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MECHANICAL AND PHYSICAL PROPERTIES OF TEAK LEAVES WASTE/POLYURETHANE COMPOSITES FOR PARTICLE-BOARD APPLICATION

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Abstract. A teak leaf waste/polyurethane composite has been made for particle-board application. Some variation treatments were carried out on the use of polyurethane as the matrix with the range of 0.04-0.20 (w/w). Mechanical and physical properties have been tested on the composites produced. The test results showed the highest compressive strength of the sample reaches 38.5 MPa for polyurethane fraction of 0.14 (w/w). The composite has a density of 1261 kg.m^{-3} which is in accordance with the result of density on teak. The physical properties have been also investigated and it was found that the lowest value of water absorption is 1.38%. This result indicated that composites can be used as an eco-friendly industrial scale instead of wood.

Keywords: composite, polyurethane, teak leaf, mechanical properties.

1. Introduction

In the last few decades, the wood demand in the world always grows with the total demand for 2020 is projected attain to 515 million m^3 [1]. To fulfill this demand growth, one of the most likely efforts is composite engineering. Composite engineering promises organic and inorganic waste utilization as a synthesized-material having excellent properties relating the woods, such as mechanical strength, water absorption and so on [2-6]. Furthermore, the composite engineering is also a solution for the raising of environment awareness causing the high deforestation in several countries [2-3]. In addition, the utilization of natural waste and fibers for some composites has been widely studied and developed from various countries including Indonesia, Malaysia, Thailand, India, Brazil and Japan [2-4].

Some research relating to the polymers and plastics is focused on developing fibers, plastics, adhesives, composites, textiles, paper from renewable resources derived mostly from agricultural wastes containing lignocellulosic [4]. One of the efforts done is maximizing some natural fibers such as sisal, hemp, bamboo and agricultural wastes such as leaves, coconut fibers, rice husk, peanut shells and straw also can be used as composite reinforcements [5-6]. Most developing countries like Indonesia, some agricultural wastes usually are used for compost production. Composites of these natural fibers have several advantages such as easy to process, environmentally, good acoustic properties, low energy processes, good mechanical and thermal properties, water resistance, non-corrosive, non-toxic and naturally degradable [7].

Further, several studies related to the leaves waste composites have been done, such as Masturi *et al*. developed composite from leaves waste and paper waste using polyvinyl acetate as matrix and the compressive strength reaches to 45.60 MPa [2]. Campanella *et al*. successfully made a composite of Northern Red Oak leaves with plant oil-based resins [6], while Thakur *et al* have also developed biopolymer composite using ligno-cellulosic of *Hibiscus Sabdariffa* [8]. A different method has been performed by Nongman *et al* having created a composite by lamination method of banana leaf fiber and adhesive [5].

In this work, we developed a composite of teak leaves waste with polyurethane matrix. The teak leaves was chosen since its utilization only as compost having a low add value as well its availability is very abundant in Indonesia. The aim of this study is to produce a teak leaves waste composite having physical and mechanical properties making a chance to be used on an eco-friendly industrial scale.

2. Materials and Methods

The teak leaves waste of Semarang Indonesia was used, while diisocyanates and polyol as polyurethane (PU) polymerization materials were purchased in Multi Kimia Inc, Semarang, Indonesia.

For first, the leaves waste was dried and crushed using a blender machine to form a powder. Then, the powder was sieved using 700 mesh. In the other hand, the diisocyanates and polyol were mixed using magnetic stirrer for 5 minutes with ratio 1:1 to form polyurethane and mixed with the teak leaves waste powder with varied composition while the leaves teak powder was kept at 12 g weight. The PU/teak leaves waste mixture was stirred using mixer machine to homogenize the mixture, then pressed into a cylindrical mold at pressure of 3 tons for 15 minutes. Having been pressed, the composite was removed from the mold and dried for 24 hours at room temperature.

The investigation to the content of the teak leaves powder cellulose bonding to polyurethane and detecting spectral changes in teak leaves waste composite was performed using FTIR Spectrometer Frontier type (Perkin Elmer) in a spectral range of 400-4000 cm^{-1}. Meanwhile, the identification of the mechanical properties was measured by a compressive strength test using Sociedad Espanola anonima Ibertest instrument. In this test, the sample is cut into cubic shape and was measured using testing machine. In addition, to ensure the water resistance of the composite, the identification of the physical properties was performed by density measurement, further, water absorption test was done by soaking this sample in the water for 24 h to obtain the weight of water absorbed into the composite compared to the initial weight.

3. Result and Discussion

3.1. Compressive Strength Test

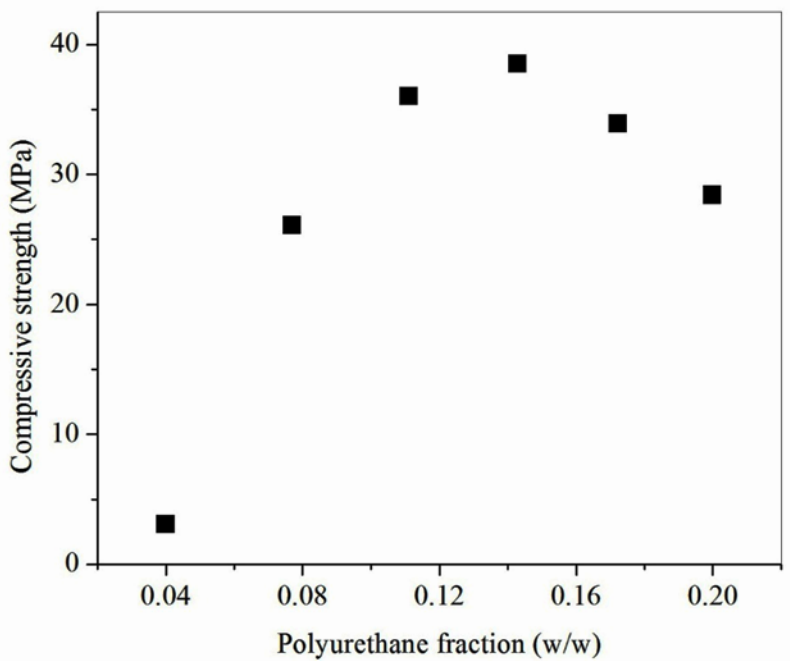
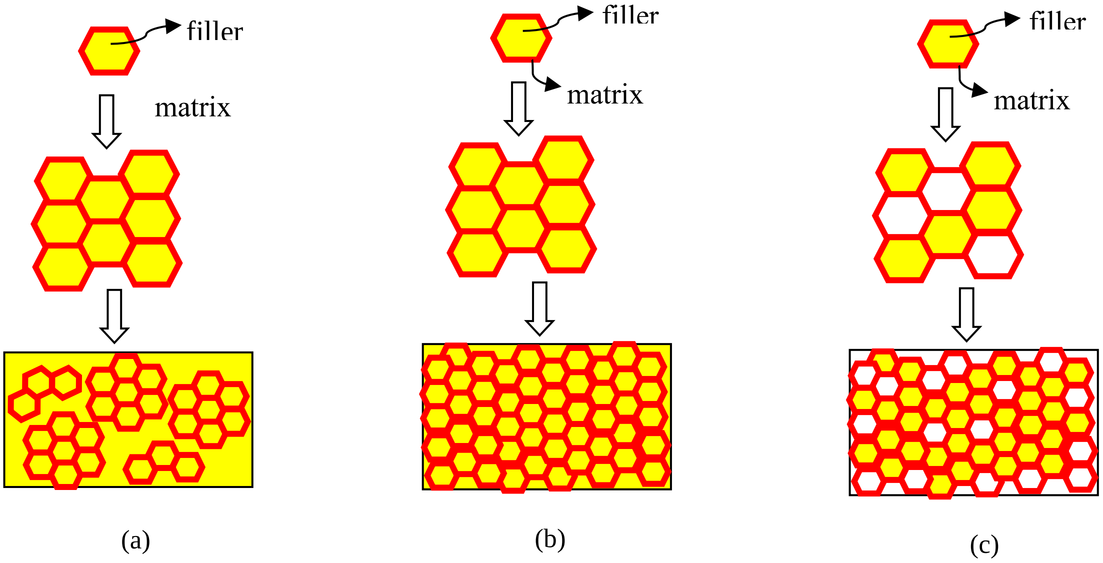


Figure 1. Compressive strength of teak leave waste composite as polyurethane content (w/w) and the compressive strength attains at maximum for 0.14 fraction (w/w).

Figure 1 shows the effect of polyurethane addition on composite compressive strength. The highest compressive strength is achieved as 38.5 MPa for polyurethane fraction of 0.04 (w/w).

This resulting strength is 86% higher than that of polyurethane fraction of 0.04 (w/w). The increase in compressive strength is closely related to the interaction between the surface of the particles and the polymer. The composites with 0.04 and 0.08 (w/w) have relatively low compressive strength regarding there are still many teak leaf particles not binding to the polymer matrix due to. When the polyurethane fraction was added to 0.14 (w/w), the composite attains the highest compressive strength. This shows that all matrices of the polymer exactly interact with all the teak powder particles so that the composite becomes more compact and withstands higher loads such as the mechanisms in **Figure 2**.

Figure 2(a) illustrates the bonding of leaves teak particles and polyurethane polymers at a mass varied of 0.04-0.08 (w/w). Preliminarily all polymer matrices are not filled by particles. Having been pressed to form a composite, some of the diffused particles do not bind to the matrix. The presence of leaf particles not binding to the matrix affects the ability of the composite in holding the load so it becomes easily fragile and broken. In **Figure 2(b)**, a composite with polyurethane content of 0.14 (w/w) shows that all teak leaf particles are precisely bind to a polymer matrix, that is called as optimum fraction of polyurethane in this composite. This matrix bond is capable of forwarding the load in all directions so that the resulting composite is stronger and withstand maximum load [9]. Further, as **Figure 2(c)** the composites with a polyurethane content of 0.17-0.20 (w/w) show some parts of polyurethane not binding to the filler, so that many avoids from polyurethane polymer appear and it result in the decreasing of the compressive strength [10-11].



(a) (b) (c)

Figure 2. Filler particle distribution scheme and polymer matrix. (a) low compressive strength, (b) high compressive strength, (c) low compressive strength

Using the explanation above, the added mass of polyurethane improve the composite compressive strength. However, the larger mass otherwise makes the reduction of compressive strength, as in **Figure 1** the maximum strength is the composite with the polyurethane fraction of 0.14 (w/w). According to Jaya *et al.*, polyurethane is a hydrophobic matrix and teak leaf powder is a hydrophilic filler so that when the polymer was added much more than the filler there will be a very poor adhesion force between surfaces [12]. Further, the raising of the strength until certain fraction is related to the maximum contact amount between the polymer and solid waste particle [13].

The most important interaction in the composite is predominantly emerged between the polyurethane and the cellulose in teak leaves. According to Alemdar and Sain, the cellulose in plant fibers has important role in reinforcing material in composite applications [14]. The presence of cellulose in teak leaves can be investigated through the FTIR spectrum in **Figure 3.**

Figure 3(a) and **Figure 3(b)** show the FTIR spectrum of the pure cellulose obtained from Ramirez-Flores et al [15] and the teak leaves respectively, with the comparison can be shown in **Table 1**. The FTIR spectrum of pure cellulose and leaves teak powder cellulose's has a significant difference at peak 1631 cm^{-1}. The peak of 1631 cm^{-1} indicates absorption band of hydrogen forming a hydrogen bridge from hydroxyl groups and oxygen atoms from glucose monomer and usually found at a cellulose fiber [16]. The peak of 1631 cm^{-1} is not found at pure cellulose but it found at cellulose of natural fibers [17-20].

The peak of 2929 and 2300 cm^{-1} indicates a strong C-H bond in cellulose polymers [21]. While, peak of 3401 cm^{-1} shows the stretching of the O-H group bonds [22]. Further, the peak of 780 cm^{-1} and 1438 cm^{-1} shows the C-H bonds connecting the glucose units of cellulose. The peak at 1234 cm^{-1} indicates that there is a C-O-C group which is the structure of lignin. The absorption area occurring at 1076 cm^{-1} indicates the presence of a C-O bond in the cellulose component [12].

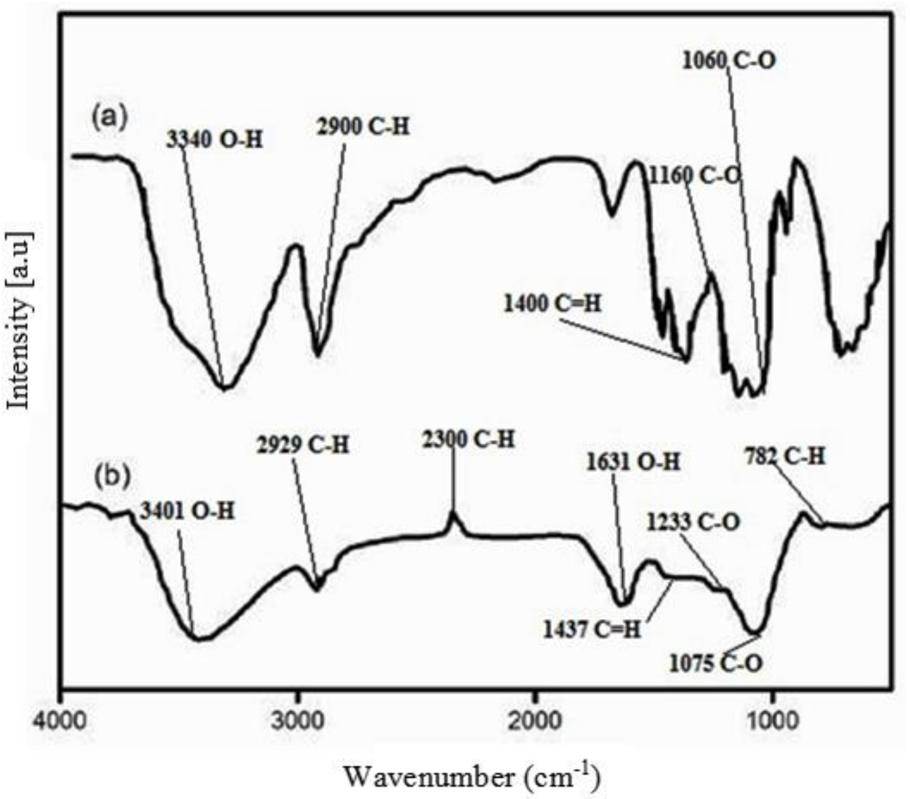


Figure 3. Spectrum of cellulose of: (a) pure cellulose obtained from Ramirez-Flores [13] b. cellulose of teak leaves powder in this work where many peaks of both celluloses have more similarities.

Using the FTIR analysis, it can be shown that cellulose and lignin components are found in the teak leaves. This leave can compete the conventional fibers such as glass fiber regarding its high performance as reinforcing material for thermosetting and thermoplastic matrices with their naturally degradable superior properties [23].

Table 1. Comparison of the peak absorption at pure cellulose and cellulose of teak leaves powder

Bond	Wavenumber (cm^{-1})	
	Pure cellulose (cm^{-1}) [15]	Cellulose of teak leaves powder (cm^{-1})
O-H	3340 – 3500	3401
C-H	2800 – 2920	2929
C-O-C	1160	1233
C-O	1035 – 1060	1075
C=H	1400	1437

The interaction arising between the teak leaves and the polymer is a van der Waals interaction. This interaction was confirmed by several FTIR spectrum shifts as shown in **Figure**

4. The shifts in spectrum are appeared due to the vibrating atoms as a result of the energy of disruption of atoms closing each other.

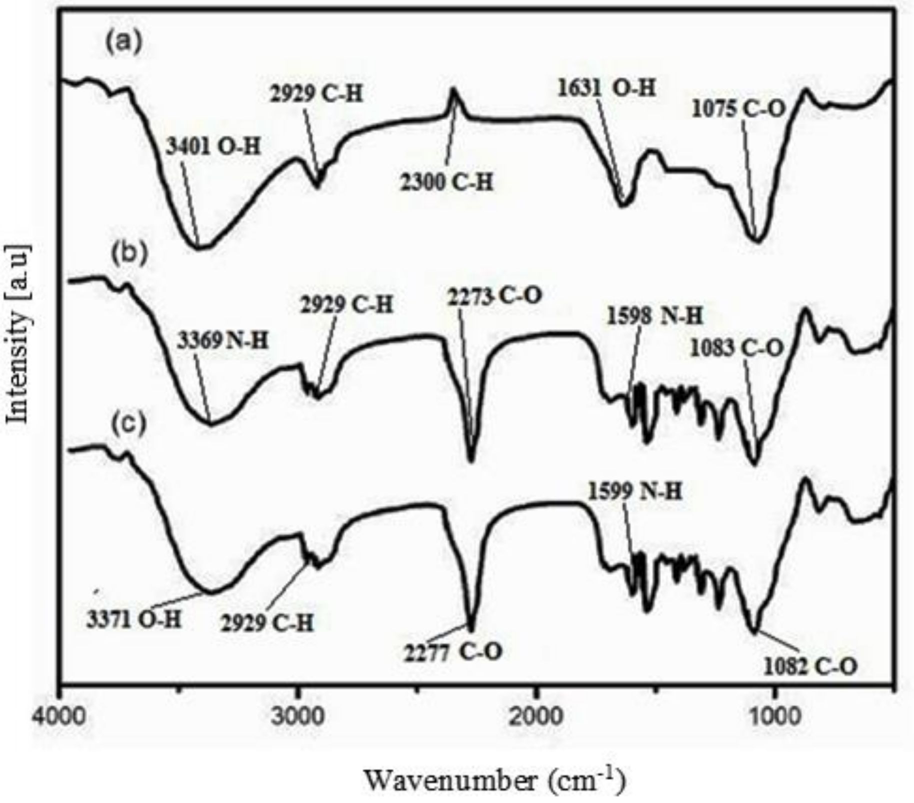


Figure 4. FTIR spectrum a. teak leaves, b. polyurethane, c. PU/teak leave composite

Polyurethane has two functional groups, i.e. carbonyl and amine groups. The spectrum shift starts to occur of 3369 cm^{-1} to 3371 cm^{-1} indicates the O-H bond. This shift is due to the amine group of the polyurethane was disturbed by hydrogen bonding in the hydroxyl group of leaves and forming a hydrogen bridge [24]. Further, the shift of 2273 cm^{-1} to 2277 cm^{-1} shows the carbonyl group of polyurethane undergoes a spectral shift. This is due to the disturbance of the C-H leaves bonding to the carbonyl group forming the O-H bridges. The peak at 1598 cm^{-1} shifting to 1599 cm^{-1} also indicates the amine group in polyurethane disturbed by the O-H bonds of the teak leaves. Furthermore, the shift also occurs at the carbonyl peak of 1083 cm^{-1} to 1082 cm^{-1} due to disturbance of the leave's C-O bond of the peak of 1075 cm^{-1} (Figure **5**). Similar results were confirmed by a previous work that PVA-sepiolite composite which derived its van der Waals interactions between silanol groups and carbon C-O and shifted from 1088 cm^{-1} to 1085 cm^{-1} [25].

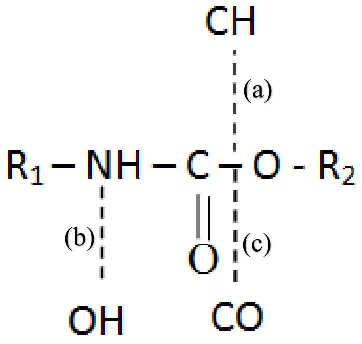


Figure 5. Van Der Waals interactions on the teak leaf waste composites, dashed lines illustrate the Van der Waals interaction area of (a) CH and carbonyl group (b) OH and amine group, and (c) CO and carbonyl group.

3.2. Density Test

Figure 6 shows the density of composite samples with polyurethane variation ranges from 825 to 1261 kg.m^{-3}. The density value is proportional to the increase in PU content. The lowest density value is found in the sample with a polyurethane fraction (w/w) of 0.04, while the highest density value is in the sample with 0.20 of polyurethane fraction (w/w). It can be explained when the polyurethane polymer content rises and the filler is fixed, then the number of links produced by the polymer increases. Hence, the increased links result in stronger bonds to bind the filler so that it gets closer and density increases [7].

The density of 0.08 (w/w) is in accordance with the results of the density value of teak that has been investigated by Wahyudi et al. obtaining the teak density values have a density of 1000 - 1200 kg.m^{-3} with an average value of 1160 kg.m^{-3} [17]. In this study, the resulted-density value is higher than that of Martins et al work producing particle board with density of 510 - 570 kg.m^{-3} [18]. The composites produced in this work have a similar density to the results of the Raju et al. producing some composites with densities ranging from 1100 to 1320 kg.m^{-3} [19]. Further, Butylina et al. have produced wood fibre-polypropylene composite having density ranged from 1040 to 1140 kg.m^{-3} [20].

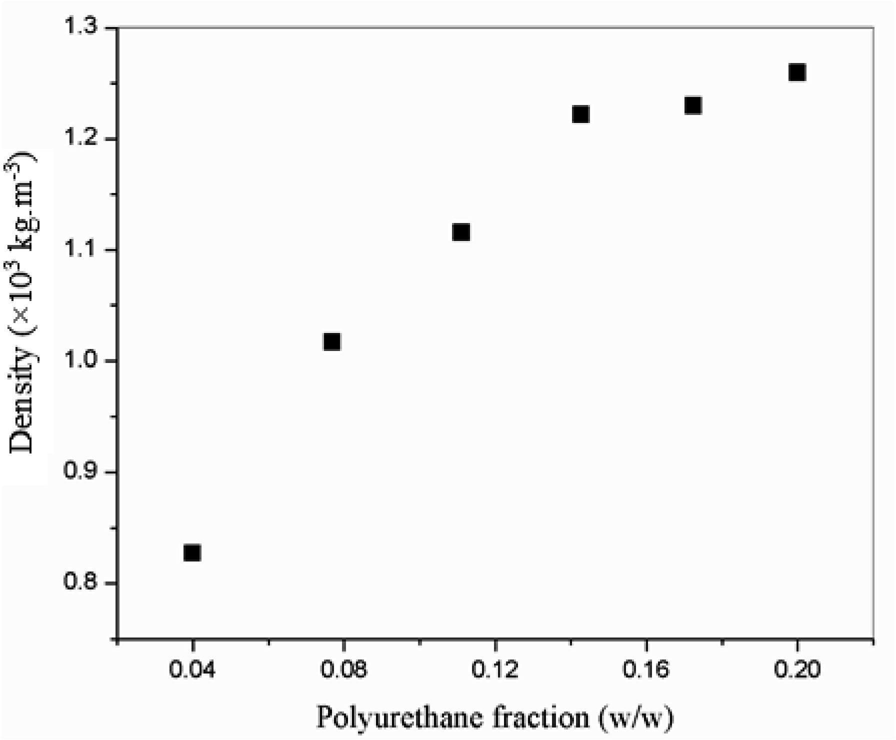


Figure 6. Composite density as polyurethane fraction (w/w) function

The density data shows that this composite belong to the high-density particle board category because it has a density more than 0.80 g/cm^3 in accordance with the standard designation 1554-67 by American Society [26]. Additionally, the composite has also meet the Indonesia National Standard of 03-2105-2006 where particleboard density having values in the range of 400 - 900 kg.m^{-3} [27].

3.3. Water Absorption

The value of water absorption was measured to determine the ability of leaves teak waste composite to absorb the water after 24 hours of soaking. The highest water absorption rate reaches 107.35% in composites with polyurethane fraction of 0.04 (w/w). After soaking for 24 hours, the composite undergoes thick development until the composite becomes cracked and broken due to the cavities emerging between matrix and filler particles. These cavities then absorb a lot of water and further it can affect the dimensional stability of the composite. Additionally, the hydrophilic filler also absorbs a lot of water during the immersion.

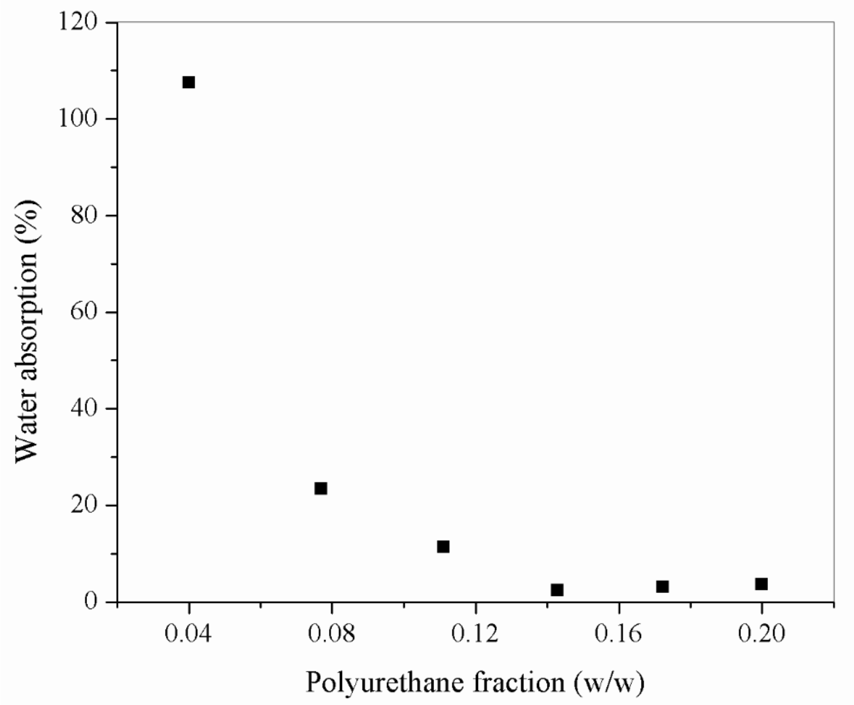


Figure 7. Water absorption of composite tends to zero for larger polyurethane fraction (w/w)

Figure 7 shows the water absorption value was decreasing with the increase of polyurethane fraction. The large fraction tends to produce composites with lower water absorption. It can be explained as the cavities among the teak leaves has been fulfilled by large amount of the polyurethane. Hence, the amount of water adsorbed decreases.

