EVALUATION AND APPLICATION OF THE INVASIVE WEED MIKANIA MICRANTHA AS AN ALTERNATIVE REINFORCEMENT IN RECYCLED HIGH DENSITY POLYETHYLENE

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In this study *Mikania micrantha* particle (MP) and fiber (MF) were added to recycled high density polyethylene (rHDPE) for producing natural fiber (or particle) reinforced plastic composites (NFRPC) by the flat-platen pressing process. The results showed that the flexural strength and stiffness of NFRPC were significantly improved through incorporating *M. micrantha* particle and fiber. Higher aspect ratio of reinforcement displayed stronger mechanical properties. The vertical density profile in composites significantly influenced the mechanical properties of NFRPC. A conventional V-shaped profile and a uniform vertical density profile (homo-profile) were observed in MP and MF based NFRPC, respectively. Additionally, with increasing lignocellulose content, a more uniform vertical density profile and higher wood screw holding strength were observed. These results indicate *M. micrantha* particle and fiber are excellent reinforcements for NFRPC applications.

Keywords: Mikania micrantha; Natural fiber reinforced plastic composite (NFRPC); Flat-platen pressing; Aspect ratio; Density profile

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INTRODUCTION

Mikania micrantha (Asteraceae), an invasive weed originating from Central and South America, seriously damages agricultural land, orchards, pastures, and forests across Asia and Africa. Because of its vigorous, rapid, and rampant growth, *M. micrantha* is called the "mile-a-minute weed" and is listed as one of the top 10 worst weeds in the world (Holm *et al.* 1977). The first record of the species in the eastern hemisphere is from 1844, when it was cultivated at Hong Kong Zoological and Botanical Gardens (Wang *et al.* 2003). *M. micrantha* is an herbaceous vine that reproduces readily through both sexual and vegetative means. It propagates by wind-dispersed achenes and stem fragments that root easily at the nodes (Barreto and Evans 1995). Once established, this weed smothers other plant species nearby and then kills the smothered plants by cutting out the sunlight (Zhang *et al.* 2004).

To control or eliminate the infestation of *M. micrantha*, a number of physical, chemical, and biological methods have been employed (Chen *et al.* 2003; Cock *et al.*

2000; Kuo *et al.* 2002). Of these, consecutive-cutting is one of the most simple and effective methods, but this method simultaneously causes a huge amount of lignocellulosic waste, such as stem fragments. Without suitable treatment, the resulting fragments may reproduce through vegetative means and further invade ecosystems. Therefore, one of the unprecedented challenges in consecutive-cutting is how to utilize the lignocellulosic residues. A previous study reported that velvet leaf (*Abutilon theophrasti*), which is also a weed and an agricultural problem like *M. micrantha*, could be used as a source of high quality plant fiber (Reddy and Yang 2008). Therefore, natural fiber reinforced plastic composites (NFRPC) made with *M. micrantha* could potentially become one of the most useful methods for its beneficial utilization.

The wood-plastic composite (WPC), a kind of NFRPC, is reported to belong to one of the most dynamic sectors in the plastics industry (Rothlin 2007). The global WPC market has been experiencing double-digit growth in North America and Europe (Ashori 2008; Lei and Wu 2010), and the volume of WPC is predicted to increase from 129,000 tons in 2008 to 427,000 tons in 2014 (Lampinen 2010). Accordingly, the demand for lignocellulose as a raw material for WPC and other wood productions is gradually increasing, which intensifies the worldwide scarcity of forest resource in many countries. Due to greater environmental awareness, research on the development of composites produced by various waste materials is being actively carried out in the past years (Ashori and Nourbakhsh 2010; Nourbakhsh and Ashori 2010; Yao *et al.* 2008). The use of agro-forest waste as a reinforcement for WPC production can alleviate the shortage of wood resources and is advantageous to the economy and the environment.

Thus, the aim of this study was to develop and utilize *M. micrantha* as alternative raw materials for NFRPC. Systematic experiments were carried out to realize the effects of *M. micrantha* morphology and loading content on the mechanical properties of NFRPC. In addition, the dynamic mechanical properties of NFRPC were also determined in this study.

