

Research Article

Physical, Mechanical, and Thermal Properties and Characterization of Natural Fiber Composites Reinforced Poly(Lactic Acid): Miswak (*Salvadora Persica* L.) Fibers

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7000 years ago, miswak fiber (MF) was used as a toothbrush for oral care. However, since the emergence of plastic materials, it monopolized the oral care industry. The increment of plastic products also promotes accumulation of plastic wastes after its disposal. Thus, many researchers have turn to biodegradable products to reduce this problem. The aim of this study is to investigate the chemical, physical, and mechanical properties of MF as reinforcement in composites that are suitable to replace the toothbrush materials. The MF was reinforced in PLA composite with different weight percentage (0%, 10%, 20%, and 30%) and undergoes several types of testing. The chemical results show that there were high presence of cellulose in the fiber which could act as medium to transfer stress load equally from fiber to matrix. However, the results show low cellulosic contents in MF that affects the poor interfacial bonding between fiber and matrix. Physical properties shows a positive indication to be used as a toothbrush handle. As the fiber content increases, the density also increased. SEM micrographic illustrated the presence of voids as the cause for reduction in mechanical properties of composites. The mechanical results show the proposed material is comparable to the materials used in commercial applications. As for the thermal result, the TGA test melting point of the proposed composite material was comparable to the pure PLA, which means the proposed material can use similar processing temperature as PLA. DSC shows that Tg of PLA/MF composite is found to be similar to Tg in loss modulus of composites. DMA finding found that PLA/MF30 have the highest storage modulus 2062 MPa and the lowest tan δ 0.6 among PLA/MF composites. This concludes that there is a possibility of using these materials as an alternative in composites and increase the fiber strength by using pretreatments and/or compatibilizer.

1. Introduction

In the past decades, natural materials have been used to manufacture personal hygiene products. In the early 1900s, chemists had found that the substance known as “celluloid” could be shaped into practical shapes, which are suitable for toothbrush handles. In the early twentieth century, manufacturers started to make use of nylon and other plastics to

replace celluloid materials when plastic was invented [1]. Due to its 100% waterproof property, nylon had gain higher attraction and replaces celluloid in 1938 [2, 3]. Since then, plastic has dominated the hygiene industry and was commonly used to manufacture toothbrushes as shown in Figure 1. However, plastics are well known as a nonbiodegradable material, which promotes landfills after its disposal. Every single toothbrush that was manufactured decades ago



FIGURE 1: Conventional toothbrush.

could still be found around us as a piece of trash, somewhere on the planet [2, 3]. Furthermore, the American Dental Association stated that everyone should change their toothbrushes every three to four months. Hence, this had further increased the amount of wastes produced from the toothbrush disposal, where approximately 23 billion toothbrushes were annually discarded [3, 4]. Many toothbrushes were nonrecyclable due to the materials used during production. Biodegradable or biobased plastics were not always better for the environment than traditional plastics, either because they do not break down as well or because they have their own complicated environmental footprints [3, 5]. However, any alternative that decreases the overall amount of material and packaging used is a positive move. An alternative action is to incorporate natural fiber into polymer matrix to produce composite material for toothbrush handles.

The development of biodegradable materials has attracted many interests of researchers globally. Recently, a new toothbrush that uses biodegradable bamboo toothbrushes with PLA-based compostable bristles has been produced and available in the market [6]. This had arisen the need of toothbrush waste reduces in our environment. The biodegradable and renewable aliphatic polyesters are one of the most promising biodegradable plastic materials. Polylactic acid (PLA) polymer is one of the most intensively developed and commercially available due to its biodegradability, renewability, and high strength to weight ratio. There were many studies that had extensively used PLA as the polymer matrix for composite material for numerous applications and purposes [7]. In addition, PLA were also used for food packaging, tissue engineering, household engineering, and drug delivery application [8], and it was commonly used for biomedical applications due to its biocompatibility, biodegradability, and nontoxic features [9].

In the midst of competitive materials and processing cost of biopolymer, PLA polymer exhibits attractive physical and mechanical properties such as good clarity, barrier proper-

ties, superior modulus, and strength performances [10]. However, the cost of the raw material of PLA is relatively higher compared to conventional synthetic materials, thus demoted the willingness of industrialists to use it. PLA also exhibits drawbacks such as sensitive to heat distortion temperature and moisture, as well as low resistance to hydrolysis and flexibility, which makes it harder to use PLA as the composite's matrix [11].

Fiber reinforcements are also one of the factors that could increase the composite's mechanical strength. Several experimental studies had combined PLA with a variety of natural fibers such as flax, kenaf, hemp, and bamboo, in order to create biodegradable composites [12–17]. From these studies, it was found that natural fiber reinforcement in polymer composites was able to reduce the production cost while still retaining its biodegradability [18]. There was one study made by Bajpai et al., which had developed new tribological material and better wear behavior of the composites by incorporating 20 wt. % of sisal, nettle, and *Grewia optiva* fibers into the polymer composite [19]. Oksman et al. [20] study had shown that flax fiber-reinforced PLA composite has 50% higher tensile strength as compared with flax/polypropylene composite [20]. Meanwhile, in Nishino's study, it was found that kenaf fiber reinforcement of up to 70 wt. % resulted in high strength of biodegradable composite [21]. All of these studies show that natural fiber reinforcements in composites could potentially increase the mechanical performance of the polymer composites.

In this technologically advanced era, the conventional toothbrush was deemed to be more expensive compared to the cost of miswak, especially in regions where miswak tree was grown locally. Figure 2 shows the processing of the miswak tree to fibers which could be used as a reinforcement material. 7000 years ago, Babylonians (the Greeks and Romans) used chewing sticks as a toothbrush as shown in Figure 3, which then gradually changes to the current plastic toothbrush [22].

Miswak is becoming more popular in Muslim countries, particularly in various African and Arab nations. It was also known as the natural toothbrush. The World Health Organization has also suggested of using miswak for oral hygiene due to its antibacterial effect [23]. There are more than 180 plant species that can be used as a natural toothbrush. These species differ from each other on the basis of appearance, scent, texture, and taste. The plant sources vary around the world, with neem (*Azadirachta indica*) being the most widely used in India; lime tree (*Citrus aurantifolia*) and orange tree citrus (*Citrus sinensis*) being used in West Africa; Senna (*Cassia vennea*) being used in other parts of Africa; and Arak (*Salvadora persica*) being used in the Middle East [24]. Furthermore, neem is a native plant of India, and Arak (*S. persica*) is widely used in Saudi Arabia; miswak could be found in Indonesia, Malaysia, Australia, Sri Lanka, Burma, Pakistan, and Africa [25]. Miswak is popular for a variety of reasons, including its inexpensive cost of production and easy access to the source [26] and most importantly its antibacterial effects. *Salvadora persica* or miswak fiber (MF) could be used as an alternative fiber reinforcement for polymer composites. The miswak has been used for

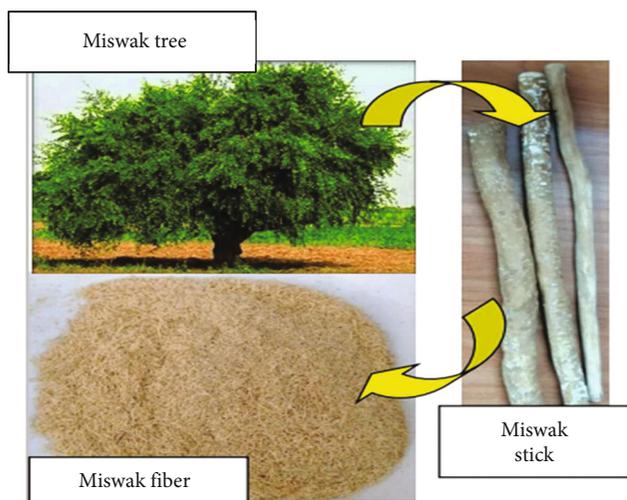


FIGURE 2: Miswak fiber obtained from miswak stick that originated from miswak tree.



FIGURE 3: Frayed miswak stick.

centuries, and there is an Islamic etiquette in using miswak as stated in past studies [27]. Cleansing effects and oral health protection provided by the miswak fibers during mechanical cleaning and releases of beneficial chemicals from fibers, respectively, have proven its high value as a composite fiber reinforcement with antibacterial properties [28]. Miswak fibers have been reported having more than one type of antimicrobial agents that inhibit positive and negative bacteria that help enhance in plaque removal [29]. Despite the fact that there have been several articles on PLA and natural fiber composites, there was no study which discusses on the effect of miswak (*Salvadora persica*) fiber as a reinforcement fiber in PLA matrix. Natural fiber not only reduces the cost of the material, but they also encourage disintegration and biodegradation, with the added benefit of antibacterial characteristics. This advantage is potentially useful for a wide range of application, especially in dental care. Therefore, the aim of this study was to characterize the mechanical, physical, and thermal properties of the miswak fiber-reinforced polymer composites.

