

Research Article

Oil Palm Mesocarp Fiber as New Lignocellulosic Material for Fabrication of Polymer/Fiber Biocomposites

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New biocomposites consisting of poly (butylene succinate) (PBS) and various content (0–70 wt%) of oil palm mesocarp fiber (OPMF) or oil palm empty fruit bunch fiber (OPEFBF) were fabricated by melt blending and subsequently hotpress moulding. The tensile, flexural, and impact properties of those biocomposites were evaluated and compared. Enhancement of flexural modulus of 200 or 150% was observed with PBS biocomposite loaded with 70 wt% of OPMF or OPEFBF. PBS/OPMF biocomposites exhibited higher values of tensile, flexural and impact strengths, and tensile and flexural moduli than those of PBS/OPEFBF biocomposites. These results indicated that OPMF feature better reinforcing agent for PBS as compared to that of OPEFBF.

1. Introduction

Poly (butylene succinate) (PBS) is a biodegradable polymer synthesized by the polycondensation of 1, 4-butanediol and succinic acid [1]. It has excellent mechanical properties, melt processing capability, and biodegradability [2]. It can be degraded by microorganisms in compost, soil, and seawater [1, 3]. However, relatively high production cost of PBS as compared to nonbiodegradable polymer has limited many of its applications. Therefore, blending of PBS with low cost natural fiber may produce the required materials at competitive price [3].

Utilization of natural fibers in composite preparation offer several advantages compared to those of synthetic fibers which include low density, low cost, renewability, and biodegradability [4, 5]. Natural fibers like kenaf [3, 6], jute [2, 5], coir [7], silk [8], bamboo [9], sisal [10], and oil palm empty fruit bunch [11–13] have been extensively studied for fabrication of polymer/fiber composites. Rozman et al. [11] reported

that incorporation of the OPEFBF into polypropylene matrix resulted in an improved tensile modulus but reduced in tensile strength, elongation at break, and impact strength with increasing fiber content.

Biomass of OPMF and OPEFBF are lignocellulosic materials produced in palm oil mill after the oil extraction process. OPEFBF is obtained from the fruit bunch after removal of oil palm fruits whereas OPMF is residue obtained from oil palm fruits after the oil extraction [14]. They are either left in the palm oil mill as wastes or used as boiler fuel to produce steam and to generate power [13, 15]. The current utilization of these fibers however has created huge environmental pollution to the environment. Due to this reason, this research was carried out to utilize them in a better way as filler for biocomposites preparation. Additionally, the utilization of these fibers in biocomposites preparation will be an added-value of oil palm biomass in the future. Other authors have reported in the past the blending of OPEFBF with high density polyethylene [11], polypropylene [12] and polycaprolactone [13] to fabricate

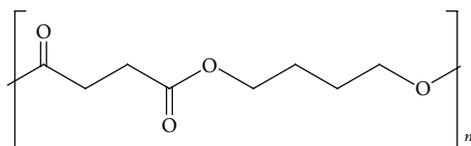


FIGURE 1: Molecular structure of PBS.

TABLE 1: Chemical and mechanical properties of OPMF and OPEFBB [14].

	OPMF	OPEFBB
Lignin (%)	11	19
Cellulose (%)	60	65
Ash content (%)	3	2
Tensile strength (MPa)	80	248
Young's modulus (MPa)	500	2000
Elongation at break (%)	17	14

polymer/fiber composites but, so far, there is no literature on reinforcing PBS. In addition, the utilization of OPMF in fabrication of polymer/fiber composite has not been reported previously.

The aim of this paper is to study the potential of OPMF as new lignocellulosic material for fabrication of polymer/fiber biocomposite via mechanical properties comparison between PBS/OPMF and PBS/OPEFBB biocomposites. In order to produce price competitive material, the amount of fibers used was increased up to 70 wt% for fabrication of PBS biocomposites.

2. Experimental

2.1. Materials. Poly (butylene succinate) (PBS), under trade name of BIONOLLE 1903MD was purchased from Showa Denko, Japan. It has a density of 1.26 g/cm³ and melting point of ~115°C. Its molecular structure is shown in Figure 1. OPEFBB and OPMF were kindly supplied by Sabutek (M) Sdn. Bhd., Malaysia and FELDA Seriting Hilir Oil Palm Mill, Malaysia, respectively. The chemical and mechanical properties of OPMF and OPEFBB are given in Table 1.