MATERIALS AND METHODS

Lignocellulosic Materials

Mikania micrantha was sampled from individual farmland in Yunlin County in June, 2010. *M. micrantha* particle (MP) was prepared by hammer mill and sieved into four fractions, -6+16, -16+30, -30+60, and -60 mesh. A refiner was utilized to prepare *M. micrantha* fiber (MF). Sugi (*Cryptomeria japonica*) particle (SP) and fiber (SF), as reference wood materials, were also prepared by the above-mentioned methods. The morphological characteristics of lignocelluloses were analyzed by a stereo microscope (M500, Motic, China) equipped with a digital CCD camera (STC-TC83USB-A, Sentech, Japan) and image analysis software (MultiCam EZ 2007, Shengtek, Taiwan). The aspect ratio of lignocelluloses was calculated according to their morphological characteristics.

Plastics

Recycled high density polyethylene (rHDPE; melting point: 130° C; melt flow index (MFI): 4.2 g/10 min; density: 940 kg/m³) was kindly supplied by Horng Gee Co., Ltd (Changhua, Taiwan). The plastic pellets were ground in an attrition mill to reduce their particle size to less than 20 mesh before composite processing.

Composite Processing

To manufacture the NFRPC, the flat-platen pressing process was applied according to our previous paper (Lee *et al.* 2010). Lignocelluloses (MP or MF) and rHDPE powder with different weight ratios (50/50, 60/40, and 70/30 wt%) were batch mixed by paddle-type mixer at room temperature for 5 min. The mixture was hand-formed by a forming box into homogeneous single-layer mats and then pressed by two-step processing: (1) hot pressing at 180°C for 7.5 min and (2) finished by cold pressing for 10 min. The expected density of NFRPC was 750 \pm 50 kg/m³. The format of the NFRPC was 300 mm × 200 mm with 12 mm thickness.

Determining the Composites Properties

The density, water absorption, thickness swelling, wood screw holding strength, internal bond, and flexural properties were determined according to the ASTM D-790, D-1037, and D-2395. In brief, modulus of rupture (MOR) and modulus of elasticity (MOE) were obtained by the three-point static bending test with a loading speed of 5.12 mm/min and a span of 192 mm. Wood screw holding strength and internal bond were tested at a tensile speed of 1.5 and 2 mm/min, respectively. All samples were conditioned at 23°C and 50% relative humidity for 2 weeks before testing. At least five specimens of each blend were tested.

Evaluation of the Vertical Density Profile

The vertical density profiles of specimens were analyzed by a QTRS-01X X-ray density profiler (Quintek Measurement Systems, Knoxville, TN, USA). Specimens (50 mm \times 25 mm and 12 mm thickness) were scanned in the thickness direction and the measurements were taken at 0.04-mm intervals. Density was determined based on the relationship with X-ray attenuation. Absorption of the X-ray energy is determined in a controlled range and is related to the actual sample density. The equipment was calibrated to the actual sample density. Three specimens of each blend were tested.

Dynamic Mechanical Analysis

The dynamic mechanical properties of the NFRPC were performed in a single-cantilever bending mode (DMA 8000, PerkinElmer, USA) at a heating rate of 2°C/min and a frequency of 1 Hz. The storage modulus (E') and loss tangent (Tan δ) were recorded over a range of temperature from -180°C to 130°C. The dimension of the sample was 30 mm × 12 mm with 4 mm thickness.

Scanning Electron Microscopy

Scanning electron microscopy (SEM) was used to examine the morphology of lignocelluloses and plastics in NFRPC. After the internal bond test, SEM images of the fracture surfaces of NFRPC were captured with a Hitachi scanning electron microscope (TM-1000, Japan) with an accelerating voltage of 15 kV. The samples were viewed perpendicular to the fractured surface.

Analysis of Variance

All results were expressed as mean \pm standard deviation (SD). The significance of difference was calculated by Scheffe's test; *P* values < 0.05 were considered significant.