2. Experimental Details

2.1. Materials. Polylactic acid (PLA) (grade 2003D) produced by Nature Works, USA, was used and purchased at Polycomposite Sdn Bhd. The density, melting temperature, and glass transition temperature of PLA were 1.24 g/cm^3 , 160°C , and $55\text{-}60^\circ\text{C}$, respectively. Miswak stick was bought from Al-Khair Natural Products, Malaysia.

2.2. Biocomposite Preparation. Miswak fiber (MF) was prepared by chopping the stick into smaller pieces and then crushed the pieces into short fibers. The short fibers were then grind into powder and then dried in an oven to remove moisture. The dried MF was then sieved to get fibers below mesh 30. PLA pallets and MF were mixed by using Brabender Internal Mixer to obtain the composite blends. The temperature and rotor speed were fixed at 160°C and 50 rpm, respectively. The composite was prepared with varying weight composition ranging from 0 to 30 wt. % of MF content as shown in Table 1.

The composite blends were preheated for 4 minutes at 160°C before going to full-press. It was then hot pressed for four minutes at 160°C using a Vecho Vation 40 tonne compression molding. The $30 \text{ cm} \times 30 \text{ cm}$ composite plate was then placed in a cold press between two steel plates, where it was cooled for 4 minutes at 25°C . Figure 4 depicts the compression molding machine. Finally, the composite plate was cut into smaller samples suitable for testing based on the ASTM Standard.

2.3. Materials Characterization Process

2.3.1. Board Density. The composite samples were weighed in air using digital weighing scale and in water using the densimeter, MD-200S Mirage. The difference in weight of the samples in two different mediums will give the weight of water displaced by the samples, and this weight in gm was converted to volume in cm^3 . Ten samples were cut into square shape with the dimension of $(10 \times 10 \times 3) \text{ mm}$. The solid square piece was then polished with 1200 grade sandpaper in order to make smooth and shiny surface. The density test was performed according to ASTM D4018 by using a weighing scale, and the dimensions were measured using Mitutoyo Digital Vernier calipers. The volume was calculated from the measured dimension. The density was then recorded for all samples and presented in Table 2 under Section 3.1 Density of composite.

The density was calculated using the equation

$$\text{Density (g/cm}^3\text{)} = \frac{\text{Mass}}{\text{Volume}}. \quad (1)$$

2.3.2. Chemical Analysis Test. Preliminary chemical analysis was performed by means of solvent extraction as well as weighing procedure after 2 hours of heating at 105°C for water content. The test used TAPPI standard procedure to determine the extractive (T204), cellulose (T203), holocellulose [30], and lignin (T222) content of the fiber composition.

TABLE 1: Formulation of PLA/MF composites.

Sample code	Matrix PLA (wt. %)	Reinforcement MF (wt. %)
PLA	100	0
PLA/MF10	90	10
PLA/MF20	80	20
PLA/MF30	70	30



FIGURE 4: Compression molding.

TABLE 2: Properties of PLA/MF composites density.

Specimens	Density (g/cm ³)	Density increment, %
PLA	1.240	—
PLA/MF10	1.255	1.21%
PLA/MF20	1.283	2.23%
PLA/MF30	1.308	1.95%

2.3.3. Water Absorption. Ten composite samples with the dimensions of (10 × 10 × 3) mm were prepared according to ASTM D570 for each different composites composition. All samples were oven dried at 60°C for 24 hours. After oven drying, the samples were cooled in desiccators over granulated silica gel before water absorption test was conducted. The test was conducted by submerging the specimens in distilled water for 72 hours and measured the increase in weight in every one hour as compared to the original oven dry weight of the specimen. Two specimens of each type formulations were tested in an adjusted room temperature of 27°C,

and the average result was recorded. Water absorption was calculated as

$$\text{Water absorption (\%)} = \frac{(W_1 - W_0)}{W_0} \times 100, \quad (2)$$

where W_0 is the weight of specimens before immersion and W_1 is the weight of specimens after 72 hours of immersion in distilled water.

2.3.4. Thickness Swelling. Ten specimens of (10 × 10 × 3) mm³ samples of each composite's formulations were prepared according to ASTM D570 for the testing of thickness swelling. The samples were measured before as T_0 and T_1 as after by using Digital Vernier calipers (Mitutoyo) with 0.01 cm accuracy. The thickness swelling was calculated according to the Equation (3):

$$\text{Thickness swelling (\%)} = \frac{(T_1 - T_0)}{T_0} \times 100, \quad (3)$$

where T_0 is the thickness of specimens before immersion and T_1 is the thickness of specimens after 72 hours of immersion in distilled water.

2.3.5. Scanning Electron Microscope (SEM). Scanning electron microscope (Hitachi S-3400 N) equipped with energy dispersive X-ray under an accelerating voltage of 15 kV at an emission current of 58 μA was used to observe the fracture surface after the tensile test. The fracture ends of the samples were mounted on an aluminum stub and coated with a thin layer of gold to avoid electrostatic charging during examination.

2.3.6. Tensile Properties. The tensile testing of the composite was conducted using 5kN Bluehill INSTRON Universal Testing Machine. The test was carried out according to ASTM D638-14. The samples were cut into dog bone shape by a plastic molder machine with the specifications of length of 165 mm, width of 13 mm, and thickness of 3 mm, respectively. The crosshead speed was set at 2.0 mm/min, and the composites were gripped at a 30-mm gauge length. Five samples were kept in a conditioning room, and the test was run at 20.5°C with the relative humidity (RH) at 48%.

2.3.7. Flexural Properties. Flexural test of the composite was performed using 5kN Bluehill INSTRON Universal Testing Machine. The test was carried out according to ASTM D790-17 three-point bending test. The samples were cut with the specifications of length of 127 mm, width of 12.7 mm, and thickness of 3 mm, respectively. The crosshead speed was set at 2 mm/min with a support span-to-depth ratio of 16:1. Five samples were kept in a conditioning room, and the test was run at 20.5°C with the relative humidity (RH) at 48%.

2.3.8. Fourier Transform Infrared Spectrometry (FTIR). Using an FTIR (Nicolet iS10 Thermo Scientific) analyzer, the chemical functional group contained in the MF was discovered. The infrared (IR) spectra were analyzed using

OMNIC software and the attenuated total reflectance (ATR) technique. The sample was prepared as a powder and scanned 32 times with a resolution of 2 cm^{-1} over a wavenumber range of 500 cm^{-1} to 4000 cm^{-1} .

2.3.9. Thermogravimetric Analysis (TGA). The samples were analyzed (TGA) on a Mettler Toledo TGA/DSC 1HT Stare System (Switzerland) from 30 to 600°C at a heating rate of $10^\circ\text{C}/\text{min}$ and nitrogen gas as its medium. TGA is a method for determining a material's thermal stability and volatile component fraction by measuring the weight change as the sample is heated at a constant rate.

2.3.10. Differential Scanning Calorimetry (DSC). The heat capacity of the composites was measured using differential scanning calorimetry. The specimens were examined to differential scanning calorimetry on a Mettler Toledo TGA/DSC 1HT Stare System (Switzerland) from 30 to 200°C at a heating rate of $10^\circ\text{C}/\text{min}$ in nitrogen gas.

2.3.11. Dynamic Mechanical Analysis (DMA). TA Instrument Q800 was used to perform dynamic mechanical analysis (DMA) in accordance with ASTM D4065. In a nitrogen environment, samples of $60\text{ mm} \times 12\text{ mm}$ for each composite structure were placed in 3-point bending mode at a frequency of 1 Hz and an amplitude of $15\text{ }\mu\text{m}$ at $25\text{--}105^\circ\text{C}$ with a continuous heating rate of $5^\circ\text{C}/\text{min}$.

3. Results and Discussion

3.1. Density of PLA/MF Composites. Table 2 shows that density of PLA/MF composites, and it found density increment with increases of MF contents; this is found aligned with previous study [31]. It was found that the MF30 sample has the highest density value. This was expected since there was a higher portion of relatively high-density MF that replaced the PLA matrix [32]. However, nonlinear response on density increment was found in this study. This can be foreseen by high porosities in the composites due to poor interfacial bonding between fiber and matrix. The voids, as a result from ineffective fiber wetting, make the composite have a lower density than it should be. Hence, there will be a high reduction of strength properties shown on the MF30 specimen.

3.2. Analysis of Chemical Composition of Miswak Fiber. Table 3 shows the chemical composition of various natural fiber from previous studies, as well as the results of a preliminary chemical study of the MF. It was reported that agricultural lignocellulosic biomass typically contains between 10% and 25% of lignin, 20% to 30% of hemicellulose, and 40% to 50% of cellulose [33]. Carbohydrates (cellulose and hemicellulose) constitute the majority of the chemical components of wood, with cellulose accounting for 40% to 50% of the dry weight and hemicelluloses for 25% to 35%. Additionally, hardwood lignin make up between 10% and 20% of the overall lignin composition [34].