2.2. Preparation of Fibers. OPMF and OPEFBB were first washed by soaking in distilled water for 24 hour and then rinsed with hot water (60°C) and acetone prior to drying at 60°C in an oven. This process was carried out to remove some impurities from the fibers. The dried fibers were then ground, sieved into size of 150–300 μm, and stored in a sealed polyethylene bag for biocomposites fabrication.

2.3. Preparation of PBS Biocomposites. PBS, OPMF and OPEFBB were dried in an oven at 60°C prior to processing. PBS biocomposites were prepared by melt blending of PBS with various amount (0–70 wt%) of OPMF or OPEFBB in a Brabender internal mixer at 120°C with 50 rpm rotor speed. PBS pellet was first introduced into the mixing chamber to melt for 2 min. Subsequently, fibers were added to the mixing

TABLE 2: Formulations of PBS biocomposites.

Code	PBS (wt%)	OPMF (wt%)	OPEFBB (wt%)
PBS	100	0	0
PBS/10 fiber*	90	10	10
PBS/20 fiber*	80	20	20
PBS/30 fiber*	70	30	30
PBS/40 fiber*	60	40	40
PBS/50 fiber*	50	50	50
PBS/60 fiber*	40	60	60
PBS/70 fiber*	30	70	70

* fiber = OPMF or OPEFBB.

chamber and continued mixing for another 13 min. These compounded materials were then compressed into 1 or 3 mm thick film by hydraulic hotpress also at 120°C under pressure of 150 kg/cm² for 5 min, followed by cold pressing at 30°C for 5 min. The formulations for PBS biocomposites fabrication were given in Table 2.

2.4. Mechanical Properties. Tensile and flexural tests for PBS and PBS biocomposites were carried out by a Universal Testing Machine (Instron 4302). Load cell of 1 kN was used and the tests were performed at 25°C. Specimens for tensile and flexural tests were cut according to the standard of ASTM D638-5 and D790, respectively. Crosshead speed of 5 mm/min was used for tensile test. Meanwhile, crosshead speed of 1.3 mm/min and support span length of 48 mm were used for flexural test. Their results were expressed in term of tensile or flexural strength, tensile, or flexural modulus and elongation at break. Unnotched Izod impact test for PBS and PBS biocomposites was carried out following the ASTM D256 using an IZOD Impact Tester equipped with a 7.5 J pendulum at 25°C. Five specimens were tested for all the tests and the average values were reported. Their standard deviations are expressed in error bar.

2.5. Scanning Electron Microscopy (SEM). The surface morphologies of OPMF, OPEFBB and tensile fracture surfaces of PBS and PBS biocomposites were recorded by a JOEL JSM-6400 scanning electron microscope operated at 15 kV. The scanning electron micrographs were recorded at the magnification of 200–300x. All the samples were coated with gold by a Bio-rad coating system before viewing to avoid samples' charging.

3. Results and Discussion

PBS biocomposites loaded with various amount (0–70 wt%) of OPMF or OPEFBB were successfully fabricated by melt blending and their tensile, flexural, and impact properties were examined and reported. Initially, PBS biocomposites loaded with 80 wt% of fibers were also fabricated in our experiment but later excluded from our experiment because the mixing is poor due to insufficient PBS to wet the fiber (fibers and PBS still remained as separated phase after melt blending). Consequently, maximum loading of fibers were

TABLE 3: Tensile strength, tensile modulus, and elongation at break of PBS, PBS/OPMF, and PBS/OPEFBB biocomposites.