The absorption value of the leaves teak waste composite produced is better than to that of several leaves waste composites having been developed by several authors, such as Al-Sulaiman et al. produced palm fiber reinforced composites with the water absorption is between 2.3 to 5.8% [28], and Batiancela et al. developed a wood particle from tea leaves waste and obtained the water absorption about 23-40% [29]. Therefore, in accordance to the several properties above, this composite has properness in competing the woods.

4. Conclusion

We have successfully developed a teak leaves-waste composite utilized as particleboard. The mechanical and physical properties of the composites have been investigated. The test results showed a composite compressive strength attains 38.5 MPa. The highest density is 1261 kg.m^{-3} that is in accordance to the American Society standard. The lowest value of water absorption is 1.38%. In addition, we have successfully approximated the van der Waals interaction with the spectrum shifts of the FTIR spectroscopy as a confirmation of the compressive strength enhancement.

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Declaration of conflicting interests

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Reviewer: 1

Comments to the Author
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2. Other applications of POLYURETHANE should be cited: https://doi.org/10.1007/s42114-017-0009-y; Macromol. Chem. Phys., 2019, 220, 1800567; https://doi.org/10.1007/s42114-018-0067-9; Polymer, 2018, 158, 381-390; Macromol. Mater. Eng., 2019, 304, 1900010.
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Also the discussion about water absorption is very limited, and I would say naïve, please be more specific.

The Conclusions are basically a repetition of the Abstract, which is not appropriate. Please indicate at which point you are and where do you suppose to be able to go from there.

In Figure 1, did you test only one sample per category? Also in terms of the significance of the study this appears quite limited. Please comment in the text on the possible reliability of results.

In Figure 2, the writings “matrix” and “filler” are hardly visible. Please rework it.

Also, please check thoroughly your level of English, since many sentences are not well understandable. A number of issues are evidenced in the review.

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Associate Editor(s)' Comments to Author:

Associate Editor: OGIHARA , Shinji
Comments to the Author:
(There are no comments.)

Response to Reviewer(s)' Comments to manuscript entitled: MECHANICAL AND PHYSICAL PROPERTIES OF TEAK LEAVES WASTE/POLYURETHANE COMPOSITES FOR PARTICLE-BOARD APPLICATION.

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I think so. We can also use a DSC analysis test to evaluated the interaction between polymers and fillers as has been investigated by Dong et al (2018). However, in this study we only focus on characterization using FT-IR.

Dong, M. *et al.* (2018) ‘Thermoplastic polyurethane-carbon black nanocomposite coating: Fabrication and solid particle erosion resistance’, *Polymer*. Elsevier Ltd, 158, pp. 381–390. doi: 10.1016/j.polymer.2018.11.003.

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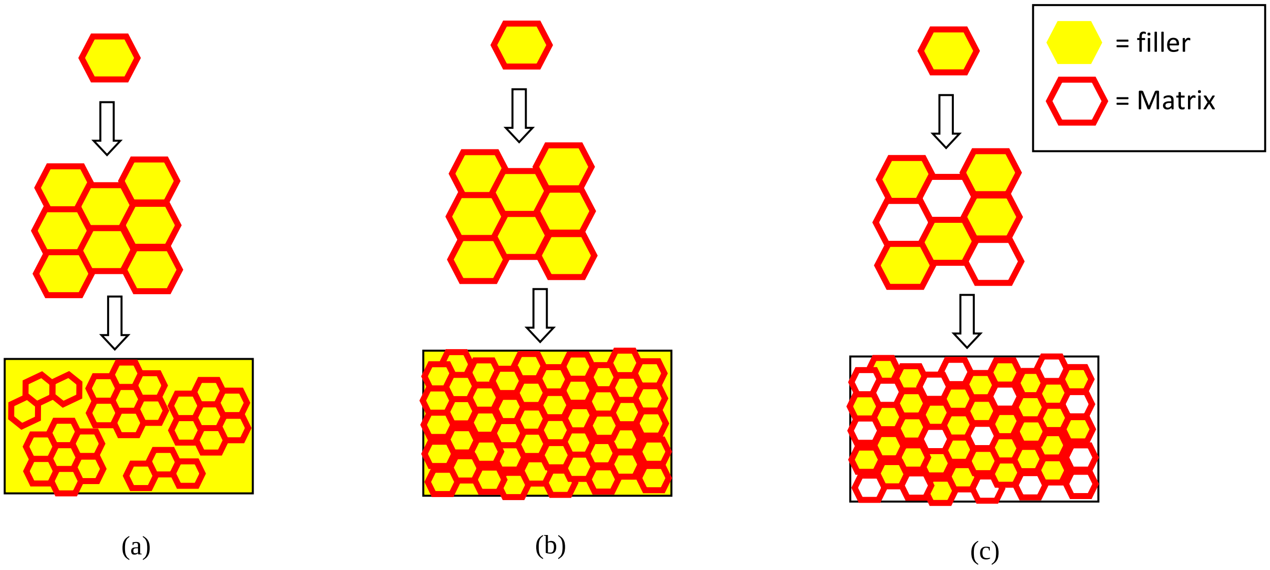

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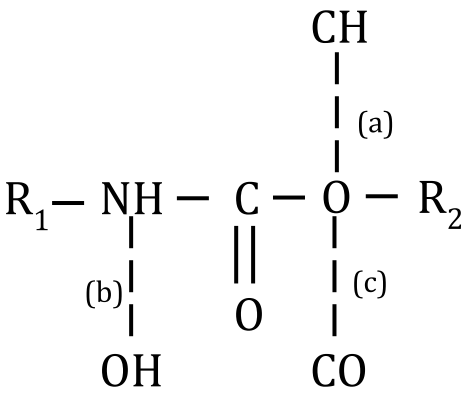

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Polyurethane is a thermosetting polymer which does not melt when heated, more resistant to acids and bases, has a high adhesive power due to the presence of highly reactive isocyanate functional groups. Furthermore, the addition of nanoparticles in polyurethane can improve thermal stability and mechanical properties of composites.

You do not present any photographs either of cracked samples or of the panels obtained, and this is a main limitation. I may understand they might not look amazing, but this is research after all. Please add some photographic documentation.

Sample testing is handled directly by the testing lab and at that time we did not take and ask for documentation. We have a composite sample documentation and the results of the water absorption test.

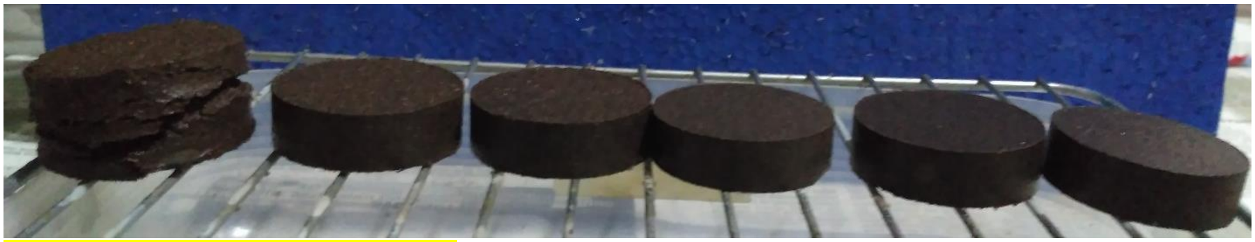

Figure 1. Sample after soaking 24 hour

Figure 2. Sample Teak leaves waste composite/polyurethane

Also the discussion about water absorption is very limited, and I would say naïve, please be more specific.

we have reviewed this section again and have corrected it.

Water absorption values were measured to determine the ability of teak leaf composite to absorb water after 24 hours of soaking. The highest water absorption rate reaches 107.35% in composites with a polyurethane fraction of 0.04 (w/w). After being immersed for 24 hours, the composite undergoes thick development until the composite becomes cracked and broken due to the cavity that appears between the matrix and the filler particles absorbs a lot of water and can further affect the stability of the dimensions of the composite. In addition, teak leaves as a hydrophilic filler also absorb a lot of water during immersion.

Figure 7 shows the water absorption value was decreasing with the increase of polyurethane fraction. Large fractions tend to produce composites with lower water absorption. the lowest water absorption was achieved in samples that had the most optimal compressive strength at fraction 0.14 (w/w). The cavity formed is very small because the pressing process in the fabrication of composites and filler particles is distributed evenly on the matrix so that only a little water is absorbed

The Conclusions are basically a repetition of the Abstract, which is not appropriate. Please indicate at which point you are and where do you suppose to be able to go from there.

we have fixed it by including what we have obtained and further research plans to improve the composite. We have successfully developed a teak leaves-waste composite utilized as particleboard. The mechanical and physical properties of the composites have been investigated. The test results showed a composite compressive strength attains 38.5 MPa. The highest density is 1261 kg.m^{-3} that is in accordance to the American Society standard. The lowest value of water absorption is 1.38%. In addition, we have successfully approximated the van der Waals interaction with the spectrum shifts of the FTIR spectroscopy as a confirmation of the compressive strength enhancement. However, the properties of organic composite are hygroscopic so they are vulnerable to being the target of fungal growth and susceptible to fire as an application in building fields. These problems will be focus of further research by applying coating technology approaches.

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Page 14 line 5: when you say about 0.08 density, you mean the relative amount, I suppose. Please rephrase. We have checked and replace it.

The density in samples with a composition of 0.08 (w/w) accordance with teak wood that has been studied by Wahyudi *et al.*, in the range of 1000-1200 kg / m3 with an average value of 1160 kg.m^{-3} [17].

Regards,

Masturi

MECHANICAL AND PHYSICAL PROPERTIES OF TEAK LEAVES WASTE/POLYURETHANE COMPOSITES FOR PARTICLE-BOARD APPLICATION

Masturi[1,*], **WN Jannah**[1], **RM Maulana**[1], **T Darsono**[1], **Sunarno**[1], **S Rustad**[2]
[1] Physics Department of State University of Semarang,
D7 Building 2nd Floor, Sekaran, Gunungpati, Semarang, Indonesia
[2] Dian Nuswantoro University, Semarang
Nakula Street, Semarang, Indonesia

*Email: masturi@mail.unnes.ac.id

Abstract. A teak leaf waste/polyurethane composite has been made for particle-board application. Some fraction variations were performed on the use of polyurethane as a matrix with a range of 0.04-0.20 (w/w). Mechanical and physical properties have been tested on the composites produced. The test results showed the highest compressive strength of the sample reaches 38.5 MPa for polyurethane fraction of 0.14 (w/w). The composite has a density of 1261 kg.m^{-3} which is in accordance with the result of density on teak. The physical properties have been also investigated and it was found that the lowest value of water absorption is 1.38%. This result indicated that composites can be used as an eco-friendly industrial scale instead of wood.

Keywords: composite, polyurethane, teak leaf, mechanical properties.

1. Introduction

In the last few decades, the wood demand in the world always grows with the total demand for 2020 is projected attain to 515 million m^3 [1]. One of the most likely efforts to fulfill the high demand for wood is composite engineering. Composite engineering promises organic and inorganic waste utilization as a synthesized-material having excellent properties relating the woods, such as mechanical strength, water absorption and so on [2]–[6]. Furthermore, the composite engineering is also a solution for the raising of environment awareness caused by the high deforestation in several countries [2-3]. In addition, the utilization of natural waste and fibers for some composites has been widely studied and developed from various countries including Indonesia, Malaysia, Thailand, India, Brazil and Japan [2-4].

In recent years, the use of composites is very widespread in the electronics, automotive and aviation industries, construction field, multipurpose applications such as particle boards, energy storage, heat and electricity insulators, ship bodies and aircraft, organic field effect-transistors, antennas, etc. due to their high mechanical strength , good thermal stability, anti-corrosion, high dielectric constant, light weight and easy to produce large scale [7-13]. Some research relating to the polymers and plastics is focused on developing fibers, plastics, adhesives, composites, textiles, paper from renewable resources derived mostly from agricultural wastes containing lignocellulosic [4]. One of the efforts done is maximizing some natural fibers such as sisal, hemp, bamboo and agricultural wastes such as leaves, coconut fibers, rice husk, peanut shells and straw also can be used as composite reinforcements [5-6]. Most developing countries like Indonesia, some agricultural wastes usually are used for compost production. Composites of these natural fibers have several advantages such as being easy to process, environmentally, good acoustic properties, low energy processes, good mechanical and thermal properties, water resistance, non-corrosive, non-toxic and naturally degradable [7].

Further, several studies related to the leaves waste composites have been done, such as Masturi *et al*. developed composite from leaves waste and paper waste using polyvinyl acetate as matrix and the compressive strength reaches to 45.60 MPa [2]. Campanella *et al*. successfully made a composite of Northern Red Oak leaves with plant oil-based resins [6], while Thakur *et al.* have also developed biopolymer composite using ligno-cellulosic of *Hibiscus Sabdariffa* [14]. A different method has been performed by Nongman *et al.* having created a composite by lamination method of banana leaf fiber and adhesive [5]. Senthilkumar *et al*. were fabricated pineapple leaf fiber / Polyester composites which have a tensile strength reached to 33.13 MPa. Good mechanical properties related to the high content of cellulose in pineapple leaf fibers so that it can transfer a sizable load from the matrix to the fiber effectively [15]. In addition, Melkanu *et al*., have investigated the mechanical properties and water absorption in sisal / Polyester fiber composites with treated fibers and without alkaline sodium hydroxide treatment. Compressive strength in sisal fibers treated better than untreated fibers with low water absorption advantages [16].

In this work, we developed a composite of teak leaves waste with polyurethane matrix. The teak leaves have been chosen since its utilization only as compost having a low added value as well as its availability is very abundant in Indonesia. Polyurethane as a thermosetting polymer was selected owing does not melt when heated, more resistant to acids and bases, has a high adhesive power due to the presence of highly reactive isocyanate functional groups, has good erosion resistance so it can be used as an anti-erosion film and coating technology [17], has an isotropic and controllable electromagnetic response, and has a negative permittivity value which is potentially applied to the electronics field [18], [19]. Furthermore, the addition of nanoparticles in polyurethane can improve thermal stability and mechanical properties of composites. Polyuretane is also used as a textile coating on inkjet-printed flexible and washable field-effect transistors (FETs) on textile technology [20]. The aim of this study is to produce a Teak leaves waste composite having a high compressive strength and low water absorption making a chance to be used on an eco-friendly industrial scale.

2. Materials and Methods

The teak leaves waste of Semarang Indonesia was used, while isocyanates and polyol as polyurethane (PU) polymerization materials were purchased in Multi Kimia Inc, Semarang, Indonesia.

For first, the leaves waste was dried and crushed using a blender machine to form a powder. Then, the powder was sieved using 700 mesh. In the other hand, the isocyanates and polyol were mixed using magnetic stirrer for 5 minutes with ratio 1:1 to form polyurethane and mixed with the teak leaves waste powder with varied composition while the leaves teak powder was kept at 12 g weight. The PU/teak leaves waste mixture was stirred using mixer machine to homogenize the mixture, then pressed into a cylindrical mold at pressure of 5 MPa for 15 minutes. Having been pressed, the composite was removed from the mold and dried for 24 hours at room temperature.

The investigation to the content of the teak leaves powder cellulose bonding to polyurethane and detecting spectral changes in teak leaves waste composite was performed using FTIR Spectrometer Frontier type (Perkin Elmer) in a spectral range of 400-4000 cm^{-1}. Meanwhile, the identification of the mechanical properties was measured by a compressive strength test using Sociedad Espanola Anonima Ibertest instrument. In this test, the sample is cut into cubic shape and was measured using testing machine. In addition, to ensure the water resistance of the composite, the identification of the physical properties was performed by density measurement, further, water absorption test was done by soaking this sample in the water for 24 h to obtain the weight of water absorbed into the composite compared to the initial weight.

3. Results and Discussion

3.1. Compressive Strength Test

Teak leaves waste/polyurethane composite have been fabricated successfully as shown **Figure 1.a. Figure 1.b.** shows the effect of polyurethane addition on composite compressive strength. The highest compressive strength is achieved as 38.5 MPa for polyurethane fraction of 0.04 (w/w). This resulting strength is 86% higher than that of polyurethane fraction of 0.04 (w/w). The increase in compressive strength is closely related to the interaction between the surface of the particles and the polymer. The composites with 0.04 and 0.08 (w/w) have relatively low compressive strength regarding there are still many teak leaf particles not binding to the polymer matrix due to. When the polyurethane fraction was added to 0.14 (w/w), the composite attains the highest compressive strength. This shows that all matrices of the polymer exactly interact with all the teak powder particles so that the composite becomes more compact and withstands higher loads such as the mechanisms in **Figure 2**.

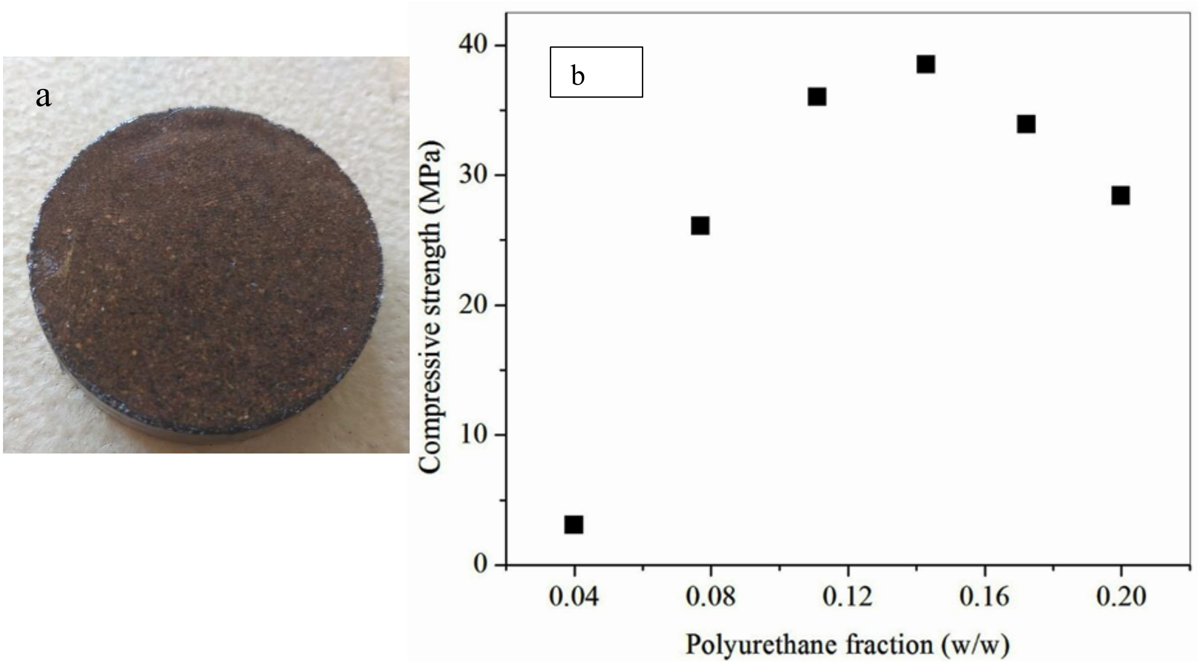

Figure 1.a Teak leaves waste/polyurethane composite**, 1.b.** Compressive strength of teak leave waste composite as polyurethane content (w/w) and the compressive strength attains at maximum for 0.14 fraction (w/w).

Figure 2(a) illustrates the bonding of leaves teak particles and polyurethane polymers at a mass varied of 0.04-0.08 (w/w). Preliminarily all polymer matrices are not filled by particles. Having been pressed to form a composite, some of the diffused particles do not bind to the matrix. The presence of leaf particles not binding to the matrix affects the ability of the composite in holding the load so it becomes easily fragile and broken. In **Figure 2(b)**, a composite with polyurethane content of 0.14 (w/w) shows that all teak leaf particles are precisely bind to a polymer matrix, that is called as optimum fraction of polyurethane in this composite. This matrix bond is capable of forwarding the load in all directions so that the resulting composite is stronger and withstand maximum load [21]. This is as reported by Chen *et al.* in his research on composite polyurethane / Graphene Oxide (GO). The addition of GO in the Polyurethane matrix is spread uniformly and can improve the mechanical properties of the composite up to 167% [22]. Further, as **Figure 2(c)** the composites with a polyurethane content of 0.17-0.20 (w/w) show some parts of polyurethane not binding to the filler, so that many voids from polyurethane polymer appear and it result in the decreasing of the compressive strength [23-24].

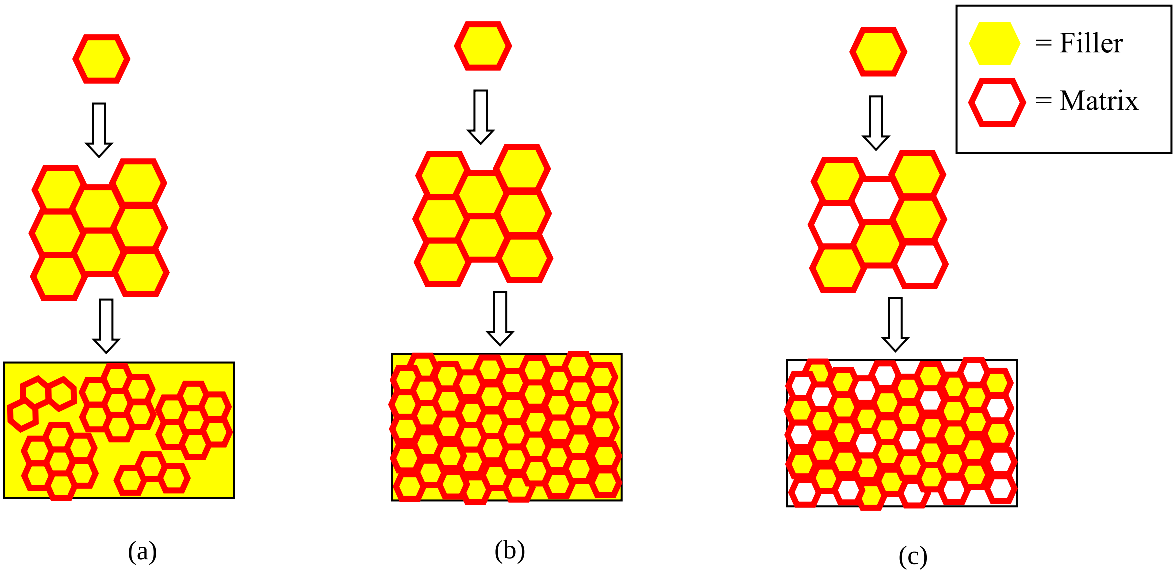

Figure 2. Filler particle distribution scheme and polymer matrix. (a) low compressive strength, (b) high compressive strength, (c) low compressive strength

Using the explanation above, the added mass of polyurethane can improve the composite compressive strength. However, the larger mass otherwise makes the reduction of compressive strength, as in **Figure 1** the maximum strength is the composite with the polyurethane fraction of 0.14 (w/w). According to Jaya *et al.*, polyurethane is a hydrophobic matrix and teak leaf powder is a hydrophilic filler so that when the polymer was added much more than the filler there will be a very poor adhesion force between surfaces [25]. Further, the raising of the strength until certain fraction is related to the maximum contact amount between the polymer and solid waste particle [26].

The most important interaction in the composite is predominantly emerged between the polyurethane and the cellulose in teak leaves. According to Alemdar and Sain, the cellulose in plant fibers has important role in reinforcing material in composite applications [27]. The presence of cellulose in teak leaves can be investigated through the FTIR spectrum in **Figure 3.**

Figure 3(a) and **Figure 3(b)** show the FTIR spectrum of the pure cellulose obtained from Ramirez-Flores *et al.* [28] and the teak leaves respectively, with the comparison can be shown in **Table 1**. The FTIR spectrum of pure cellulose and leaves teak powder cellulose's has a significant difference at peak 1631 cm^{-1}. The peak of 1631 cm^{-1} indicates absorption band of hydrogen forming a hydrogen bridge from hydroxyl groups and oxygen atoms from glucose monomer and usually found at a cellulose fiber [29]. The peak of 1631 cm^{-1} is not found at pure cellulose but it found at cellulose of natural fibers [30–33].

The peak of 2929 and 2300 cm^{-1} indicates a strong C-H bond in cellulose polymers [34]. While, peak of 3401 cm^{-1} shows the stretching of the O-H group bonds [35]. Further, the peak of 780 cm^{-1} and 1438 cm^{-1} shows the C-H bonds connecting the glucose units of cellulose. The peak at 1234 cm^{-1} indicates that there is a C-O-C group which is the structure of lignin. The absorption area occurring at 1076 cm^{-1} indicates the presence of a C-O bond in the cellulose component [25].

