

Article

Potential of Using Natural and Synthetic Binder in Wood Composites

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Abstract: The physical and mechanical properties of particleboard bonded with different cooking percentages of seaweed (*Kappaphycus alvarezii*), with different percentages of starch and different percentages of adhesives, and wood plastic composite (WPC) from High-Density Polyethylene (HDPE) with *Acacia mangium* wood powder were determined in this study. Seaweed mixed with different percentages of sulfuric acid (30%, 50%, 70%) and Sodium Hydroxide (70%, 50%, 30%) were prepared as a binder for particleboard. For a starch-based binder, different percentages of starch (10%, 15%, 20%) were prepared, before producing particleboard with different amounts of binder (20%, 25%, 30%). As for WPC, wood powders were bonded with different percentages of HDPE content (70%, 80%, 90%). Results indicated that WPC at 90% HDPE shows the best performance in the water absorption (0.07%) and thickness swelling test (2.54%). Starch-based particleboard recorded the highest Modulus of Elasticity (MOE) value (1115.07 N/mm² at 15% starch with 30% amount of binder), while WPC (90% HDPE) and starch-based particleboard (10% starch with 20% amount of binder) both recorded the highest Modulus of Rupture (MOR) at the same value, which is 7.84 N/mm². Starch-based particleboard has a better internal bond, which is 0.05 N/mm². However, seaweed-based particleboard has a higher density value, which is 0.6 g/cm³.

Keywords: recycle; renewable; wood composite; particleboard; seaweed; wood plastic composite; starch; high-density polyethylene; *Acacia mangium*; *Kappaphycus alvarezii*



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1. Introduction

Wood can be defined as a complex biological structure, a composite of many chemistries and cell types acting together to serve the needs of a living plant [1]. The formation of wood is by a variety of plants, including many that do not attain tree stature [2]. A tree is generally defined as a woody plant of 4–6 m or more in height and characterized by a single trunk rather than several stems. Produced by many botanical species, including both gymnosperms and angiosperms, wood is available in various colors and grain patterns. Wood is known as the principal strengthening and nutrient-conducting tissue of trees and other plants. It is one of the most abundant and versatile natural materials in the world [3]. It is also strong in relation to its weight, is insulating to heat and electricity, and has desirable acoustic properties. Furthermore, it imparts a feeling of “warmth” not possessed by competing materials, such as metals or stone, and it is relatively easily worked.

Although solid wood materials were popular for decades, the inconsistency of natural wood properties, which varies even between trees of the same species and the same trees, had limited the efficiency of wood material in applications. Therefore, wood composites have risen to prominence in the wood processing industry. Generally, wood composite, also known as engineered wood product, can be classified into a wide range of products, such as laminated beams to fiberboard [4]. In terms of application, wood-based composites are used both structurally and non-structurally, from panels for interior design purposes

to panels for exterior uses such as support structures in building [4]. The wood-based panels are also becoming an integral part of the wood products market, as these panels are widely used in construction of furniture, cabinets, and joinery, as well as their ability to be “engineered” in order to meet specific performances and requirements.

Wood composites comprises a wide range of products including plywood, laminated veneer lumber (LVL), parallel-strand lumber (PSL), glulam, laminated wood-non-wood composites, fiberboard, particleboard, oriented strandboard (OSB), WPC, etc., which can be produced from the breakdown of wood with defects, small diameter timbers, invasive species, as well as wood residues from small trees [4,5]. Many research works have been completed to produce wood composites, also known as “green composites”, from wood waste materials, such as recycled fibers and particles [6–8]. However, it is important that the recycled wood waste used as composite materials is free from preservatives, as they release hazardous compounds into the environment, especially when in contact with fire [9]. Adhesives/binders for wood composites are mainly produced from petroleum-based polymers. The main wood binders used in the bonding of wood composites are phenol-formaldehyde, urea-formaldehyde, and melamine formaldehyde. These are commonly utilized wood binders which possess high mechanical and stable physical properties, which accounts for approximately 70% of the world demand for formaldehyde, as reported by [10]. However, due to increasing awareness that these synthetic resins are a threat to human health and causes pollution to the environment, the acceptable level of these formaldehyde resins have gradually reduced over the years. The emission of formaldehyde from finished wood composite products are connected to the hazardous volatile organic compounds (VOCs) and it was classified as “carcinogenic to human” in 2004 [7,11]. Since then, a significant number of studies has been carried out by using agricultural crops, which includes corn, tapioca, cassava, wheat, rice, and others, which are available in large quantities [12].

Bio-based binders are one of the biodegradable polymers that are derived from natural polymers, such as starch, protein, and cellulose. However, these polymers cannot form an adhesive or act as binder alone due to their hydrophilic nature. In other words, these biodegradable adhesives are predominantly water soluble, which limits their outdoor applications or use in a moisture-rich environment [13]. The binder form has limited long-term stability, which is affected by the water absorption, ageing-caused retrogradation, poor mechanical properties, and low processability [14]. Therefore, plasticizers such as glycerol, vinegar, and water are added to enhance the binding properties and to decrease the water uptake problems of bio-based adhesive [12,15]. Apart from that, natural polymers such as tannins, lignin, and carbohydrates are also one of the popular ingredients for bio-based binders, which has been extensively discussed by Pizzi et al. [16]. The authors also reviewed the use of other pioneer biomaterials for binders, such as cashew nutshell liquid, vegetable oils, fungal mycelium bonding, and even wood friction welding techniques, which have yet to be utilized in the industry [16].

Seaweed is known as multicellular, macroscopic, and marine algae, which can be classified into three groups, i.e., Rhodophyceae, Chlorophyceae, and Phaeophyceae [17,18]. Seaweed gum has potential industrial uses as a binder [18]. *Kappaphycus alvarezii*, which is from the family of Rhodophyceae, is cultivated as main source of raw material for the carrageenan industry [19]. In addition, it is a suitable glue material and alternative for urea formaldehyde resin for wood composites, due to its environmentally friendly properties. Therefore, an alternative to urea formaldehyde as binder in wood composite industry is required to make ‘eco-friendly’ product that causes no harm to humans—*Kappaphycus alvarezii* adhesive is the alternative.

Wood plastic composites (WPCs) are composite materials made from wood which usually come in different forms, and mixed with recycled plastic materials, which act as thermoplastic resin. This material is still new in the family of composite materials, where natural fiber or filler, such as wood powder or wood fiber, is mixed with a thermoplastic resin, such as high-density polyethylene (HDPE), polyethylene terephthalate (PET),

polypropylene (PP), poly (vinyl chloride) (PVC), or polyethylene (PE). Natural fiber or filler has a lower density, less abrasiveness, and lower cost compared to the ancient synthetic fillers. Furthermore, WPCs are renewable and biodegradable. Around 100 companies worldwide have been identified to be involved in WPC manufacturing [20].

WPCs are the newest composite products that manufactured by the mixture of wood fiber or wood powder together with thermoplastics polymer, such as recycled or virgin plastics. Wood plastic composite usually contain 50 to 60% of wood and the balance is plastics. As the wood component is cheaper, the WPC products were continually refined that contain higher proportional of woods. In Europe, there is at least one manufacturer that uses a four-to-one wood-to-plastic ratio. Successful products include molding, millwork, flooring, and curved furniture or automobile parts.

Nowadays, solid waste recycling is done due to concerns about the environment. Nowadays, many industries are becoming interested in these WPCs, as there are many products that are manufactured with plastics. We know that ancient plastics are very hard and not easily decomposed or degraded in surrounding. Research had been performed to prove that plastics will never get degraded and will remain in soil for many years. Polymers need hundreds of years to decompose, yet degrade in normal environmental conditions [21].