Fiber content, (wt%)	Tensile strength, (MPa)		Tensile modulus, (MPa)		Elongation at break, (%)	
	OPMF	OPEFBB	OPMF	OPEFBB	OPMF	OPEFBB
0	37.31 ± 0.91*		248.90 ± 4.40		470.00 ± 55.20	
10	25.55 ± 1.60	25.67 ± 0.39	301.50 ± 6.41	313.20 ± 2.40	19.60 ± 2.87	17.33 ± 1.29
20	19.91 ± 0.95	19.69 ± 1.01	349.60 ± 8.36	382.00 ± 29.60	13.72 ± 1.37	13.40 ± 1.38
30	17.43 ± 0.69	16.98 ± 0.88	375.33 ± 10.89	372.93 ± 42.86	9.93 ± 0.90	9.81 ± 1.73
40	16.74 ± 1.19	15.55 ± 0.34	421.90 ± 38.60	425.20 ± 42.80	7.10 ± 0.57	6.18 ± 1.96
50	15.50 ± 1.04	14.32 ± 0.46	474.20 ± 4.24	469.90 ± 54.20	4.73 ± 0.22	4.70 ± 1.00
60	14.56 ± 0.66	12.85 ± 1.28	470.50 ± 21.70	412.00 ± 48.80	3.35 ± 0.06	3.08 ± 0.42
70	13.86 ± 0.73	10.80 ± 0.83	94.80 ± 7.60	77.00 ± 25.46	2.50 ± 0.29	2.38 ± 0.10

*Standard deviation.

fixed at 70 wt%. It is interesting to note that the tensile strength of PBS biocomposite loaded with 70 wt% of OPMF or OPEFBB is 90 or 48% higher than that of low density polyethylene (7.30 MPa) [5]. This suggests that those biocomposites may have potential application as packaging materials.

3.1. Mechanical Properties

3.1.1. Tensile Properties. The tensile strength, tensile modulus, and elongation at break of PBS, PBS/OPMF and PBS/OPEFBB biocomposites are summarized in Table 3. Neat PBS has a tensile strength of 37.10 MPa and it has decreased after melt blended with OPMF or OPEFBB due to the poor interfacial adhesion between the hydrophobic PBS and hydrophilic fibers [7].

From Table 3, it is noted that the tensile strength of the PBS biocomposites reduces drastically (32–47%) as fibers loading increase from 0–20 wt% and beyond that (30–70 wt%), the reduction is rather small. It is suggested that at low fibers loading, fibers are unable to disperse evenly in the PBS matrix, lead to high stress concentrations, and cause the bond between fiber and matrix to break, hence resulting in a dramatic reduction in tensile strength. Meanwhile, as fibers content increase, fibers may disperse more evenly and oriented, and the stress is more evenly distributed, thus the reduction in tensile strength is rather small.

Although OPEFBB has 3 times higher tensile strength than that of OPMF as shown in Table 1, it is noted that the tensile strengths of PBS/OPMF and PBS/OPEFBB biocomposites are very similar to each other up to 50 wt% fibers content. As fibers content increase to 60 or 70 wt%, it is interesting to note that the tensile strength of PBS/OPMF biocomposite is 15 or 28% higher than that of the PBS/OPEFBB biocomposite. This may due to the fact the OPMF is softer and flexible than that of OPEFBB, therefore has higher ability to disperse more evenly and oriented in the PBS matrix, consequently resulted in higher tensile strength. This observation also suggests that OPMF has better reinforcing ability than that of OPEFBB.

Neat PBS shows a tensile modulus of 248.90 MPa and it increases with increasing fibers content up to 50 wt%, and then leveled off. It is noted that PBS/OPMF biocomposites show higher tensile modulus than those of PBS/OPEFBB biocomposites. Enhancement in tensile modulus is probably

due to the fibers itself which have higher stiffness or modulus than that of polymer. Meanwhile, the decreased in tensile modulus at high fibers loading (60–70 wt%) probably due to the decreased in wettability between PBS and fibers resulted from the PBS content not sufficient to wet the surrounding fibers during composite fabrication, leading to poor load transfer between PBS and fibers [7, 16].

PBS is ductile and flexible in nature, so high value of elongation at break (EB) of 470% is noted. The EB of PBS biocomposites has reduced to around 2.50 or 2.38% upon addition of 70 wt% of OPMF or OPEFBB. The decreased in EB is due to the destroyed structural integrity of PBS by fibers and the rigid structure of fibers [17]. Furthermore, as fiber contents are increased to high extend, the fibers tend to agglomerate and imply poor PBS/fiber adhesion which will then promote the formation of microcracks at the interface, resulting in quicker fracture than that of neat PBS [17]. This result is in good agreement with the result reported by Habibi et al. [18] where the authors claimed that the EB decreased upon fiber addition for composites regardless the nature of the fiber.