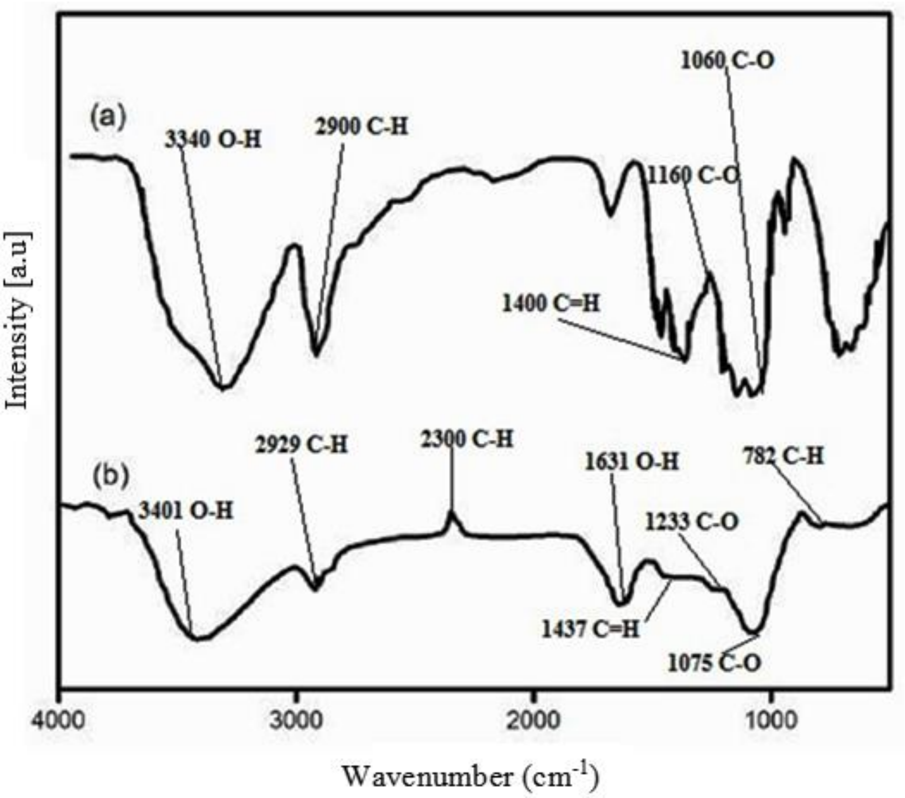

Figure 3. Spectrum of cellulose of: (a) pure cellulose obtained from Ramirez-Flores [13] b. cellulose of teak leaves powder in this work where many peaks of both celluloses have more similarities.

Using the FTIR analysis, it can be shown that cellulose and lignin components are found in the teak leaves. This leave can compete the conventional fibers such as glass fiber regarding its high performance as reinforcing material for thermosetting and thermoplastic matrices with their naturally degradable superior properties [36].

Table 1. Comparison of the peak absorption at pure cellulose and cellulose of teak leaves powder

Bond	Wavenumber (cm^{-1})	
	Pure cellulose (cm^{-1}) [15]	Cellulose of teak leaves powder (cm^{-1})
O-H	3340 – 3500	3401
C-H	2800 – 2920	2929
C-O-C	1160	1233
C-O	1035 – 1060	1075
C=H	1400	1437

The interaction arising between the teak leaves and the polymer is a van der Waals interaction. This interaction was confirmed by several FTIR spectrum shifts as shown in **Figure**

4. The shifts in spectrum are appeared due to the vibrating atoms as a result of the energy of disruption of atoms closing each other.

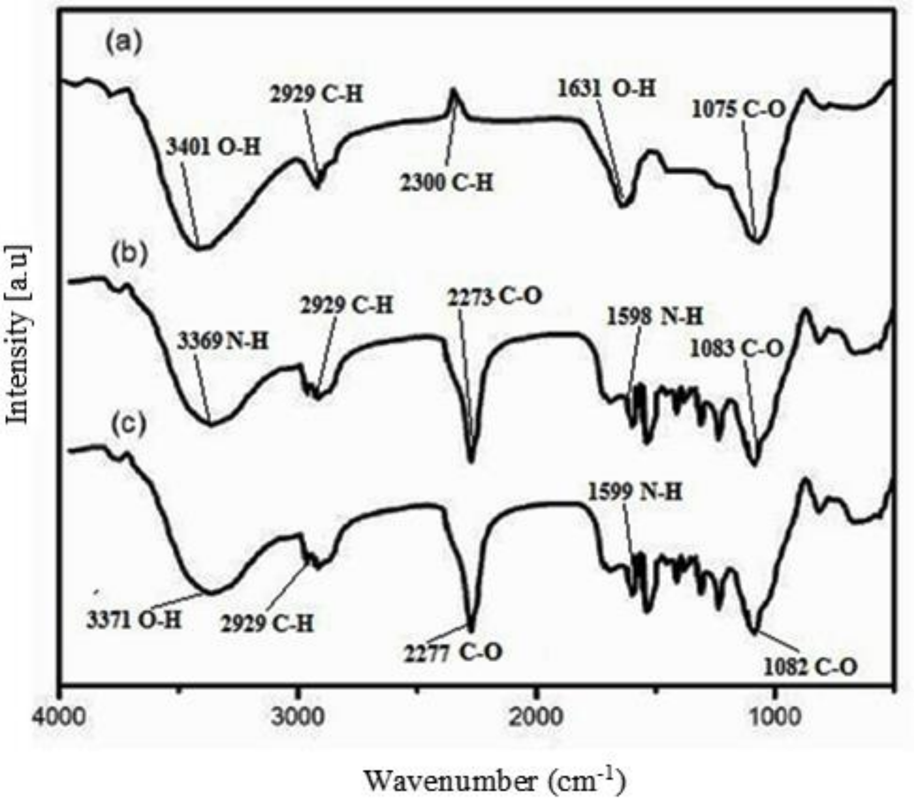

Figure 4. FTIR spectrum a. teak leaves, b. polyurethane, c. PU/teak leave composite

Polyurethane has two functional groups, i.e. carbonyl and amine groups. The spectrum shift starts to occur of 3369 cm^{-1} to 3371 cm^{-1} indicates the O-H bond. This shift is due to the amine group of the polyurethane was disturbed by hydrogen bonding in the hydroxyl group of leaves and forming a hydrogen bridge [37]. Further, the shift of 2273 cm^{-1} to 2277 cm^{-1} shows the carbonyl group of polyurethane undergoes a spectral shift. This is due to the disturbance of the C-H leaves bonding to the carbonyl group forming the O-H bridges. The peak at 1598 cm^{-1} shifting to 1599 cm^{-1} also indicates the amine group in polyurethane disturbed by the O-H bonds of the teak leaves. Furthermore, the shift also occurs at the carbonyl peak of 1083 cm^{-1} to 1082 cm^{-1} due to disturbance of the leave's C-O bond of the peak of 1075 cm^{-1} (Figure **5**). Similar results were confirmed by a previous work that PVA-sepiolite composite which derived its van der Waals interactions between silanol groups and carbon C-O and shifted from 1088 cm^{-1} to 1085 cm^{-1} [38].

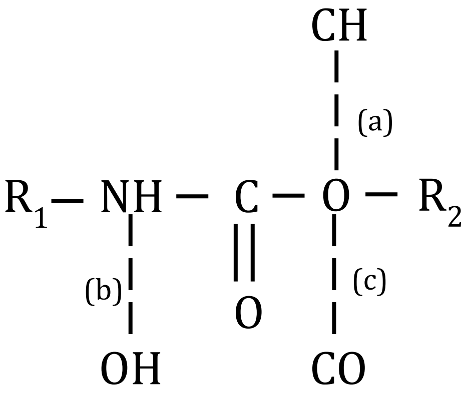

Figure 5. Van Der Waals interactions on the teak leaf waste composites, dashed lines illustrate the Van der Waals interaction area of (a) CH and carbonyl group (b) OH and amine group, and (c) CO and carbonyl group.

3.2. Density Test

Figure 6 shows the density of composite samples with polyurethane variation ranges from 825 to 1261 kg.m^{-3}. The density value is proportional to the increase in PU content. The lowest density value is found in the sample with a polyurethane fraction (w/w) of 0.04, while the highest density value is in the sample with 0.20 of polyurethane fraction (w/w). It can be explained when the polyurethane polymer content rises and the filler is fixed, then the number of link produced by the polymer increases. Hence, the increased links result in stronger bonds to bind the filler so that it gets closer and density increases [39].

The density in samples with a composition of 0.08 (w/w) accordance with teak wood that has been studied by Wahyudi *et al.*, in the range of 1000-1200 kg m^{3} with an average value of 1160 kg.m^{-3} [30]. In this study, the resulted-density value is higher than that of Martins *et al.* work producing particle board with density of 510 - 570 kg.m^{-3} [31]. The composites produced in this work have a similar density to the results of the Raju *et al.* producing some composites with densities ranging from 1100 to 1320 kg.m^{-3} [32]. Further, Butylina *et al.* have produced wood fibre-polypropylene composite having density ranged from 1040 to 1140 kg.m^{-3} [33].

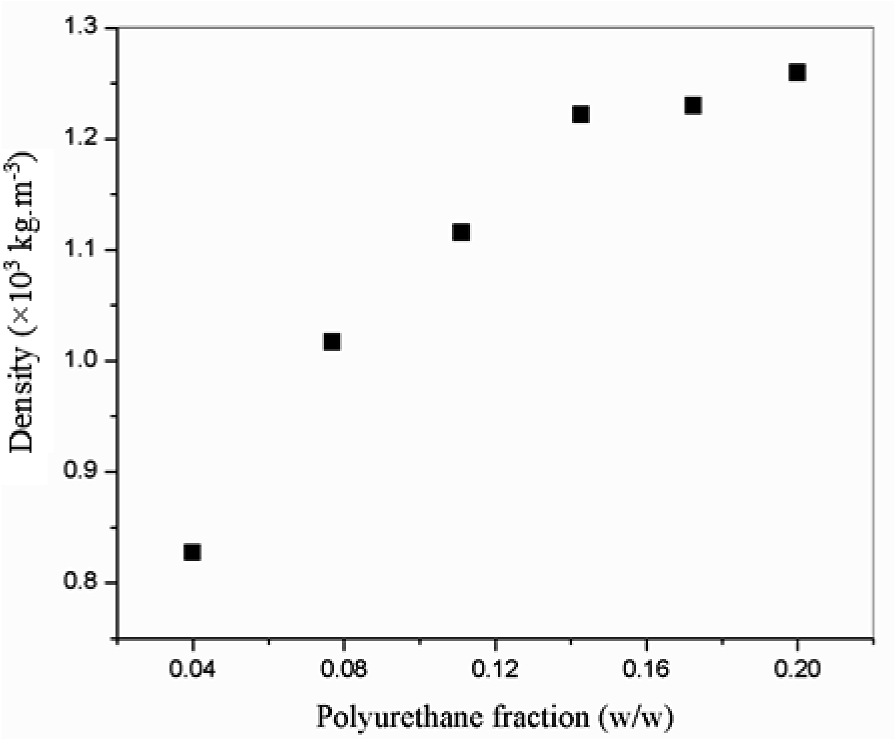

Figure 6. Composite density as polyurethane fraction (w/w) function

The density data shows that this composite belong to the high-density particle board category because it has a density more than 0.80 g/cm^3 in accordance with the standard designation 1554-67 by American Society [40]. Additionally, the composite has also meet the Indonesia National Standard of 03-2105-2006 where particleboard density having values in the range of 400 - 900 kg.m^{-3} [41].

3.3. Water Absorption

Water absorption values were measured to determine the ability of teak leaf composite to absorb water after 24 hours of soaking. The highest water absorption rate reaches 107.35% in composites with a polyurethane fraction of 0.04 (w/w). After being immersed for 24 hours, the composite undergoes thick development until the composite becomes cracked and broken due to the cavity that appears between the matrix and the filler particles absorbs a lot of water and can further affect the stability of the dimensions of the composite. In addition, teak leaves as a hydrophilic filler also absorb a lot of water during immersion.

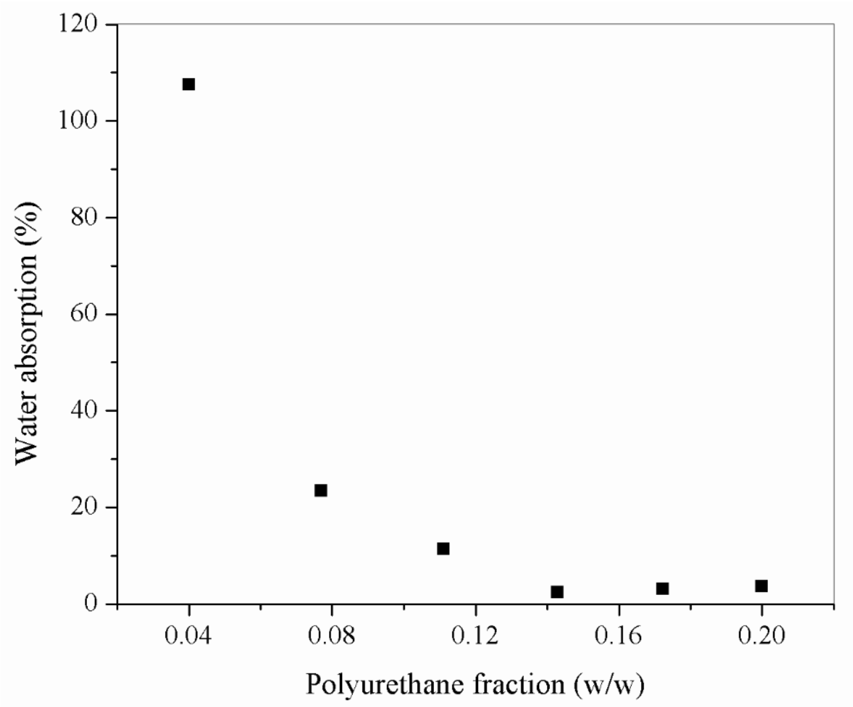

Figure 7. Water absorption of composite tends to zero for larger polyurethane fraction (w/w)

Figure 7 shows the water absorption value was decreasing with the increase of polyurethane fraction. Large fractions tend to produce composites with lower water absorption. the lowest water absorption was achieved in samples that had the most optimal compressive strength at fraction 0.14 (w/w). The cavity formed is very small because the pressing process in the fabrication of composites and filler particles is distributed evenly on the matrix so that only a little water is absorbed.

The absorption value of the leaves teak waste composite produced is better than to that of several leaves waste composites having been developed by several authors, such as Al-Sulaiman *et al.* produced palm fiber reinforced composites with the water absorption is between 2.3 to 5.8% [42], and Batiancela *et al.* developed a wood particle from tea leaves waste and obtained the water absorption about 23-40% [43]. Therefore, in accordance to the several properties above, this composite has properness in competing the woods.

4. Conclusion

We have successfully developed a Teak leaves-waste composite utilized as particleboard. The mechanical and physical properties of the composites have been investigated. The test results showed a composite compressive strength attains 38.5 MPa. The highest density is 1261 kg.m^{-3} that is in accordance to the American Society standard. The lowest value of water absorption is 1.38%. In addition, we have successfully approximated the van der Waals interaction with the spectrum shifts of the FTIR spectroscopy as a confirmation of the compressive strength enhancement. However, the properties of organic composite are hygroscopic so they are vulnerable to being the target of fungal growth and susceptible to fire as an application in building fields. These problems will be focus of further research by applying coating technology approaches.

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Declaration of conflicting interests

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Advanced Composites Letters - Decision on Manuscript ID ACM-20-0085.R1

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28 June 2020

Dear Dr. Masturi:

Manuscript ID ACM-20-0085.R1 entitled "MECHANICAL AND PHYSICAL PROPERTIES OF TEAK LEAVES WASTE/POLYURETHANE COMPOSITES FOR PARTICLE-BOARD APPLICATION" which you submitted to Advanced Composites Letters, has been reviewed. The comments of the reviewer(s) are included at the bottom of this letter.

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Reviewer(s)' Comments to Author:

Reviewer: 2

Comments to the Author
The problem of this paper is basically in English expression. It needs to be completely rewritten, and checked for spelling and sense of the sentences. There has been limited improvement from the previous version.
Another big problem concerns Table 6: I don't understand why there is no linearity between density and amount of polyurethanes. Can you please provide some optical microscope images, e.g., to show how the density relates to the amount of resin?

Reviewer: 1

Comments to the Author
It can be recommended for publishing.

Associate Editor(s)' Comments to Author:

Associate Editor: OGIHARA , Shinji
Comments to the Author:
(There are no comments.)

Response to Reviewer(s)' Comments to manuscript entitled: MECHANICAL AND PHYSICAL PROPERTIES OF TEAK LEAVES WASTE/POLYURETHANE COMPOSITES FOR PARTICLE-BOARD APPLICATION, about the density and the requirement of SEM images, and the problem English expression in the manuscript.

Dear Editor,

Thank you very much for the comment. Below is the response to the reviewer’s comment about density and the requirement of SEM images, and we have included them in the revised-manuscript. We also have made up the English expression in the manuscript.

For the linearity between density and number of polyurethanes. We have checked using linear fitting and it was obtained the linearity relation as: as; $y = 2.6378x + 0.7855$, where x and y are polyurethane fraction and composite density respectively, with $R^2 = 0.91157$ as shown in Figure 6. This explanation has been added to the manuscript.

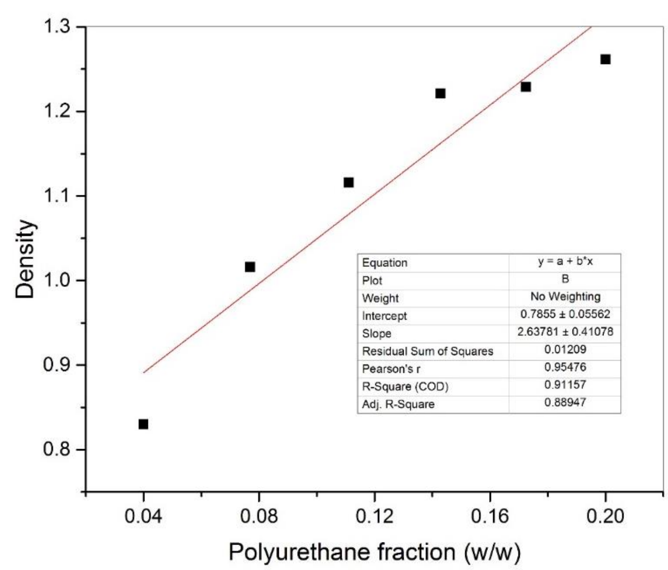

Figure 6. Composite density as polyurethane fraction (w/w) function, and it was linearity relation as: $y = 2.6378x + 0.7855$, where x and y are polyurethane fraction and composite density respectively, with $R^2 = 0.91157$.

We also show the SEM images of three composites, i.e. 1st, 3rd and 5nd samples to confirm the density relation to the polyurethane content. It was obtained that the higher the polyurethane content, the denser the distance filler mixture and the matrix. This explanation has been also included at the end of Result and Discussion of this manuscript.

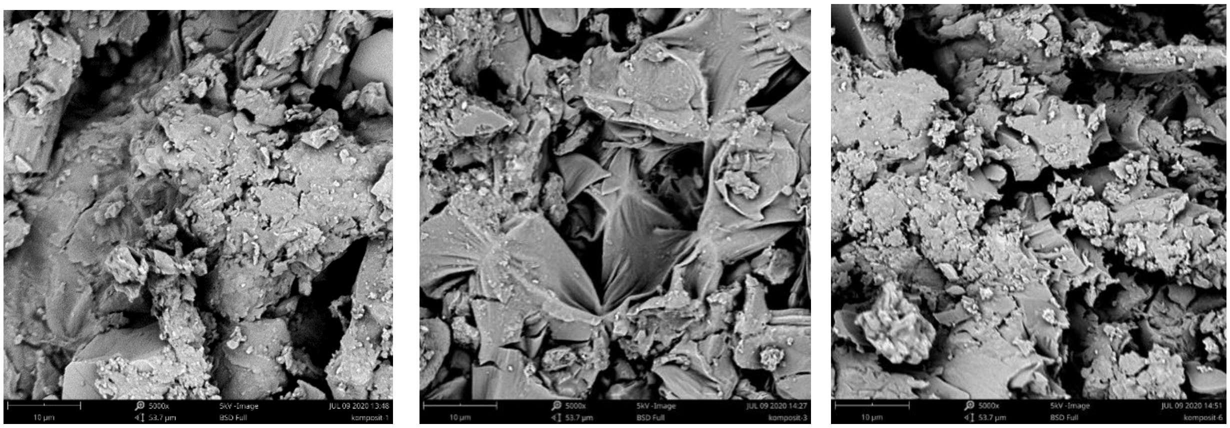

Figure 8. SEM of composite (a) 0.04 (w/w); (b) 0.12 (w/w); (c) 0.20 (w/w).

Thank you.

Regards,

Masturi

MECHANICAL AND PHYSICAL PROPERTIES OF TEAK LEAVES WASTE/POLYURETHANE COMPOSITES FOR PARTICLE-BOARD APPLICATION

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Abstract. A teak leaf waste/polyurethane composite has been made for particle-board application. Some fraction variations are performed on the use of polyurethane as a matrix with a range of 0.04-0.20 (w/w). Mechanical and physical properties have been tested on the composites produced. The test results showed the highest compressive strength of the sample reaches 38.5 MPa for polyurethane fraction of 0.14 (w/w). The composite has a density of 1261 kg.m^{-3} which is in accordance with the result of density on teak. The physical properties have been also investigated and it was found that the lowest value of water absorption is 1.38%. This result indicated that composites can be used as an eco-friendly industrial scale instead of wood.

Keywords: composite, polyurethane, teak leaf, mechanical properties.

1. Introduction

In the last few decades, total global wood demand for 2020 is projected to reach 515 million m^3 [1]. Composite technique is one of the possible ways to fulfill the demand. Composite engineering promises organic and inorganic waste utilization as a synthesized-material having excellent properties relating the woods, such as mechanical strength, water absorption, density, etc. [2-6]. Furthermore, the composite engineering is also a solution for the raising of environment awareness caused by the high deforestation in several countries [2-3]. In addition, the utilization of natural waste and fibers for some composites has been widely studied and developed from various countries including Indonesia, Malaysia, Thailand, India, Brazil and Japan [2-4].

In recent years, the use of composites is very widespread in the electronics, automotive and aviation industries, construction field, multipurpose applications such as particle boards, energy storage, heat and electricity insulators, ship bodies and aircraft, organic field effect-transistors, antennas, etc. due to their high mechanical strength , good thermal stability, anti-corrosion, high dielectric constant, light weight and easy to produce large scale [7-13]. Several study about the polymers and plastics is focused on developing fibers, plastics, adhesives, composite textiles, paper from renewable resources derived mostly from agricultural wastes containing lignocellulosic [4]. Considering that agricultural waste is only used for compost production In most developing countries such as Indonesia, researcher also focused on utilization some natural fibers such as sisal, hemp, bamboo and agricultural wastes, e.g. leaves, coconut fibers, rice husk, peanut shells and straw as filler in polymer composite [5-6].Composites of these natural fibers have several advantages such as being easy to process, environmentally, good acoustic properties, low energy processes, good mechanical and thermal properties, water resistance, non-corrosive, non-toxic and naturally degradable [7].

Further, several studies related to the leaves waste composites have been done, such as Masturi *et al*. developed composite from leaves waste and paper waste using polyvinyl acetate as matrix and the compressive strength reaches to 45.60 MPa [2]. Campanella *et al*. successfully made a composite of Northern Red Oak leaves with plant oil-based resins [6], while Thakur *et al.* have also developed biopolymer composite using ligno-cellulosic of *Hibiscus Sabdariffa* [14]. A different method has been performed by Nongman *et al.* having created a composite by lamination method of banana leaf fiber and adhesive [5]. Senthilkumar *et al*. were fabricated pineapple leaf fiber / Polyester composites which have a tensile strength reached to 33.13 MPa. Good mechanical properties related to the high content of cellulose in pineapple leaf fibers which can transfer a sizable load from the matrix to the fiber effectively [15]. In addition, Melkanu *et al*., have investigated the mechanical properties and water absorption in sisal / Polyester fiber composites with treated fibers and without alkaline sodium hydroxide treatment. Compressive strength in sisal fibers treated better than untreated fibers with low water absorption advantages [16].

In this work, we developed a composite of teak leaves waste with polyurethane matrix. The teak leaves have been chosen since its utilization only as compost having a low added value as well as its availability is very abundant in Indonesia. Polyurethane as a thermosetting polymer was selected owing does not melt when heated, more resistant to acids and bases, has a high adhesive power due to the presence of highly reactive isocyanate functional groups, has good erosion resistance which can be used as an anti-erosion film and coating technology [17], has an isotropic and controllable electromagnetic response, and has a negative permittivity value which is potentially applied to the electronics field [18-19]. Furthermore, the addition of nanoparticles in polyurethane can improve thermal stability and mechanical properties of composites. Polyurethane is also used as a textile coating on inkjet-printed flexible and washable field-effect transistors (FETs) on textile technology [20]. The aim of this study is to produce a Teak leaves waste composite having a high compressive strength and low water absorption making a chance to be used on an eco-friendly industrial scale.

2. Materials and Methods

The teak leaves waste of Semarang Indonesia was used, while isocyanates and polyol as polyurethane (PU) polymerization materials were purchased in Multi Kimia Inc, Semarang, Indonesia.