According to the chemical analysis, this miswak fiber comprises 5.09% lignin, 28.12% hemicellulose, and 21.81% cellulose. The test also indicated extractives as a component.

Other than cellulose, hemicellulose, and lignin, the extraneous components in wood include extractives and ash. Extractives are the extraneous material that is soluble in neutral solvents and amounts from 4 to 10% of the dry weight of typical wood in temperate climate species. However, extractives may make up as much as 20% of the dry weight of wood from tropical species [34]. Miswak fiber comprises 20.1% of the extractives composition, according to the results of lignocellulosic biomass composition testing using the TAPPI T204 standard. It suggests that miswak fiber can be classified as tropical series wood. Miswak fiber, or *S. persica* as it is known scientifically, is a well-established plant with high drought resilience that grows in dry to arid regions of the tropics and subtropics [35].

Natural fiber's main composition has a significant impact on how well composites perform. To examine and ascertain the impact of miswak fiber on polylactic acid polymer, however, substantial experiments are required. To better comprehend the impact of fiber composition on the mechanical and thermal characteristics of composite, tests like X-diffraction to detect the crystalline structure may be helpful. To date, it has been shown that miswak fiber has a lower cellulose concentration than other natural fibers; however, Pettersen [34] noted that cellulose from wood is difficult to separate in pure form because it is closely linked to lignin and hemicellulose. However, according to TAPPI T203 or ASTM Standard D 1103, the majority of alpha cellulose is produced after the holocellulose is treated with 17.5 percent NaOH [34, 36]. On the other hand, hemicellulose can be found mostly in the primary wall of the fiber, and lignin is an amorphous in nature incorporated into cellulose structure to enhance the strength of the wood [37]. MF has the highest hemicellulose contents as compared to other natural fiber and the hemicellulose binding microfibrils in the fiber, which provides structural reinforcement [38]. Lignin acts as glue between individual cells in the cell wall, and it provides flexibility, properties, or structure of the fiber [38].

The main component that gives fibers their stiffness, strength, and favorable thermal qualities is cellulose. However, a number of factors, including fiber particle size, fiber orientation, and fiber processing conditions, may have an impact on composite characteristics. To learn more about the qualities of composite materials, greater research on the impact of fiber sizes, orientation, and processing conditions on those attributes is required. Table 3 shows the comparison of chemical composition of miswak with other type of fibers.

3.3. Water Absorption and Thickness Swelling of PLA/MF Composites. Figure 5 shows the level of water absorption of PLA/MF composites. The aim of this study was to investigate the water absorption characteristic of the PLA/MF composites. It is significant to understand the behaviors of the composites in moist surrounding. This is also because natural fibers are sensitive to water; thus, this characteristic will definitely affect the strength of the composite when it is highly exposed to water. Other factors that could affect the composite physical properties were voids/pores, volume fraction of fibers, humidity, viscosity of matrix, and temperature [43].

TABLE 3: Constituents of miswak fiber and other natural fibers.

Fiber	Cellulose (wt. %)	Hemicellulose (wt. %)	Lignin (wt. %)	Reference
Miswak	21.81	28.12	5.09	Present study
Kenaf	70	19	3	[39]
Sugar palm	37.3	4.71	17.93	[40]
EFB	45	21	23	[41]
Hemp	70.2-74.4	17.9-22.4	3.7-5.7	[37]
Flax	71	18.6-20.6	2.2	[37]
Jute	61-71.5	13.6-20.6	12-13	[37]
Sisal	45	14.2	20.5	[42]

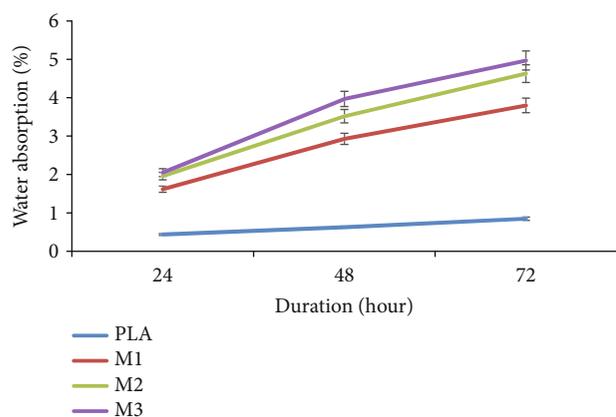


FIGURE 5: Water absorption level of PLA/MF composites.

In Figure 5, the level of water absorbs by sample for PLA, PLA/MF10, and PLA/MF20/PLA/MF30 are 0.93%, 1.34%, 1.36%, and 1.42%. Neat PLA was expected to record the lowest water absorption due to its hydrophobic nature. This was expected since PLA/MF30 had the highest cellulose content in the miswak fiber, which increases the penetration of water by capillary mechanism [44]. The increase in water absorption for hydrophilic fiber in composite samples was predicted since the PLA/MF molten flow fills the mold before it crystallizes during composite processing. When this happened, some air was retained, resulting in water penetration into the voids of the samples [45].

In Figure 6, the thickness swelling for PLA, PLA/MF10, PLA/MF20, and PLA/MF30 are 0.08%, 0.75%, 0.76%, and 0.80%. The slight increasing thickness of the neat PLA samples proves that water diffuses into the samples. PLA/MF30 composite had the highest thickness swelling due to its high fiber content. The increase in swelling of the specimens was resulted due to the presence of voids and the hydrophilic nature of the fibers that attracts water. Similar findings was reported by [46], where his study found that there was an increase in the level of swelling of the PP/miswak powder samples as the fiber content increases.

3.4. Scanning Electron Microscope of PLA/MF Composites. Figures 7(a)–7(c) show the SEM micrographics for different MF ratio (10–30 wt. %) reinforcement in PLA composite. In the 10 wt. % of MF reinforcement composite (Figure 7(a)),

ineffective mixing of fibers and matrix causes the formulation of voids. Multiple variables, including manufacturing and processing conditions, fiber volume percentage, and fiber length, can lead to void development in polymer composites. Further elements that may impact void content include matrix material, moisture content, and processing conditions [43, 46, 47]. The appearance of MF has disturbed the homogeneous flow of molten resin, creating meso- and microvoids. Nonhomogeneous air permeability of natural MF entrapped more air and reduced matrix bonding strength. The voids act as a stress concentration spot and lead to earlier failure during loading.

On the other hand, presences of void between fiber and matrix (Figure 7(b)) show ineffective interfacial bonding between hydrophilic matrix and hydrophobic natural fiber. Besides, poor fiber wetting due to poor matrix adhesion on impurities-riches fiber's surface shall be visible. This resulted a full detachment of matrix from fibers during applied tensile forces.

In Figure 7(c), there was a presence of fiber fracture and fiber pull-out. The micrographs showed that the composite breaks due to fiber-to-fiber interaction in the composite matrix. This observation was similar to the previous study [48]. From this analysis, PLA/MF composites were expected to have low mechanical properties due to conditions such as improper fiber handling, poor fiber-matrix interaction, and matrix detachment.

3.5. Tensile Properties of PLA/MF Composites. Table 4 and Figure 8 shows the tensile properties of PLA/MF samples. PLA with MF10 showed the highest tensile strength and Young's modulus values of 33.7 MPa and 2.9 GPa, respectively. The lowest tensile strength and Young's modulus values were observed from PLA with 30 wt. % fiber content as shown in Table 4.

Tensile strength was found to have dropped by 35% as a result of the addition of 10% MF to polymer. The absence of adequate polymer reinforcement provided by the MF, as seen in Figure 7(a), is what causes the composite's decreased tensile strength. The parameters used in composite fabrication led to the occurrence of voids. When fibers are incorporated into a composite material, air or volatile substances may get trapped in the material. As a result, the fiber spacing between resin-rich regions can cause microvoids to form in the composite along the individual fibers, which has a

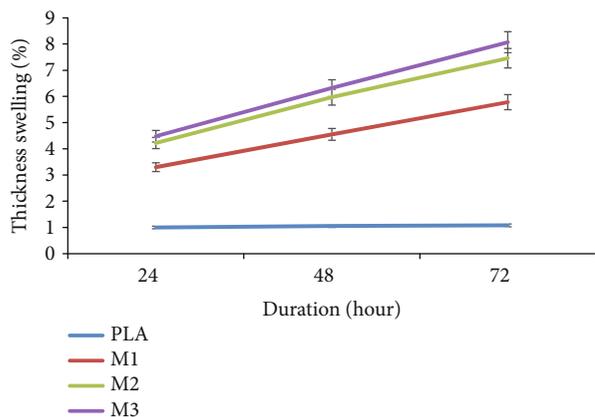


FIGURE 6: Thickness swelling level of PLA/MF composites.