As the solution to reduce the amount of waste plastic materials and wood waste, an experiment to produce wood plastic composite using waste plastic materials and wood waste, where wood powder is used as the natural filler and HDPE as the thermoplastic resin must be conducted. HDPE is used as resin to replace the usage of UF as the UF release formaldehyde gases which are harmful to our health and wood waste from *Acacia mangium* species were collected and processed into wood powder, as *Acacia mangium* is a fast-growing species and easily found. Wood products act as an important bioproducts in storing carbon dioxide. A study carried out by Kazulis et al. [22] found that a cubic meter of particleboard and OSB circumvent 720 kg of carbon dioxide, whereas a cubic meter of MDF saves 820 kg of carbon dioxide.

This study presents the physical and mechanical properties of particleboard bonded with different cooking percentages of seaweed (*Kappaphycus alvarezii*), as well as particleboard bonded with different percentages of starch with different percentages of adhesive and wood plastic composite (WPC) using High-Density Polyethylene (HDPE) and *Acacia mangium* wood powder.

2. Materials and Methods

2.1. Production of Wood Particles

Plantation species *Acacia mangium* was utilized in this study, which was obtained and processed into wood particles. Firstly, wood chips were formed from debarked logs or sawn timbers at various sizes through FOMA/BX wood-chipper machine. Then, laboratory scale machineries such as the BX466 knife-ring flaker were used to obtain wood flakes, and lastly, vibrator screener machine was used to obtain proper sizes of *Acacia mangium* wood particles ranging between 1.25 mm to 3.15 mm thick.

2.2. Seaweed Binder Particleboard

The process began with the cooking of seaweed was cut into length ranging from 0.5 cm to 2 cm, weighed 300 g and placed inside a pot. After that, 100 mL of mixed solution at various amount of 1 M NaOH and H₂SO₄ solution were poured inside the pot, in accordance with percentage ratio shown in Table 1, whereby 100 mL was equal to 100%.

In the production of seaweed-based binder particleboard, liquid sodium hydroxide was prepared. In total, 1 M 500 mL solution for both sodium hydroxide (NaOH) with 95% purity and sulfuric acid (H₂SO₄) with 95% purity were prepared as the seaweed cooking solution [23]. Sodium hydroxide in solid form weighed 4.21 g, using the weighing balance, was poured inside 100 mL distilled water, and was stirred in order to prepare sodium hydroxide 1 M 100 mL solution. The seaweed was cooked (80 °C for 30–60 min) until the

formation of gel can be observed [24]. The solution was stirred continuously during the cooking session until the seaweed completely transformed into liquid form.

Table 1. Volume (mL) of mixed chemical solution for different chemical cooking percentages.

Seaweed Chemical Cooking Percentage	Volume of Chemical Solution (mL)	
	1 M Sulfuric Acid (H ₂ SO ₄)	1 M Sodium Hydroxide (NaOH)
S 30:70	30	70
S 50:50	50	50
S 70:30	70	30

Seaweed is insoluble in cold water, but it dissolved faster in hot boiling water due to its thermoplastic properties, which had the ability to be retransformed into liquid form even after turning into solid by reheating the material. Four different types of glue blending were shown in Table 2, whereby a mixture of *Acacia mangium* wood particles, urea formaldehyde (UF), and 2% ammonium chloride as hardener were blended.

Table 2. Four types of seaweed binder with modifications using acid and alkali.

Type of Binders	Modification
100% UF (control)	100% Urea Formaldehyde (100% UF) + Wood Particles + 2% ammonium chloride Hardener
S 30:70	Seaweed Binder with 30% Acid: 70% Alkali + Wood Particles
S 50:50	Seaweed Binder with 50% Acid: 50% Alkali + Wood Particles
S 70:30	Seaweed Binder with 70% Acid: 30% Alkali + Wood Particles

After that, the mixture was distributed in a metal mold in accordance with size of 300 mm × 300 mm and using a 10 mm thick steel stopper. Metal plate and aluminum foil were placed underneath the steel compressor. Prior to mat forming process when the mat underwent a cold-pressing process for 3 min with pressure of 80 kg/cm² using a cold-pressing machine.

A cold-compressed mat was placed inside hot press machine (165 °C, with pressure of 80 kg/cm² for 15 min) to cure the resin inside the mat. Temperature of 165 °C and 80 kg/cm² pressure were applied both on surfaces of the mat during the process, in order to enhance the curing rate of the seaweed binder and forming compact particleboard whereby the particles were bonded strongly to each other. The particleboards formed after the hot-pressing process were labeled and left for the conditioning process.

2.3. Starch-Based Binder Particleboard

Starch-based binder was prepared separately in which the ingredients of tapioca starch, glycerol, vinegar, and water were mixed and heated with a hot plate. Due to the thermoplastic properties of binder manufactured from starch, the process of mixing with wood particles was done before the binder cooled to below 70 °C. Three different percentages of starch (10%, 15%, and 20%) were incorporated into the production of starch-based binder to determine the effect of different amount of starch in the binder through application on the particleboard. Table 3 below summarized the percentage ratio of starch and adhesive content in particleboard.

Table 3. Composition of particleboard with different percentages of starch with different percentages of adhesive content.

Code	Composition
UF20A	20% Urea-formaldehyde binder + wood particles
UF25A	25% Urea-formaldehyde binder + wood particles
UF30A	30% Urea-formaldehyde binder + wood particles
10S20A	10% starch + 20% starch-based binder + wood particles
10S25A	10% starch + 25% starch-based binder + wood particles
10S30A	10% starch + 30% starch-based binder + wood particles
15S20A	15% starch + 20% starch-based binder + wood particles

Table 3. *Cont.*

Code	Composition
15S25A	15% starch + 25% starch-based binder + wood particles
15S30A	15% starch + 30% starch-based binder + wood particles
20S20A	20% starch + 20% starch-based binder + wood particles
20S25A	20% starch + 25% starch-based binder + wood particles
20S30A	20% starch + 30% starch-based binder + wood particles

A 300 mm × 300 mm × 10 mm particleboard was produced with different amount of binder content (20%, 25% and 30%) mixed with particles and pressed at a temperature of 165 °C, at 135 MPa pressure for 5 min. The board were then left to condition for 3 days before dimensional stability and physical and mechanical testing.

2.4. Wood Plastic Composite (WPC)

In the production of WPC, the billets were debarked to remove the bark from the tree trunk. Next, the billets are chipped to more than 5 cm in size using a chipper machine, followed by chips being flaked using a flaker machine to produce flakes smaller than 5 cm in size. Then, the flakes were screened using the vibrator screener machine to separate the flakes and fibers. After the flaking process, the sieve machine was used to filter any other particles and to collect wood powder only. Finally, the wood powder was dried in the oven for 24 h to achieve less than 2% moisture content.

Clean consumed HDPE bottles were cut into a range of 0.5 cm to 1.0 cm sized chips randomly, because the smaller the HDPE, the easier it is for the HDPE to melt and mix well with the wood powder (WP).

The size of the test piece for tensile strength was produced according to [25], with 160 mm × 14 mm × 3 mm for the length, width, and thickness respectively. Meanwhile, the size of the test piece for the Modulus of Elasticity (MOE) and Modulus of Rupture (MOR) were produced according to the [26], with 160 mm × 20 mm × 3 mm for the length, width, and thickness and the size of the test piece for thickness swelling and water absorption were based on [27], with 70 mm × 25 mm × 3 mm.

The WP and HDPE were mixed manually according to the ratio stated in Table 4 so as to produce a homogenous composite. The mixture was placed in the mold to form a uniform mat. The mold used was 160 mm × 14 mm × 3 mm, 160 mm × 20 mm × 3 mm, and 70 mm × 25 mm × 3 mm, for the length, width, and thickness. Then, the mold was kept under the hot press machine with a temperature of 170 °C and a pressure of 0.5 N/mm² for 15 min.

Table 4. WP-HDPE compositions.