For first, the leaves waste was dried and crushed using a blender machine to form a powder. Then, the powder was sieved using 700 mesh. In the other hand, the isocyanates and polyol were mixed using magnetic stirrer for 5 minutes with ratio 1:1 to form polyurethane and mixed with the teak leaves waste powder with varied composition while the leaves teak powder was kept at 12 g weight. The PU/teak leaves waste mixture was stirred using mixer machine to homogenize the mixture, then pressed into a cylindrical mold at pressure of 5 MPa for 15 minutes. Having been pressed, the composite was removed from the mold and dried for 24 hours at room temperature.

The investigation to the content of the teak leaves powder cellulose bonding to polyurethane and detecting spectral changes in teak leaves waste composite was performed using FTIR Spectrometer Frontier type (Perkin Elmer) in a spectral range of 400-4000 cm^{-1}. Meanwhile, the identification of the mechanical properties was measured by a compressive strength test using Sociedad Espanola Anonima Ibertest instrument. In this test, the sample is cut into cubic shape and was measured using testing machine. In addition, to ensure the water resistance of the composite, the identification of the physical properties was performed by density measurement, further, water absorption test was done by soaking this sample in the water for 24 h to obtain the weight of water absorbed into the composite compared to the initial weight. Surface morphology has also been investigated using Phenom pro X desktop SEM with EDX with a current of 20 mA for 90 seconds. Samples were coated with gold (Au Pd) using the Quorum Sc7620 Sputter Coater tool.

3. Result and Discussion

3.1. Compressive Strength Test

Teak leaves waste/polyurethane composite have been fabricated successfully as shown **Figure 1.a. Figure 1.b.** shows the effect of polyurethane addition on composite compressive strength. Composites 0.04 and 0.08 (w/w) have relatively low compressive strength because many teak leaves particles not binding to the polymer matrix. The highest compressive strength achieved 38.5 MPa for the polyurethane fraction 0.14 (w/w). The resulting strength is 86% higher than polyurethane fraction 0.04 (w/w) which is closely related to the interaction between the surface of the particles and the polymer as in **Figure 2**. In polyurethane fraction 0.14 (w/w), all polymer matrix exactly interact with all teak leaves particles, composite is more compact and can sustains a higher load.

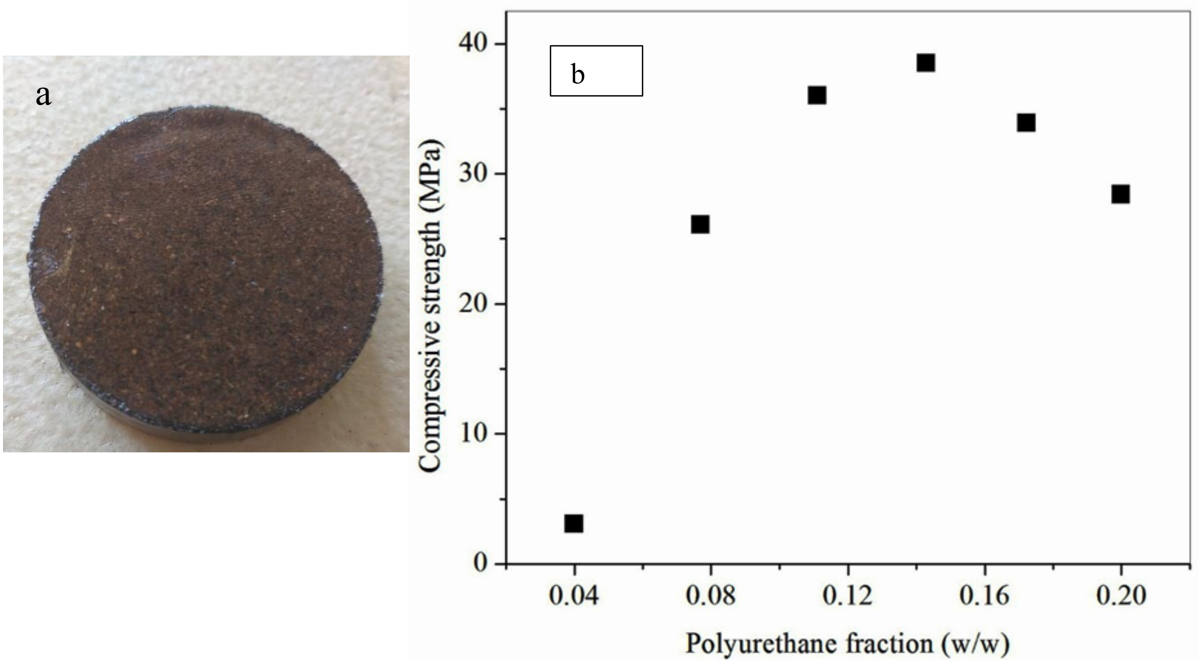

Figure 1.a Teak leaves waste/polyurethane composite**, 1.b.** Compressive strength of teak leave waste composite as polyurethane content (w/w) and the compressive strength attains at maximum for 0.14 fraction (w/w).

Figure 2(a) illustrates the bonding of leaves teak particles and mass varied of polyurethane polymers at 0.04-0.08 (w/w). When the mixture of filler and polymer is pressed to form a composite, the polyurethane polymer partially interacts with leaf particles and some particles diffuse which not interact with each other. It can act as cracking initiator and composite might be brittle. In **Figure 2 (b)**, a composite 0.14 (w/w) is called the optimal fraction because all the teak leaves particles are precisely bound to the polymer matrix. The matrix bond forward load in all directions, composite be stronger and resistant to the maximum load [21]. Chen *et al.* also reported in their research about composite polyurethane/graphene oxide (GO) where the addition of GO in the polyurethane matrix can improve the mechanical properties of the composite up to 167% [22]. Further, as **Figure 2(c)** the composites with a polyurethane content of 0.17-0.20 (w/w) show some parts of polyurethane not binding to the filler, appear many voids from polyurethane polymer and compressive strength decreases again [23-24].

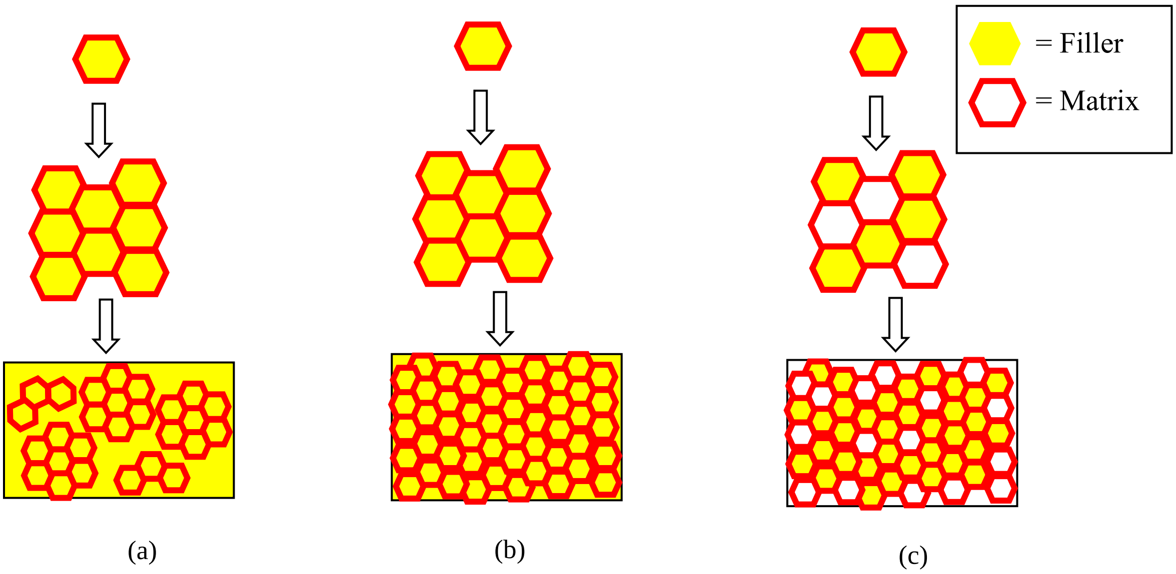

Figure 2. Filler particle distribution scheme and polymer matrix. (a) low compressive strength, (b) high compressive strength, (c) low compressive strength

Using the explanation above, the added mass of polyurethane can improve the composite compressive strength [25]. However, the larger of added polyurethane content can reduce the compressive strength. The compressive strength-produced does not increase continuously but reaches the maximum value as in **Figure 1**. According to Jaya *et al.*, when the polymer was added much more than the filler there will be a very poor adhesion force between surfaces due to polyurethane is a hydrophobic matrix and teak leaves powder is a hydrophilic filler [25].

Further, the raising of the strength until certain fraction is related to the maximum contact amount between the polymer and solid waste particle [26].

The most important interaction in the composite is predominantly emerged between the polyurethane and the cellulose in teak leaves. According to Alemdar and Sain, the cellulose in plant fibers has important role in reinforcing material composite applications [27]. The presence of cellulose in teak leaves can be investigated through the FTIR spectrum in **Figure 3.**

Figure 3(a) and **Figure 3(b)** show the FTIR spectrum of the pure cellulose obtained from Ramirez-Flores *et al.* [28] and the teak leaves respectively, with the comparison can be shown in **Table 1**. The FTIR spectrum of pure cellulose and leaves teak powder celluloses has a significant difference at peak 1631 cm^{-1}. The peak of 1631 cm^{-1} indicates absorption band of hydrogen forming a hydrogen bridge from hydroxyl groups and oxygen atoms from glucose monomer and usually found at a cellulose fiber [29]. The peak of 1631 cm^{-1} is not found at pure cellulose but it found at cellulose of natural fibers [30–33].

The peak of 2929 and 2300 cm^{-1} indicates a strong C-H bond in cellulose polymers [34]. While, peak of 3401 cm^{-1} shows the stretching of the O-H group bonds [35]. Further, the peak of 780 cm^{-1} and 1438 cm^{-1} shows the C-H bonds connecting the glucose units of cellulose. The peak at 1234 cm^{-1} indicates that there is a C-O-C group which is the structure of lignin. The absorption area occurring at 1076 cm^{-1} indicates the presence of a C-O bond in the cellulose component [25].

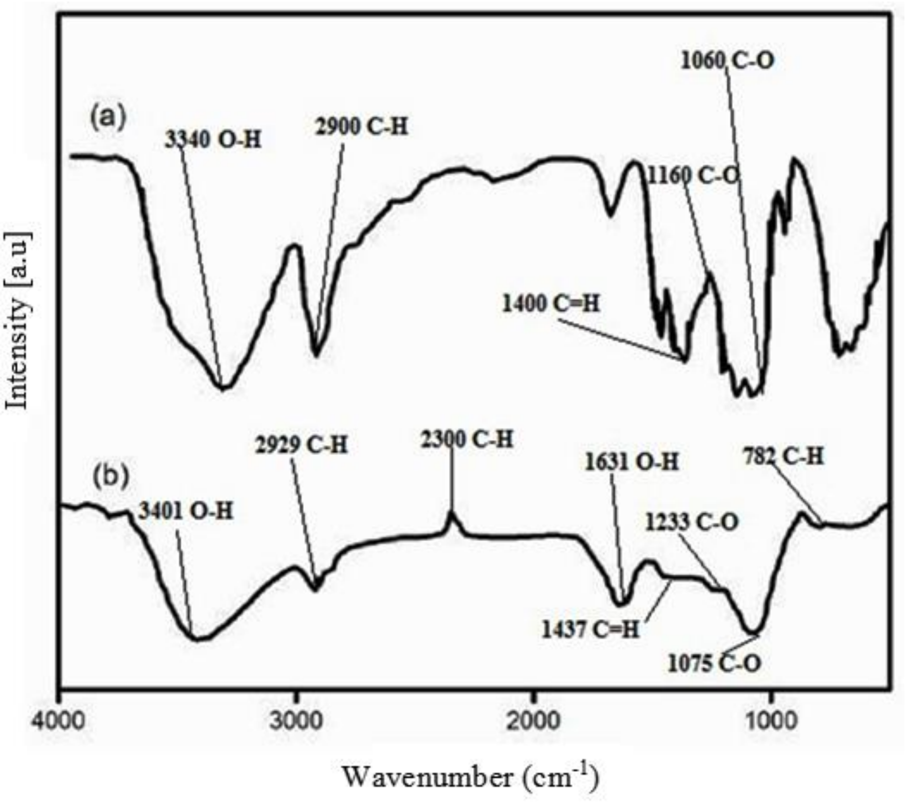

Figure 3. Spectrum of cellulose of: (a) pure cellulose obtained from Ramirez-Flores [13] b. cellulose of teak leaves powder in this work where many peaks of both celluloses have more similarities.

Using the FTIR analysis, it can be shown that cellulose and lignin components are found in the teak leaves. This leave can compete the conventional fiber such as glass fiber regarding its high performance as reinforcing material for thermosetting and thermoplastic matrices with their naturally degradable superior properties [36].

Table 1. Comparison of the peak absorption at pure cellulose and cellulose of teak leaves powder

Bond	Wavenumber (cm^{-1})	
	Pure cellulose (cm^{-1}) [15]	Cellulose of teak leaves powder (cm^{-1})
O-H	3340 – 3500	3401
C-H	2800 – 2920	2929
C-O-C	1160	1233
C-O	1035 – 1060	1075
C=H	1400	1437

The interaction arising between the teak leaves and the polymer is a van der Waals interaction. This interaction was confirmed by several FTIR spectrum shifts as shown in **Figure**

4. The shifts in spectrum are appeared due to the vibrating atoms as a result of the energy of disruption of atoms closing each other.

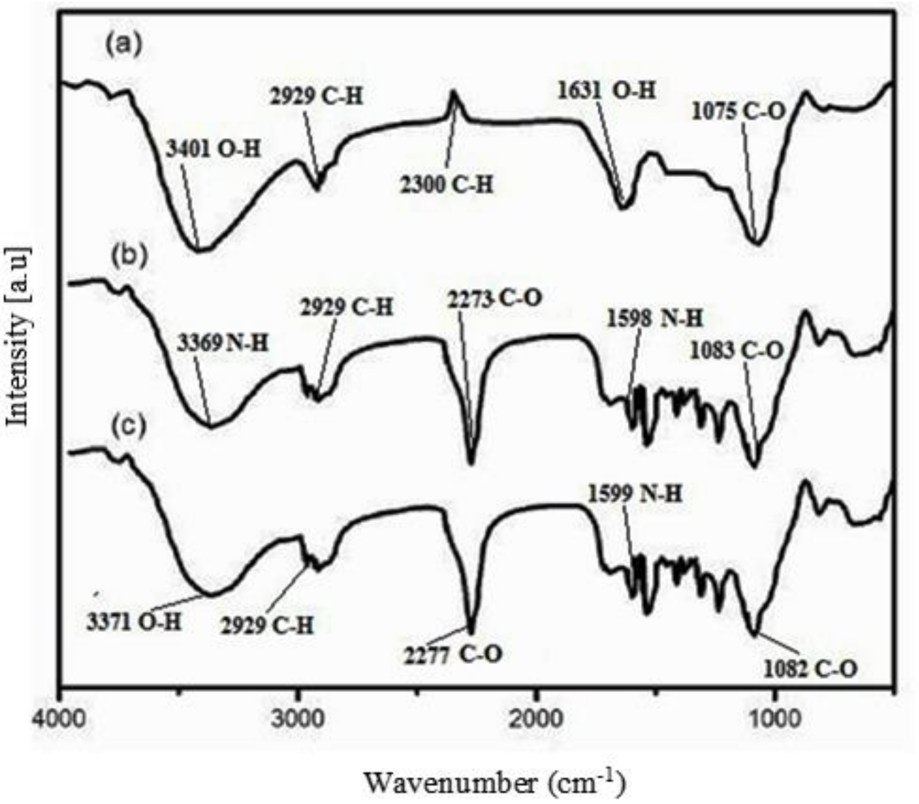

Figure 4. FTIR spectrum a. teak leaves, b. polyurethane, c. PU/teak leave composite

Polyurethane has two functional groups, i.e. carbonyl and amine groups. The spectrum shift starts to occur of 3369 cm^{-1} to 3371 cm^{-1} indicates the O-H bond. This shift is due to the amine group of the polyurethane was disturbed by hydrogen bonding in the hydroxyl group of leaves and forming a hydrogen bridge [37]. Further, the shift of 2273 cm^{-1} to 2277 cm^{-1} shows the carbonyl group of polyurethane undergoes a spectral shift. This is due to the disturbance of the C-H leaves bonding to the carbonyl group forming the O-H bridges. The peak at 1598 cm^{-1} shifting to 1599 cm^{-1} also indicates the amine group in polyurethane disturbed by the O-H bonds of the teak leaves. Furthermore, the shift also occurs at the carbonyl peak of 1083 cm^{-1} to 1082 cm^{-1} due to disturbance of the leave's C-O bond of the peak of 1075 cm^{-1} (Figure **5**). Similar results were confirmed by a previous work that PVA-sepiolite composite which derived its van der Waals interactions between silanol groups and carbon C-O and shifted from 1088 cm^{-1} to 1085 cm^{-1} [38].

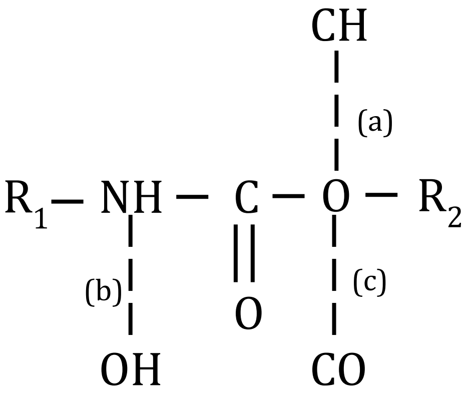

Figure 5. Van Der Waals interactions on the teak leaf waste composites, dashed lines illustrate the Van der Waals interaction area of (a) CH and carbonyl group (b) OH and amine group, and (c) CO and carbonyl group.

3.2. Density Test

Figure 6 shows the density of composite samples with polyurethane variation ranges from 825 to 1261 kg.m^{-3}. Considering that the interpretation data shows a coefficient of determination of 91.15%, there is a linear relationship between the amount of polyurethane and the density of the composite. Similar results were obtained by Afzaludin *et al.* on sugar palm / glass fiber reinforced thermoplastic polyurethane [39]. The lowest density value is found in the sample with a polyurethane fraction (w/w) of 0.04, while the highest density value is in the sample with 0.20 of polyurethane fraction (w/w). It can be explained when the polyurethane polymer content rises and the filler is fixed, then the number of links produced by the polymer increases. Hence, the increased links result in stronger bonds to bind the filler which can increase the density [40].

The density in samples with a composition of 0.08 (w/w) accordance with teak wood that has been studied by Wahyudi *et al.*, in the range of 1000-1200 kg m^{-3} with an average value of 1160 kg.m^{-3} [30]. In this study, the resulted-density value is higher than the research of Martins *et al.* producing particle board with density of 510 - 570 kg.m^{-3} [31]. The composites produced in this work have a similar density to the results of the Raju *et al.* producing some composites with densities ranging from 1100 to 1320 kg.m^{-3} [32]. Further, Butylina *et al.* have produced wood fibre-polypropylene composite having density ranged from 1040 to 1140 kg.m^{-3} [33].

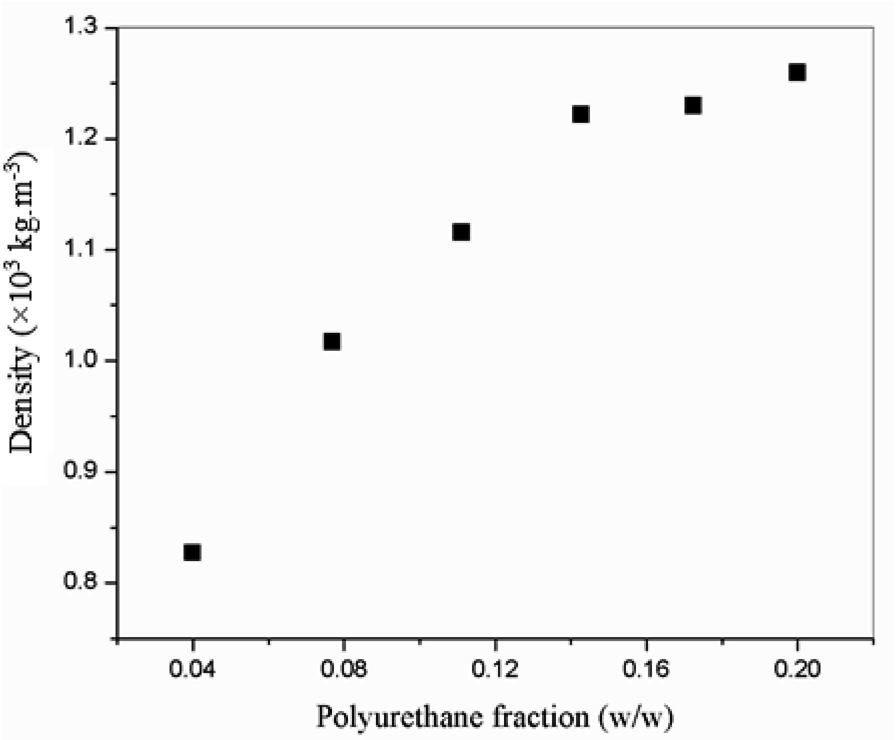

Figure 6. Composite density as polyurethane fraction (w/w) function

The density data shows that this composite belong to the high-density particle board category due to has a density more than 0.80 g/cm^3 in accordance with the standard designation 1554-67 by American Society [41]. Additionally, the composite has also meet the Indonesia National Standard of 03-2105-2006 where particleboard density having values in the range of 400 - 900 kg.m^{-3} [42].

3.3. Water Absorption

Figure 6 shows the density of composite samples with polyurethane variation ranges from 825 to 1261 kg.m-3. Considering the interpretation data showing a coefficient of determination, R^2=0.91157, there is a linear relationship between the polyurethane content and the composite density. Similar results were obtained by Afzaludin et al. on sugar palm / glass fiber reinforced thermoplastic polyurethane [38]. The lowest density value is found in the sample with a polyurethane fraction (w/w) of 0.04, while the highest density value is in the sample with 0.20 of polyurethane fraction (w/w). It can be explained when the polyurethane polymer content rises and the filler is fixed, then the number of links produced by the polymer increases. Hence, the increased links result in stronger bonds to bind the filler which can increase the density [39].

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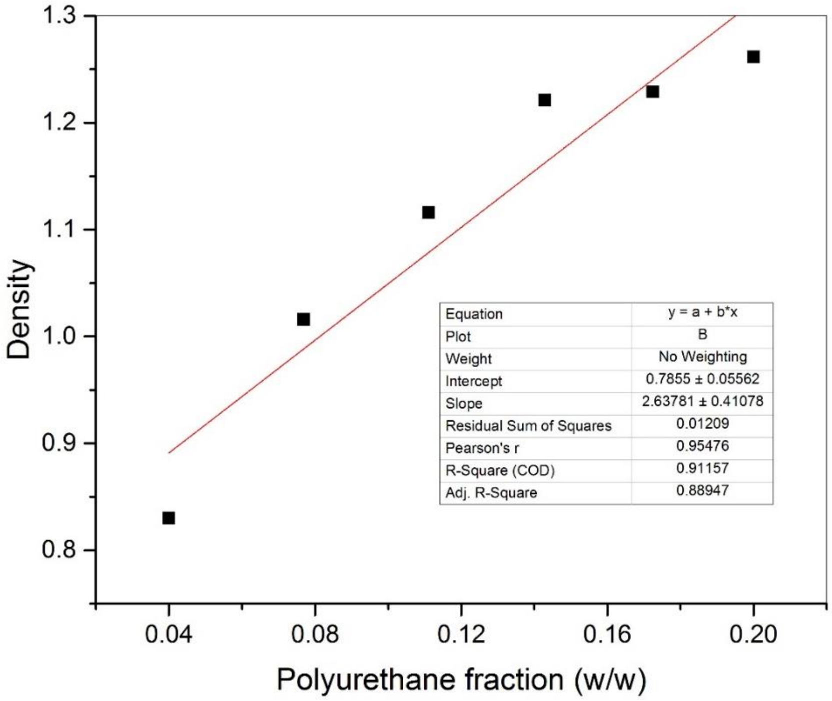

Figure 6. Composite density as polyurethane fraction (w/w) function, and it was linearity relation as; y=2.6378x+0.7855, where x and y are polyurethane fraction and composite density respectively, with R^2=0.91157.

The density data shows that this composite belong to the high-density particle board category due to has a density more than 0.80 g/cm3 in accordance with the standard designation 1554-67 by American Society [40]. Additionally, the composite has also meet the Indonesia National Standard of 03-2105-2006 where particleboard density having values in the range of 400 - 900 kg.m-3 [41].