3.1.2. Flexural Properties. The flexural strengths and moduli of PBS and PBS biocomposites filled with various content of OPMF or OPEFBB are shown in Figures 2 and 3, respectively. From Figure 2, neat PBS shows a flexural strength of 37.58 MPa and it has decreased to 27.26 and 21.53 MPa or reduction of 27 and 43% upon addition of 70 wt% of OPMF or OPEFBB, respectively. The reduction in flexural strength may be ascribed to insufficient wetting of fiber by the polymer matrix with the presence of the fiber bundles and more fiber-fiber interactions as reported by Mohd Ishak et al. [19]. Although the flexural strength of PBS biocomposites decreases with increasing fibers content; however, PBS/OPMF biocomposites still exhibit higher flexural strength than those of PBS/OPEFBB biocomposites at all fiber loading. For instance, the flexural strength of PBS/70 OPMF biocomposite is 27% higher over PBS/70 OPEFBB biocomposite. This result once again proves that OPMF may have better interfacial adhesion and compatibility with PBS matrix and therefore produce stronger materials.

In contrast to flexural strength, increasing trend is observed in the flexural modulus of PBS biocomposites with increasing fibers content of up to 70 wt%. The PBS/OPMF

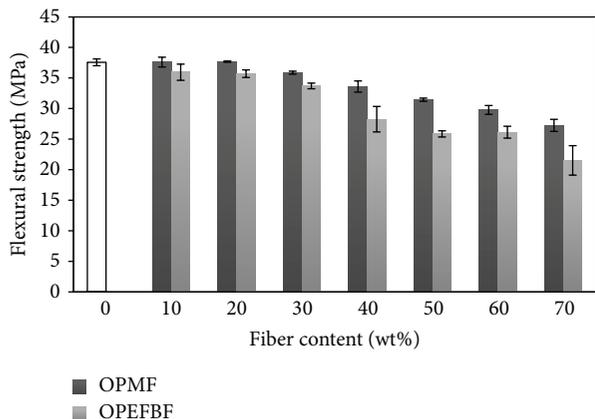


FIGURE 2: Flexural strength of PBS, PBS/OPMF and PBS/OPEFBB biocomposites.

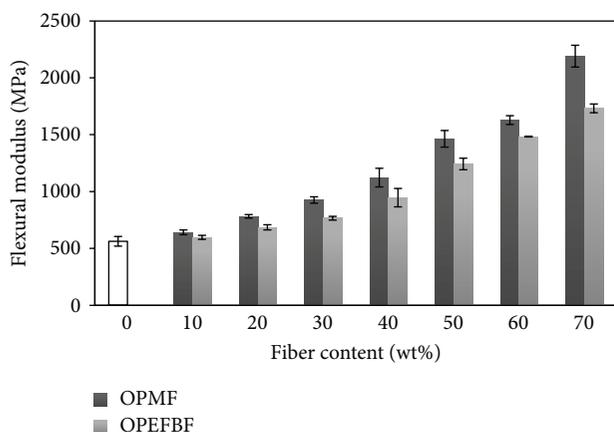


FIGURE 3: Flexural modulus of PBS, PBS/OPMF, and PBS/OPEFBB biocomposites.

biocomposites show higher flexural moduli than those of PBS/OPEFBB biocomposites at all fiber loading. Improvements of 200 and 150% are noted for PBS/70 OPMF and PBS/70 OPEFBB, respectively as compared to that of neat PBS. Lee et al. [6] reported that when natural fibers were incorporated into the polymer matrix, the segmental movement of the polymer chains was hindered; hence, the composites become stiffer and improve their flexural moduli.