RESULTS AND DISCUSSION

Effects of Aspect Ratio of Lignocellulose on the Physical and Mechanical Properties of NFRPC

It is well known that the dimensional characteristics play a dominant role in finding the suitability of any fibrous raw material (Ashori and Bahreini 2009). Thus, a better realization of the morphology of lignocellulose is necessary for developing NFRPC. The aspect ratios (length to diameter ratio) of the *M. micrantha* particle (MP) and fiber (MF) are shown in Table 1. Among all lignocelluloses, MF displayed the highest aspect ratio (26.23), while the value of MP ranged from 2.31 to 4.38, which gradually decreased with increasing mesh number. Table 1 also shows the physical and mechanical properties of NFRPC with varying aspect ratio. The density of all NFRPC ranged from 726 to 779 kg/m^3 , and there were no significant differences among them. After 24 h of water immersion, NFRPC made with -6+16 mesh MP and MF presented the lowest water absorption. Interestingly, NFRPC made with MF showed higher thickness swelling (5.1%) than all other NFRPC made with MP. Das et al. (2000) reported that the adsorbed water in the composite could reside in three main regions: the cell wall, the cell lumen, and the voids between the lignocellulose and polymer matrix. Of these, the water located at the cell wall would cause the most dimensional change. Accordingly, these results implied that the water in NFRPC made with MF was located mainly in the cell wall of lignocelluloses.

· ·		Fiber			
Properties	-6+16	-16+30	-30+60	-60	
	[4.38]*	[3.30]	[2.43]	[2.31]	[26.23]
Density (kg/m ³)	738 ^A	726 ^A	750 ^A	776 ^A	779 ^A
	(19)	(11)	(7)	(9)	(92)
Water absorption (%)	11.5 ^C	15.6 ^{AB}	16.3 ^A	15.7 ^{AB}	12.2 ^{BC}
	(0.6)	(1.3)	(2.0)	(2.6)	(1.4)
Thickness swelling (%)	2.3 ^B	2.5 ^B	2.5 ^B	2.0 ^B	5.1 ^A
	(0.3)	(0.3)	(0.4)	(0.1)	(0.5)
Wood screw holding strength (N)	623 ^{AB}	630 ^{AB}	589 ^{BC}	524 ^C	697 ^A
	(73)	(20)	(33)	(52)	(46)
Internal bond (MPa)	2.0 ^A	2.0 ^A	1.9 ^A	1.4 ^B	0.4 ^C
	(0.2)	(0.2)	(0.1)	(0.2)	(0.1)
MOR (MPa)	9.1 ^B	8.6 ^B	8.7 ^B	7.7 ^B	18.0 ^A
	(1.1)	(0.4)	(0.6)	(0.5)	(1.8)
MOE (GPa)	0.77 ^B	0.74 ^B	0.69 ^B	0.62 ^B	1.56 ^A
	(0.11)	(0.06)	(0.05)	(0.04)	(0.13)

Table 1. Effects of *M. micrantha* Morphology on the Physical and Mechanical

 Properties of NFRPC

* Aspect ratio of reinforcement (length/diameter).

Values within parentheses are SD.

Particle and fiber contents in NFRPC: 50 wt%.

Different superscript letters within a row indicate significant differences (n = 5, P < 0.05).

On the other hand, as can be seen in Table 1, NFRPC made with higher aspect ratio of lignocellulose, especially MF, resulted in a stronger wood screw holding strength. Similarly, NFRPC made with MF also showed the highest MOR (18.0 MPa) and MOE (1.56 GPa) among all NFRPC. These results are in good agreement with previous reports (Ashori and Nourbakhsh 2009; La Mantia and Morreale 2011) and reveal that the higher aspect ratio of the fibrous raw material provided superior strength and stiffness for composites (Bouafif *et al.* 2009; Migneault *et al.* 2008; Stark and Rowlands 2003). However, the internal bond, one of the important mechanical properties that provides direct information on the interfacial adhesion of the composites, showed an opposite trend to that of wood screw holding strength and flexural properties. The NFRPC made with MP showed a significantly stronger internal bond (ranged from 1.4 to 2.0 MPa) than that made with MF (0.4 MPa).

One possible reason for this behavior may be related to the MF associated with higher total surface area as compared to MP. The mechanical properties of NFRPC are not only influenced by the aspect ratio of the lignocellulose but also by interfacial adhesion and stress transfer between reinforcement and polymer matrix (Ashori and Nourbakhsh 2009). In other words, at the same loading content, composites manufactured with small particles or fibers decreased the wettability and interfacial adhesion, resulting in poor mechanical properties. The SEM micrographs of fracture surface of NFRPC are presented in Fig. 1. They clearly show that NFRPC was made with a larger particle size, and the particles were more uniformly distributed in the polymer matrix (Fig. 1A–C). In contrast, when small size MP (-60 mesh) or MF was used in NFRPC, the surface area of lignocelluloses increased.