3.4. Scanning Electron Microscopy

Morphological structure using SEM from composites 0.04; 0.12; 0.20 (w/w) was shown in **Figure 8**. There is a cavity or gap between the filler mixture and the matrix. Based on data analysis, the gap value between molecules in the composite 0.04 (w/w) is 13 µm. The low concentration of filler results in agglomeration in certain parts and gaps between the mixture appear large enough which can act as cause of cracking and breakage when subjected to a load [45]. This was confirmed by the compressive strength and density results on the composite 0.04 (w/w) resulting in the lowest compressive strength and density. The resulting gap will be narrower when the concentration of polyurethane polymers higher and the density increase [46]. The gap value in the composite 0.12 (w/w) is 4.65 µm and decrease when the polyurethane concentration increases which the gap distance 2.96 µm in the composite 0.20 (w/w). The morphological structure of the composite accordance with the trends obtained when measuring the density value of the sample. The hydroxyl group in cellulose plays an important role in the mechanism of forming bonds between molecules with polymer chains [47]. Interaction between cellulose molecules and polyurethane polymers which can increase the density and decrease the gap between the filer and the polymer [46].

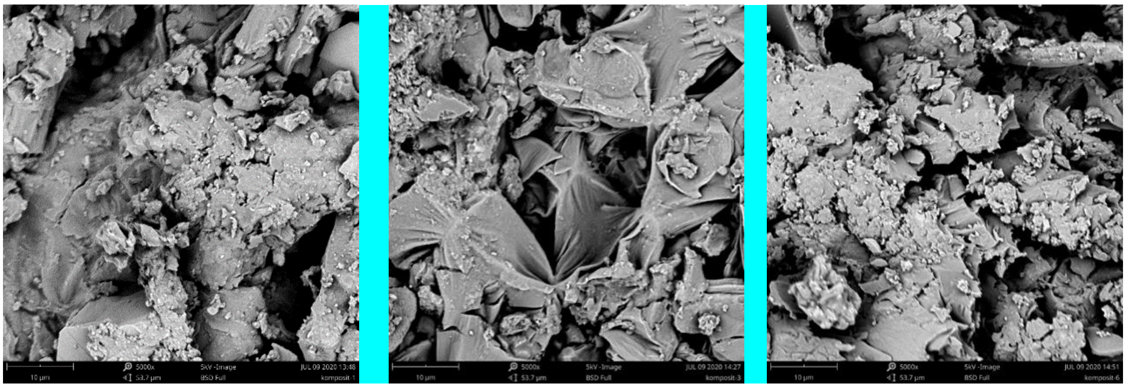

Figure 8. SEM of composite (a) 0.04 (w/w); (b) 0.12 (w/w); (c) 0.20 (w/w).

4. Conclusion

We have successfully developed a Teak leaves-waste composite utilized as particleboard. The mechanical and physical properties of the composites have been investigated. The test results showed a composite compressive strength attains 38.5 MPa. The highest density is 1261 kg.m^{-3} that is in accordance to the American Society standard. The lowest value of water absorption is 1.38%. In addition, we have successfully approximated the van der Waals interaction with the spectrum shifts of the FTIR spectroscopy as a confirmation of the compressive strength enhancement. Morphological structure shows that the gap between mixture decreases as the polyurethane fraction increases according to the density of the composite. However, the properties of organic composite are hygroscopic so they are vulnerable to being the target of fungal growth and susceptible to fire as an application in building fields. These problems will be focus of further research by applying coating technology approaches.

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Declaration of conflicting interests

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Advanced Composites Letters - Decision on Manuscript ID ACM-20-0085.R2

Advanced Composites Letters <onbehalfof@manuscriptcentral.com> Sun, Aug 9, 2020 at 7:28 PM
Reply-To: acm@sagepub.com
To: masturi@mail.unnes.ac.id

9 August 2020

Dear Dr. Masturi:

Manuscript ID ACM-20-0085.R2 entitled "MECHANICAL AND PHYSICAL PROPERTIES OF TEAK LEAVES WASTE/POLYURETHANE COMPOSITES FOR PARTICLE-BOARD APPLICATION" which you submitted to Advanced Composites Letters, has been reviewed. The comments of the reviewer(s) are included at the bottom of this letter.

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Once again, thank you for submitting your manuscript to Advanced Composites Letters and I look forward to receiving your revision.

Sincerely,
Professor Igor Guz
Advanced Composites Letters
acm@sagepub.com

Reviewer(s)' Comments to Author:

Reviewer: 2

Comments to the Author
A number of problems has been solved and this work becomes more suitable for publication. However, there are still a number of issues that need attention.

Please offer more details about compressive tests in the text, such as the maximum load of the machine and the speed of loading.

In Figure 1, how many samples did you test per amount of teak leaves filling? If the answer is "one", the study has considerable limitations. Of course, you may establish which is the right amount of filling to start with. If you tested more that one sample per amount of filling please provide average and standard deviation.

I don't understand Figure 2, how it can provide more information about the composite. Teak leaves powder has no relation with graphene oxide distribution, I suggest that teak leaves would tend much more to agglomerate and not bond properly.

Also, the text at a number of points is not very understandable. I suggest the places on which attention should be placed, also clarifying that the whole paper needs to be carefully read once more and rewritten to convey the right sense.

In particular:
Abstract: what do you mean by "This result indicated that composites can be
used as an eco-friendly industrial scale instead of wood"? Please rephrase.
Introduction line 2: "Composite technique is one of the possible ways to fulfill the
demand". i don't understand what "composite technique" is. The production of composites, may be?
Page 5 line -3: please replace "were fabricated" by "have fabricated".
Page 6 line 9: please replace "owing" with "because it"
P.16 line 7: please replace "accordance" with "is in agreement"
P. 17 line 2: please replace "having" with "has"
P. 20 line 4: please replace "decrease" with "decreases"
P. 20 line 11: please replace "filer" with "filler"

Associate Editor(s)' Comments to Author:

Associate Editor: OGIHARA , Shinji
Comments to the Author:
(There are no comments.)

Response to Reviewer(s)' Comments to manuscript entitled: MECHANICAL AND PHYSICAL PROPERTIES OF TEAK LEAVES WASTE/POLYURETHANE COMPOSITES FOR PARTICLE-BOARD APPLICATION

Dear Editor,

Thank you very much for the comment (Reviewer 2). Below are the responses of the reviewer comments and we have also updated them in the manuscript.

Regards,

Masturi

Reviewer: 2

Comments to the Author

1. A number of problems has been solved and this work becomes more suitable for publication. However, there are still a number of issues that need attention.
   Please offer more details about compressive tests in the text, such as the maximum load of the machine and the speed of loading.

Response:

The identification of the mechanical properties was measured by a compressive strength test using Sociedad Espanola Anonima Ibertest Instrument with maximum force of 250 kN and speeds of movements was 0.05 kN/s.

2. In Figure 1, how many samples did you test per amount of teak leaves filling? If the answer is "one", the study has considerable limitations. Of course, you may establish which is the right amount of filling to start with. If you tested more that one sample per amount of filling please provide average and standard deviation.

Response:

In Figure 1, the test is carried out once per sample and handled by the testing lab technician. We do not include the error bar values.

3. I don't understand Figure 2, how it can provide more information about the composite. Teak leaves powder has no relation with graphene oxide distribution, I suggest that teak leaves would tend much more to agglomerate and not bond properly.

Response:

Figure 2(a) illustrates the bonding of leaves teak particles and mass varied of polyurethane polymers at 0.04-0.08 (w/w). When the mixture of filler and polymer is pressed to form a composite, the polyurethane polymer partially interacts with leaf particles and some particles diffuse which not interact with each other. It can act as cracking initiator and composite might be brittle. The addition of polyurethane content at 0.12-0.14 (w/w) can improve the composite compressive strength. As shown in **Figure 2 (b)**, a composite 0.14 (w/w) is called the optimal fraction because all the teak leaves particles are precisely bound to the polymer matrix. The matrix bond forward load in all directions, composite be stronger and resistant to the maximum load [21]. However, the compressive strength-produced does not increase continuously but reaches the maximum value. Further, as **Figure 2(c)** the composites with a polyurethane content of 0.17-0.20 (w/w) show some parts of polyurethane not binding to the filler, appear many voids from polyurethane polymer, teak leaves would tend to agglomerate and not bond properly which can reduce compressive strength [23-24]. According to Jaya et al., when the polymer was added much more than the filler there will be a very poor adhesion force between surfaces due to polyurethane is a hydrophobic matrix and teak leaves powder is a hydrophilic filler [24]. Further, the raising of the strength until certain fraction is related to the maximum contact amount between the polymer and solid waste particle [25]

4. Also, the text at a number of points is not very understandable. I suggest the places on which attention should be placed, also clarifying that the whole paper needs to be carefully read once more and rewritten to convey the right sense.

   In particular:

   Abstract: what do you mean by "This result indicated that composites can be used as an eco-friendly industrial scale instead of wood"? Please rephrase.

Response:

We mean that the teak leaves waste composites are potentially to replace wood raw materials due to in accordance American Society Standard Designation 1554-67 and SNI 03-2105-2006, can reduce the rate of deforestation and solve the problem of waste. We have replaced with: “This result indicated that the composites-produced are potentially to replace wood raw material in eco-friendly industrial scale.”

5. Introduction line 2: "Composite technique is one of the possible ways to fulfill the demand". I don't understand what "composite technique" is. The production of composites, may be?

Response:

Composite engineering is an engineering technology that can compose two or more elements with different properties to produce new raw materials. In this case, the substitute raw material in question is particle board which can replace wood raw material

6. Page 5 line -3: please replace "were fabricated" by "have fabricated".
   Page 6 line 9: please replace "owing" with "because it"
   P.16 line 7: please replace "accordance" with "is in agreement"
   P. 17 line 2: please replace "having" with "has"
   P. 20 line 4: please replace "decrease" with "decreases"
   P. 20 line 11: please replace "filer" with "filler"

Response:

We have checked and replace it in the manuscript.

MECHANICAL AND PHYSICAL PROPERTIES OF TEAK LEAVES WASTE/POLYURETHANE COMPOSITES FOR PARTICLE-BOARD APPLICATION

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Abstract. A teak leaf waste/polyurethane composite has been made for particle-board application. Some fraction variations are performed on the use of polyurethane as a matrix with a range of 0.04-0.20 (w/w). Mechanical and physical properties have been tested on the composites produced. The test results showed the highest compressive strength of the sample reaches 38.5 MPa for polyurethane fraction of 0.14 (w/w). The composite has a density of 1261 kg.m^{-3} which is in accordance with the result of density on teak. The physical properties have been also investigated and it was found that the lowest value of water absorption is 1.38%. This result indicated that the composites-produced are potentially to replace wood raw material in eco-friendly industrial scale.

Keywords: composite, polyurethane, teak leaf, mechanical properties.

1. Introduction

In the last few decades, total global wood demand for 2020 is projected to reach 515 million m^3 [1]. Composite technique is one of the possible ways to fulfill the demand. Composite engineering promises organic and inorganic waste utilization as a synthesized-material having excellent properties relating the woods, such as mechanical strength, water absorption, density, etc. [2-6]. Furthermore, the composite engineering is also a solution for the raising of environment awareness caused by the high deforestation in several countries [2-3]. In addition, the utilization of natural waste and fibers for some composites has been widely studied and developed from various countries including Indonesia, Malaysia, Thailand, India, Brazil and Japan [2-4].

In recent years, the use of composites is very widespread in the electronics, automotive and aviation industries, construction field, multipurpose applications such as particle boards, energy storage, heat and electricity insulators, ship bodies and aircraft, organic field effect-transistors, antennas, etc. due to their high mechanical strength , good thermal stability, anti-corrosion, high dielectric constant, light weight and easy to produce large scale [7-13]. Several study about the polymers and plastics is focused on developing fibers, plastics, adhesives, composite textiles, paper from renewable resources derived mostly from agricultural wastes containing lignocellulosic [4]. Considering that agricultural waste is only used for compost production In most developing countries such as Indonesia, researcher also focused on utilization some natural fibers such as sisal, hemp, bamboo and agricultural wastes, e.g. leaves, coconut fibers, rice husk, peanut shells and straw as filler in polymer composite [5-6].Composites of these natural fibers have several advantages such as being easy to process, environmentally, good acoustic properties, low energy processes, good mechanical and thermal properties, water resistance, non-corrosive, non-toxic and naturally degradable [7].

Further, several studies related to the leaves waste composites have been done, such as Masturi *et al*. developed composite from leaves waste and paper waste using polyvinyl acetate as matrix and the compressive strength reaches to 45.60 MPa [2]. Campanella *et al*. successfully made a composite of Northern Red Oak leaves with plant oil-based resins [6], while Thakur *et al.* have also developed biopolymer composite using ligno-cellulosic of *Hibiscus Sabdariffa* [14]. A different method has been performed by Nongman *et al.* having created a composite by lamination method of banana leaf fiber and adhesive [5]. Senthilkumar *et al.* have fabricated pineapple leaf fiber / Polyester composites which have a tensile strength reached to 33.13 MPa. Good mechanical properties related to the high content of cellulose in pineapple leaf fibers which can transfer a sizable load from the matrix to the fiber effectively [15]. In addition, Melkanu *et al*., have investigated the mechanical properties and water absorption in sisal / Polyester fiber composites with treated fibers and without alkaline sodium hydroxide treatment. Compressive strength in sisal fibers treated better than untreated fibers with low water absorption advantages [16].

In this work, we developed a composite of teak leaves waste with polyurethane matrix. The teak leaves have been chosen since its utilization only as compost having a low added value as well as its availability is very abundant in Indonesia. Polyurethane as a thermosetting polymer was selected because it does not melt when heated, more resistant to acids and bases, has a high adhesive power due to the presence of highly reactive isocyanate functional groups, has good erosion resistance which can be used as an anti-erosion film and coating technology [17], has an isotropic and controllable electromagnetic response, and has a negative permittivity value which is potentially applied to the electronics field [18-19]. Furthermore, the addition of nanoparticles in polyurethane can improve thermal stability and mechanical properties of composites. Polyurethane is also used as a textile coating on inkjet-printed flexible and washable field-effect transistors (FETs) on textile technology [20]. The aim of this study is to produce a Teak leaves waste composite having a high compressive strength and low water absorption making a chance to be used on an eco-friendly industrial scale.

2. Materials and Methods

The teak leaves waste of Semarang Indonesia was used, while isocyanates and polyol as polyurethane (PU) polymerization materials were purchased in Multi Kimia Inc, Semarang, Indonesia.

For first, the leaves waste was dried and crushed using a blender machine to form a powder. Then, the powder was sieved using 700 mesh. In the other hand, the isocyanates and polyol were mixed using magnetic stirrer for 5 minutes with ratio 1:1 to form polyurethane and mixed with the teak leaves waste powder with varied composition while the leaves teak powder was kept at 12 g weight. The PU/teak leaves waste mixture was stirred using mixer machine to homogenize the mixture, then pressed into a cylindrical mold at pressure of 5 MPa for 15 minutes. Having been pressed, the composite was removed from the mold and dried for 24 hours at room temperature.

The investigation to the content of the teak leaves powder cellulose bonding to polyurethane and detecting spectral changes in teak leaves waste composite was performed using FTIR Spectrometer Frontier type (Perkin Elmer) in a spectral range of 400-4000 cm^{-1}. Meanwhile, the identification of the mechanical properties was measured by a compressive strength test using Sociedad Espanola Anonima Ibertest Instrument with maximum force of 250 kN and speeds of movements was 0.05 kN/s. In this test, the sample is cut into cubic shape and was measured using testing machine. In addition, to ensure the water resistance of the composite, the identification of the physical properties was performed by density measurement, further, water absorption test was done by soaking this sample in the water for 24 h to obtain the weight of water absorbed into the composite compared to the initial weight. Surface morphology has also been investigated using Phenom pro X desktop SEM with EDX with a current of 20 mA for 90 seconds. Samples were coated with gold (Au Pd) using the Quorum Sc7620 Sputter Coater tool.

3. Result and Discussion

3.1. Compressive Strength Test

Teak leaves waste/polyurethane composite have been fabricated successfully as shown **Figure 1.a. Figure 1.b.** shows the effect of polyurethane addition on composite compressive strength. Composites 0.04 and 0.08 (w/w) have relatively low compressive strength because many teak leaves particles not binding to the polymer matrix. The highest compressive strength achieved 38.5 MPa for the polyurethane fraction 0.14 (w/w). The resulting strength is 86% higher than polyurethane fraction 0.04 (w/w) which is closely related to the interaction between the surface of the particles and the polymer as in **Figure 2**. In polyurethane fraction 0.14 (w/w), all polymer matrix exactly interact with all teak leaves particles, composite is more compact and can sustains a higher load.

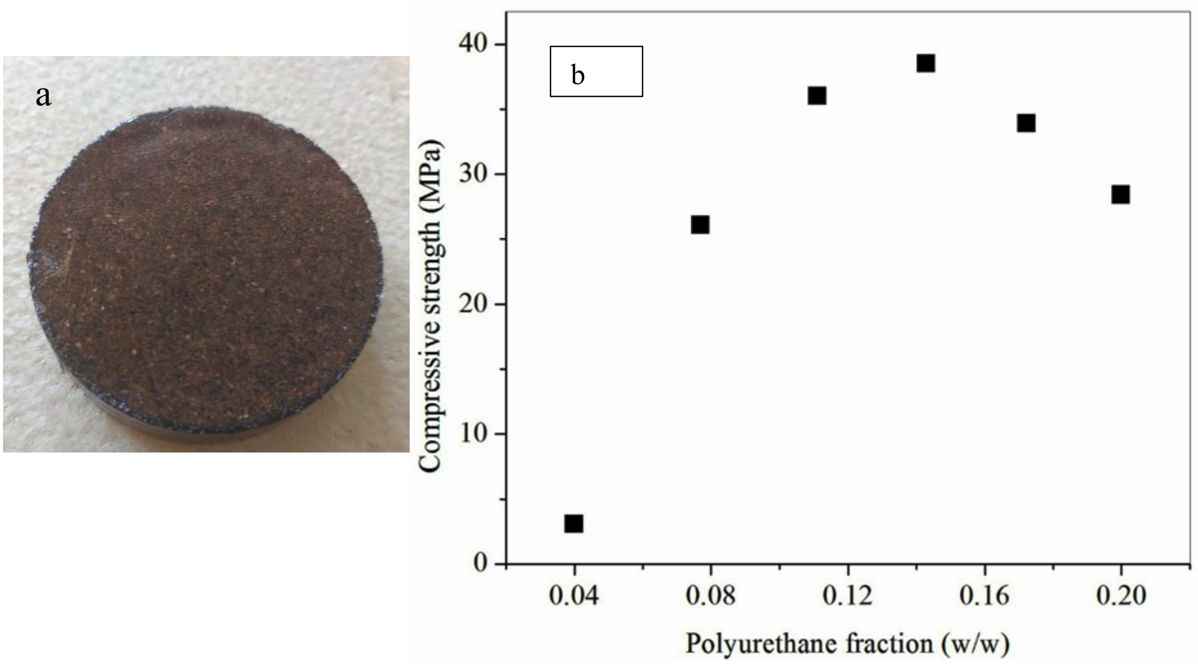


Figure 1.a Teak leaves waste/polyurethane composite**, 1.b.** Compressive strength of teak leave waste composite as polyurethane content (w/w) and the compressive strength attains at maximum for 0.14 fraction (w/w).

Figure 2(a) illustrates the bonding of leaves teak particles and mass varied of polyurethane polymers at 0.04-0.08 (w/w). When the mixture of filler and polymer is pressed to form a composite, the polyurethane polymer partially interacts with leaf particles and some particles diffuse which not interact with each other. It can act as cracking initiator and composite might be brittle. The addition of polyurethane content at 0.12-0.14 (w/w) can improve the composite compressive strength. As shown in **Figure 2 (b)**, a composite 0.14 (w/w) is called the optimal fraction because all the teak leaves particles are precisely bound to the polymer matrix. The matrix bond forward load in all directions, composite be stronger and resistant to the maximum load [21]. However, the compressive strength-produced does not increase continuously but reaches the maximum value. Further, as **Figure 2(c)** the composites with a polyurethane content of 0.17-0.20 (w/w) show some parts of polyurethane not binding to the filler, appear many voids from polyurethane polymer, teak leaves would tend to agglomerate and not bond properly which can reduce compressive strength [23-24]. According to Jaya et al., when the polymer was added much more than the filler there will be a very poor adhesion force between surfaces due to polyurethane is a hydrophobic matrix and teak leaves powder is a hydrophilic filler [24]. Further, the raising of the strength until certain fraction is related to the maximum contact amount between the polymer and solid waste particle [25].

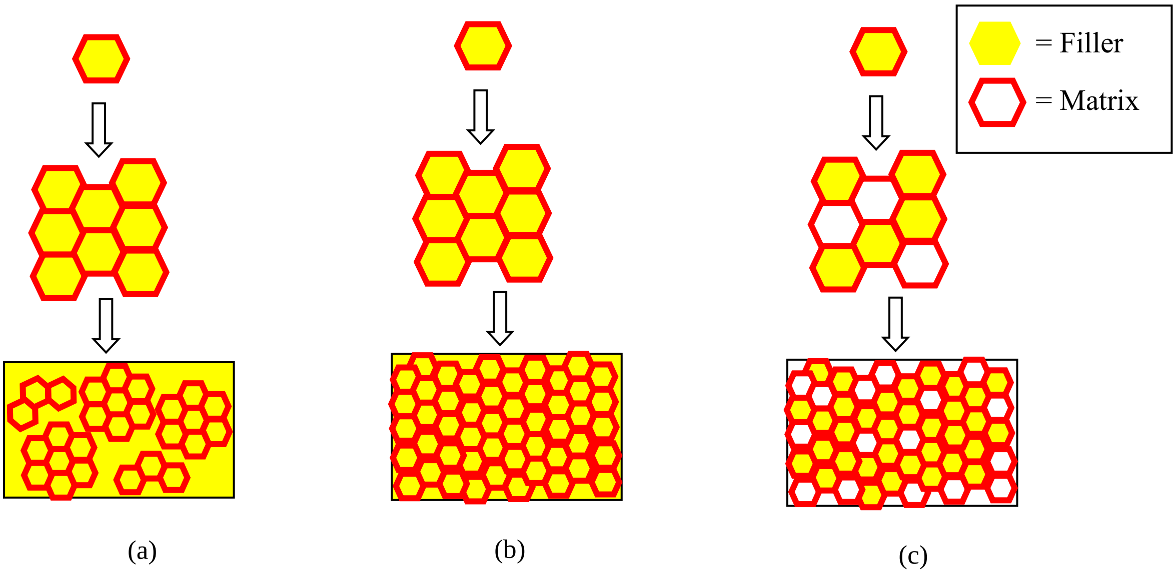


Figure 2. Schematic of the particle distribution of filler and polymer matrix. (a) low compressive strength, (b) high compressive strength, (c) low compressive strength

The most important interaction in the composite is predominantly emerged between the polyurethane and the cellulose in teak leaves. According to Alemdar and Sain, the cellulose in plant fibers has important role in reinforcing material composite applications [26]. The presence of cellulose in teak leaves can be investigated through the FTIR spectrum in **Figure 3.**

Figure 3(a) and **Figure 3(b)** show the FTIR spectrum of the pure cellulose obtained from Ramirez-Flores *et al.* [27] and the teak leaves respectively, with the comparison can be shown in **Table 1**. The FTIR spectrum of pure cellulose and leaves teak powder celluloses has a significant difference at peak 1631 cm^{-1}. The peak of 1631 cm^{-1} indicates absorption band of hydrogen forming a hydrogen bridge from hydroxyl groups and oxygen atoms from glucose monomer and usually found at a cellulose fiber [28]. The peak of 1631 cm^{-1} is not found at pure cellulose but it found at cellulose of natural fibers [30–33].

The peak of 2929 and 2300 cm^{-1} indicates a strong C-H bond in cellulose polymers [33]. While, peak of 3401 cm^{-1} shows the stretching of the O-H group bonds [34]. Further, the peak of 780 cm^{-1} and 1438 cm^{-1} shows the C-H bonds connecting the glucose units of cellulose. The peak at 1234 cm^{-1} indicates that there is a C-O-C group which is the structure of lignin. The absorption area occurring at 1076 cm^{-1} indicates the presence of a C-O bond in the cellulose component [24].

Using the FTIR analysis, it can be shown that cellulose and lignin components are found in the teak leaves. This leave can compete the conventional fiber such as glass fiber regarding its high performance as reinforcing material for thermosetting and thermoplastic matrices with their naturally degradable superior properties [35].

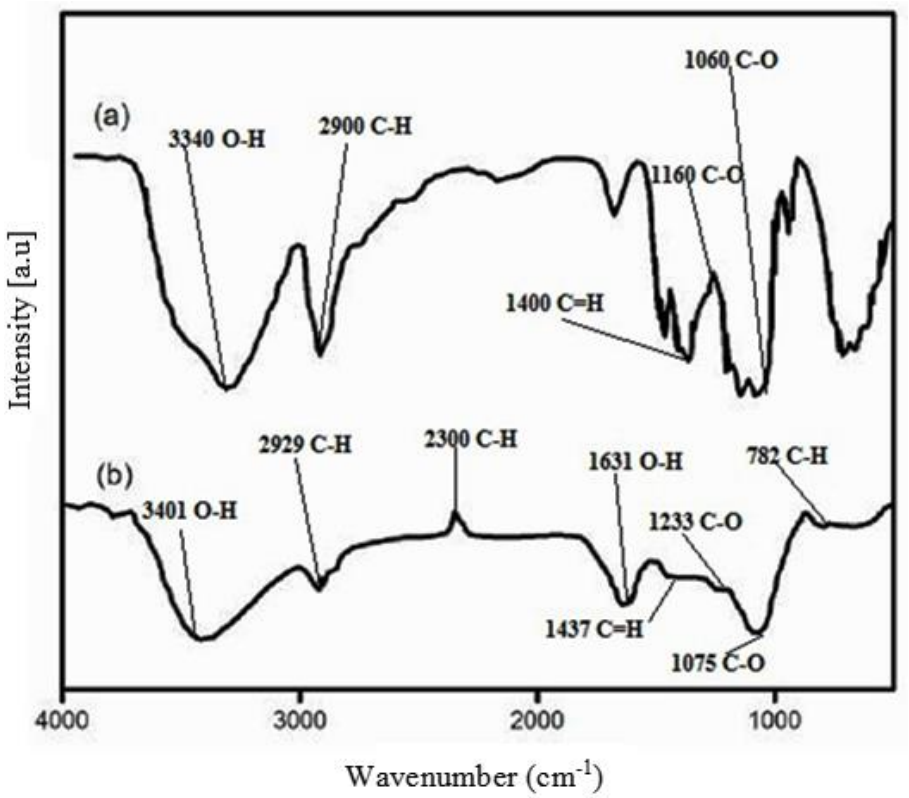


Figure 3. Spectrum of cellulose of: (a) pure cellulose obtained from Ramirez-Flores [13] b. cellulose of teak leaves powder in this work where many peaks of both celluloses have more similarities.