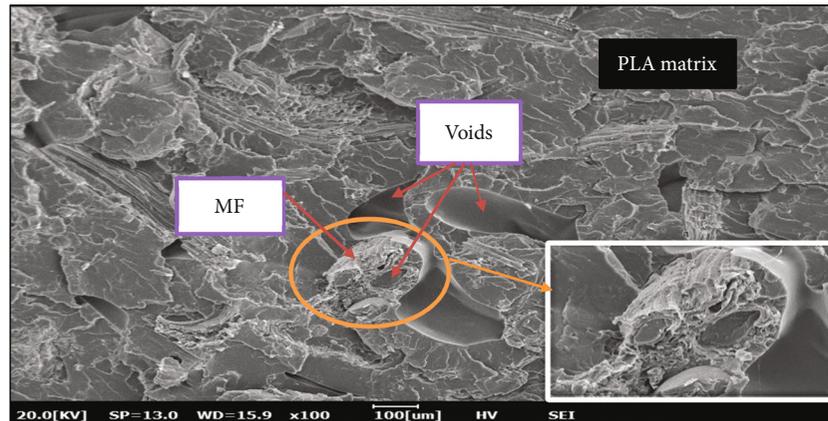
negative impact on the tensile properties of the composites. The adherence of the fibers to the matrix is another element that lowers the tensile strength of PLA/MF composite.

When fibers were added to a matrix, it was anticipated that the resulting fiber-reinforced composite will be stiffer than a pure polymer; thus, it could carry the load applied to the composite. However, as shown by SEM in Figures 7(b) and 7(c), the condition of poor adhesion between the fiber and the matrix had led to a reduction in tensile characteristics. According to study made by Mazur et al., PLA composites made with natural particles (Wood, Bamboo, and Cork), inadequate fiber/matrix adhesion, and empty spaces in the material had a detrimental impact on the material's mechanical characteristics [49]. Pongtanayut et al. discovered that the insertion of soft phase rubber reduced the tensile strength of PLA matrix, suppressing PLA crystallinity and resulting in decreased tensile strength [50]. However, by adding 10 wt. % MF to polymers results in increased Young's modulus values. The MF reinforcements, regardless of the amount of fiber content, have shown better strength but small increment (<5%) on the tensile modulus. All of the blends had similar Young's modulus values to the pure PLA as the modulus improvement is only ranging from 1.05% to 3.19%. It is widely known that adding any mineral or filler to a composite will improve its stiffness or modulus.

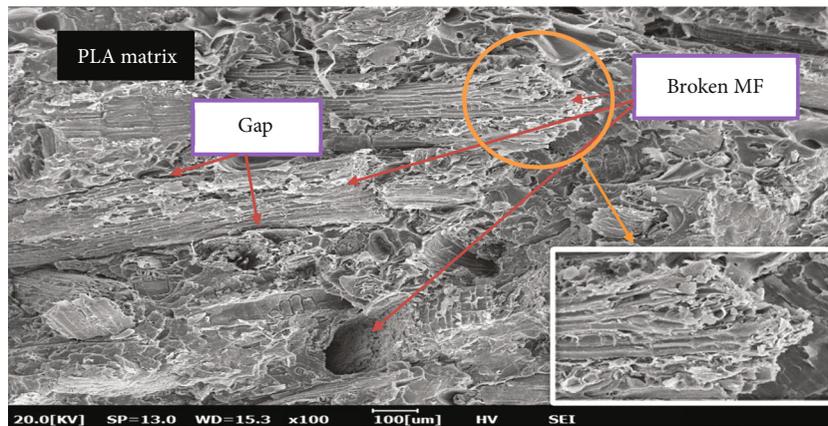
Studies on the enhancement in Young's modulus brought on by adding wood flour (WF) materials up to 7 wt. % were discovered by Zhang et al. in [51] with 2wt. % WF/resin recording the highest Young's modulus at 323.8MPa [51]. Similar increment in tensile modulus of composite was found by Sherwani et al. [52] in which untreated sugar palm fiber/glass fiber-reinforced PLA hybrid composite tensile modulus increased with the addition of GF [52]. Table 4 displays the elongation at break as a function of MF content. The abrupt decrease in elongation upon break is a result of miswak fiber's stiffening action. Similar behavior is consistent with studies conducted by Morreale et al. [53] that discovered the addition of WF into biodegradable polyester will decrease the elongation at break of the samples [53].

In this research, the Brabender internal mixer screw speed was maintained at 50 rpm to blend PLA pellets with MF. According to [54], increasing the microscrew compounder's speed had a negative influence on the tensile and impact parameters of all composite materials. When PEG was added to virgin PLA at a screw speed of 200 rpm, it was discovered that the yield strength and modulus value decreased. Sample PLA/PEG/POSS composite compounded at a screw speed of 100 rpm, however, showed higher tensile and impact results. As it was discovered, PLA/kenaf bast and PLA/kenaf core exhibit improved results on flexural strength of 60 rpm rotation speed, and impact strength higher value was measured with PLA/kenaf bast and PLA/kenaf core at 70 rpm rotation speed. Jaafar et al. [55] investigated the varying speeds of extrusion (60, 70, and 80 rpm) and showed various behavior of fiber dispersion. The effect of screw speeds of 40, 80, and 140 rpm extrude on the mechanical characteristics of PLA/HNT composite has been examined by Venkatesh et al. [56]. The PLA/HNT 5 wt. % nanocomposite compounded at 140 rpm showed improved HNT dispersion in the PLA matrix, leading to higher mechanical characteristics. Whereas processing speed does not significantly affect the tensile characteristics of virgin PLA, Galvez et al. [57] have found that adding ATBC 10 wt. % causes a reduction in tensile values of 36% for 60 rpm speed and 50 percent for 150 rpm speed. Tensile modulus was reduced by 36% and 44% at 60 and 70 rpm, respectively. Nevertheless, it should be highlighted that the bra-bender's screw speed had a detrimental effect on the mechanical characteristics of the PLA/MF composite.

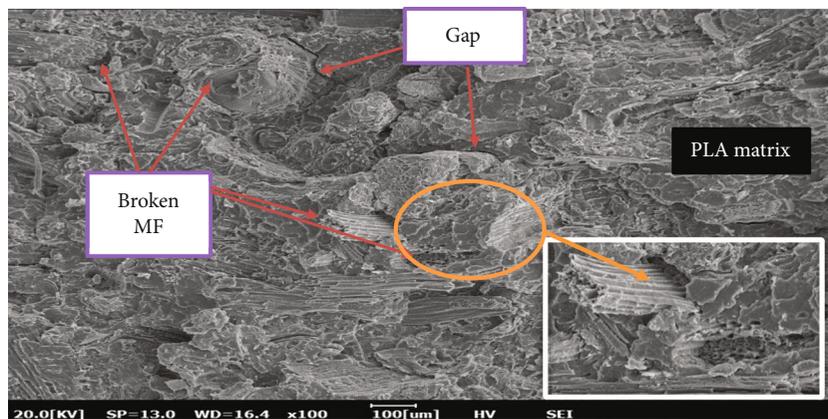
The technique used to manufacture composites has an influence on the performance of natural fiber-reinforced polymer composites. The settings used during the compression molding process to create short natural fiber composites have a big impact on the composite's mechanical characteristics. When creating composites via compression molding, studied the compounding process between polymer and short natural fiber. According to research, compression molding holds materials for an average of 4 to 15 minutes. To produce superior mechanical characteristics of natural fiber composites, it is vital to choose the right molding pressure, temperature, and holding time. The impact of manufacturing factors on the strength of biodegradable composite was researched by Rubio-Lopez et al. [58]. Studies have shown that a large reduction in production time may result in a decrease in the manufacturing cost of composite materials. Compression molding preheating time of 2 minutes and heating under pressure for 3 minutes produced tensile strengths more than 100 MPa. The mechanical characteristics of kenaf fiber polymer composite were studied by Bernard et al. [59] to see how temperature and speed of compression molding affected such qualities. Tensile strength increased with the addition of 10 weight percent unidirectional kenaf fiber, and impact characteristics were shown to worsen with compression at 230°C and 10 minutes of holding. In this PLA/MF studies, the compression molding parameter was discovered after 4 min of preheating, complete pressing, and cooling at 160°C. The parameter was discovered by analyzing the state of the extracted



(a) PLA/MF10



(b) PLA/MF20



(c) PLA/MF30

FIGURE 7: SEM micrograph of PLA/MF tensile fractured surfaces of different loading (a) PLA/MF10 wt. %, (b) PLA/MF20 wt. %, and (c) PLA/MF30 wt. % with 100 magnifications.

sample, which had a well melted flat surface and no evidence of trapped air.