Code	WP-HDPE Composition Based on Percentage Weight	
	Wood Powder (WP) Content (%)	HDPE Content (%)
100H	-	100
90H	10	90
80H	20	80
70H	30	70

2.5. Comparison of Physical Properties for Different Binders

2.5.1. Density

The standard utilized for seaweed particleboards and starch particleboards density test was JIS A 5908 [28], where the length, width, and thickness of the test pieces were measured by using a digital vernier caliper. The length, width, and thickness of every test piece were multiplied to calculate the volume (m³). Every test piece was weighed by utilizing the estimated parity to acquire mass. From that point onward, every data from volume and mass were used to calculate the density of the test pieces. The dimension of

test pieces for this test was 100 mm × 100 mm. The thickness, length, width, and mass were measured to the nearest 0.05 mm, 0.1 mm, 0.1 mm, and 0.1 g, respectively. In addition, the thickness was determined to the closest 0.01 g/cm³. The formula for density test is shown in Equation (1):

$$\text{Density} = m/v \text{ (g/cm}^3\text{)} \quad (1)$$

where m = mass (g) and v = volume (cm³).

2.5.2. Thickness Swelling

The thickness swelling test for WPC, starch-based binder particleboard, and seaweed-based binder particleboard were conducted by using different standards, as shown in Table 5. WPC had different dimension of test piece, while test pieces dimension for both starch-based binder particleboard and seaweed-based binder particleboard were like each other.

Table 5. Thickness swelling and water absorption tests standards for different type of composite materials.

Type of Composite Material	Testing Standard	Dimension of Test Pieces (Length × Width × Thickness)
Wood plastic composite (WPC)	[26]	70 mm × 25 mm × 3 mm
Starch-based binder particleboard	[28]	50 mm × 50 mm × 10 mm
Seaweed-based binder particleboard	[28]	50 mm × 50 mm × 10 mm

Thickness in the center of a test piece of starch-based binder particleboard and seaweed-based binder were measured to the nearest 0.05 mm by using micrometer. After that, the test piece was immersed about 3 cm below water (20 ± 1 °C, 24 h). The test piece was taken out after 24 h and the final thickness was measured.

Test pieces of WPC were conditioned in oven for 24 h with temperature of 50 ± 5 °C, followed by initial thickness measurement before immersion inside water for 24 h. After 24 h, the test pieces were being reconditioned in oven (50 ± 5 °C, 24 h). The final measurement of thickness was taken after the reconditioning process. Calculation of thickness swelling percentages for the green materials was obtained by referring to Equation (2):

$$\text{Thickness swelling} = t_2 - t_1/t_1 \times 100 \text{ (\%)} \quad (2)$$

where t_1 is the thickness (mm) of test pieces before immersion in water and t_2 is the thickness (mm) of test pieces after immersion in water.

2.5.3. Water Absorption

Water absorption test for WPC, starch-based binder particleboard, and seaweed-based binder particleboard were also conducted by using different standards, as shown in Table 5. Specimens of starch-based binder particleboard and seaweed-based binder were weighed and measured to a precision of 0.001 g and 0.001 mm, respectively, before being submerged inside water. After that, the test pieces were immersed in water (20 ± 1 °C, 24 h). The weight of the test piece before and after immersion was recorded and the percentage of water absorption was calculated.

Both thickness swelling and water absorption tests for WPC have a similar procedure, whereby specimens were conditioned in an oven for 24 h with a temperature of 50 ± 5 °C, followed by a weighing process to obtain the initial weight before immersion inside water for 24 h and after being reconditioned in oven (50 ± 5 °C, 24 h). The final weight was taken after the reconditioning procedure is completed. Water absorption percentages for the green materials is expressed in percentage in Equation (3):

$$\text{Water absorption} = W_t - W_o/W_o \times 100 \text{ (\%)} \quad (3)$$

where W_o is the oven-dry weight of the specimen (g) and W_t is the weight of the specimen at a given immersion time (g).

2.6. Comparison of Mechanical Properties for Different Binders

2.6.1. Bending Properties

A three-point bending test setup was employed in accordance with [27] to determine the Modulus of Elasticity (MOE) and Modulus of Rupture (MOR) of WPC with dimension of 160 mm × 20 mm × 3 mm, while particleboard manufactured from seaweed and starch binder referring to bending test of [28] had a dimension of 200 mm × 50 mm × 10 mm. The specimens were positioned flatwise on the Universal Testing Machine (UTM), with the load applied at speed of 10 mm/min. The maximum load (P) was recorded, and MOE and MOR of the wood composites were calculated by using Equations (4) and (5), respectively:

$$\text{MOE} = PL^3/4bh^3d \text{ (N/mm}^2\text{)} \quad (4)$$

$$\text{MOR} = 3PL/2bh^2 \text{ (N/mm}^2\text{)} \quad (5)$$

where P is the maximum load (N) applied on the specimens, L is the span of the specimen (mm), b is the width of specimens (mm), h is the thickness of specimens (mm), and d is the deflection of specimens (mm).

2.6.2. Internal Bond (IB) Strength

Internal bond test for the seaweed particleboard and starch particleboard was conducted as per [28], while the test pieces were cut in size of 50 mm × 50 mm. The test pieces were adhered to steel blocks by using hot-melt glue and a tension load was connected vertically to the board face. From that point onward, the maximum load (P') at the time of failed force (breaking heap of opposite rigidity of the board) was measured. In this test, the tension loading speed was set to 1 mm/min. Lastly, the internal bond was calculated according to Equation (6):

$$\text{Internal bond} = P'/2bL \text{ (N/mm}^2\text{)} \quad (6)$$

where P' is the maximum load (N) at the time of the failing force, b is the width (mm), and L is the length (mm) of the sample.

2.6.3. Tensile Strength

Tensile strength testing for WPCs was done according to [25] with a specified size of 160 mm × 14 mm × 3 mm for length, width, and thickness in the form of a dumbbell shape. The tensile properties were determined using the universal testing machine (UTM) and Equation (7):

$$\text{Tensile strength} = \text{Max load (N)}/\text{Area (mm}^2\text{)} \text{ (N/mm}^2\text{)} \quad (7)$$

2.7. Statistical Analysis

Data analysis was performed using the post-hoc test in analysis of variance (ANOVA) to determine the significant differences ($p \leq 0.05$) between the parameters. The different alphabets (a, b, c, and d) displayed beside the average values in the following graphs show significant differences among the parameters being compared. As for the standard deviations, they were obtained from the total samples (N = 30) of each parameter, where it measures the spread of the data distribution.

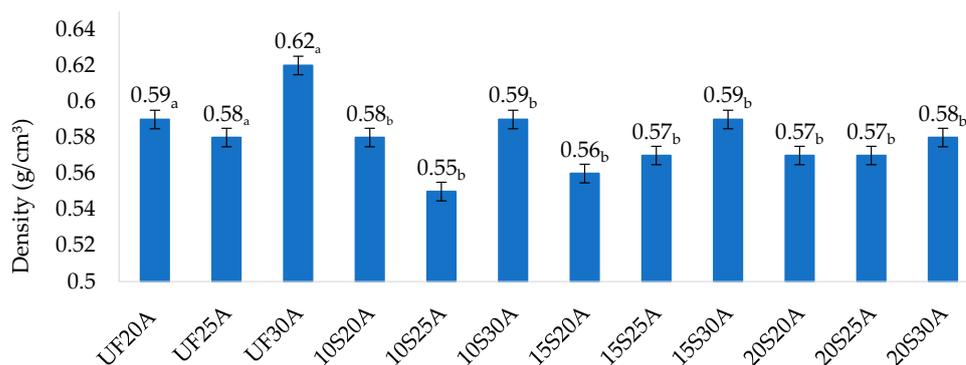
3. Results and Discussion

3.1. Density

Density is one of fundamental properties of wood composite panel products, while density determines the index strength of the boards itself [29–31]. In general, density

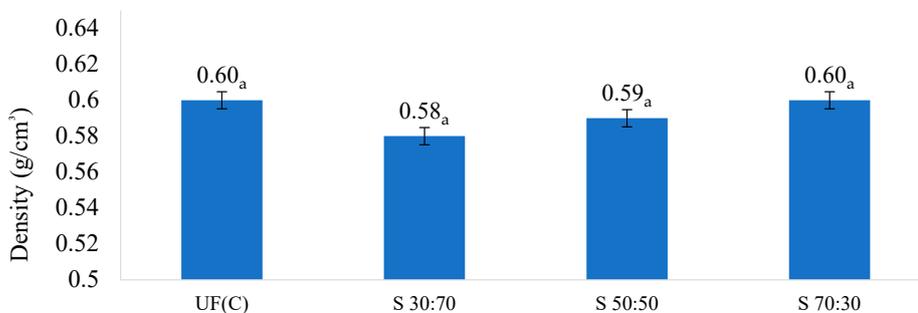
demonstrates the presence of the genuine number of wood particles in a unit volume of wood composite panel and measured by dry load of wood composite panel partitioned by volume of the panel [32].