It is possible to observe that the polymer matrix was not continuously distributed anymore and that the lignocelluloses may be in immediate contact with one another, resulting in numerous cavities (Fig. 1D) or incomplete inter-fiber bonding (Fig. 1E) in NFRPC.

The presence of insufficient matrix contact confirms that there was poor interfacial adhesion and stress transfer (Adhikary *et al.* 2008). Hence, the NFRPC made with -60 mesh MP and MF exhibited a weaker internal bond than others. Moreover, the vertical density profile of NFRPC may be the other reason for this occurrence. In general, composites with higher core density are associated with a superior internal bond (Wong *et al.* 1999).

The NFRPC made with MF displayed lower core density and a weaker internal bond than those made with MP (Fig. 2), and *vice versa*. Accordingly, the vertical density profile of NFRPC was responsible for the internal bond of the composites.

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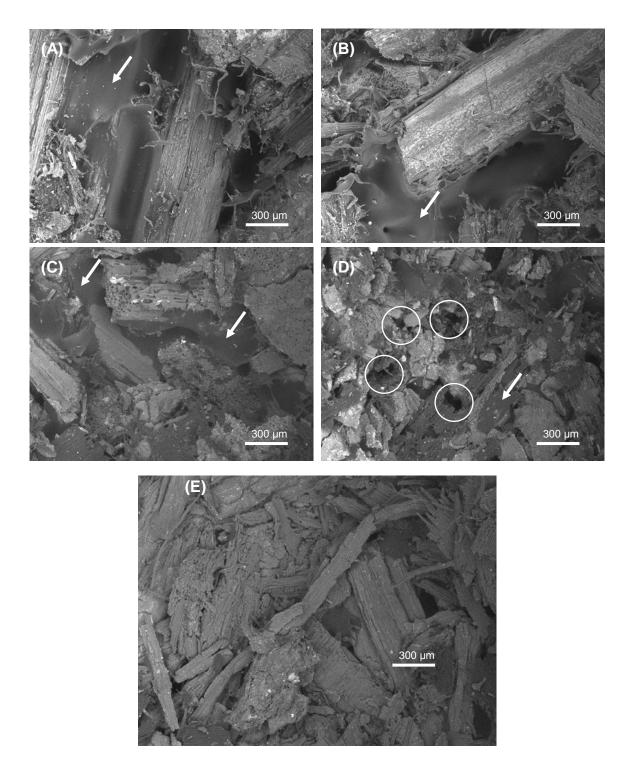


Fig. 1. SEM micrographs of the fracture surface of NFRPC made with various particle sizes. (A) -6+16 mesh; (B) -16+30 mesh; (C) -30+60 mesh; (D) -60 mesh; (E) fiber. Particle and fiber contents in NFRPC: 50 wt%. Arrows indicate rHDPE matrices; circles indicate cavities

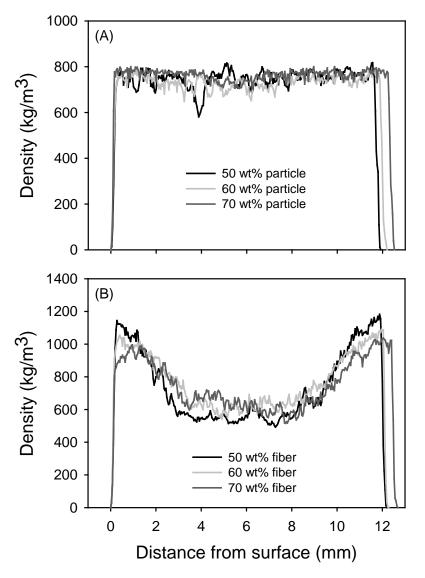


Fig. 2. Vertical density profiles of NFRPC made with various (A) particle (-6+16 mesh) and (B) fiber contents

Effects of Aspect Ratio of Lignocellulose on the Dynamic Mechanical Properties of NFRPC