3.6. Flexural Properties of PLA/MF Composites. An INSTRON universal testing machine was used to determine the flexural strengths of PLA/MF, and the results are shown in Table 5. The link between the flexural characteristics of the polymers and the miswak fiber was shown in Figure 9. For neat PLA, the maximum flexural strength and modulus

were displayed as 95.6 MPa and 3.4 GPa, respectively. The maximum flexural strength and modulus with 10 wt. % MF were 66.1 MPa and 3.2 GPa, respectively. It was discovered that PLA with MF offered noticeably weaker flexural characteristics. Pure PLA was stiffer and more brittle than PLA/MF composite. The flexural characteristics of the composites were influenced by the polymers' inherent strengths [60]. According to Doh et al. [60], the inclusion of 10% liquefied wood in liquefied wood polymer composites (LWPC)

TABLE 4: Tensile properties of the composite's samples.

Sample	Strength (MPa)	Tensile	
		Young modulus (GPa)	Elongation at break (%)
PLA	52.1	2.85	2.64
PLA/MF10	33.7	2.94	1.69
PLA/MF20	28.4	2.88	1.74
PLA/MF30	24.1	2.89	1.49

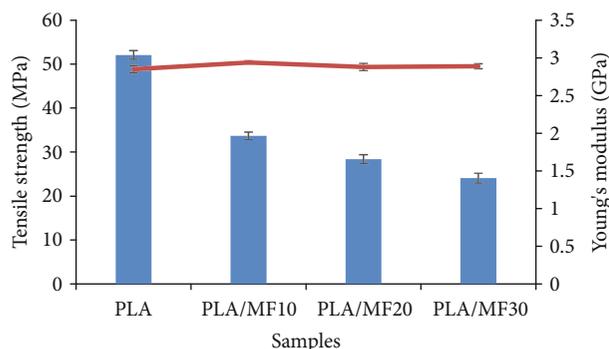


FIGURE 8: Tensile strength and modulus of PLA/MF composites.

TABLE 5: Flexural properties of composite samples.

Sample	Strength (MPa)	Flexural
		Young's modulus (GPa)
PLA	95.6	3.4
PLA/MF10	66.1	3.2
PLA/MF20	57.1	2.8
PLA/MF30	49.5	2.9

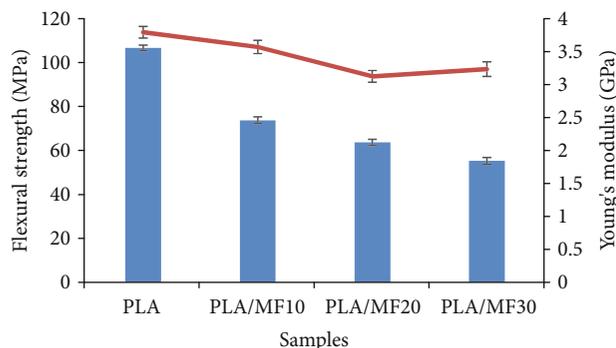


FIGURE 9: flexural strength and the flexural modulus of PLA/MF composites.

increased the flexural MOE and MOR of polymer with higher melt index. This is because the high flow value of the polymer helps in blending the polymer and the liquid wood powder during the compounding process at a specific temperature, increasing the flexural characteristics [60].

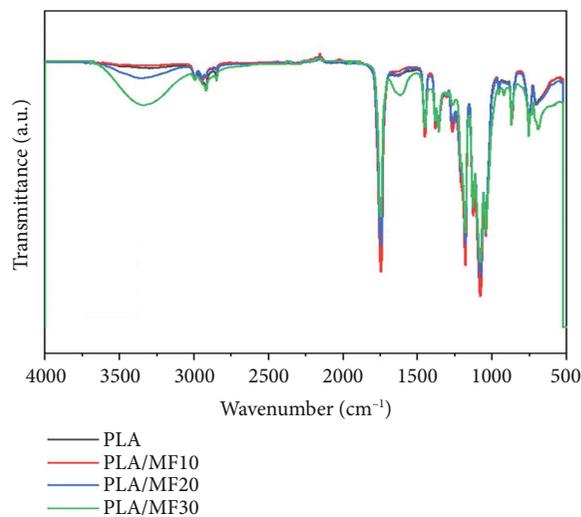


FIGURE 10: IR spectra of PLA/MF composites.

Fiber orientation is another aspect that impacts the flexural characteristics. As the fiber orientation becomes more parallel to the direction of the load applied, the greater the flexural qualities since the phenomena happening during the flexural test depend on the location, with the top layer being subjected to compression and the lower layer to tension [49]. However, Golmakani et al. [61] discovered the opposite conclusion in their investigation into the impact of wood powder and nanoclay particle content on polyethylene matrix. They discovered that when wood floor was added in amounts of 40 wt. % and 50 wt. % in composites without nanoclay, the flexural strength values were 40.78 MPa and 45.87 MPa, respectively, which may be regarded as having strong flexural qualities [61].

Flexural strength and modulus were decreased for composite materials when fiber content was increased by 10%, 20%, and 30%. When compared to plain PLA, PLA/MF10, PLA/MF20, and PLA/MF30, all decreased by roughly 30.8%, 40.2%, and 48.2% as compared to neat PLA, respectively. As shown in (Figures 7(a)–7(c)), the increase in porosity of the composites brought on by insufficient interfacial bonding is what leads to the drop in characteristics. The outcomes match those of the PLA composite with natural fibers work as determined by SEM [49]. Yusoff et al. [62] found that while the flexural strength of kenaf-coir/PLA was lower than kenaf-bamboo-coir/PLA and bamboo-coir/PLA, it had the highest flexural modulus, which was 70% higher than the other combinations, as the impurities in bamboo fiber caused fiber-matrix interfacial failure and decreased the stiffness of the composites [62].

3.7. Fourier Transform Infrared Spectrometry (FTIR) of PLA/MF Composites. Figure 10 shows the IR spectra of PLA polymer and PLA/MF. The sample's primary absorbance was found in low-wave number areas, particularly 500 cm^{-1} to 1800 cm^{-1} . In a previous study, similar findings were reported [63]. The spectra of three different types of fiber loading in matrix were almost identical, as shown in Figure 9. The information about the functional groups in

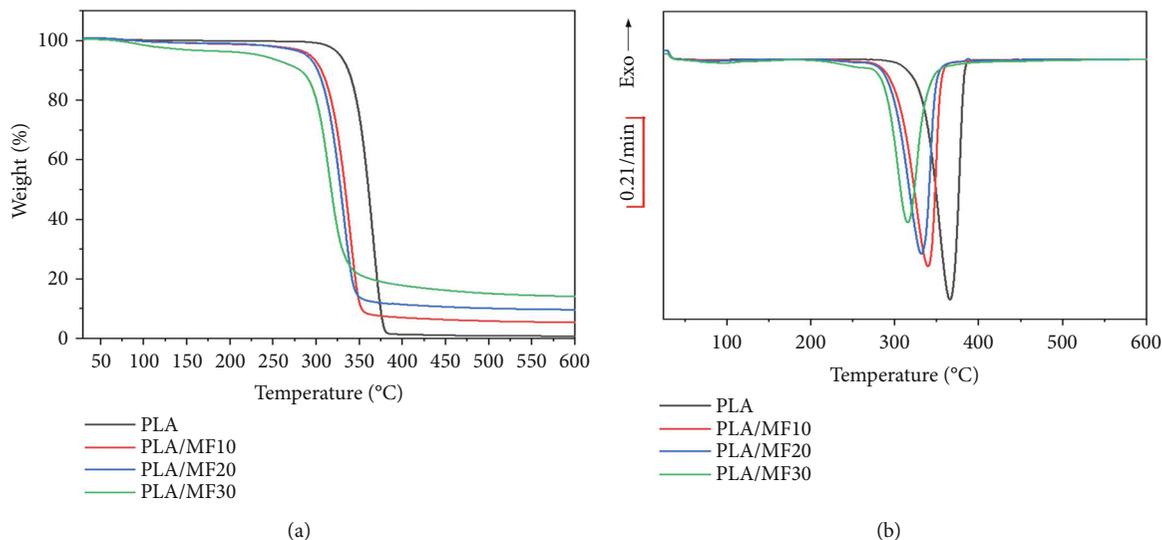


FIGURE 11: (a) TGA curve of PLA/MF composite. (b) DTG curve of PLA/MF composite.

TABLE 6: Thermal degradation temperature of PLA/MF composites.