For starch-based binder particleboard, the density values from [33] did not fluctuate much in their average as shown in Figure 1A. Based on Figure 1A, it can be seen that a density at UF30A amount of adhesive content gives the most astounding normal value for every type of adhesive, which were 0.62 g/cm³ for UF and 0.59 g/cm³ and 0.58 g/cm³ for 10S20A and 20S30A, respectively. The increasing average value for adhesive content should result in increasing the average value of density, such as in 15S, which gave values in the range of 0.56 g/cm³ to 0.59 g/cm³, while 20S gave values in the range of 0.57 g/cm³ to 0.58 g/cm³ when the adhesive content increases by 5%.



Different Percentages of Starch with Different Amounts of Adhesive Content

(A) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters



Different Ratios of Acid to Alkali in Seaweed-based Adhesive Particleboard

(B) The same alphabet (a) displayed beside the average values show no significant differences among the parameters

Figure 1. Average density values for (A) starch-based adhesive particleboard, (B) seaweed-based adhesive particleboard.

In 10S and urea-formaldehyde glues, there were somewhat different in that the average value decreased when the amount of adhesive content was 25A and increased again at 30A. The results recorded were from 0.59 g/cm³ in 20% of adhesive content, decreasing marginally to 0.58 g/cm³ when it achieved 25A and increasing again to 0.62 g/cm³ in 30% of adhesive content for UF. Moreover, 10S have values recorded from 0.58 g/cm³ to 0.55 g/cm³ and ultimately 0.59 g/cm³ for adhesive content that increases from 20A to 25A to 30A. No significant difference ($p \leq 0.05$) was found between the parameters. The conceivable explanation for this was that the test pieces picked random were generally at

the edge of the board produced where materials were less compact, and therefore, it gives lower density.

For seaweed particleboard, the average density in Figure 1B demonstrated that UF (C) has the highest density mean value (0.60 g/cm^3), followed by S 70:30 (0.60 g/cm^3), S 50:50 (0.59 g/cm^3), and S 30:70 (0.58 g/cm^3) [29]. The statistical analysis showed no significant difference at $p \leq 0.05$. All kinds of adhesives achieved the targeted JIS standard [28], whereby the Japanese Industrial Standard set the scope of density of particleboard between $0.4\text{--}0.9 \text{ g/cm}^3$. There are a few factors that can impact the density of particleboard, such as density of wood, the number of particles (grams) in mat forming, pressing pressure, resin content, and other additives [33]. This result is validated with the manual mixing process, whereby the particles were not enough; thus, targeted density could not be accomplished.

UF (C) particleboard has a higher mean of density because the holding between particles is stronger because of the addition of ammonium chloride as hardener, thus making the board to have higher density. The low mean of density for S 50:50 (0.59 g/cm^3) and S 70:30 (0.67 g/cm^3) were lower than the targeted density, which was 0.60 g/cm^3 , which may be caused by the springback of the boards because of swelling during the conditioning process according to [34]. Meanwhile, S 70:30 indicated the minimal mean of density with higher rates of sulfuric acid (70%) in seaweed adhesive cooking produce weak binder for particleboard. A higher concentration of acid in seaweed adhesives decreased the binding properties of the seaweed adhesive; hence, the particles were not strongly bonded and has a higher rate of springback, thus resulting in low density of the particleboard.

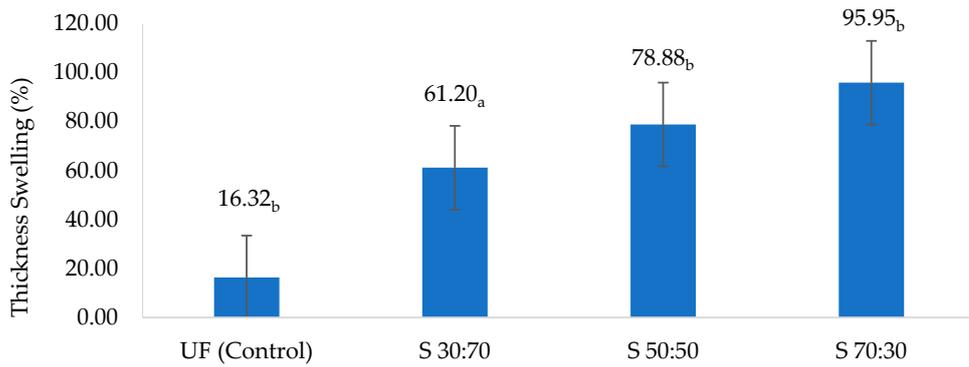
In conclusion, both seaweed particle and starch particleboard have almost similar results. For this testing, seaweed particleboard shows a better performance than starch particleboard, where S 30:70 obtained 0.58 g/cm^3 ; meanwhile, 10S25A obtained 0.55 g/cm^3 from starch particle, marking the lowest performance for density testing.

3.2. Thickness Swelling

In comparison to wood composites, WPC 90H, 80H, and 70H had the lowest thickness swelling percentages (2.54%, 3.32%, and 3.67% respectively), as shown in Figure 2A, followed by particleboard bonded with starch-based adhesives particleboard (Figure 2B) 15S30A (71.18%), 10S25A (71.57%), 20S30A (75.01%), 10S30A (76.14%), 15S25A (78.49%), 20S25A (78.75%), 20S20A (80.31%), 15S20A (82.39%), and 10S20A (84.42%) [33]. Conversely, particleboard bonded with seaweed presented in Figure 2C recorded the highest percentages of thickness swelling, whereby S 30:70, S 50:50, and S 70:30 recorded 61.20%, 78.88%, and 95.95%, respectively [35]. For WPC and starch-based adhesives particleboard, no significant difference was found between the parameters. As for seaweed-based particleboard, S 30:70 was statistically different from all other parameters.

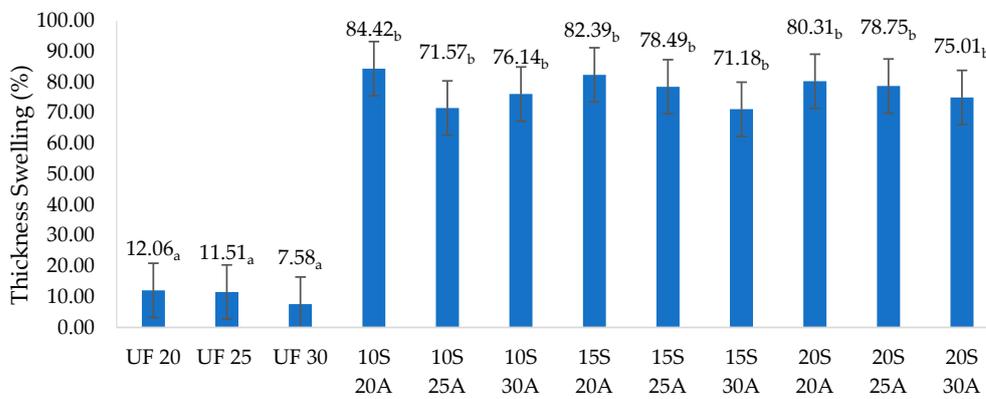
WPC recorded the lowest thickness swelling percentage because it contained HDPE, which exhibit the hydrophobic characteristic that resulted in zero permeability of water. Thickness swelling percentage of starch-based adhesive particleboard was heavily influenced by poor dispersal rate of starch-based adhesive when mixed with wood particles, and weak bonding between neighboring polymer caused the particleboard to swell [36]. Seaweed-based adhesive particleboard recorded the highest thickness swelling percentage due to hydrophilic properties of seaweed, which tends to absorb a higher amount of water.