The storage modulus (*E'*) should be used in assessing the molecular basis of mechanical properties of materials due to its sensitivity to structural changes, such as interfacial adhesion between the lignocellulose and the polymer matrix (Hong *et al.* 2008). Figure 3 shows the typical dynamic flexural storage modulus and loss tangent (tan δ) curves for neat rHDPE and NFRPC made with various particle sizes as a function of temperature. There were significant increases in the storage modulus of rHDPE matrix incorporating all sizes of MP at a temperature below the glass transition temperature (T_g)

of NFRPC (-117°C). This behavior is primarily attributed to the increased matrix stiffness with the reinforcing effect imparted by the MP. Furthermore, the NFRPC with higher aspect ratio showed stronger storage modulus (Fig. 3A). This result is in good agreement with a previous report (Ashori and Nourbakhsh 2009) that revealed that higher aspect ratio was associated with better stress transfer between the matrix and the fibers. Additionally, the stiffness of neat rHDPE was temperature-independent at temperatures below -20°C, while the stiffness of NFRPC dropped quickly. This drop of E' was assigned to the nature of lignocelluloses (Dufresne *et al.* 2003; Jebrane *et al.* 2011).

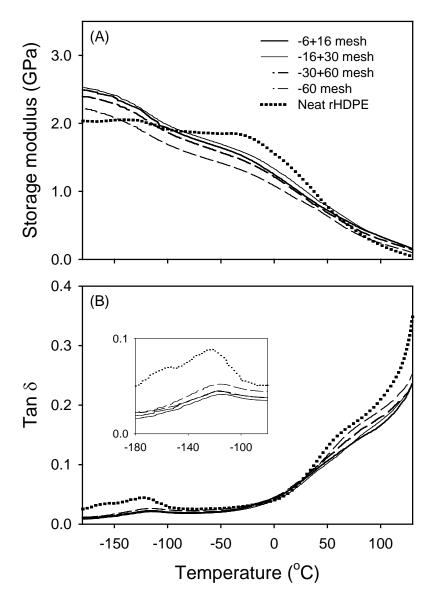


Fig. 3. Effects of *M. micrantha* morphology on (A) storage modulus and (B) Tan δ of NFRPC. Particle content in NFRPC: 50 wt%

Tan δ , a measure of material-related damping properties, is an indication of molecular motions existing in the materials. Molecular motion at the interface contributes to damping or energy dissipation of the composites (Hong *et al.* 2008). In other words, strong interaction between the lignocellulose and polymer matrix tends to decrease the mobility of the molecular chain at the interface, and hence reduces the tan δ values. Figure 3B shows the tan δ of neat rHDPE and NFRPC made with various MP sizes as a function of temperature. The result suggested the MP size was not associated with the tan δ value of NFRPC. However, all NFRPC displayed a decreased magnitude of damping peak compared to neat rHDPE. This was expected, because the lignocelluloses usually have superior elastic modulus to general plastics, which carry a greater extent of stress and allow only a small part of it to strain the interface, resulting in less energy dissipation at the interface between the lignocellulose and polymer matrix (Mohanty et al. 2006; Sombatsompop et al. 2003). Moreover, compared with the neat rHDPE, all T_g of NFRPC made with MP were shifted from -122°C to -117°C. This is primarily attributed to the presence of chemical impurities in the rHDPE matrix, which might cause polar-polar interactions between the rHDPE and MP molecules, resulting in the segmental immobilization of matrix chains at the fiber surface (Adhikary et al. 2008; Sombatsompop et al. 2003; Hong and Wool 2005).

Effects of Lignocellulose Content on the Mechanical Properties of NFRPC

The effect of lignocellulose content on the mechanical properties, such as wood screw holding strength, internal bond, MOR, and MOE, are given in Fig. 4. The neat rHPDE showed a higher wood screw holding strength than NFRPC made with MP or MF. In other words, the presence of lignocellulose in the polymer matrix resulted in decreased wood screw holding strength. Note, however, as the MP content increased, the wood screw holding strength increased slightly. In contrast, this mechanical property gradually decreased with increased MF content. Furthermore, the trend of internal bond of all NFRPC decreased when the MP or MF content increased from 50 to 70 wt%. This tendency may be attributable to decreased wettability, interfacial adhesion, and stress transfer between lignocellulose and polymer matrix with increasing fiber content. Ashori and Nourbakhsh (2009) reported that the impact strength of agro-residue-reinforced polypropylene composites decreased with increasing agro-residue content. Lin et al. (2002) also reported that the more lignocellulose content was utilized, the less polymer matrix content and insufficient bonded area existed in the composites, leading to a lower internal bond. On the other hand, as shown in Fig. 4A and 4B, it is also observed that all NFRPC made with MF exhibited an inferior internal bond compared to others made with MP at the same loading content. This tendency may be due to the effect of the vertical density profile of the NFRPC. In general, the vertical density profile of the composites produced by conventional hot pressing resembles a flat "V-shape" (Lee et al. 2010; Wong et al. 1999). As shown in Fig. 2B, the density profile of NFRPC made with MF resembled a typical V-shape. Oppositely, uniform density profiles, *i.e.* equal face and core density, were observed for all NFRPC made with MP (Fig. 2A). Wong et al. (1999) reported the composites exhibiting higher core density showed a stronger internal bond. Thus, compared to MF, all NFRPC made with MP displayed a higher core density and a stronger internal bond.