Sample	Initial degradation, (°C)	Final degradation, (°C)	Residue (wt. %)
PLA	336.38	367.32	0.86
PLA/MF10	292.42	341.64	5.52
PLA/MF20	265.77	333.36	9.78
PLA/MF30	266.98	317.27	14.23

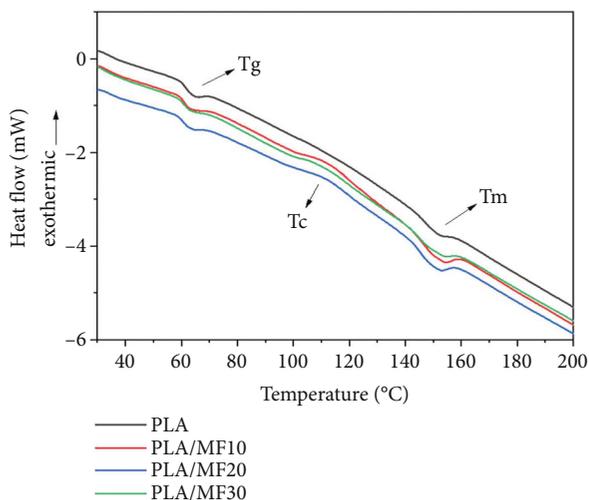


FIGURE 12: DSC curve of PLA/MF composite.

the sample is revealed by FTIR spectra. The PLA characteristic peaks, on the other hand, were found ranging in between 2800 cm^{-1} and 3000 cm^{-1} , as shown in Figure 9. The peaks at 2994 cm^{-1} correspond to O-H stretching to the carboxylic acid and 2944 cm^{-1} to the alkane's C-H stretching. The spectrum of the PLA/MF composite resem-

bled that of Wisam et al. [64], with primary peaks almost identical in wavenumbers (2995 cm^{-1} , 2944 cm^{-1} , 1749 cm^{-1} , and 1184 cm^{-1}) [64].

The peak at 1746 cm^{-1} corresponds to the C=O stretching of lignin esters or lactone groups [65, 66], the peak at 1451 cm^{-1} corresponds to the C-H bending of cellulose and hemicellulose alkane or methyl groups, and the peak at 1180 cm^{-1} corresponds to the C-O stretching of aliphatic ether groups of hemicelluloses. The peak at 1042 cm^{-1} corresponds to the sulfoxide group's S=O stretching, while the peak at 755 cm^{-1} corresponds to the 1, 2-disubstituted group's C-H bending. The interaction between the PLA/MF in this study was primarily physical, as no significant peaks or obvious shifting were observed when compared to the neat PLA spectrum [67]. The major peaks of MF fiber were not observed in the fiber-reinforced PLA composites, possibly because they were covered by other strong PLA peaks, given that the amount of fiber was small in comparison to the PLA polymer matrix. The presence of these peaks at 1746 cm^{-1} , 1451 cm^{-1} , 1381 cm^{-1} , 1359 cm^{-1} , 1266 cm^{-1} , 1180 cm^{-1} , 1127 cm^{-1} , 1042 cm^{-1} , and 755 cm^{-1} indicated that the fiber had been introduced into the polymer matrix.

3.8. Thermogravimetric Analysis (TGA). TGA testing was used to investigate the thermal degradation of the PLA/MF composite. Figures 11(a) and 11(b) show the mass lost at each stage as a function of temperature, with the residual mass at 600°C and its derivative mass analysis.

The first degradation showed a mild weight loss at less than 100°C for each sample due to moisture loss, as shown in Figure 10(b) [15, 70]. According to the FTIR results, as shown in Figures 11(a) and 11(b), the thermal degradation of the PLA/MF composite included complex reactions such as the breakage of C-H and C-O bonds [69].

Figure 11(a) illustrates a single step of transition occurred for plain PLA and PLA/MF composites. The single step was described as the decomposition of cellulose in PLA/

TABLE 7: DSC analysis of PLA/MF composites.

Samples	T _g (°C)	T _c (°C)	T _m (°C)
PLA	57.27	—	138.91
PLA/MF10	56.57	95.84	139.19
PLA/MF20	56.09	91.19	139.75
PLA/MF30	56.39	94.55	138.42

MF composite; a sharp transition occurred over a temperature range of 300°C to 400°C [40, 70]. The decomposition of PLA was a shift that occurred at the initial temperature of 330°C-360°C. As can be seen in Figure 11(a), the miswak fiber has an effect on the thermal stability of the composites as the step mainly occurred over a temperature range of 325°C to 350°C. Hence, the use of MF has resulted in a lower thermal degradation temperature and increase in residual value as shown in Table 6.

Maximum degradation is illustrated in Figure 11(b) and as final degradation temperature in Table 6. The composite decomposed quickly at 300°C to 400°C, as shown in Figure 11(b). Figure 11(b) also shows the thermal degradation on the PLA molecular chain caused the maximum decomposition rate peak around 360°C. Moreover, it was observed that there was no more mass loss after 400°C, indicating that the molecular chain had begun to carbonize at this point [68]. All PLA/MF composite maximum decomposition rate peak occurred around 300°C to 350°C. For the composition with high MF contents (30 wt. %), it seems obvious that the degradation is shifted to lower temperature. It indicates that the addition of MF had reduced the composite thermal stability as it was also found that MF's thermal degradation temperature (166°C) was found to be lower than PLA's (336°C) and could be one of the causes for lower thermal degradation temperature. The finding revealed that the addition of natural fiber affects the matrix by accelerating the thermal degradation of PLA as fiber contains a carbonyl group as observed in the FTIR test [71–73]. All of these observations further proved that the MF shows an effect on the thermal stability of the PLA matrix.

In addition to MF, the composites' initial degradation temperature has decreased slightly, and the composites' final thermal decomposition solid residue has increased, as shown in Table 6. The much high solid residue generated from hemicellulose and lignin contain in MF can be attributed to the charring, while the full decomposition of cellulose may be attributed to quick devolatilization reaction that led to very few solid residues left as such in PLA/MF10 [74]. Furthermore, the MF thermal decomposition solid residue was found to be 37% higher than PLA (0.86%). As a result, the thermal decomposition solid residue has increased, while the composite initial decomposition temperature decreased.

3.9. Differential Scanning Calorimetry (DSC). The thermal characteristics of plain PLA and PLA-based composites were characterized using DSC measurements. The thermograms that are produced are shown in Figure 12, and the numerical values are listed in Table 7. A melt-peak in the thermograms suggests the presence of crystalline domains.

All composites had comparable glass temperatures (T_g); however, they were somewhat lower than plain PLA. Molecular properties, compatibility, and composition all impact the glass transition temperature [75]. Glass transition temperature can also be influenced by poor compatibility with additional fiber [68]. As a result of the poor compatibility between the MF and PLA, the glass transition temperature was decreased. T_g of particulate packed polymers did not alter much as a result of the creation of a hard interphase because the surface area of micron-sized fiber particles is too small to induce large quantities of interphase to occur [76, 77].

In the DSC heating scans, all the PLA-based composites showed faint exothermic peaks just before the endotherm peak associated with melting is attributed to the crystallization [78]. However, no obvious crystallization peak was found for plain PLA. This indicates that the crystallization rate of PLA was very slow and temperature 90°C was too low to induce PLA crystallization within a short time due to low chain mobility. The MF/PLA composites however observed a faint exothermic peak from 90°C to 120°C as addition of fiber slightly induce PLA crystallization in composite.

Melting temperatures for PLA-based composites increased somewhat, suggesting that crystal sizes grew in the sequence MF30 < MF10 < MF20, as bigger crystals melt at higher temperatures than smaller crystallites [79]. Table 7 shows that the melting temperatures of pure PLA and PLA/MF composites are 138°C to 140°C, as illustrated in Figure 12. Spiridon et al. [79] discovered exothermic peaks in cellulose fiber/PLA samples using a similar DSC heating scan [79].

3.10. Dynamic Mechanical Analysis (DMA). The effect of MF loading on the viscoelastic properties of PLA composites was investigated using DMA. Figure 13 depicts the temperature dependence of dynamic mechanical properties, storage modulus in Figure 13(a), loss modulus in Figure 13(b), and tan δ in Figure 13(c) of PLA and MF/PLA composites.

The storage modulus, loss modulus, and tan delta curves as a function of temperature were examined using a dynamic mechanical analyzer at elevated temperatures. The viscoelastic behavior of materials can disclose the polymeric system's structure. PLA and all composite samples have a typical storage modulus curve illustrated in Figure 13(a), as found in semicrystalline polymers, with high storage modulus at low temperatures and a sharp decline at glass transition temperature, reaching the second plateau, which corresponds to the rubbery state. The storage modulus of all samples is provided in Table 8 at two distinct temperatures (above and below the glass transition temperature). PLA-based composites had a greater storage modulus in glassy areas than plain PLA, which is especially noticeable in composites with MF30 with 30 wt. % fiber content. Only MF30 had a slightly greater storage modulus in the rubbery state than pure PLA, indicating that PLA/MF composites had improved thermal-mechanical stability at extreme temperatures [80]. The improvement in storage modulus was characterized by the restricted polymer chain mobility because of the addition of lignocellulosic fiber. Similar behavior for PLA-based composites has been reported due to the incorporation of