3.1.3. Impact Properties. Figure 4 shows the impact strength of un-notched samples of PBS, PBS/OPMF, and PBS/OPEFBB biocomposites. It is evident that the impact strength of PBS biocomposites is lower than that of neat PBS regardless of type and content of fibers. PBS/OPMF biocomposites show higher degree of impact strength as compared to those of PBS/OPEFBB biocomposites. PBS is ductile or plastic like in nature, can absorb more energy during impact failure, and, hence, has high impact strength. However, the presence of OPMF or OPEFBB in the PBS matrix has turned the ductile behavior of PBS to brittle as indicates by decreasing of impact strength of about 33–84% or 36–89%, respectively. According to Mat Taib [20], the introduction of stiff fiber in a ductile

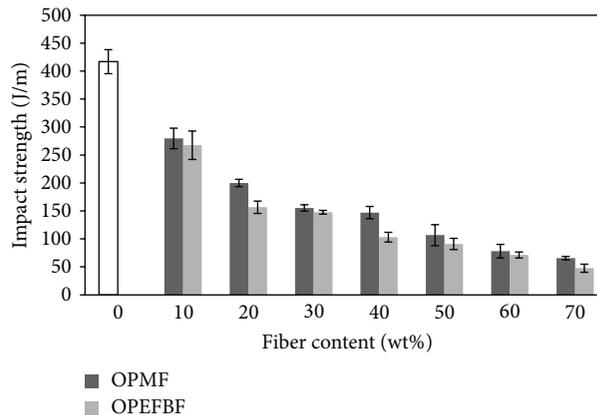


FIGURE 4: Impact strength of PBS, PBS/OPMF, and PBS/OPEFBB biocomposites.

matrix will restrict the segmental motion of the polymer chains, which will consequently limit the deformability of the matrix phase, resulting in low impact strength.

3.2. Morphology. SEM analysis is performed to evaluate the interfacial adhesion between PBS matrix and OPMF or OPEFBB. Figure 5 shows the SEM micrographs of OPMF, OPEFBB, and tensile fractured surfaces of PBS and PBS biocomposites.

PBS exhibits smooth fracture surface. OPMF and OPEFBB also show smooth surfaces but with some impurities covering their surface. Once they are melt blended together, 2 phases (PBS and fibers) are obviously seen as shown in Figures 5(d)–5(g). The presence of voids on the fractured surfaces of the PBS biocomposites which resulted from fiber pull outs is observed. In addition, gaps are also visible between the PBS and OPMF as shown in Figures 5(d)–5(e). The presence of voids and gaps are evidences of the poor interfacial adhesion resulting from the lack of compatibility between hydrophilic fibers and hydrophobic PBS. Poor interfacial adhesion can then act as a stress concentration point upon exertion of external forces and consequently result in premature failure due to poor stress transfer from matrix to the fibers [21].

At low fibers loading (Figures 5(d)–5(e)), it can be clearly seen that the fibers are not evenly distributed throughout the PBS matrix and the gaps between PBS and fibers are large; however as fibers content increase (Figures 5(f)–5(g)), fibers become more evenly distributed and oriented in the PBS matrix. This may be the reason why drastic reductions in tensile strength and elongation at break are observed at low fibers loading but rather small at high fiber loading as discussed in Section 3.1.1.

For comparison proposes, the tensile fracture surface micrograph of PBS biocomposite filled with 70 wt% of OPMF or OPEFBB is presented in Figures 5(g) or 5(h), respectively. The smooth and clean fiber surface shown in Figure 5(h) indicates a weak interaction or poor adhesion between OPEFBB and PBS. Meanwhile, rough fiber surface and few fiber breakages are observed in Figure 5(g), which indicates better

