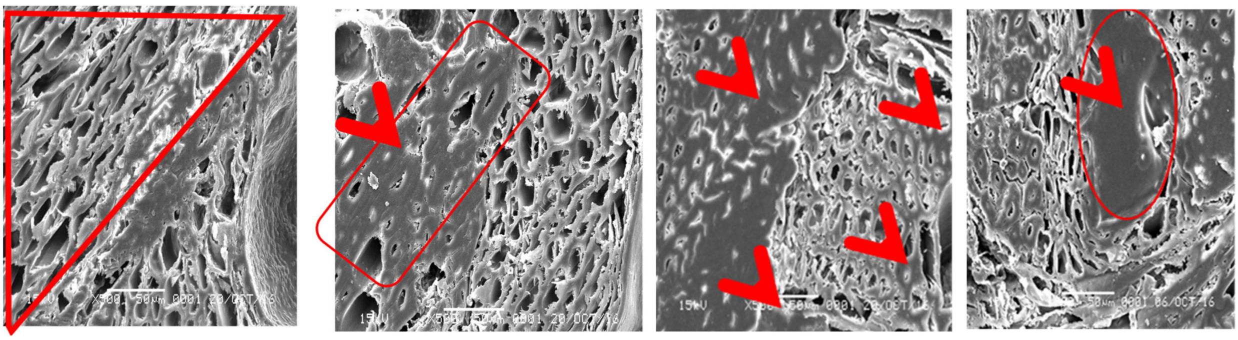
Figure 8. Internal bond of the particleboard. The bars indicate the standard deviation.

For the treatment of the UF/MDI ratio, the internal bond values ranged from 0.5–0.8 N/mm^2. The lowest and highest values were obtained on boards with a ratio of 55/45 and 100/0, respectively. The presence of the MDI adhesive resulted in a lower internal bond value when compared to that without the MDI mixture. This was due to the low moisture content of the particles used to accommodate the UF adhesive. This study was conducted using a moisture content of 5% particles.

Furthermore, the MDI performance will be better in sufficient water. This is because there will be a reaction with molecules containing active hydrogen to produce basic polyurethane and polyurea molecules. Active hydrogen sources can bind hydroxyl groups in wood, wood extractives, or wood resins and their moisture content. Wood has a chemical functional group known as a hydroxyl group. Meanwhile, the MDI on the isocyanate group (–N=C=O) reacts with the hydroxyl group to form a urethane chain. A combination of nonpolar factors and MDI aromatic components are resistant to hydrolysis [35]. The statistical analysis showed that at the 95% confidence interval, the two treatments produced significantly different influences on the internal bond parameters. Most of the IBs were consistent with the JIS A 5908-2003 standard which requires a minimum value of 0.15 N/mm^2 [24] except for the pressing temperature treatment of 160 °C. In terms of the EN 312-2010 standard, all of the boards manufactured in the UF/MDI ratio and the boards with the pressing temperature treatment of 140 °C meet the minimum IB requirements of 0.45 N/mm^2 for the board type P5, i.e., load-bearing for use in humid conditions, and the board in the 100/0 ratio even type P7 (heavy duty load-bearing boards for use in humid conditions)—0.75 N/mm^2 [25].

3.9. Scanning Electron Microscopy Analysis

The morphology of the particleboard surface using UF adhesive and UF/MDI adhesive combination was observed by scanning electron microscopy (SEM). The UF/MDI ratios observed in this research were 100/0, 85/15, and 70/30 (Figure 9). The hot-pressing process resulted in densification on the cell wall of the wood particles as shown in the triangle area in Figure 9a. This was described as the cells being flattened. The particleboard became more compact, and the density tended to increase. The particleboard bonded by UF only showed that UF was not distributed evenly as shown in the rectangle area and narrow in Figure 9b. A more even distribution of MDI in particles can be seen in Figure 9c,d. This analysis proved that adding MDI in a UF/MDI adhesive combination was required to create a more even distribution of adhesive by filling the cell cavities among the particles. It was concluded that the addition of MDI adhesive in the manufacture of a particleboard UF/MDI adhesive combination was able to improve its dimensional stability and bending properties.



(**a**) (**b**) (**c**) (**d**)

Figure 9. SEM analysis of the particleboards: (**a**) densification on the cell wall of wood particles and (**b**) UF/MDI adhesive combination ratios 100/0, (**c**) 85/15, and (**d**) 70/30 in 500× magnification.

4. Conclusions

In summary, the increased pressing temperature resulted in increased thickness swelling and water absorption. The resulting swelling thickness value ranges from 12.1–19.9% On the contrary, the MOE, MOR, and internal bond values decreased. The increase in the MDI ratio on the UF/MDI adhesive combination successfully improved the water absorption, thickness swelling, and modulus of rupture (MOR) values of the particleboards. Compared to several related studies, it was shown that the presence of the MDI adhesive in the UF/MDI adhesive combination showed an improvement in thickness swelling, MOE, MOR, and IB values. However, in this study, the IB value produced decreased. The optimum temperature to obtain the particleboard's physical and mechanical properties was 140 °C. The UF/MDI ratio of 85/15 was determined for an optimum combination at this temperature. The increasing ratio of MDI in the UF/MDI adhesive combination resulted in a more even distribution of adhesives. Consequently, the particleboard's water absorption, thickness swelling, and MOR properties were improved.

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