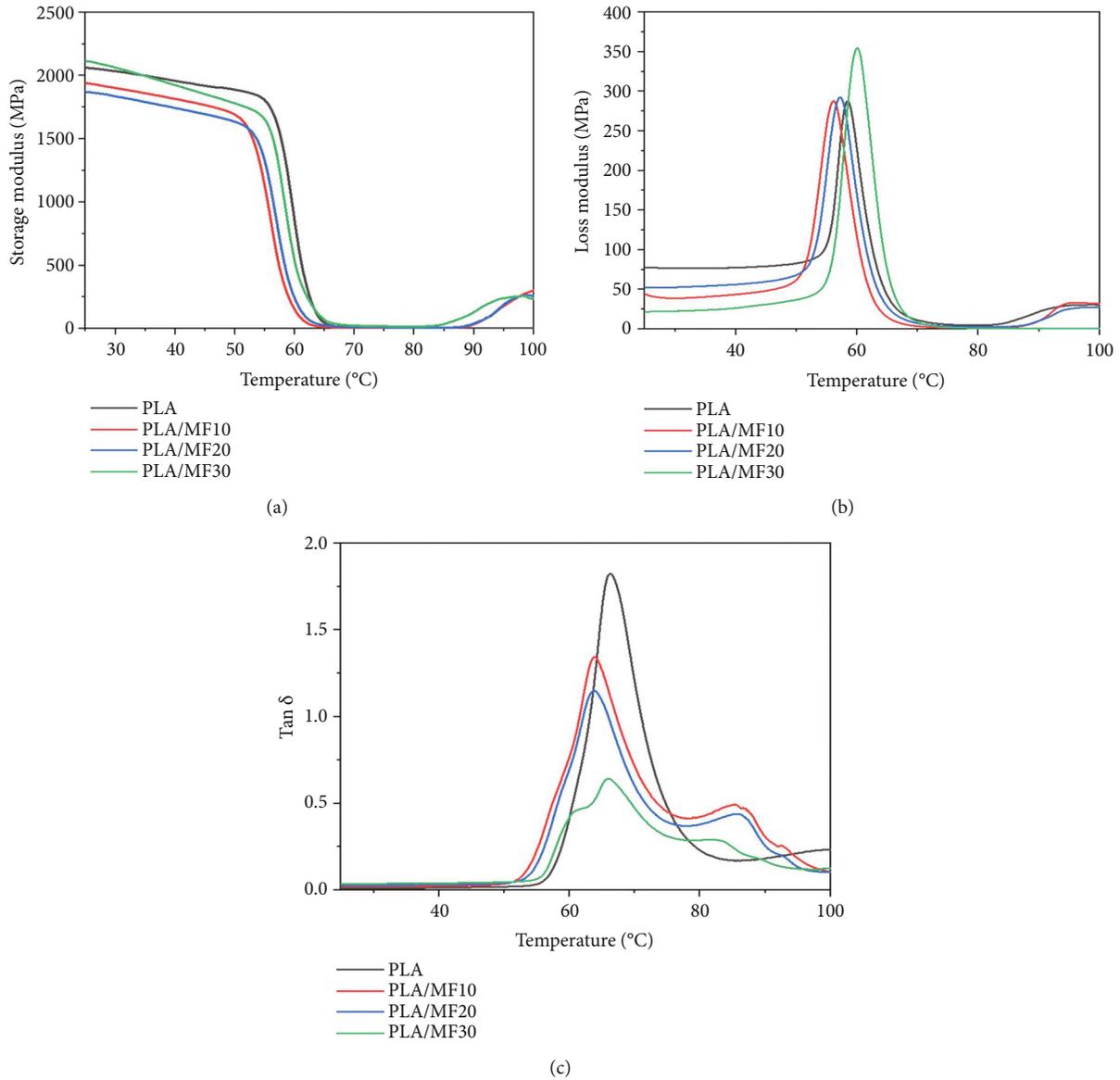


FIGURE 13: (a) Storage modulus curve of PLA/MF composites with different fiber loading (10 wt. %-30 wt. %). (b) Loss modulus curve of PLA/MF composites with different fiber loading (10 wt. %-30 wt. %). (c) Tan Delta curve of PLA/MF composites with different fiber loading (10 wt. %-30 wt. %).

TABLE 8: Storage modulus, loss modulus, and tan δ of PLA/MF composites.

Composites	Storage modulus (MPa)		Peak height of tan δ curve	Tg (°C) from tan δ curve	Tg (°C) from loss modulus curve
	30 °C	100 °C			
PLA	2034	4.35	1.8	66	60
MF10	1901	302.93	1.3	63	56
MF20	1835	260.08	1.1	63	57
MF30	2062	236.32	0.6	65	58

different concentration of lignocellulosic fiber into PLA matrix [80]. In glass transition region, it was observed that the E'' peak of PLA composite was higher than PLA/MF composites illustrated in Figure 13(b). The PLA/MF com-

posites all had a similar level of E'' peak, showing that adding fiber into the polymer chain disrupts PLA chain movement, which is linked to the matrix chains' reduced mobility.

TABLE 9: Comparison mechanical properties of PLA/MF with thermoplastic based material.

Type of material	Tensile strength (MPa)	Young's modulus (GPa)	Flexural strength (MPa)	Flexural modulus (GPa)	Fabrications	Characteristics	Reference
PLA/MF	24-34	2-3	49-66	2-3.2	Melt mixing, compression molding	(i) High stiffness (ii) High flexural strength (iii) Lightweight	Present study
PLA	45-62	1.-2.8	63-79	2.8-3.7	Extrusion, injection molding	(i) High strength (ii) High stiffness (iii) Biodegradable	[99-101]
Recycled PLA	52-64	0.012-0.014	0.001-0.004	NA	Extrusion, 3D printing	(i) Drastic decrease in tensile after 9th recycle (ii) Good phase shape (iii) Low processing temperature	[102, 103]
PP	30-45	2-3	20-35	1-4	Rotating twin screw extruder	(i) Flexible (ii) Lightweight (iii) Toughness	[104-106]
Recycled PP	25-40	0.6-0.8	30-40	3-4	Rotating twin screw extruder	(i) Stiffer (ii) Brittle (iii) High bending	[105, 106]
Bamboo/PP	10-15	2-4	45-50	2-3.5	Melt mixing and compression molding	(i) Lightweight (ii) High flexural strength (iii) Hybrid has better tensile strength	[107, 108]
PE	15-17	2-3	19-20	1-2	Rotating twin screw extruder	(i) Chemical resistant (ii) Impact resistance (iii) Resist wear and fatigue	[107]
LDPE	10-15	0-1	20-30	0.3-0.4	Rotating twin screw extruder	(i) Toughness (ii) Flexible (iii) Lightweight	[83, 107]

The placement of the tan delta curve's peak value as the glass transition temperature of samples suggests that DSC measurements yielded similar glass transition temperatures. In Figure 13(b) after 60°C, the composite softened, the modulus began to decrease, and after 70°C, the composite was in a constant state due to α -relaxation of the amorphous regions in PLA [81]. Furthermore, the amplitude of the tan delta peak indicates the energy dissipation caused by the chain mobility of the polymer segments at the corresponding temperature as illustrated in Figure 13(c). The tan delta peak value was reduced when fillers were added to PLA, indicating stiffening action of fillers in composites. The peak value of tan delta in composites decreased as the fiber content of the composite increased. The lower tan delta peak values for PLA/MF30 composites compared to PLA/MF10 composites can be attributed to a greater interfacial connection between the MF and PLA matrix due to more uniform fiber dispersion as better fiber-matrix contact can constrain polymer chains, resulting in less energy loss [82]. This finding matches that of Shojaeiarani et al., who found decreased tan delta intensity in PLA-based composites reinforced with cellulose crystal [80].

4. Comparison of the Mechanical Characteristics of MF/PLA Composite with Thermoplastic-Based Materials

Most researchers utilize the ASTM D638 standard to assess the tensile characteristics and the ASTM D790 standard to determine the stiffness for the material used for dental applications like toothbrushes or biomedical applications like implants [46, 83-89].

The mechanical characteristics of PLA/MF composite are compared in Table 9 to materials that are often used to make biomedical, dental, or packaging products, such as PP, PE, TPE, recycled polymer, and bamboo-based PP composite material [88-95].

The table demonstrates that the majority of the tensile and flexural characteristics of PLA/MF are within the specified application's range. This demonstrates that adding MF to PLA polymer will not reduce the initial strength of the commercial polymer or PLA based composite; rather, it will only cause a decrease in PLA's properties, which could be brought on by a number of factors, including the strength of the fiber reinforcement, the processing environment, and the interfacial adhesion between the fiber and matrix.

TABLE 10: Challenge injection molders deals with PLA polymer.