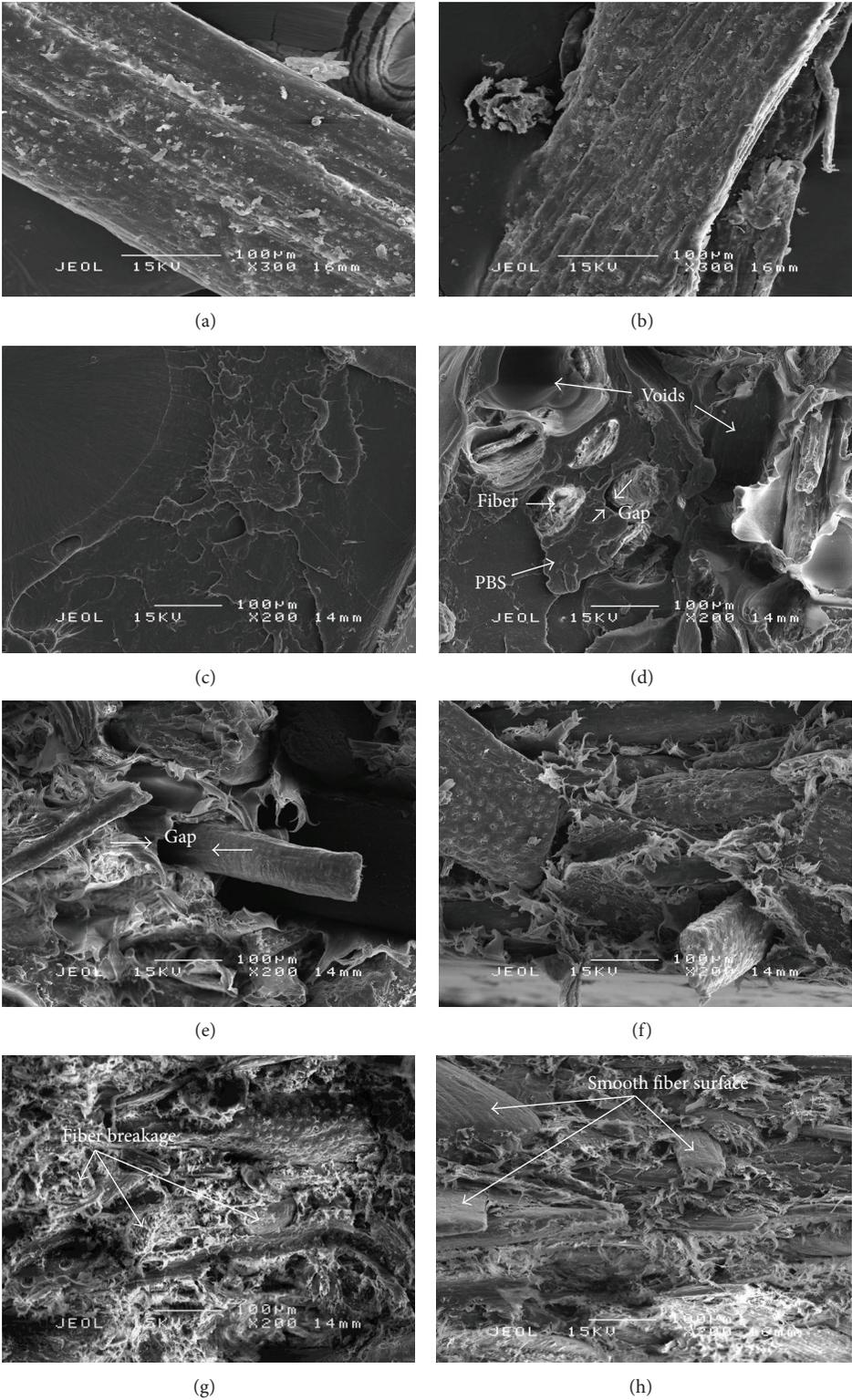


FIGURE 5: SEM micrographs of (a) OPME, (b) OPEFBF, (c) PBS, (d) PBS/10 OPME, (e) PBS/30 OPME, (f) PBS/50 OPME, (g) PBS/70 OPME, and (h) PBS/70 OPEFBF.

interfacial adhesion between PBS and OPMF. This may be the reason why PBS/OPMF biocomposites especially at high OPMF loading exhibit higher tensile, flexural and impact strengths, and tensile and flexural moduli than those of PBS/OPEFB biocomposites.

4. Conclusions

In this study, PBS biocomposites loaded with high amount of fibers (0–70 wt%) were successfully fabricated by melt blending technique. Flexural and tensile moduli of PBS biocomposites were enhanced by the present of fibers but an opposite trend was observed for elongation at break, tensile, flexural, and impact strengths. Throughout the study, PBS/OPMF biocomposites possessed higher tensile, flexural and impact strengths, and tensile and flexural moduli than those of PBS/OPEFB biocomposites. It was evident that OPMF could be a potential new lignocellulosic material for fabrication of polymer/fiber biocomposites. Furthermore, the prepared biocomposites may have potential as packaging materials since they have a better tensile strength than that of polyethylene.

Conflict of Interests

The authors declare that there is no conflict of interests with any financial organization regarding the materials discussed in the paper.

Acknowledgment

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