Table 1. Comparison of the peak absorption at pure cellulose and cellulose of teak leaves powder

Bond	Wavenumber (cm^{-1})	
	Pure cellulose (cm^{-1}) [15]	Cellulose of teak leaves powder (cm^{-1})
O-H	3340 – 3500	3401
C-H	2800 – 2920	2929
C-O-C	1160	1233
C-O	1035 – 1060	1075
C=H	1400	1437

The interaction arising between the teak leaves and the polymer is a van der Waals interaction. This interaction was confirmed by several FTIR spectrum shifts as shown in **Figure 4**. The shifts in spectrum are appeared due to the vibrating atoms as a result of the energy of disruption of atoms closing each other.

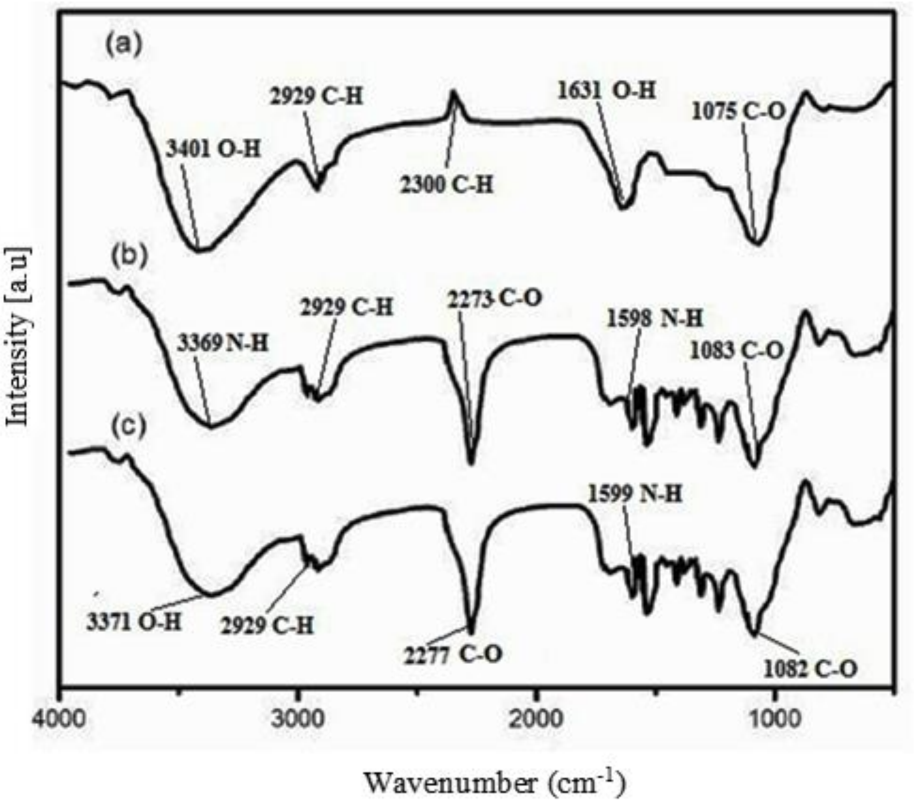


Figure 4. FTIR spectrum a. teak leaves, b. polyurethane, c. PU/teak leave composite

Polyurethane has two functional groups, i.e. carbonyl and amine groups. The spectrum shift starts to occur of 3369 cm^{-1} to 3371 cm^{-1} indicates the O-H bond. This shift is due to the amine group of the polyurethane was disturbed by hydrogen bonding in the hydroxyl group of leaves and forming a hydrogen bridge [36]. Further, the shift of 2273 cm^{-1} to 2277 cm^{-1} shows the carbonyl group of polyurethane undergoes a spectral shift. This is due to the disturbance of the C-H leaves bonding to the carbonyl group forming the O-H bridges. The peak at 1598 cm^{-1} shifting to 1599 cm^{-1} also indicates the amine group in polyurethane disturbed by the O-H bonds of the teak leaves. Furthermore, the shift also occurs at the carbonyl peak of 1083 cm^{-1} to 1082 cm^{-1} due to disturbance of the leave's C-O bond of the peak of 1075 cm^{-1} (Figure **5**). Similar results were confirmed by a previous work that PVA-sepiolite composite which derived its van der Waals interactions between silanol groups and carbon C-O and shifted from 1088 cm^{-1} to 1085 cm^{-1} [37].

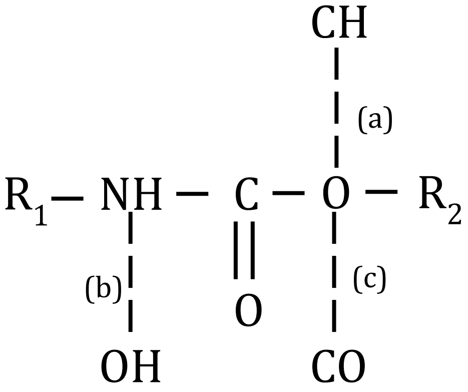
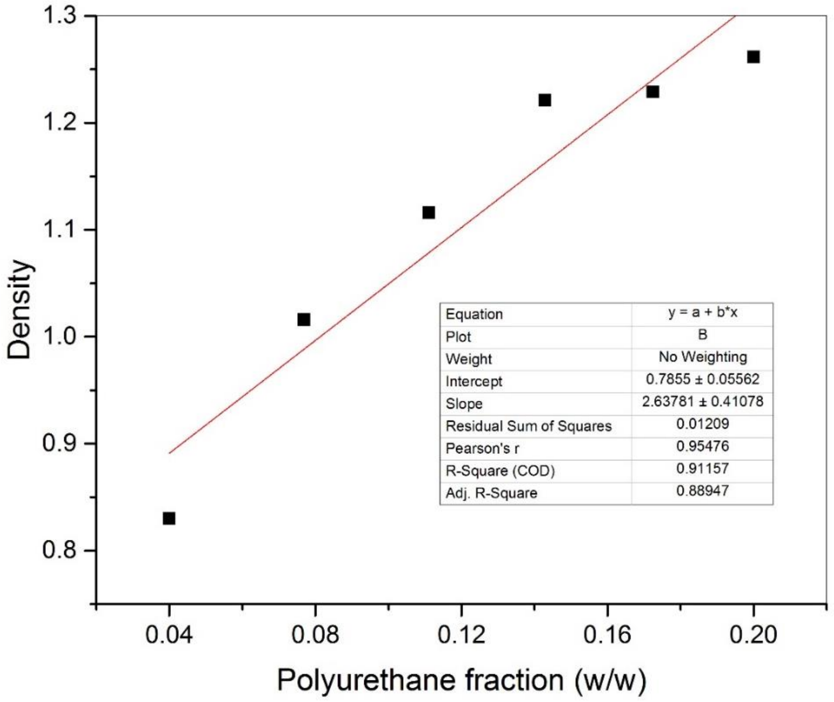


Figure 5. Van Der Waals interactions on the teak leaf waste composites, dashed lines illustrate the Van der Waals interaction area of (a) CH and carbonyl group (b) OH and amine group, and (c) CO and carbonyl group.

3.2. Density Test

Figure 6 shows the density of composite samples with polyurethane variation ranges from 825 to 1261 kg.m^{-3}. Considering the interpretation data showing a coefficient of determination, $R^2 = 0.91157$, there is a linear relationship between the polyurethane content and the composite density. Similar results were obtained by Afzaludin *et al.* on sugar palm / glass fiber reinforced thermoplastic polyurethane [38]. The lowest density value is found in the sample with a polyurethane fraction (w/w) of 0.04, while the highest density value is in the sample with 0.20 of polyurethane fraction (w/w). It can be explained when the polyurethane polymer content rises and the filler is fixed, then the number of links produced by the polymer increases. Hence, the increased links result in stronger bonds to bind the filler which can increase the density [39].

The density in samples with a composition of 0.08 (w/w) accordance with teak wood that has been studied by Wahyudi *et al.*, in the range of 1000-1200 kg m^{-3} with an average value of 1160 kg.m^{-3} [29]. In this study, the resulted-density value is higher than the result of Martins *et al.* producing particle board with density of 510 - 570 kg.m^{-3} [30]. The composites produced in this work have a similar density to the results of the Raju *et al.* producing some composites with densities ranging from 1100 to 1320 kg.m^{-3} [31]. Further, Butylina *et al.* have produced wood fibre-polypropylene composite having density ranged from 1040 to 1140 kg.m^{-3} [32].



Equation	y = a + b*x
Plot	B
Weight	No Weighting
Intercept	0.7855 ± 0.05562
Slope	2.63781 ± 0.41078
Residual Sum of Squares	0.01209
Pearson's r	0.95476
R-Square (COD)	0.91157
Adj. R-Square	0.88947

Figure 6. Composite density as polyurethane fraction (w/w) function, and it was linearity relation as; $y = 2.6378x + 0.7855$, where x and y are polyurethane fraction and composite density respectively, with $R^2 = 0.91157$.

The density data shows that this composite belong to the high-density particle board category due to has a density more than 0.80 g/cm^3 in accordance with the standard designation 1554-67 by American Society [40]. Additionally, the composite has also meet the Indonesia National Standard of 03-2105-2006 where particleboard density having values in the range of 400 - 900 kg.m^{-3} [41].

3.3. Water Absorption

Water absorption values were measured to determine the ability of teak leaf composite to absorb water after 24 hours of soaking. The highest water absorption rate reaches 107.35% in composites with a polyurethane fraction of 0.04 (w/w). After being immersed for 24 hours, the composite undergoes thick development until the composite becomes cracked and broken due to the cavity that appears between the matrix and the filler particles absorbs a lot of water and can further affect the stability of the dimensions of the composite. In addition, teak leaves as a hydrophilic filler also absorb a lot of water during immersion.

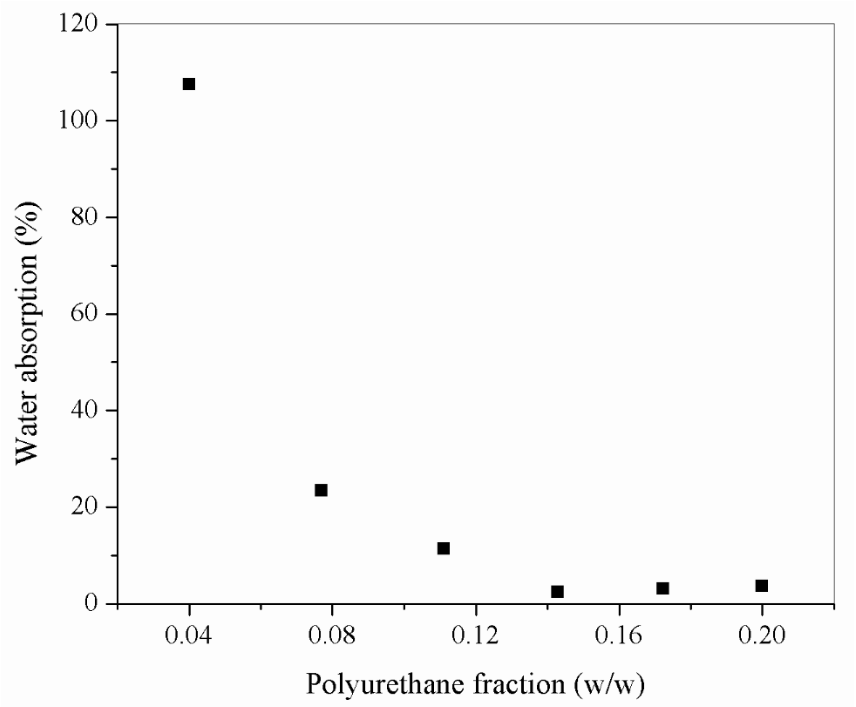


Figure 7. Water absorption of composite tends to zero for larger polyurethane fraction (w/w)

Figure 7 shows the water absorption value was decreasing with the increase of polyurethane fraction. At high fractions, it tends to produce composites with lower water absorption. the lowest water absorption was achieved in samples having the most optimal compressive strength at fraction 0.14 (w/w). The cavity formed is very small because the pressing process in the fabrication of composites and filler particles is distributed evenly on the matrix and absorbed a little water.

The absorption value of the leaves teak waste composite produced is better than to that of several leaves waste composites has been developed by several authors, such as Al-Sulaiman *et al.* produced palm fiber reinforced composites with the water absorption is between 2.3 to 5.8% [42], and Batiancela *et al.* developed a wood particle from tea leaves waste and obtained the water absorption about 23-40% [43]. Therefore, in accordance to the several properties above, this composite has properness in competing the woods.

3.4. Scanning Electron Microscopy

Morphological structure using SEM from composites 0.04; 0.12; 0.20 (w/w) are shown in **Figure 8**. There is a cavity between the filler mixture and the matrix. Based on data analysis, the gap value between molecules in the composite 0.04 (w/w) is 13µm. The low concentration of filler results in agglomeration in certain parts and gaps between the mixture appear large enough which can act as cause of cracking and breakage when subjected to a load [44]. This was confirmed by the compressive strength and density results on the composite 0.04 (w/w) resulting in the lowest compressive strength and density. The resulting gap will be narrower when the concentration of polyurethane polymers higher and the density increase [45]. The gap value in the composite 0.12 (w/w) is 4.65 µm and decreases when the polyurethane concentration increases which the gap distance 2.96 µm in the composite 0.20 (w/w). The morphological structure of the composite accordance with the trends obtained when measuring the density value of the sample. The hydroxyl group in cellulose plays an important role in the mechanism of forming bonds between molecules with polymer chains [46]. Interaction between cellulose molecules and polyurethane polymers which can increase the density and decreases the gap between the filler and the polymer [45].

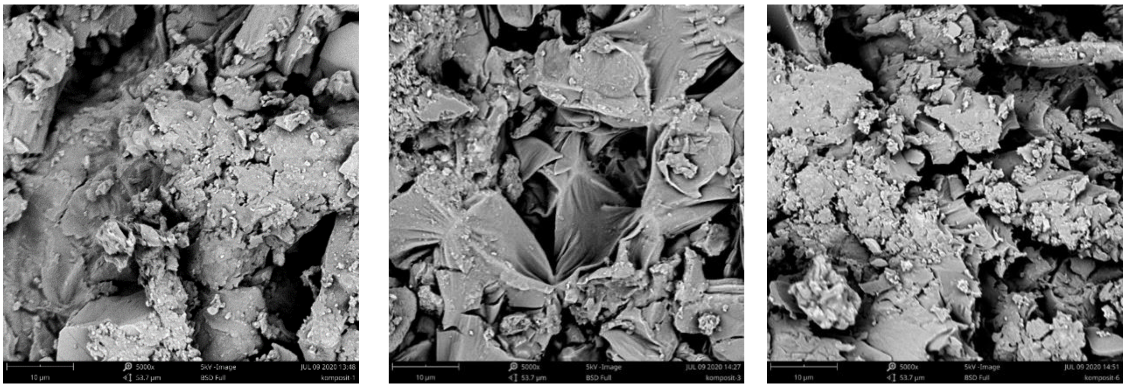


Figure 8. SEM of composite (a) 0.04 (w/w); (b) 0.12 (w/w); (c) 0.20 (w/w).

4. Conclusion

We have successfully developed a Teak leaves-waste composite utilized as particleboard. The mechanical and physical properties of the composites have been investigated. The test results showed a composite compressive strength attains 38.5 MPa. The highest density is 1261 kg.m^{-3} that is in agreement to the American Society standard. The lowest value of water absorption is 1.38%. In addition, we have successfully approximated the van der Waals interaction with the spectrum shifts of the FTIR spectroscopy as a confirmation of the compressive strength enhancement. Morphological structure shows that the gap between mixture decreases as the polyurethane fraction increases according to the density of the composite. However, the properties of organic composite are hygroscopic so they are vulnerable to being the target of fungal growth and susceptible to fire as an application in building fields. These problems will be focus of further research by applying coating technology approaches.

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Mechanical and physical properties of teak leaves waste/polyurethane composites for particleboard application

Masturi[1], WN Jannah[1], RM Maulana[1], T Darsono[1], Sunarno[1] and S Rustad[2][AQ2]

Abstract

A teak leaf waste/polyurethane composite has been made for particleboard application. Some fraction variations are performed on the use of polyurethane as a matrix with a range of 0.04–0.20 (w/w). Mechanical and physical properties have been tested on the composites produced. The test results showed that the highest compressive strength of the sample reaches 38.5 MPa for polyurethane fraction of 0.14 (w/w). The composite has a density of 1261 kg m^{-3} which is in accordance with the result of density on teak. The physical properties have been also investigated and it was found that the lowest value of water absorption is 1.38%. This result indicated that the composites produced are potentially to replace wood raw material in eco-friendly industrial scale.

Keywords

composite, polyurethane, teak leaf, mechanical properties

Introduction

In the last few decades, total global wood demand for 2020 is projected to reach 515 million m^3.[1] Composite technique is one of the possible ways to fulfill the demand. Composite engineering promises organic and inorganic waste utilization as a synthesized material having excellent properties [AQ4] relating the woods, such as mechanical strength, water absorption, density, and so on.[2–6] Furthermore, the composite engineering is also a solution for the raising of environment awareness caused by the high deforestation in several countries.[2,3] In addition, the utilization of natural waste and fibers for some composites has been widely studied and developed from various countries including Indonesia, Malaysia, Thailand, India, Brazil, and Japan.[2–4]

In recent years, the use of composites is very widespread in the electronics, automotive and aviation industries, construction field, multipurpose applications such as particleboards, energy storage, heat and electricity insulators, ship bodies and aircraft, organic field-effect transistors (FETs), antennas, and so on due to their high mechanical strength, good thermal stability, anticorrosion, high dielectric constant, lightweight, and easy to produce large scale.[7–13] Several studies about the polymers and plastics are focused on developing fibers, plastics, adhesives, composite textiles, and paper from renewable resources derived mostly from agricultural wastes containing lignocellulosic.[4] Considering that agricultural waste is only used for compost production in most developing countries such as Indonesia, researcher also focused on utilization of some natural fibers such as sisal, hemp, bamboo, and agricultural wastes, for example, leaves, coconut fibers, rice husk, peanut shells,

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and straw as filler in polymer composite.[5,6] Composites of these natural fibers have several advantages such as being easy to process, environmentally, good acoustic properties, low energy processes, good mechanical and thermal properties, water resistance, noncorrosive, nontoxic, and naturally degradable.[7]

Further, several studies related to the leaves waste composites have been done, such as Masturi et al. developed composite from leaves waste and paper waste using polyvinyl acetate as matrix and the compressive strength reaches to 45.60 MPa.[2] Campanella et al. successfully made a composite of Northern Red Oak leaves with plant oil-based resins,[6] while Thakur et al. have also developed biopolymer composite using lignocellulosic of *Hibiscus Sabdariffa*.[14] A different method has been performed by Nongman et al. having created a composite by lamination method of banana leaf fiber and adhesive.[5] Senthilkumar et al. have fabricated pineapple leaf fiber/polyester composites which have a tensile strength reached to 33.13 MPa. Good mechanical properties related to the high content of cellulose in pineapple leaf fibers can transfer a sizable load from the matrix to the fiber effectively.[15] In addition, Melkamu et al. have investigated the mechanical properties and water absorption in sisal/polyester fiber composites with treated fibers and without alkaline sodium hydroxide treatment. Compressive strength in sisal fibers treated better than untreated fibers with low water absorption advantages.[16]

In this work, we developed a composite of teak leaves waste with polyurethane matrix. The teak leaves have been chosen since its utilization only as compost having a low added value as well as its availability is very abundant in Indonesia. Polyurethane as a thermosetting polymer was selected because it does not melt when heated, is more resistant to acids and bases, has a high adhesive power due to the presence of highly reactive isocyanate functional groups, has good erosion resistance which can be used in anti-erosion film and coating technology,[17] has an isotropic and controllable electromagnetic response, and has a negative permittivity value which is potentially applied to the electronics field.[18,19] Furthermore, the addition of nanoparticles in polyurethane can improve thermal stability and mechanical properties of composites. Polyurethane is also used as a textile coating on inkjet-printed flexible and washable FETs on textile technology.[20] The aim of this study is to produce a teak leaves waste composite having a high compressive strength and low water absorption making a chance to be used on an eco-friendly industrial scale.

Materials and methods

The teak leaves waste of Semarang Indonesia was used, while isocyanates and polyol as[AQ5] polyurethane (PU) polymerization materials were purchased in Multi Kimia Inc., Semarang, Indonesia.

First, the leaves waste was dried and crushed using a blender machine to form a powder. Then, the powder was sieved using 700 mesh. On the other hand, the isocyanates and polyol were mixed using a magnetic stirrer for 5 min with a ratio of 1:1 to form polyurethane and mixed with the teak leaves waste powder with varied composition while the leaves teak powder was kept at 12 g weight. The PU/teak leaves waste mixture was stirred using mixer machine to homogenize the mixture, then pressed into a cylindrical mold at a pressure of 5 MPa for 15 min. Having been pressed, the composite was removed from the mold and dried for 24 h at room temperature.

The investigation to the content of the teak leaves powder cellulose bonding to polyurethane and detecting spectral changes in teak leaves waste composite was performed using Fourier transform infrared (FTIR)[AQ6] Spectrometer Frontier type (Perkin Elmer[AQ7]) in a spectral range of 400–4000 cm^{-1}. Meanwhile, the identification of the mechanical properties was measured by a compressive strength test using Sociedad Espanola Anonima Ibertest Instrument with a maximum force of 250 kN and speed of movements was 0.05 kN s^{-1}. In this test, the sample is cut into cubic shape and was measured using testing machine. In addition, to ensure the water resistance of the composite, the identification of the physical properties was performed by density measurement;[AQ8] further, water absorption test was done by soaking this sample in the water for 24 h to obtain the weight of water absorbed into the composite compared to the initial weight. Surface morphology has also been investigated using Phenom pro X desktop scanning electron microscopy (SEM)[AQ9] with EDX[AQ10] with a current of 20 mA for 90 s. Samples were coated with gold (Au Pd) using the[AQ11] Quorum Sc7620 Sputter Coater tool.

Results and discussion

Compressive strength test

Teak leaves waste/polyurethane composite have been fabricated successfully as shown in Figure 1(a). Figure 1(b) shows the effect of polyurethane addition on composite compressive strength. Composites 0.04 and 0.08 (w/w) have relatively low compressive strength because many teak leaves particles do not bind to the polymer matrix. The highest compressive strength achieved 38.5 MPa for the polyurethane fraction 0.14 (w/w). The resulting strength is 86% higher than polyurethane fraction 0.04 (w/w) which is closely related to the interaction between the surface of the particles and the polymer as shown in Figure 2. In polyurethane fraction 0.14 (w/w), all polymer matrix exactly interacts with all teak leaves particles, and composite is more compact and can sustain a higher load.

Figure 2(a) illustrates the bonding of leaves teak particles and mass varied of polyurethane polymers at 0.04–0.08 (w/w). When the mixture of filler and polymer is pressed to

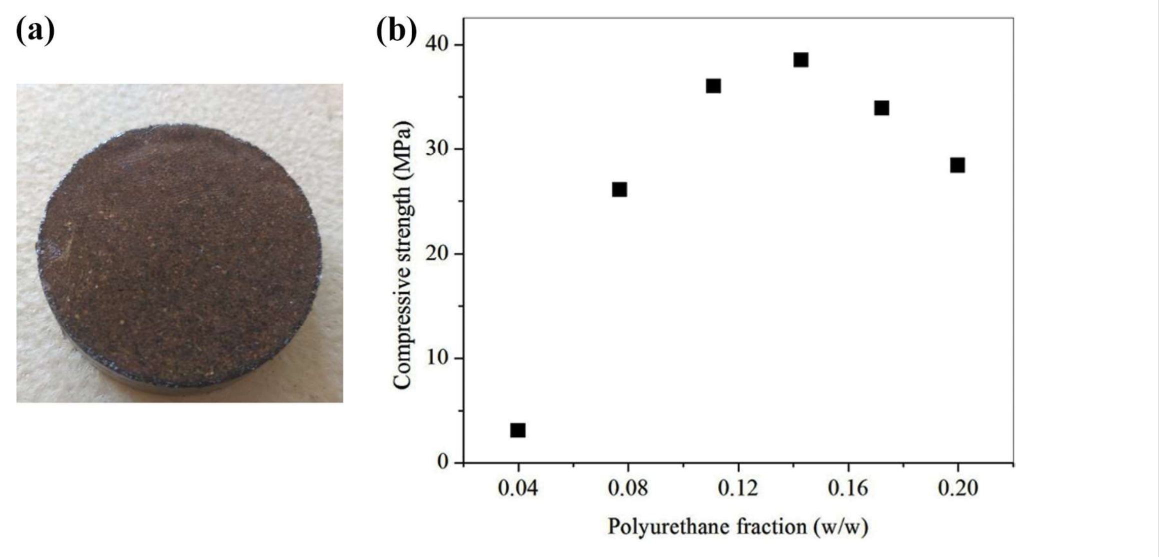

Figure 1. (a) Teak leaves waste/polyurethane composite. (b) Compressive strength of teak leaves waste composite as polyurethane content (w/w) and the compressive strength attain at maximum for 0.14 fraction (w/w).