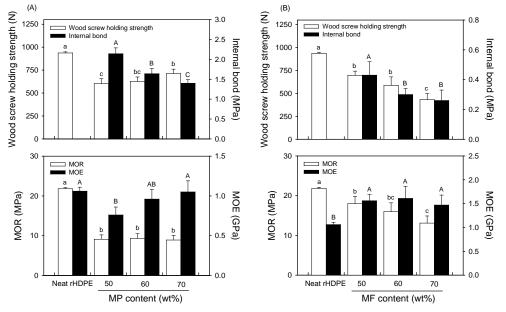


Fig. 4. Effects of *M. micrantha* (A) particle (-6+16 mesh) and (B) fiber contents on the mechanical properties of NFRPC. Results are mean \pm SD (*n* = 5). Bars with different letters indicate significant differences among groups (*P* < 0.05)

In this study, the presence of lignocellulose (MP or MF) in the polymer matrix decreased the flexural strength, but the changing trends differed between MP- and MFbased NFRPC. In the case of NFRPC made with MP, the addition of 50 wt% MP abruptly decreased the MOR from 21.8 MPa (neat rHDPE) to 9.4 MPa and then leveled off when more MP contents were introduced (Fig. 4). In contrast, the flexural strength of NFRPC made with MF gradually decreased from 21.8 MPa to 13.1 MPa at 70 wt% MF content. In general, plant fibers reinforce plastics because of their relatively high strength and stiffness (Suryanegara et al. 2009). Xue et al. (2007) reported the flexural strength of aspen fiber/PP composite increased in a parabolic curve with respect to the fiber content, with the maximum value occurring at 40 wt%. The mechanical properties of the composites are negatively affected by various factors, including poor fiber dispersion caused by intermolecular hydrogen bonding, and the wide polarity differences of the surfaces that retard the interaction of polymer/fiber bonding (Harper and Wolcott 2004; Maldas et al. 1989; Raj et al. 1989). The decline in MOR may be attributable to aggregation of the woody materials. Poor interfacial adhesion leads to composites with poor mechanical properties (Bengtsson et al. 2005; Bouza et al. 2008; Hung and Wu 2010; Mansour et al. 2008; Najafi et al. 2006). With the MOE, it is worth noting that the modulus of NFRPC made with 50 wt% MP was lower than that of neat rHDPE. In addition, the modulus gradually increased with increasing MP content. In contrast, all NFRPC made with MF showed superior modulus to neat rHDPE. This result revealed MF displayed a significant reinforcement effect without chemical additives. This is common in composites and usually results from better interfacial adhesion between the polymer matrix and reinforcements (Krouit et al. 2010).

Figure 5 shows the storage modulus and tan δ of neat rHDPE and NFRPC with various particle and fiber contents as a function of temperature. As shown in Fig. 5A, the

results revealed that the storage modulus increased with increasing particle content from 50 to 60 wt%, and then leveled off. In addition, the storage modulus of NFRPC made with MF displayed a similar tendency. The storage modulus increased significantly when 50 wt% MF was introduced into rHDPE, and then flattened out. These results were similar to the results obtained in the flexural test (Fig. 4) and agreed with the reports in the literature (Shah *et al.* 2008; Singh and Mohanty 2007). A possible explanation is that the reinforcement effect imparted by the particle and fiber allowed a greater degree of stress transfer at the interface between the polymer matrix and lignocellulose. Moreover, the decreased magnitude of damping peak and up-shift of T_g were also observed in the NFRPC made with MP and MF. However, with increasing lignocellulose content, there were no significant changes in both properties.