In fact, thickness swelling of wood composite was highly influenced by several factors, such as the distribution and quality of adhesive, suitability, moisture content, and chemical composition of the material itself [30]. This explained why seaweed-based adhesive particleboard recorded the highest mean value of thickness swelling percentage compared to starch-based adhesive particleboard and WPC; seaweed had a higher moisture content in comparison with starch and HDPE.



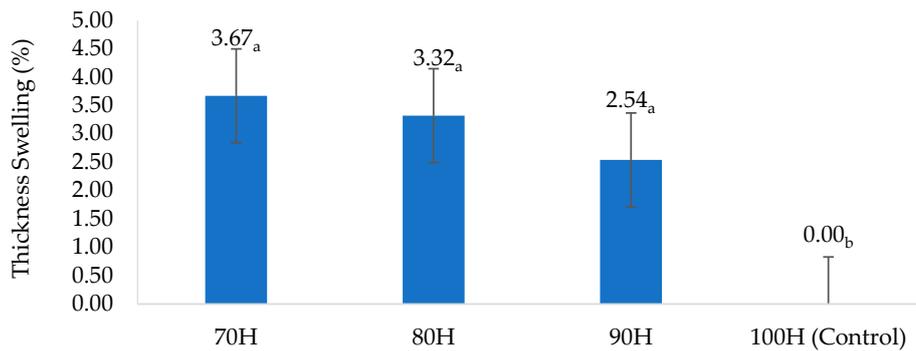
Different Ratios of Acid to Alkali in Seaweed-based Adhesive

(A) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters



Different Percentage of Starch with Different Amounts of Adhesive Content

(B) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters



Different Ratios of High-Density Polyethylene (HDPE) and Wood Powder

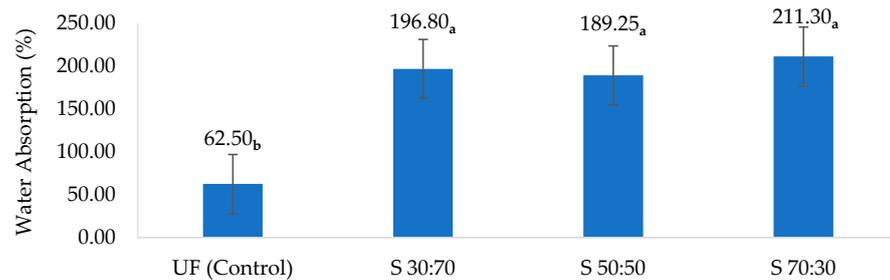
(C) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters

Figure 2. Average thickness swelling for (A) seaweed-based adhesive particleboard, (B) starch-based particleboard, (C) WPC.

3.3. Water Absorption

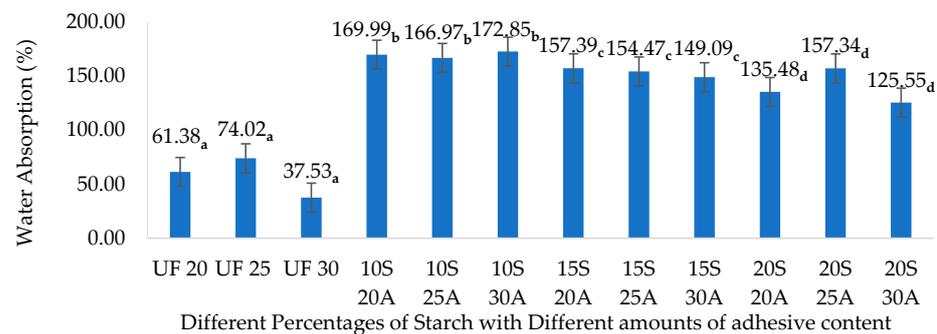
The results displayed in Figure 3A–C indicated that WPC 90H, 80H, and 70H have the lowest water absorption percentages (0.07%, 0.11%, and 0.81%, respectively); fol-

lowed by particleboard bonded with starch-based adhesive 20S30A (125.55%), 20S20A (135.48%), 15S30A (149.09%), 20S25A (157.34%), 15S20A (157.39%), 15S25A (154.47%), 10S205A (166.97%), 10S20A (169.99%), and 10S30A (172.85%) [33]. Particleboard bonded with seaweed documented the highest percentage of water absorption, whereby S 50:50, S 30:70, and S 70:30 recorded 189.25%, 196.80%, and 211.30%, respectively [35]. As for statistical evaluation, no significant difference was found between the parameters for seaweed-based particleboard. Significant differences were found between 10%, 15%, and 20% starch for starch-based particleboard. Moreover, 70H was found to be statistically different from all other parameters for WPC.



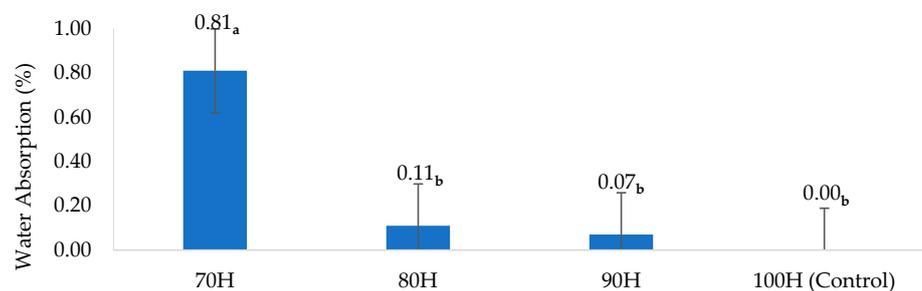
Different Ratios of Acid to Alkali in Seaweed-based Adhesive Particleboard

(A) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters



Different Percentages of Starch with Different amounts of adhesive content

(B) The different alphabets (a, b, c, and d) displayed beside the average values show significant differences among the parameters



Different Ratios of High-Density Polyethylene (HDPE) and Wood Powder

(C) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters

Figure 3. Average water absorption for (A) seaweed-based adhesive particleboard, (B) starch-based adhesive particleboard, (C) WPC.

In general, the water absorption test is a measurement method to indicate the percentage of water diffused inside the wood composite component. The water absorption rate is dependent on the density and water diffusivity of the material itself [37]. Water

diffusivity is defined as a rate at which water moves to the interior of the product from its surface, whereby it is mainly affected by the porous structure of the material and adhesive itself [37,38]. This explained why WPC recorded the lowest water absorption percentage compared to starch-based adhesive particleboard and seaweed particleboard. In depth, water absorption affected mechanical strength of wood composite, whereby a composite material that absorbed a higher amount of moisture in hygroscopic range tends to have a lower index strength [29,37].

HDPE hydrophobic properties impacted the rate of water being absorbed inside WPC, whereby WPC with higher ratio of HDPE tend to repel the water molecules better compared to WPC with a higher ratio of wood powder. Water absorption percentage of starch-based adhesive particleboard is affected by hydrophilic nature of starch, but the result displayed in Figure 3 indicated that seaweed had better water absorbing properties compared to starch. Seaweed exhibits hydrophilic characteristics, which tend to attract water molecules inside it, and thus, resulted in a high percentage of water absorption for seaweed-based adhesive particleboard.