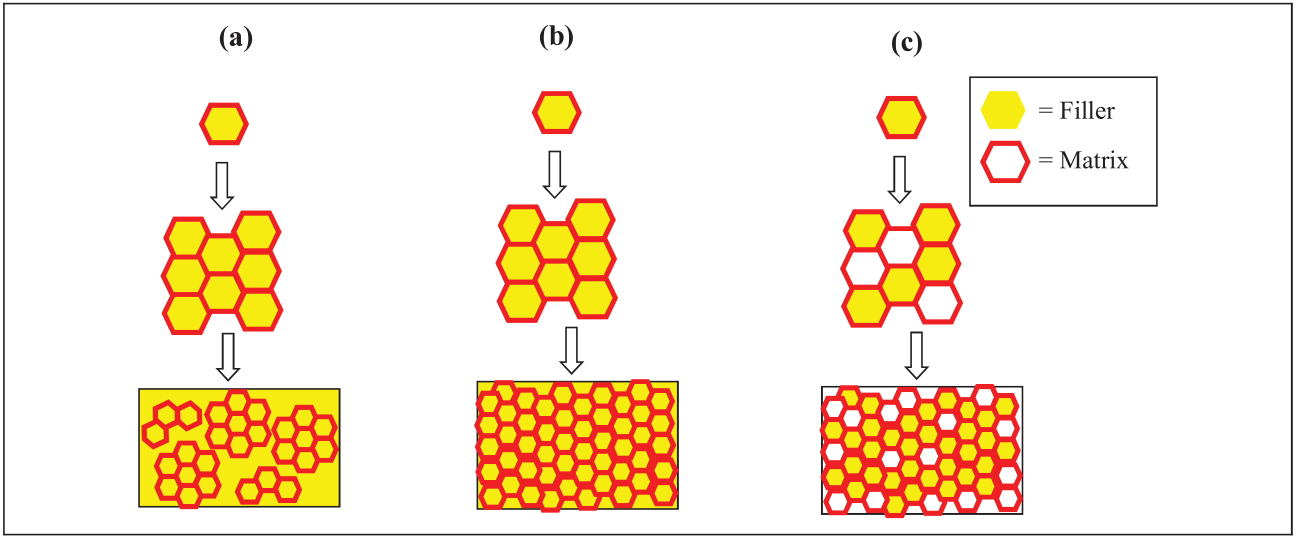

Figure 2. Schematic of the particle distribution of filler and polymer matrix. (a) Low compressive strength, (b) high compressive strength, and (c) low compressive strength.

form a composite, the polyurethane[AQ12] polymer partially interacts with leaf particles and some particles diffuse which not interact with each other. It can act as cracking initiator and composite might be brittle. The addition of polyurethane content at 0.12–0.14 (w/w) can improve the composite compressive strength. As shown in Figure 2(b), a composite 0.14 (w/w) is called the optimal fraction because all the teak leaves particles are precisely bound to the polymer matrix. The matrix bond forward load in all directions, composite be stronger and resistant to the maximum load.[21] However, the compressive strength produced does not increase continuously but reaches the maximum value. Further, as shown in Figure 2(c), the composites with a polyurethane[AQ13] content of 0.17–0.20 (w/w) show some parts of polyurethane not binding to the filler, appear many voids from polyurethane polymer, teak leaves would tend to agglomerate and not bond properly which can reduce compressive strength.[22,23] According to Jaya et al., when the polymer was added much more than the filler there will be a very poor adhesion force between surfaces because polyurethane is a hydrophobic matrix and teak leaves powder is a hydrophilic filler.[23] Further, the raising of the strength until certain fraction is related to the maximum contact amount between the polymer and solid waste particle.[24]

The most important interaction in the composite is predominantly emerged between the polyurethane and the cellulose in teak leaves. According to Alemdar and Sain, the

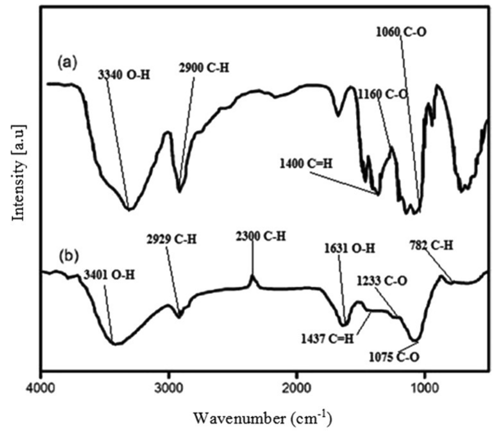

Figure 3. Spectrum of cellulose of (a) pure cellulose obtained from Ramirez-Flores[13] and (b) teak leaves powder in this work where many peaks of both celluloses have more similarities.[AQ30]

cellulose in plant fibers has important role in reinforcing material composite applications.[25] The presence of cellulose in teak leaves can be investigated through the FTIR spectrum in Figure 3.

Figure 3(a) and (b) shows the FTIR spectrum of the pure cellulose obtained from Ramirez-Flores et al.[26] and the teak leaves, respectively, with the comparison given in Table 1. The FTIR spectrum of pure cellulose and leaves teak powder celluloses has a significant difference at peak 1631 cm^{-1}. The peak of 1631 cm^{-1} indicates the absorption band of hydrogen forming a hydrogen bridge from hydroxyl groups and oxygen atoms from glucose monomer and usually found at a cellulose fiber.[27] The peak of 1631 cm^{-1} is not found at pure cellulose, but it is found at cellulose of natural fibers.[28–31]

The peak of 2929 and 2300 cm^{-1} indicates[AQ14] a strong C–H bond in cellulose polymers,[31] while peak of 3401 cm^{-1} shows the stretching of the O–H group bonds.[32] Further, the peak of 780 and 1438 cm^{-1} shows the C–H bonds connecting the glucose units of cellulose. The peak at 1234 cm^{-1} indicates that there is a C–O–C group which is the structure of lignin. The absorption area occurring at 1076 cm^{-1} indicates the presence of a C–O bond in the cellulose component.[23]

Using the FTIR analysis, it can be shown that cellulose and lignin components are found in the teak leaves. This leaf can compete with the conventional fiber such as glass fiber regarding its high performance as reinforcing material for thermosetting and thermoplastic matrices with their naturally degradable superior properties.[33]

The interaction arising between the teak leaves and the polymer is a van der Waals interaction. This interaction was confirmed by several FTIR spectrum shifts as shown in Figure 4. The shifts in spectrum are appeared due to the vibrating atoms as a result of the energy of disruption of atoms closing each other.

Table 1. Comparison of the peak absorption at pure cellulose and cellulose of teak leaves powder.

	Wavenumber (cm^{-1})	
Bond	Pure cellulose (cm^{-1})[15]	Cellulose of teak leaves powder (cm^{-1})
O–H	3340–3500	3401
C–H	2800–2920	2929
C–O–C	1160	1233
C–O	1035–1060	1075
C=H	1400	1437

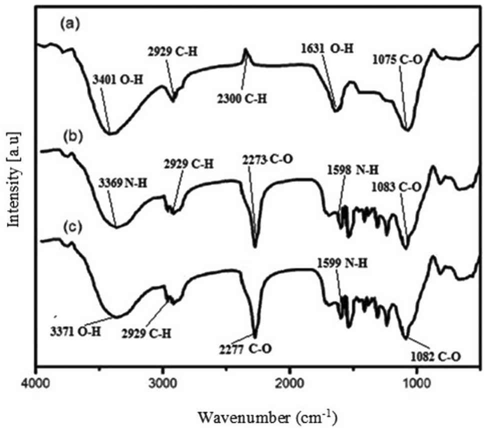

Figure 4. FTIR spectrum of (a) teak leaves, (b) polyurethane, and (c) PU/teak leave composite. FTIR: Fourier transform infrared.

Polyurethane has two functional groups, that is, carbonyl and amine groups. The spectrum shift starts[AQ15] to occur of 3369 cm^{-1} to 3371 cm^{-1} indicates the O–H bond. This shift is because the amine group of the polyurethane was disturbed by hydrogen bonding in the hydroxyl group of leaves and forming a hydrogen bridge.[34] Further, the shift of 2273 cm^{-1} to 2277 cm^{-1} shows that the carbonyl group of polyurethane undergoes a spectral shift. This is due to the disturbance of the C–H leaves bonding to the carbonyl group forming the O–H bridges. The peak at 1598 cm^{-1} shifting to 1599 cm^{-1} also indicates the amine group in polyurethane disturbed by the O–H bonds of the teak leaves. Furthermore, the shift also occurs at the carbonyl peak of 1083 cm^{-1} to 1082 cm^{-1} due to the disturbance of the leaves' C–O bond of the peak of 1075 cm^{-1} (Figure 5). Similar results were confirmed by a previous work that [AQ16] PVA-sepiolite composite derived its van der Waals interactions between silanol groups and carbon C–O and shifted from 1088 cm^{-1} to 1085 cm^{-1}.[35]

Density test

Figure 6 shows the density of composite samples with polyurethane variation ranging from 825 kg m^{-3} to 1261

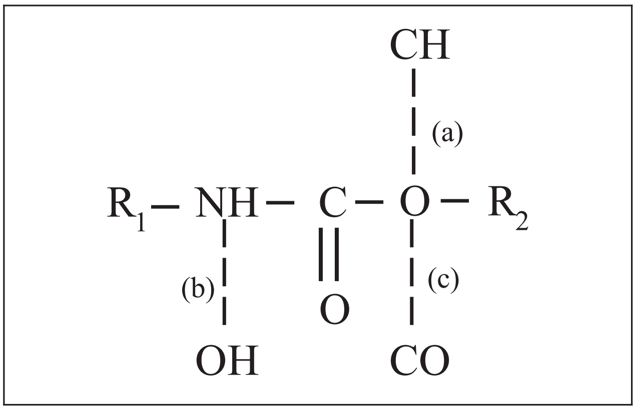

Figure 5. Van der Waals interactions on the teak leaf waste composites, dashed lines illustrate the van der Waals interaction area of (a) CH and carbonyl group, (b) OH and amine group, and (c) CO and carbonyl group.

kg m^{-3}. Considering the interpretation data showing a coefficient of determination, $R^2 = 0.91157$, there is a linear relationship between the polyurethane content and the composite density. Similar results were obtained by Afzaludin et al. on sugar palm/glass fiber-reinforced thermoplastic polyurethane.[36] The lowest density value is found in the sample with a polyurethane fraction (w/w) of 0.04, while the highest density value is in the sample with 0.20 of polyurethane fraction (w/w). It can be explained when the polyurethane polymer content rises and the filler is fixed, then the number of links produced by the polymer increases. Hence, the increased links result in stronger bonds to bind the filler which can increase the density.[37]

The density in samples with a composition[AQ17] of 0.08 (w/w), in accordance with teak wood that has been studied by Wahyudi et al., is in the range of 1000–1200 kg m^{-3} with an average value of 1160 kg m^{-3}.[38] In this study, the resulted density value is higher than the result of Martins et al. producing particleboard with a density of 510–570 kg m^{-3}.[28] The composites produced in this work have a similar density to the results of the Raju et al. producing some composites with densities ranging from 1100 kg m^{-3} to 1320 kg m^{-3}.[29] Further, Butylina et al. have produced wood fiber–polypropylene composite having a density ranging from 1040 kg m^{-3} to 1140 kg m^{-3}.[30]

The density data shows that this composite belong to the high-density particleboard category due to has a[AQ18] density more than 0.80 g cm^{-3} in accordance with the standard designation 1554-67 by American Society.[39] Additionally, the composite has also met the Indonesia National Standard of 03-2105-2006 where particleboard density having values in the range of 400–900 kg m^{-3}.[40]

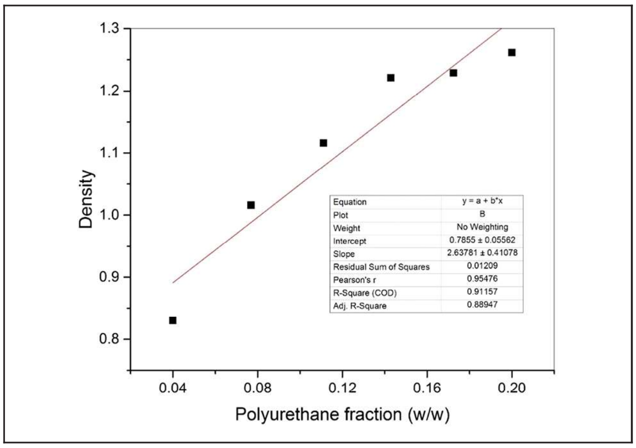

Figure 6. Composite density as polyurethane fraction (w/w) function, and it was linearity relation as; $y = 2.6378x + 0.7855$, where x and y are polyurethane fraction and composite density, respectively, with $R^2 = 0.91157$.

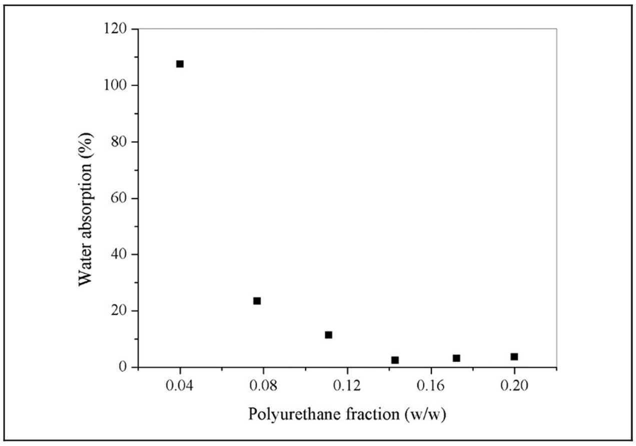

Figure 7. Water absorption of composite tends to zero for larger polyurethane fraction (w/w).

Water absorption

Water absorption values were measured to determine the ability of teak leaf composite to absorb water after 24 h of soaking. The highest water absorption rate reaches 107.35% in composites with a polyurethane fraction of 0.04 (w/w). After being immersed for 24 h, the composite undergoes thick development until the composite becomes cracked and broken because the cavity that appears between the matrix and the filler particles absorbs a lot of water and can further affect the stability of the dimensions of the composite. In addition, teak leaves as a hydrophilic filler also absorb a lot of water during immersion.

Figure 7 shows that the water[AQ19] absorption value decreases with the increase of polyurethane fraction. At high fractions, it tends to produce composites with lower water absorption. The lowest water absorption was achieved in samples having the most optimal compressive strength at fraction 0.14 (w/w). The cavity formed is very small because the pressing process in the fabrication of composites and filler particles is distributed evenly on the matrix and absorbed a little water.

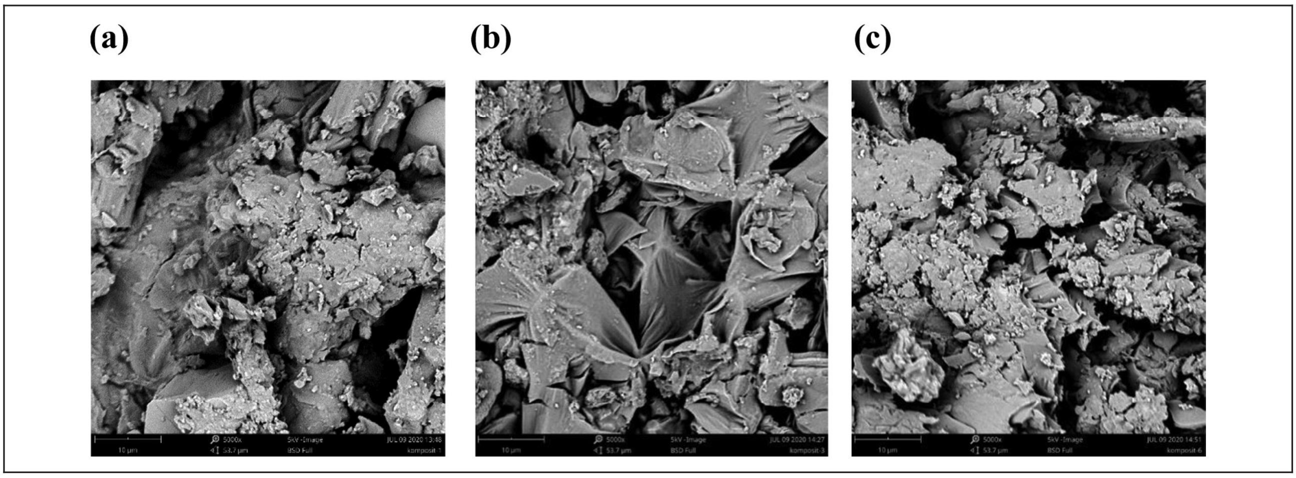

Figure 8. SEM of composite (a) 0.04 (w/w), (b) 0.12 (w/w), and (c) 0.20 (w/w). SEM: scanning electron microscopy.

The absorption value of the leaves teak waste composite produced is better than that of several leaves waste composites, which has been developed by several authors, such as Al-Sulaiman et al. produced palm fiber-reinforced composites with the water absorption ranging between 2.3% and 5.8%,[41] and Batiancela et al. developed a wood particle from tea leaves waste and obtained the water absorption about 23–40%.[42] Therefore, in accordance with the several properties above, this composite has properness in competing the woods.

Scanning electron microscopy

Morphological structure using SEM of composites 0.04, 0.12, and 0.20 (w/w) is shown in Figure 8. There is a cavity between the filler mixture and the matrix. Based on data analysis, the gap value between molecules in the composite 0.04 (w/w) is 13 µm. The low concentration of filler results in agglomeration in certain parts, and gaps between the mixture appear large enough which can act as cause of cracking and breakage when subjected to a load.[43] This was confirmed by the compressive strength and density results on the composite 0.04 (w/w) resulting in the lowest compressive strength and density. The resulting gap will be narrower when the concentration of polyurethane polymers is higher and the density increases.[44] The gap value in the composite 0.12 (w/w) is 4.65 µm and[AQ20] decreases when the polyurethane concentration increases and the gap distance 2.96 µm in the composite 0.20 (w/w). The morphological structure of the composite in accordance with the trends is obtained when measuring the density value of the sample. The hydroxyl group in cellulose plays an important role in the mechanism of forming bonds between molecules with polymer chains.[45] Interaction between cellulose molecules and polyurethane polymers can increase the density and decreases the gap between the filler and the polymer.[44]

Conclusion

We have successfully developed a teak leaves waste composite utilized as particleboard. The mechanical and physical properties of the composites have been investigated. The test results showed that a composite compressive strength attains 38.5 MPa. The highest density is 1261 kg m^{-3} which is in agreement with the American Society standard. The lowest value of water absorption is 1.38%. In addition, we have successfully approximated the van der Waals interaction with the spectrum shifts of the FTIR spectroscopy as a confirmation of the compressive strength enhancement. Morphological structure shows that the gap between mixture decreases as the polyurethane fraction increases according to the density of the composite. However, the properties of organic composite are hygroscopic so they are vulnerable to being the target of fungal growth and susceptible to fire as an application in building fields. These problems will be the focus of further research by applying coating technology approaches.

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Declaration of conflicting interests

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AQ: 20	Please approve edits made to the sentence “The gap value in the composite 0.12 (w/w) is 4.65 µm and . . . ”.
AQ: 21	Please check whether the “Acknowledgment” and “Funding” sections are OK as set.
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AQ: 24	Please note that Refs 23 to 39, Ref. 29, Refs. 40 to 46, and Ref. 22 have been changed to Refs 22 to 37, Ref. 38, Refs 39 to 45, and Ref. 46, respectively, to maintain sequential order here as well as inside the text. Please approve.
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Mechanical and physical properties of teak leaves waste/polyurethane composites for particleboard application

Masturi[1], WN Jannah[1], RM Maulana[1], T Darsono[1], Sunarno[1] and S Rustad[2][AQ2]

AQ2 = OK

Abstract

A teak leaf waste/polyurethane composite has been made for particleboard application. Some fraction variations are performed on the use of polyurethane as a matrix with a range of 0.04–0.20 (w/w). Mechanical and physical properties have been tested on the composites produced. The test results showed that the highest compressive strength of the sample reaches 38.5 MPa for polyurethane fraction of 0.14 (w/w). The composite has a density of 1261 kg m^{-3} which is in accordance with the result of density on teak. The physical properties have been also investigated and it was found that the lowest value of water absorption is 1.38%. This result indicated that the composites produced are potentially to replace wood raw material in eco-friendly industrial scale.

Keywords

composite, polyurethane, teak leaf, mechanical properties

Introduction

In the last few decades, total global wood demand for 2020 is projected to reach 515 million m^3.[1] Composite technique is one of the possible ways to fulfill the demand. Composite engineering promises organic and inorganic waste utilization as a synthesized material having excellent properties [AQ4] relating the woods, such as mechanical strength, water absorption, density, and so on.[2–6] Furthermore, the composite engineering is also a solution for the raising of environment awareness caused by the high deforestation in several countries.[2,3] In addition, the utilization of natural waste and fibers for some composites has been widely studied and developed from various countries including Indonesia, Malaysia, Thailand, India, Brazil, and Japan.[2–4]

AQ4: OK

In recent years, the use of composites is very widespread in the electronics, automotive and aviation industries, construction field, multipurpose applications such as particleboards, energy storage, heat and electricity insulators, ship bodies and aircraft, organic field-effect transistors (FETs), antennas, and so on due to their high mechanical strength, good thermal stability, anticorrosion, high dielectric constant, lightweight, and easy to produce large scale.[7–13] Several studies about the polymers and plastics are focused on developing fibers, plastics, adhesives, composite textiles, and paper from renewable resources derived mostly from agricultural wastes containing lignocellulosic.[4] Considering that agricultural waste is only used for compost production in most developing countries such as Indonesia, researcher also focused on utilization of some natural fibers such as sisal, hemp, bamboo, and agricultural wastes, for example, leaves, coconut fibers, rice husk, peanut shells,

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AQ3 = OK

[AQ1]

AQ1 = OK

and straw as filler in polymer composite.[5,6] Composites of these natural fibers have several advantages such as being easy to process, environmentally, good acoustic properties, low energy processes, good mechanical and thermal properties, water resistance, noncorrosive, nontoxic, and naturally degradable.[7]

Further, several studies related to the leaves waste composites have been done, such as Masturi et al. developed composite from leaves waste and paper waste using polyvinyl acetate as matrix and the compressive strength reaches to 45.60 MPa.[2] Campanella et al. successfully made a composite of Northern Red Oak leaves with plant oil-based resins,[6] while Thakur et al. have also developed biopolymer composite using lignocellulosic of *Hibiscus Sabdariffa*.[14] A different method has been performed by Nongman et al. having created a composite by lamination method of banana leaf fiber and adhesive.[5] Senthilkumar et al. have fabricated pineapple leaf fiber/polyester composites which have a tensile strength reached to 33.13 MPa. Good mechanical properties related to the high content of cellulose in pineapple leaf fibers can transfer a sizable load from the matrix to the fiber effectively.[15] In addition, Melkamu et al. have investigated the mechanical properties and water absorption in sisal/polyester fiber composites with treated fibers and without alkaline sodium hydroxide treatment. Compressive strength in sisal fibers treated better than untreated fibers with low water absorption advantages.[16]

In this work, we developed a composite of teak leaves waste with polyurethane matrix. The teak leaves have been chosen since its utilization only as compost having a low added value as well as its availability is very abundant in Indonesia. Polyurethane as a thermosetting polymer was selected because it does not melt when heated, is more resistant to acids and bases, has a high adhesive power due to the presence of highly reactive isocyanate functional groups, has good erosion resistance which can be used in anti-erosion film and coating technology,[17] has an isotropic and controllable electromagnetic response, and has a negative permittivity value which is potentially applied to the electronics field.[18,19] Furthermore, the addition of nanoparticles in polyurethane can improve thermal stability and mechanical properties of composites. Polyurethane is also used as a textile coating on inkjet-printed flexible and washable FETs on textile technology.[20] The aim of this study is to produce a teak leaves waste composite having a high compressive strength and low water absorption making a chance to be used on an eco-friendly industrial scale.

Materials and methods

The teak leaves waste of Semarang Indonesia was used, while isocyanates and polyol as[AQ5] polyurethane (PU) polymerization materials were purchased in Multi Kimia Inc., Semarang, Indonesia.