Material	Challenge	Causes	Solution suggestion
PLA	Flow lines (patterns or lines appear on parts)	(i) Issue in varying flow rates as molten polymer moves through mold	✓ Increase the injection pressure and speeds to ensure cavities are filled evenly
		(ii) Changes in the thickness of the molds walls	✓ Increase temperature of the molten polymer or mold preventing polymer from quick cooling
		(iii) Injection speed to slow that cause molten polymer to solidify at different rates	
	Sink marks (small depression in thicker areas of the injection molding after there is shrinkage in the finished product)	(i) Issue with pressure inside cavity (ii) Incorrect mechanism or cooling time	✓ Decrease mold temperature ✓ Allow polymer to cool and cure longer inside the mold
Vacuum voids (air pockets trapped inside an injection molded part)	(i) Insufficient holding pressure that led to molten plastic to condense inside the mold	✓ Use less viscous polymer to avoid trapping gas ✓ Increase holding pressure and time	
Warping (finished part are uneven in areas)	(i) Nonuniform cooling or varying cooling rates inside mold	✓ Cooling process are not rushed in term of time and speed	

* Source from <http://www.iconnsystems.com> and <http://www.rtpcompany.com/technical-info>.

Owing to the near resemblances in strength and stiffness as shown in Table 9, it is evident that employing MF might be more advantageous than using a conventional polymer, but modifications and more study are required to attain the necessary features and characteristics.

Additionally, PLA is one of the least expensive materials available for composite materials of biomedical or packaging, and it also has some of the finest tensile and flexural qualities among other thermoplastics [94, 96, 97]. There is a good likelihood that the material will come into contact with moisture and water while the toothbrush is being made. Injection molding is often used in the production of toothbrushes. This procedure entailed creating a mold in the shape of a toothbrush, injecting molten plastic into the mold using a hot channel nozzle, cooling the plastic, and ejecting it from the mold [98].

Currently, this procedure requires comprehension of the fundamentals of injection molding, where material choice is frequently reliant on knowledge of the maximum short-use temperature, the melt flow, and the relation between stress and temperature [109]. Therefore, in order to use the injection molding method, it is crucial to comprehend not only the mechanical side of the material but also its thermal stability. Thermoplastic materials including PP, PE, ABS, PA, and PMMA are often utilized in injection molding [109].

Bioplastic has previously caused problems for injection molders since the materials were pricey and incompatible with existing machinery. However, PLA's characteristics might make it well suited for some applications while necessitating changes for others [92, 110].

4.1. Injection Molding and PLA. We compared the mechanical properties of the current PLA/MF composites with those of a commercial material that is currently widely used in making toothbrush handles as shown in Table 9 because one of the objectives of this study was to develop composite materials containing miswak, which is renowned for its antibacterial properties, to be potentially feasible for application

similar to toothbrush handle. Understanding the optimal injection molding settings for polylactic acid (PLA)-based compounds is crucial to obtaining the best performance out of the materials [109]. PLA is a semicrystalline polyester thermoplastic. Since polyester is prone to hydrolysis, molders must take special care to ensure that the material is dry before molding. By adjusting the molding conditions, PLA may be molded similarly to other semicrystalline polyester. Thermoplastic PLA rapidly absorbs water from the atmosphere because it is hygroscopic. Even a minor quantity of moisture will hydrolyze PLA in the melt phase, causing molecular weight to drop and characteristics to be lost [79, 89, 111]. Table 10 shows factor or challenge that occurs when polymers are molded using the injection process.

Since PLA is costly, has inferior water barrier qualities to more common thermoplastics like PP, and has low heat stability, it is not utilized to produce toothbrushes [71, 110, 112–114]. However, great attempts have been made to alter PLA's performance to increase its temperature tolerance and mechanical strength as well as to adapt it to increase its uses in high-end markets [72, 110, 114–118].

The slower PLA crystallization time, which is reflected in longer molding cycles compared to commercial fossil-based polymers, is a disadvantage for the PLA injection molding application. PLA could not be injected into molds at ambient temperature, which is the norm for fossil-based polymers, since the resultant material is amorphous and has a poor storage modulus, especially at temperatures higher than its glass transition temperature. This complicates the PLA injection molding process [119, 120]. The proper mold temperature and cycle duration must be attained in order to crystallize PLA during injection molding. However, the PLA crystallization rate must be hastened throughout the process to decrease the additional cost brought on by the usage of hot molds and the lengthened time cycle. In the past, studies have looked at a variety of methods, including plasticization, nucleation, and physical blending with other polymers, in order to quickly obtain high PLA crystallinity [119,

TABLE 11: Characteristic of PLA based material by using injection molding.

Material	Approach	Application	Fabrication	Characteristics	Ref
PLA/sisal fiber	Reinforcement fiber	Bio composites material	Direct- injection molding	<ul style="list-style-type: none"> ✓ High tensile modulus ✓ High flexural modulus ✓ Better impact properties ✓ Improving heat deflection ✓ Temperature with crystallinity 	[126]
PLA/nucleating agents (LAK, talc, calcium carbonate)	Nucleating agents	Commercial application	Extrusion and injection molding	<ul style="list-style-type: none"> ✓ Percentage above 50%. ✓ Increase flexural modulus ✓ Lower impact resistance ✓ Improve strength and modulus 	[127]
PLA/nucleating agents (talc, ethylenebis-stearamide (EBS))	Nucleating agents		Extrusion and injection molding	<ul style="list-style-type: none"> ✓ Increase heat distortion temperature ✓ Decreased processing time increase crystallinity level ✓ High loads resistance ✓ Increase shrinkage with melt temperature increase ✓ Increase warpage with packing time decrease 	[121]
PLA-based bone screws	Reinforcement nanoparticles	Biomedical application	Injection molding	<ul style="list-style-type: none"> ✓ Large range of mechanical properties ✓ Improve microstructure ✓ Improve surface roughness ✓ Improved thermal properties 	[128]
PLA/TPU	Hybrid polymer		Extrusion and injection molding	<ul style="list-style-type: none"> ✓ Improved tensile and impact properties ✓ Reduction damping factor ✓ Increase deflection temperature under load 	[129]
PLA/hydroxyapatite (HA)	Surface modification of HA	Good load bearing applications	Extrusion and injection molding	<ul style="list-style-type: none"> ✓ Increase tensile modulus ✓ Increase Izod strength 	[130]
PLA/jute	Reinforcement fiber	Fully environmentally friendly material (FEFM)	Pultrusion process and injection molding		[131]

121–124]. The nucleation method is particularly appealing since it may reduce the injection molding time while barely impacting PLA stiffness and strength. Additionally, it was claimed that the nucleation technique is adaptable since it is simple to apply during extrusion and that a variety of nucleating chemicals are readily available [125]. The method for enhancing PLA characteristics by injection molding is shown in Table 11.

PLA is by far the most significant and promising bio-based and biodegradable plastic for solid applications. Even though PLA has received a lot of scientific attention, it has mostly been used in biomedical and packaging applications. The manufacturing cost of PLA will drop dramatically at industrial sizes due to a growing awareness of environmental contamination and advancements in production technology, and PLA-based composites will hold great promise for the future of the material business.

5. Conclusion and Future Scopes

Miswak fiber (MF)-reinforced PLA composites with different fiber loading were successfully prepared by melt blending. These studies have looked into how MF affects

morphologies, physical characteristics, mechanical properties, and thermal attributes. The research has proved that increasing the amount of MF slightly lowered the tensile and flexural strength and modulus of PLA/MF composites. The results might have been impacted by void presence, porosity, fiber orientation, and inadequate interfacial adhesion between the fiber and matrix composite. In addition, fiber's low adhesion profile—caused by a smaller proportion of cellulosic components and a higher proportion of hemicellulose and extractives on the fiber surface—prevents it from effectively binding with the matrix. This study discovered a nonlinear response to density increments of 10 wt. % fibers. The addition of MF increases the storage modulus in the low temperature region, which is associated with lower α -relaxation, as compared to PLA. According to DSC's TGA study, the composites' initial thermal degradation temperature has dropped considerably, while their final thermal decomposition residual ratio has risen steadily. This is as a result of MF's lower thermal decomposition temperature compared to PLA's. Also, with fabrication of the PLA/MF composite, the study's objective was accomplished, and the material's characteristics in terms of its physical, mechanical, and thermal capabilities were investigated. In the future, the

studies intend to examine the whole range of how various processing conditions—including those related to time and temperature—affect the mechanical and thermal characteristics of composite materials. Additionally, fiber processing parameters, fiber orientation, and fiber particle size can all be taken into account as variables that affect the features of the composite. In addition, fiber treatment or compatibilizing agents are considered in the endeavor to enhance the qualities of PLA/MF composites, as their effect on antibacterial and biocompatibility of the composites is still needed for research as to utilize and explore the efficacy and usefulness of miswak plant for diverse application.

Data Availability

The analysis data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare no conflict of interest.

Authors' Contributions

Conceptualization was performed by Khalina A.; methodology was contributed by Nur Diyana A.F.; validation was performed by Sapuan M.S.; formal analysis was performed by Nurazzi M.N.; investigation was performed by Aisyah H.A.; resources was contributed by Nur Diyana A.F.; data curation was performed by Nur Diyana A.F.; writing—original draft preparation was contributed by Nur Diyana A.F.; writing—review and editing was contributed by Lee C.H.; supervision was performed by Khalina A. All authors have read and agreed to the published version of the manuscript.

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