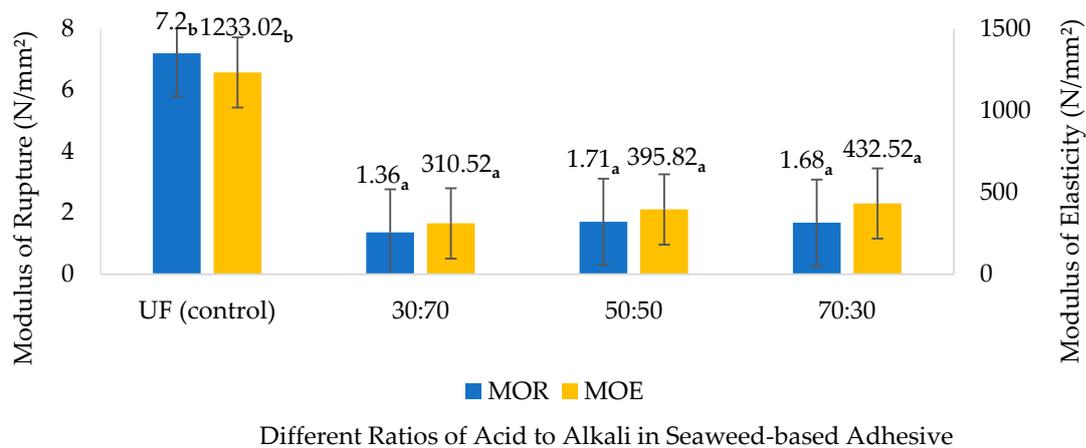
Previous studies [36] stated that the water diffusivity of starch-based adhesive could be elaborated by the hydrophilic nature of starch, whereby the starch molecule had two functional groups (OH group and C-O-C bond). In depth, the OH group is susceptible to a substitution reaction, which tend to absorb the water molecule highly, thus resulting in problem in terms of water absorption of the composite material itself. Additionally, the C-O-C bond existing in the starch molecule was also vulnerable to chain breakdown. Moreover, weak hydrogen bonds in starch triggered the breakdown of hydrogen groups, forming another hydrogen bond with water molecules, and therefore, they resulted in poor water resistance in the starch-based adhesive particleboard [39].

3.4. Bending Properties

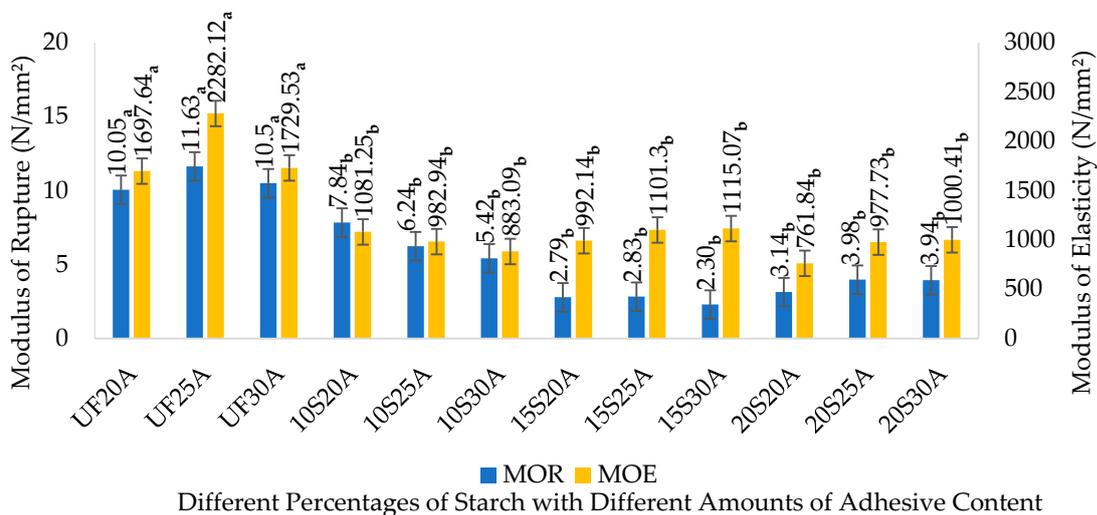
The bending strength of a product often defined from its value in terms of MOE and MOR. MOE determines the ability of the wood composites to resist elastic deformation when load was applied. A stiff material has higher MOE values in which the shape does not change easily; it only deflects by a certain amount, which is suitable in application of furniture or construction.

Unlike MOE, which measures the wood's deflection, MOR measures the ability of a materials to sustain load before ruptures occurs, which is also known to measure the ultimate strength. It represents the maximum stress experienced within the fibers when it ruptures. Figure 4A,B display the MOE and MOR of the particleboard manufactured from seaweed-based and starch-based [33] adhesives.

From Figure 4A, particleboard manufactured from seaweed-based adhesive also shows lower values of MOE compared to UF adhesive of particleboard and starch-based adhesive particleboard, which was 432.52 N/mm^2 at S 70:30 (seaweed-based adhesive with 70% H_2SO_4 and 30% NaOH), while the highest mean value of MOR was 1.71 N/mm^2 at S 50:50 (seaweed-based adhesive with 50% H_2SO_4 and 50% NaOH) [35]. Both MOE and MOR value of particleboard manufactured from seaweed-based adhesive by using different ratios of H_2SO_4 and NaOH were all lower than the strength of particleboard manufactured from urea-formaldehyde (control). The lowest MOE and MOR value of particleboard with seaweed-based adhesive was at ratio of 30:70, which was 310.52 N/mm^2 . This implicates that the higher percentage of alkali in the adhesive will cause the bonding strength of particles to weaken. This is due to the excessive duration of pressing time, which causes the over curing of seaweed-based adhesive particleboard, which affected the bonding strength between particles. Ref. [34] reported that the factors, such as pressing temperature and time as well as surrounding temperature, could have affected the strength of the particleboard.



(A) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters



(B) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters

Figure 4. Average MOE and MOR for (A) seaweed-based adhesive particleboard. (B) starch-based adhesive particleboard.

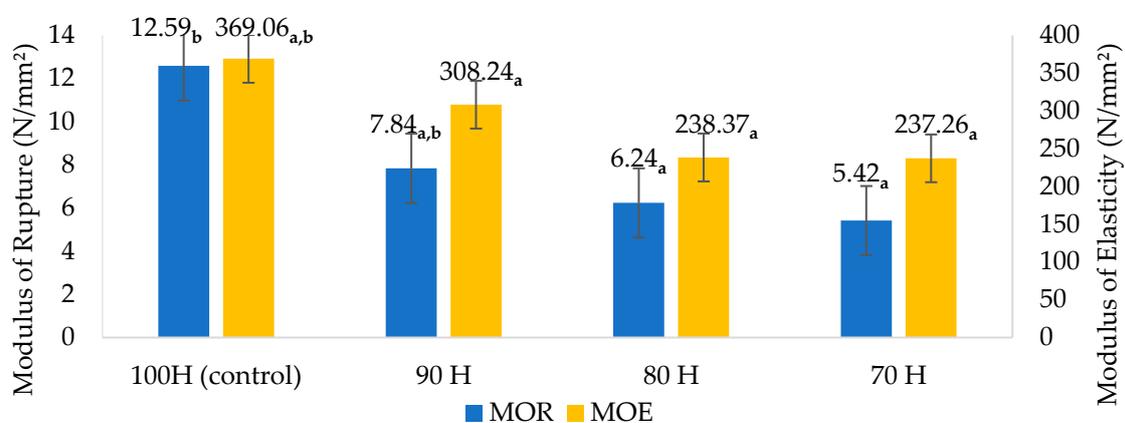
Apart from that, UF, which acts as control, also has a higher value of MOE and MOR compared to particleboard produced from starch-based adhesive, as shown in Figure 4B. The highest mean MOE value of particleboard manufactured from starch-based adhesive was 1115.07 N/mm², while particleboard bonded by using UF could reach up to 2282.12 N/mm² in MOE. However, the MOR of starch-based adhesive was highest at 10S20A, in which the value was 7.84 N/mm² and UF could achieve 11.63 N/mm². This indicates that the materials bonded with urea-formaldehyde tend to be stiffer and require high loads to deform and rupture completely. On the other hand, in the case of particleboards made from starch-based adhesive, the lower bending properties means material are flexible and elastic under stress, but this deflection could not sustain too much load which resulted in the rupture of the material. For MOE and MOR, for both seaweed-based and starch-based particleboard, no significant difference was found between the parameters.