AQ5: we use only
"polyurethane", without
"(PU)"

First, the leaves waste was dried and crushed using a blender machine to form a powder. Then, the powder was sieved using 700 mesh. On the other hand, the isocyanates and polyol were mixed using a magnetic stirrer for 5 min with a ratio of 1:1 to form polyurethane and mixed with the teak leaves waste powder with varied composition while the leaves teak powder was kept at 12 g weight. The ~~PU/ teak leaves~~ waste mixture was stirred using mixer machine to homogenize the mixture, then pressed into a cylindrical mold at a pressure of 5 MPa for 15 min. Having been pressed, the composite was removed from the mold and dried for 24 h at room temperature.

The investigation to the content of the teak leaves powder cellulose bonding to polyurethane and detecting spectral changes in teak leaves waste composite was performed using Fourier transform infrared (FTIR)[AQ6] Spectrometer Frontier type (Perkin Elmer[AQ7]) in a range of 400–4000 cm^{-1}. Meanwhile, the identi of the mechanical properties was measured by a c sive strength test using Sociedad Espanola Anonima Ibertest Instrument with a maximum force of 250 kN and speed of movements was 0.05 kN s^{-1}. In this test, the sample is cut into cubic shape and was measured using testing machine. In addition, to ensure the water resistance of the composite, the identification of the physical properties was performed by density measurement;[AQ8] further, water absorption test was done by soaking this sample in the water for 24 h to obtain the weight of water absorbed into the composite compared to the initial weight. Surface morphology has also been investigated using Phenom pro X desktop scanning electron microscopy (SEM)[AQ9] with EDX[AQ10] with a current of 20 mA for 90 s. Samples were coated with gold (Au Pd) using the[AQ11] Quorum Sc7620 Sputter Coater tool.

AQ6 = OK

AQ7 = location of
Perkin Elmer
manufacturer is:
Massachusetts, U.S.A.

AQ8 = OK

AQ9 = OK

AQ10, we
complete to
"with
energy-
dispersive
X-ray
(EDX)..."

AQ11 = we compete to
"Quorum Sc7620 Sputter
Coater tool produced by
Quantum Design AG,

Results and discussion

Compressive strength test

Teak leaves waste/polyurethane composite have been fabricated successfully as shown in Figure 1(a). Figure 1(b) shows the effect of polyurethane addition on composite compressive strength. Composites 0.04 and 0.08 (w/w) have relatively low compressive strength because many teak leaves particles do not bind to the polymer matrix. The highest compressive strength achieved 38.5 MPa for the polyurethane fraction 0.14 (w/w). The resulting strength is 86% higher than polyurethane fraction 0.04 (w/w) which is closely related to the interaction between the surface of the particles and the polymer as shown in Figure 2. In polyurethane fraction 0.14 (w/w), all polymer matrix exactly interacts with all teak leaves particles, and composite is more compact and can sustain a higher load.

Figure 2(a) illustrates the bonding of leaves teak particles and mass varied of polyurethane polymers at 0.04–0.08 (w/w). When the mixture of filler and polymer is pressed to

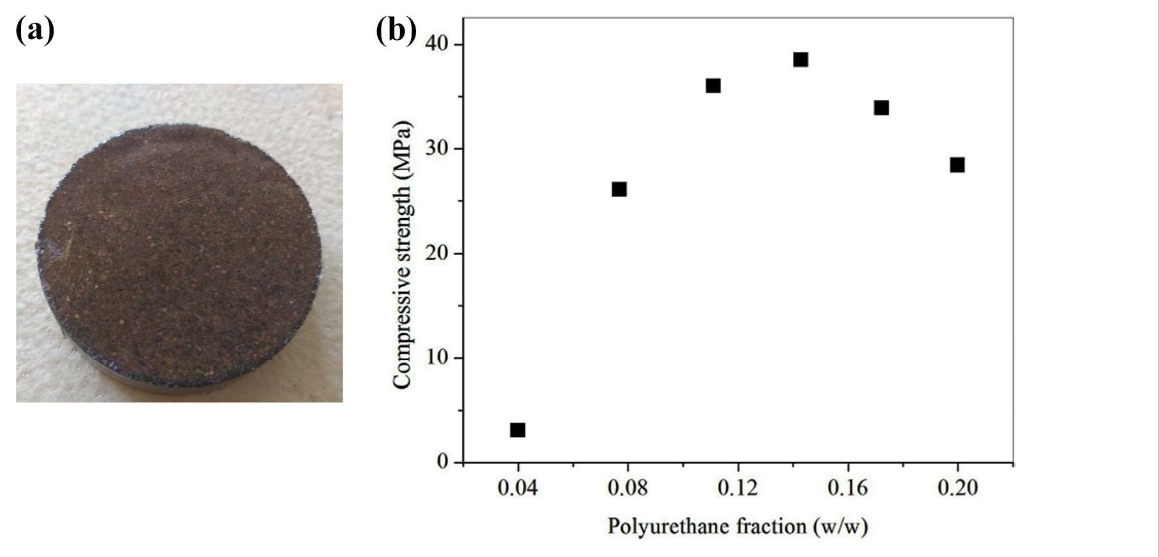


Figure 1. (a) Teak leaves waste/polyurethane composite. (b) Compressive strength of teak leaves waste composite as polyurethane content (w/w) and the compressive strength attain at maximum for 0.14 fraction (w/w).

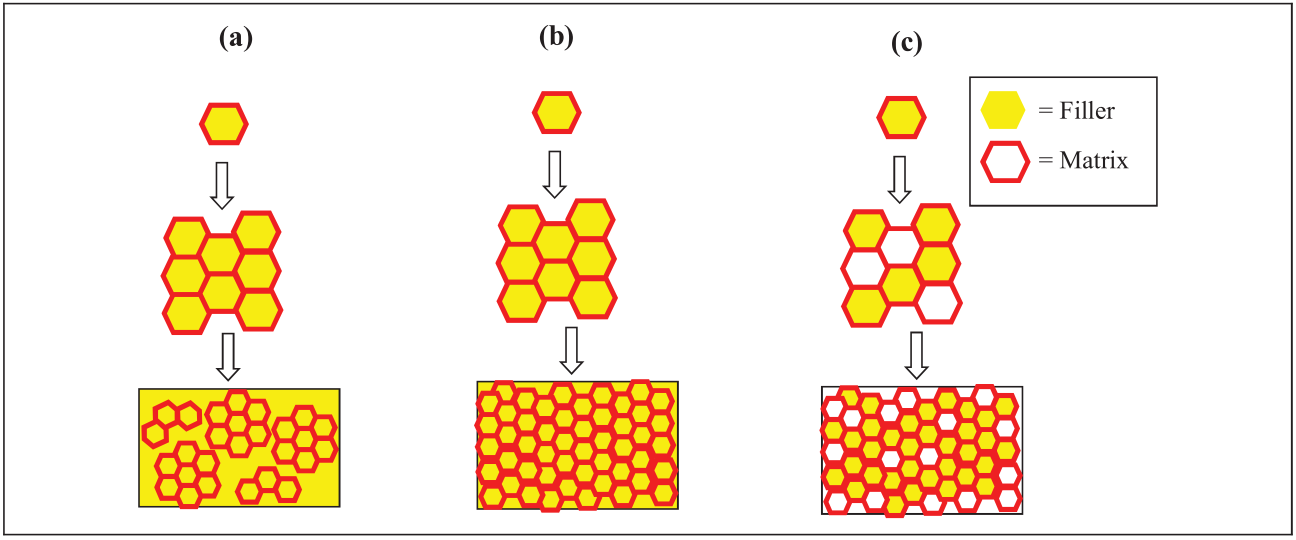


Figure 2. Schematic of the particle distribution of filler and polymer matrix. (a) Low compressive strength, (b) high compressive strength, and (c) low compressive strength.

AQ12: We may rewrite as:

"When the mixture of filler and polymer is pressed to form a composite, the polyurethane polymer partially interacts with leaf particles, and another particles diffuse out and do not result in polyurethane-leaf particle interaction."

ane[AQ12] polymer par-
and some particles diffuse
er. It can act as cracking
brittle. The addition of
4 (w/w) can improve the
As shown in Figure 2(b),
led the optimal fraction
icles are precisely bound
bond forward load in all
and resistant to the max-

imum load.[21] However, the compressive strength produced does not increase continuously but reaches the maximum value. Further, as shown in Figure 2(c), the composites with a polyurethane[AQ13] content of 0.17–0.20 (w/w) show some parts of polyurethane not binding to the filler, appear many voids from polyurethane polymer, teak leaves would tend to agglomerate and not bond properly which can reduce compressive strength.[22,23] According to Jaya et al., when the polymer was added much more than the filler there will be a very poor adhesion force between surfaces because polyurethane is a hydrophobic matrix and teak leaves powder is a hydrophilic filler.[23] Further, the raising of the strength until certain fraction is related to the maximum contact amount between the polymer and solid waste particle.[24]

The most important interaction in the composite is predominantly emerged between the polyurethane and the cellulose in teak leaves. According to Alemdar and Sain, the

AQ13: it is OK. It explains the low compressive strength occurs as result of many voids forming due to agglomeration of teak leaves

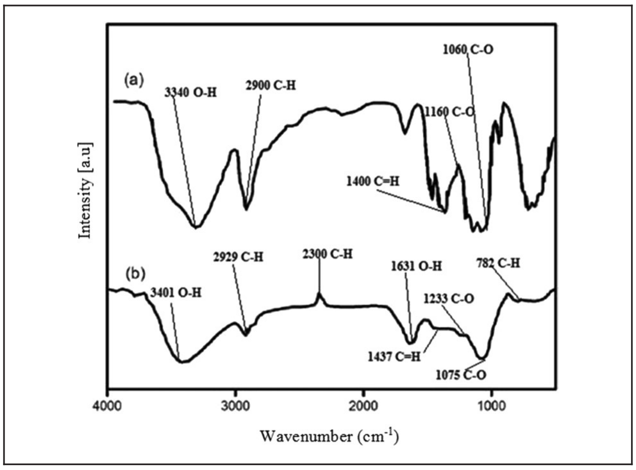


Figure 3. Spectrum of cellulose of (a) pure cellulose obtained from Ramirez-Flores[13] and (b) teak leaves powder in this work where many peaks of both celluloses have more similarities.[AQ30]

AQ30: OK

cellulose in plant fibers has important role in reinforcing material composite applications.[25] The presence of cellulose in teak leaves can be investigated through the FTIR spectrum in Figure 3.

Figure 3(a) and (b) shows the FTIR spectrum of the pure cellulose obtained from Ramirez-Flores et al.[26] and the teak leaves, respectively, with the comparison given in Table 1. The FTIR spectrum of pure cellulose and leaves teak powder celluloses has a significant difference at peak 1631 cm^{-1}. The peak of 1631 cm^{-1} indicates the absorption band of hydrogen forming a hydrogen bridge from hydroxyl groups and oxygen atoms from glucose monomer and usually found at a cellulose fiber.[27] The peak of 1631 cm^{-1} is not found at pure cellulose, but it is found at cellulose of natural fibers.[28–31]

AQ14: OK

The peak of 2929 and 2300 cm^{-1} indicates[AQ14] a strong C–H bond in cellulose polymers,[31] while peak of 3401 cm^{-1} shows the stretching of the O–H group bonds.[32] Further, the peak of 780 and 1438 cm^{-1} shows the C–H bonds connecting the glucose units of cellulose. The peak at 1234 cm^{-1} indicates that there is a C–O–C group which is the structure of lignin. The absorption area occurring at 1076 cm^{-1} indicates the presence of a C–O bond in the cellulose component.[23]

Using the FTIR analysis, it can be shown that cellulose and lignin components are found in the teak leaves. This leaf can compete with the conventional fiber such as glass fiber regarding its high performance as reinforcing material for thermosetting and thermoplastic matrices with their naturally degradable superior properties.[33]

The interaction arising between the teak leaves and the polymer is a van der Waals interaction. This interaction was confirmed by several FTIR spectrum shifts as shown in Figure 4. The shifts in spectrum are appeared due to the vibrating atoms as a result of the energy of disruption of atoms closing each other.

Table 1. Comparison of the peak absorption at pure cellulose and cellulose of teak leaves powder.

	Wavenumber (cm^{-1})	
Bond	Pure cellulose (cm^{-1})[15]	Cellulose of teak leaves powder (cm^{-1})
O–H	3340–3500	3401
C–H	2800–2920	2929
C–O–C	1160	1233
C–O	1035–1060	1075
C=H	1400	1437

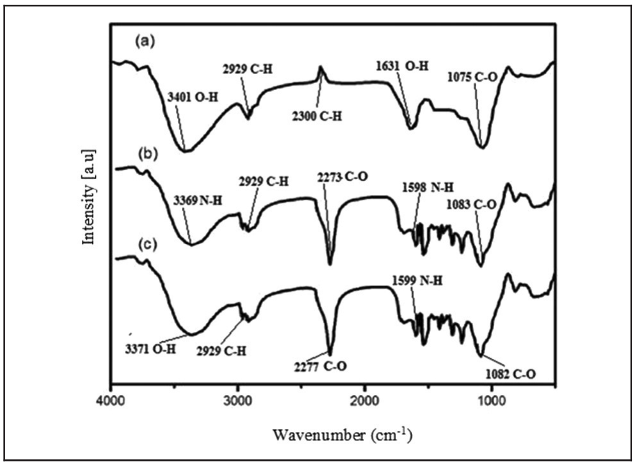


Figure 4. FTIR spectrum of (a) teak leaves, (b) polyurethane, and (c) ~~PU/teak leave composite~~. FTIR: Fourier transform infrared.

AQ15: We replace with "The spectrum shift occurs from 3369 cm-1 to 3371 cm-1 that indicates the O-H bonds."

Polyurethane has two functional groups, th nyl and amine groups. The spectrum shift star occur of 3369 cm^{-1} to 3371 cm^{-1} indicates th This shift ~~is because~~ the amine group of the polyurethane was disturbed by hydrogen bonding in the hydroxyl group of leaves and forming a hydrogen bridge.[34] Further, the shift of 2273 cm^{-1} to 2277 cm^{-1} shows that the carbonyl group of polyurethane undergoes a spectral shift. This is due to the disturbance of the C–H leaves bonding to the carbonyl group forming the O–H bridges. The peak at 1598 cm^{-1} shifting to 1599 cm^{-1} also indicates the amine group in polyurethane disturbed by the O–H bonds of the teak leaves. Furthermore, the shift also occurs at the carbonyl peak of 1083 cm^{-1} to 1082 cm^{-1} due to the disturbance of the leaves' C–O bond of the peak of 1075 cm^{-1} (Figure 5). Similar results were confirmed by a previous work that [AQ16] PVA-sepiolite composite derived its van der Waals interactions between silanol groups and carbon C–O and shifted from 1088 cm^{-1} to 1085 cm

AQ16: we complete to " ... polyvinyl alcohol (PVA)-sepiolite ..."

Density test

Figure 6 shows the density of composite samples with polyurethane variation ranging from 825 kg m^{-3} to 1261

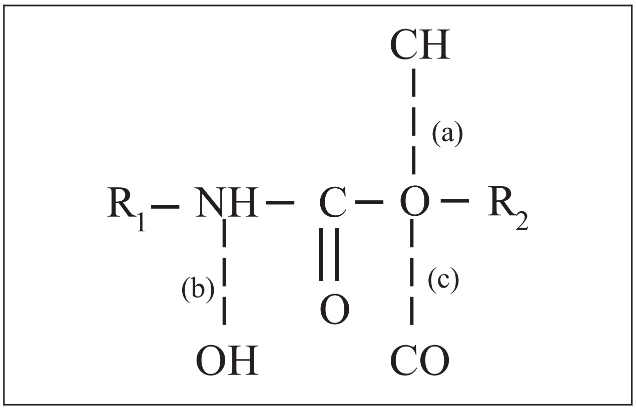


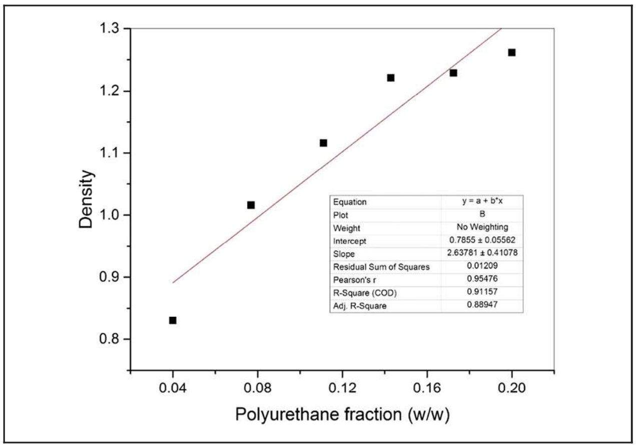
Figure 5. Van der Waals interactions on the teak leaf waste composites, dashed lines illustrate the van der Waals interaction area of (a) CH and carbonyl group, (b) OH and amine group, and (c) CO and carbonyl group.

kg m^{-3}. Considering the interpretation data showing a coefficient of determination, $R^2 = 0.91157$, there is a linear relationship between the polyurethane content and the composite density. Similar results were obtained by Afzaludin et al. on sugar palm/glass fiber-reinforced thermoplastic polyurethane.[36] The lowest density value is found in the sample with a polyurethane fraction (w/w) of 0.04, while the highest density value is in the sample with 0.20 of polyurethane fraction (w/w). It can be explained when the polyurethane polymer content rises and the filler is fixed, then the number of links produced by the polymer increases. Hence, the increased links result in stronger bonds to bind the filler which can increase the density.[37]

AQ17: OK

The density in samples with a composition[AQ17] of 0.08 (w/w), in accordance with teak wood that has been studied by Wahyudi et al., is in the range of 1000–1200 kg m^{-3} with an average value of 1160 kg m^{-3}.[38] In this study, the resulted density value is higher than the result of Martins et al. producing particleboard with a density of 510–570 kg m^{-3}.[28] The composites produced in this work have a similar density to the results of the Raju et al. producing some composites with densities ranging from 1100 kg m^{-3} to 1320 kg m^{-3}.[29] Further, Butylina et al. have produced wood fiber–polypropylene composite having a density 1 1040 kg m^{-3} to 1140 kg m^{-3}.[30] ty data shows that this composite belong to the particleboard category due to has a[AQ18] e than 0.80 g cm^{-3} in accordance with the signation 1554-67 by American Society.[39] Additionally, the composite has also met the Indonesia National Standard of 03-2105-2006 where particleboard density having values in the range of 400–900 kg m^{-3}.[40]

AQ18 = In us, this sentence is OK. The density of 0.8 g.cm-3 is in high-density particleboard category as American Society standard.



Equation	y = a + b*x
Plot	B
Weight	No Weighting
Intercept	0.7855 ± 0.05562
Slope	2.63781 ± 0.41078
Residual Sum of Squares	0.01209
Pearson's r	0.95476
R-Square (COD)	0.91157
Adj. R-Square	0.88947

Figure 6. Composite density as polyurethane fraction (w/w) function, and it was linearity relation as; $y = 2.6378x + 0.7855$, where x and y are polyurethane fraction and composite density, respectively, with $R^2 = 0.91157$.

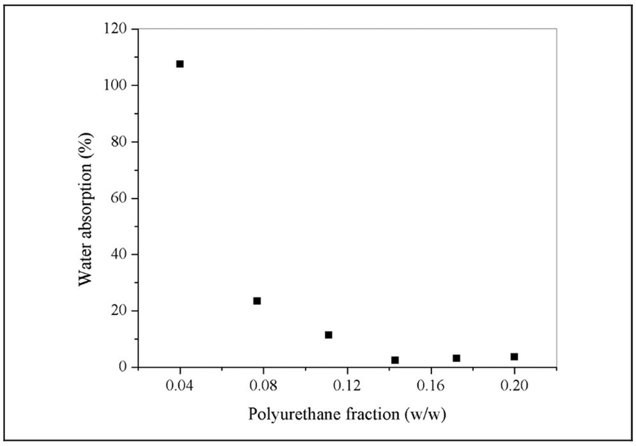


Figure 7. Water absorption of composite tends to zero for larger polyurethane fraction (w/w).

Water absorption

Water absorption values were measured to determine the ability of teak leaf composite to absorb water after 24 h of soaking. The highest water absorption rate reaches 107.35% in composites with a polyurethane fraction of 0.04 (w/w). After being immersed for 24 h, the composite undergoes thick development until the composite becomes cracked and broken because the cavity that appears between the matrix and the filler particles absorbs a lot of water and can further affect the stability of the dimensions of the composite. In addition, teak leaves as a hydrophilic filler also absorb a lot of water during immersion.

Figure 7 shows that the water[AQ19] absorption decreases with the increase of polyurethane fraction. At high fractions, it tends to produce composites with lower water absorption. The lowest water absorption was achieved in samples having the most optimal compressive strength at fraction 0.14 (w/w). The cavity formed is very small because the pressing process in the fabrication of composites and filler particles is distributed evenly on the matrix and absorbed a little water.

AQ19 = OK

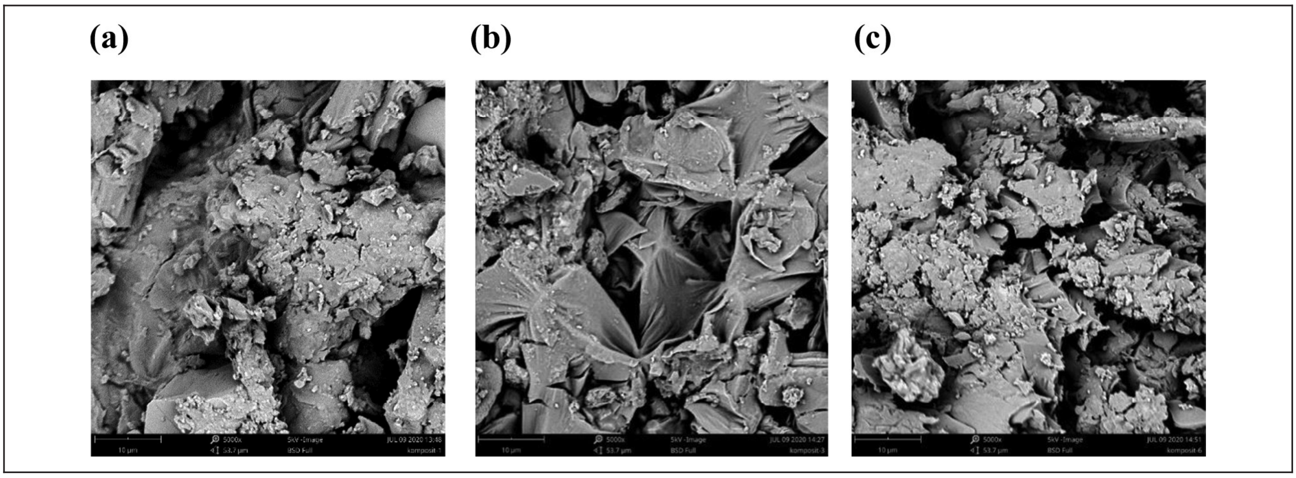


Figure 8. SEM of composite (a) 0.04 (w/w), (b) 0.12 (w/w), and (c) 0.20 (w/w). SEM: scanning electron microscopy.

The absorption value of the leaves teak waste composite produced is better than that of several leaves waste composites, which has been developed by several authors, such as Al-Sulaiman et al. produced palm fiber-reinforced composites with the water absorption ranging between 2.3% and 5.8%,[41] and Batiancela et al. developed a wood particle from tea leaves waste and obtained the water absorption about 23–40%.[42] Therefore, in accordance with the several properties above, this composite has properness in competing the woods.

Scanning electron microscopy

Morphological structure using SEM of composites 0.04, 0.12, and 0.20 (w/w) is shown in Figure 8. There is a cavity between the filler mixture and the matrix. Based on data analysis, the gap value between molecules in the composite 0.04 (w/w) is 13 µm. The low concentration of filler results in agglomeration in certain parts, and gaps between the mixture appear large enough which can act as cause of cracking and breakage when subjected to a load.[43] This was confirmed by the compressive strength and density results on the composite 0.04 (w/w) resulting in the lowest compressive strength and density. The resulting gap will be narrower when the concentration of polyurethane polymers is higher and the density increases.[44] The gap value in the composite 0.12 (w/w) is 4.65 µm and[AQ20] decreases when the polyurethane concentration increases and the gap distance 2.96 µm in the composite 0.20 (w/w). The morphological structure of the composite in accordance with the trends is obtained when measuring the density value of the sample. The hydroxyl group in cellulose plays an important role in the mechanism of forming bonds between molecules with polymer chains.[45] Interaction between cellulose molecules and polyurethane polymers can increase the density and decreases the gap between the filler and the polymer.[44]

AQ20 = OK

Conclusion

We have successfully developed a teak leaves waste composite utilized as particleboard. The mechanical and physical properties of the composites have been investigated. The test results showed that a composite compressive strength attains 38.5 MPa. The highest density is 1261 kg m^{-3} which is in agreement with the American Society standard. The lowest value of water absorption is 1.38%. In addition, we have successfully approximated the van der Waals interaction with the spectrum shifts of the FTIR spectroscopy as a confirmation of the compressive strength enhancement. Morphological structure shows that the gap between mixture decreases as the polyurethane fraction increases according to the density of the composite. However, the properties of organic composite are hygroscopic so they are vulnerable to being the target of fungal growth and susceptible to fire as an application in building fields. These problems will be the focus of further research by applying coating technology approaches.

Acknowledgment[AQ21]

AQ21 = OK

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Declaration of conflicting interests

The author(s) declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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AQ22: publication year : 2009.
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