On the other hand, to realize the efficiency of *M. micrantha* (non-wood materials) as a natural reinforcement in plastic composites, sugi particle (SP) and sugi fiber (SF) were also utilized as reference controls (wood materials). The mechanical properties of NFRPC made with SP, SF, MP, and MF are shown in Table 2. Similar wood screw holding strength was found in all types of NFRPC. However, the flexural properties of NFRPC made with MP and MF displayed dissimilar trends to SP and SF. Lower strength and modulus were presented in the NFRPC made with both non-wood materials. The chemical compositions of *M. micrantha* and sugi show perceptible differences. *M. micrantha* displayed approximately 10% less holocellulose and α -cellulose contents than sugi (data not shown). The physicomechanical properties of NFRPC have been reported to contribute to the chemical composition characteristics of lignocelluloses (Mwaikambo and Ansell 2002). Accordingly, a different mechanical behavior can therefore be expected for wood and non-wood reinforcements.

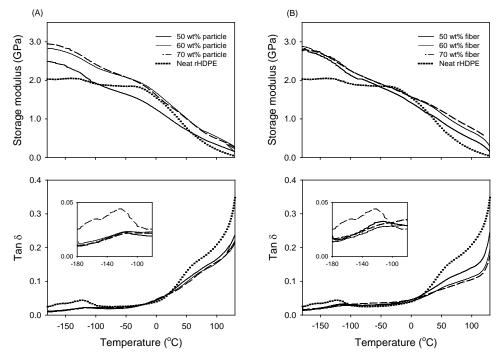


Fig. 5. Effects of *M. micrantha* (A) particle (-6+16 mesh) and (B) fiber contents on the storage modulus and tan δ of the NFRPC

	Neat rHDPE	Wood material		Non-wood material	
Properties		SP	SF	MP	MF
		[6.98]*	[6.91]	[4.38]	[26.23]
Density (kg/m ³)	796 ^A	743 ^A	749 ^A	738 ^A	779 ^A
	(5)	(47)	(56)	(19)	(92)
Wood screw holding strength (N)	937 ^A	815 ^{AB}	642 ^B	623 ^B	697 ^A
	(12)	(203)	(109)	(73)	(46)
Internal bond (MPa)	—	0.8 ^{BC}	1.3 ^B	2.0 ^A	0.4 ^C
		(0.2)	(0.5)	(0.2)	(0.1)
MOR (MPa)	21.8 ^A	18.6 ^{BC}	20.7 ^{AB}	9.1 ^D	18.0 [°]
	(1.1)	(1.4)	(0.9)	(1.1)	(1.8)
MOE (GPa)	1.06 ^C	1.47 ^B	1.98 ^A	0.77 ^D	1.56 ^B
	(0.05)	(0.09)	(0.18)	(0.11)	(0.13)

Table 2. Effects of Reinforcement Species on the Mechanical Properties of NFRPC

* Aspect ratio of reinforcement (length/diameter).

Values within parentheses are SD.

Particle (-6+16 mesh) and fiber contents in NFRPC: 50 wt%.

Different superscript letters within a row indicate significant differences (n = 5, P < 0.05).

CONCLUSIONS

M. micrantha particle and fiber were successfully introduced into NFRPC as reinforcements by the manufacturing method of forming and flat-pressing, which is similar to particleboard production. The results showed that *M. micrantha* particle and fiber were proper and attractive reinforcements in NFRPC. When incorporating 50 wt% *M. micrantha* fiber with rHDPE, the composites displayed excellent flexural stiffness. Compared with neat rHDPE, the NFRPC made with 50 wt% *M. micrantha* fiber improved the MOE by 47%. Furthermore, not only the wettability, interfacial adhesion, and stress transfer between lignocellulose and polymer matrix but also the vertical density profile of composites play a key role in mechanical properties. To our knowledge, this is the first report to address the properties of NFRPC made with *M. micrantha* particle or fiber. Despite *M. micrantha* showing slightly lower strength and stiffness in plastic composites compared to sugi (wood materials), both properties would be significantly improved by increasing aspect ratio value and loading content of *M. micrantha*. Accordingly, *M. micrantha*, a notorious eco-killer and lignocellulosic waste, shows great potential as a novel alternative reinforcement in plastic composites.

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