The stiffness of material often depends on the types of application in which only certain amount of deflection is allowable. The elasticity of starch-based adhesives was affected by the presence of glycerol, which acts as a plasticizer in the composition of the

adhesive. Ref. [40] reported that the MOE of starch-based adhesive can be improved with a decreased amount of glycerol up to 20% in the formulation. However, the incorporation of different amounts of glycerol into the formulation of a starch-based adhesive were not investigated in this paper. Furthermore, the bending strength (MOR) might be affected due to the amylose and amylopectin present in the starch composition. The heating process of starch in water during preparation of the starch-based adhesive might cause the rupture and collapse of amylose and amylopectin, which gives cohesive, flexible, and weak materials.

Nevertheless, the difference in bending properties between starch-based adhesive and seaweed adhesive as wood binder might be due to the composition of the liquid adhesive itself. Starch-based adhesive has incorporated glycerol and water as one of the plasticizers to control the brittleness and reduced the drying rate of the adhesive while in application. However, seaweed-based adhesive with different amount of sulfuric acid and sodium hydroxide does not act as a plasticizer in the adhesive. Sulfuric acid appears as a medium to breakdown the components and sodium hydroxide was used to obtain the carrageenan found in seaweed that aims to increase the viscosity of the adhesive [41]. However, too high viscosity of liquid adhesive will cause the uneven distribution of adhesive and difficulties in mixing. As for seaweed-based adhesive in this section, the viscosity was approximately 11,000 mPas [42] to 15,200 mPas, while UF has a viscosity of 17,000 mPas. Although the viscosity does not differ much, the thermoplastic properties of the seaweed adhesive cause it to harden in a shorter time and difficulties in mixing with particles will be encountered. This may lead to the uneven distribution and not fully covered surfaces of particles. Moreover, the properties of carrageenan found in the seaweed was one of the factors contributing to the difficulties in application. According to [41], different types of carrageen exhibit different gelation properties due to the distinction in sulfate groups and anhydro bridges. They reported that kappa-carrageenan found in *Kappaphycus alvarezii* seaweed exhibits rigids and brittle gels, iota-carrageenan (*Eucheuma denticulatum* seaweed) forms softer, elastic, and cohesive gels, while lambda-carrageenan could not form gels.

WPCs, on the other hand, have the highest mean value of MOE at a ratio of 10:90, which was 308.241 N/mm² (Figure 5). The mean value of MOE for HDPE (control) was 369.06 N/mm². No significant difference was found between the parameters. This indicates that materials manufactured by incorporating plastic with wood powder are flexible and elastic compared to pure plastic materials. Figure 6 shows the result of the bending test for all the ratios of wood plastic composites. 70H has the lowest elasticity compared to other ratios, and the time taken for it to break was shorter than 15 min. This was because the wood powder present in the wood plastic composites had made the bond between lignocellulosic weakened and brittle [43].



Different Ratios of High-Density Polyethylene to Wood Powder

Figure 5. Average MOE and MOR values of WPC with different alphabets (a, and b) displayed beside the average values show significant differences among the parameters.

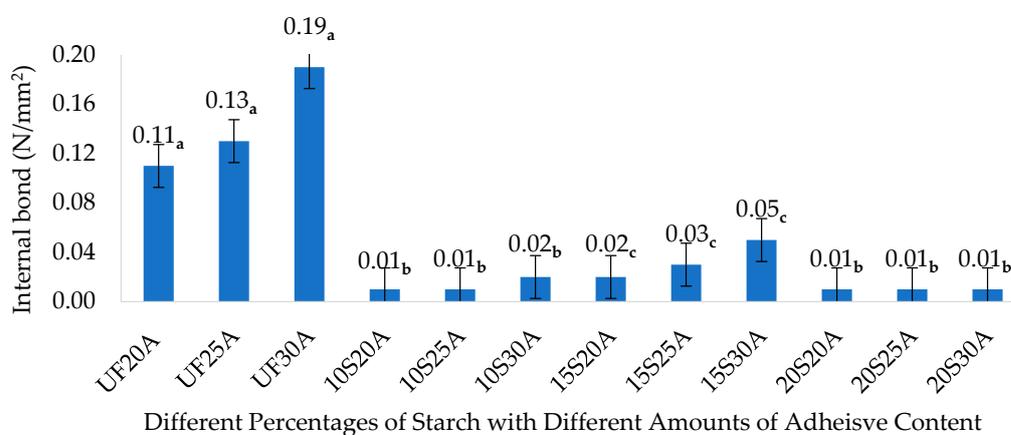


Figure 6. Bending failures of WOC with different ratios of HDPE and wood powder.

3.5. Internal Bond (IB) Strength

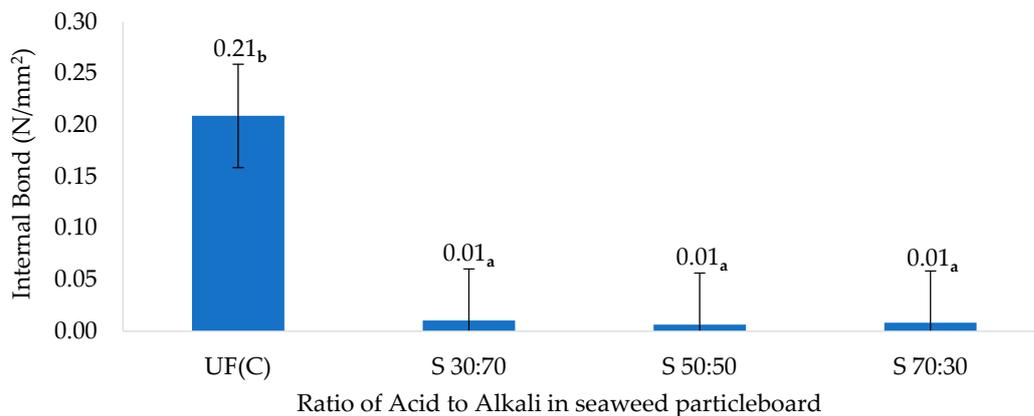
Interior holding is a typical strategy used to gauge glue holding execution in wood composite boards, though the holding quality is for the most part decided as per viability of paste application in assembling a wood composite item (Annual Book of ASTM Standards, 1989). In any case, the Annual Book of ASTM Standards [44] reported that the bonding strength of wood composite material is additionally influenced by the size of wood particles used and density of wood composite itself.

As indicated by Figure 7A, inside the different amount of adhesive in the manufactured of particleboard, the IB strength of the three consecutives amount of adhesive content 20S20A, 20S25A, and 20S30A of starch-based adhesive has the lowest average value among all, which was 0.01 N/mm^2 [33]. Moreover, the average value of particleboard manufactured by using starch-based adhesives encounters an increase from 10S to 15S, which was from 0.01 N/mm^2 to 0.02 N/mm^2 at 20%. Meanwhile, average value at 25A of adhesive content increases from 0.01 N/mm^2 to 0.03 N/mm^2 for 10S and 15S, respectively. The average value of IB strength at 10S30A adhesive content was 0.02 N/mm^2 and the value raised to 0.05 N/mm^2 in 15S30A. The 15% starch parameters were found to be statistically different from all other parameters.



(A) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters

Figure 7. Cont.



(B) The different alphabets (a, and b) displayed beside the average values show significant differences among the parameters

Figure 7. Average IB for (A) starch-based adhesive particleboard, (B) seaweed-based adhesive particleboard.

It can be seen to decrease when the amount of starch increases from 15S to 20S. The graph above demonstrated that the starch-based adhesives give better holding with 15S, and when it achieves 20S, the bonding between particles may begin to weaken, which makes the IB strength decrease to a similar level as 10S. A possible clarification to this pattern can be related with the viscosity between starch-based adhesives. Moreover, the average internal bond of seaweed-based particleboards [29] in Figure 7B also demonstrated that the performance of UF (C) were better (0.21 N/mm^2) as compared to the adhesive made from seaweed. A different acid-to-alkali ratio showed no effect on the IB strength of seaweed particleboard in which the values remain at 0.01 N/mm^2 . However, the statistical output showed no significant differences between the parameters.

As a conclusion, 15S30A from starch particleboard with 0.05 N/mm^2 shows a good performance than seaweed particleboard. Meanwhile, 10S2A0, 10S25A, 20S20A, 20S25A, and 20S30A of starch particleboard followed by seaweed particleboard obtained the lowest value at 0.01 N/mm^2 . According to [45], although starch was widely used as binders, sizing materials, glue, and pastes, its bonding capacity was still not strong enough to glue wood. Therefore, modified starches that are made from graft polymerization with vinyl acetate and butyl acrylate have been included in application of wood adhesive [46]. However, the properties and functions were still not appropriately investigated. As reported by [47], the biopolymer-based adhesives were too weak for practical use.

3.6. Tensile Strength

Figure 8 indicates that the 100H has the highest tensile, which was 18.404 N/mm^2 , followed by 90H with 9.421 N/mm^2 and 80H with 7.102 N/mm^2 . Meanwhile, the 70H has the lowest tensile, which was only 6.378 N/mm^2 . No significant difference was found between the parameters.

In [48], various results were reported for the flexural and tensile properties of the composites made from the recycled plastics. Some of the research indicated that the recycled plastics and the virgin plastics have similar properties, while other research reported superior properties and other reported lower properties. Bonding of plastic with wood happened when the plastic was melted at a certain melting point (for the case of HDPE, its melting point is $130 \text{ }^\circ\text{C}$) so that the thermoplastic could flow easily and blend with the wood materials. The flowing rate was found to be faster when the plastics had a lower melting point. In contrast to the flexural and tensile properties, all the result show lower impact strengths in composites made from recycled plastics and lignocellulosic materials in comparison to composites made from virgin plastics [49].

Figure 9A shows the flexibility of the 100H (C) when the tensile strength testing machine pulls the test piece. Figure 9B shows the different result obtained from the tensile strength testing of 100H (C). Based on Figure 8, the average value of 100H (C) has the highest value of tensile strength. Based on the observation, it was shown that the data of tensile properties of each test piece have different characteristics. A possible reason for this was because of the usage of different sources of HDPE. Industry may use different types of chemicals to produce HDPE-based plastic products.

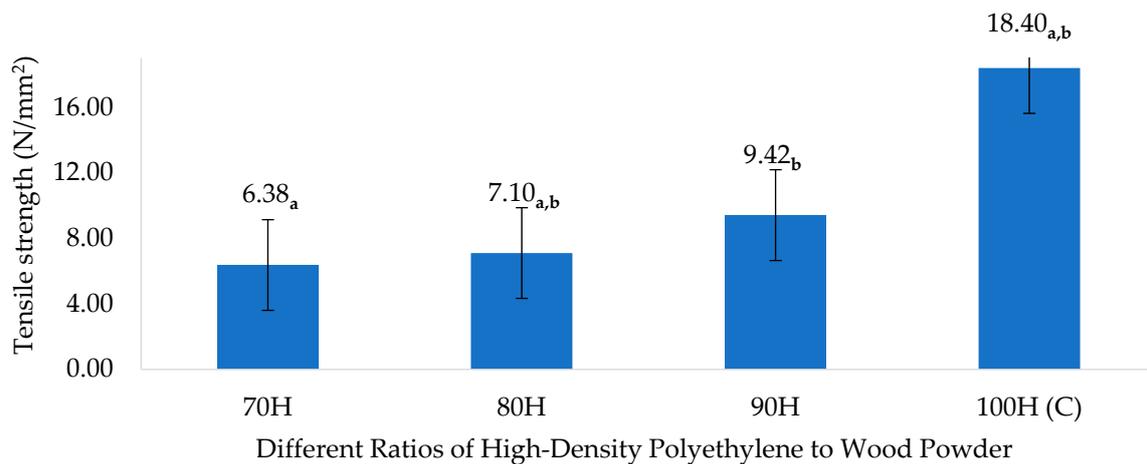


Figure 8. Average tensile strength of WPC at different ratio of HDPE and wood powder with different alphabets (a, and b) displayed beside the average values show significant differences among the parameters.

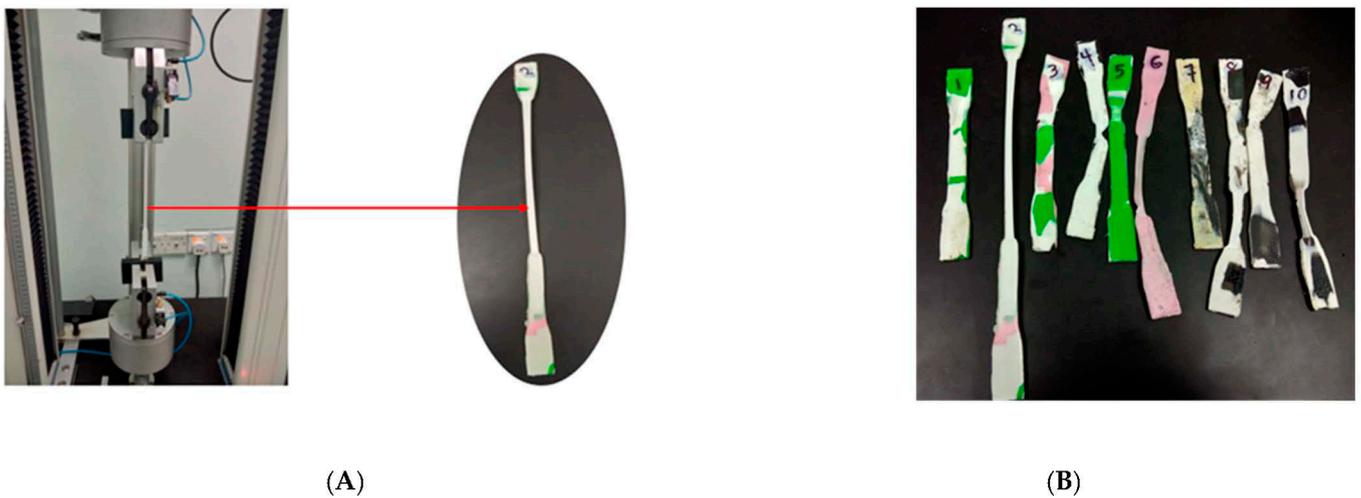


Figure 9. (A) Tensile strength test for ratio 100% of HDPE (100H). (B) Test pieces for ratio 100% of HDPE (100H) after testing.

Wood and plastic composites show a positive effect on the properties of the composites. Wood properties of the composites increases with the use of plastic, as plastic helps decreasing the moisture content, thickness swelling, and water absorption of composite. Other than that, plastic also helps improve the tensile strength of the wood [50]. Many of the researchers stated that the additional of wood powder could increase some physical, mechanical, and thermal properties, for example, rigidity, stiffness, strength, hardness, and fire resistance [51,52]. However, when the non-polar polymer matrix is mixed up with a huge amount of polar wood powder, this mixture may cause of reducing in composite phys-

ical and mechanical properties, as this happens because of the weak interfacial adhesion and lack of fiber dispersion caused by fiber agglomeration [53].

4. Conclusions

Based on the research conducted and the data in the literature, it can be concluded that the production of wood plastic composites (WPC) included in this paper contains natural materials such as wood powder blended with polymers to reduce landfill waste and provides an alternative to recycling technologies.

From this paper, it can be concluded that particleboard manufactured from bio-adhesives exhibit a hydrophilic nature when in contact with water. A high amount of water absorbed causes a swelling in thickness of particleboard as compared to WPC. Moreover, starch-based particleboard was concluded to be stiffer and able to sustain a higher stress load than seaweed-based particleboard and WPC. The huge amount of wood powder present has caused brittleness as well as a decrease in stretchability of WPC through tensile strength test.

One of the limitations faced when conducting this research was the method used to mix the wood particles with the binder. Mechanized blending was suggested for a better and more even mixture of wood particles and binder for particleboard produced in this study. However, physical and mechanical behavior of particleboard from seaweed and starch-based adhesives as well as WPC discussed in this paper could provide a basic guideline for more research to be done in this aspect, whereby a non-impact construction is presented.

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