Natural Fiber-Reinforced Hybrid Polymer Nanocomposites: Effect of Fiber Mixing and Nanoclay on Physical, Mechanical, and Biodegradable Properties

Md. S. Islam,[a,*] Mansor B. Ahmad,[a] Mahbub Hasan,[b] Sidek Abdul Aziz [c], Mohammad Jawaid,[d] M. K. Mohamad Haafiz,[e] and Siti A. H. Zakaria [a]

Combining two kinds of fibers is a potential way to improve the essential properties of natural fiber-reinforced hybrid polymer composites. Biocomposites produced from natural resources are experiencing an increase in interest due to their high demand in the market for manufacturing, in addition to environmental and sustainability issues. In this study, natural fiber-reinforced hybrid polymer nanocomposites were prepared from coir fiber, wood fiber, polypropylene, and montmorillonite nanoclay using a hot press technique. The effects of fiber mixing and montmorillonite on their physico-mechanical and biodegradable properties were subsequently investigated. Before being used, both the wood and the coir fibers were alkali-treated to reduce their hydrophilicity. The mechanical properties of the fabricated composites were measured using a universal tensile testing machine and found to be enhanced after fiber mixing and nanoclay incorporation. Fourier transform infrared spectra indicated that the characteristic peaks of the composites shifted after fiber mixing. A new peak around 470 cm^{-1} was observed in the case of the nanocomposites, which confirmed the interaction between the fiber, polymer, and montmorillonite (MMT). Scanning electron microscopic analysis revealed that MMT strongly improved the adhesion and compatibility between the fiber and polymer matrix. The combining of fibers improved the biodegradability and water absorption properties, while MMT addition had the reverse effect on the same properties of the composites.

*Contact information: a: Department of Chemistry, Faculty of Science, Universiti Putra Malaysia, Serdang, Selangor, Malaysia; b: Department of Materials and Metallurgical Engineering, Bangladesh University of Engineering and Technology, Dhaka, Bangladesh; c: Department of Physics, Faculty of Science, Universiti Putra Malaysia, Serdang, Selangor, Malaysia; d: Laboratory of Biocomposite Technology, Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia; e: School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia; *Corresponding author: msaifuli2003@yahoo.com or msaifuli2007@gmail.com*

INTRODUCTION

Awareness of environment protection issues has recently encouraged research in the usage of agriculture residues, which are abundant and cause problems during handling. Agricultural crop residues are produced abundantly in billions of tons and are available at low cost; however, only a small portion of the residue is utilized as household fuel or fertilizer, whereas the major portion is burned in the field causing air and environmental pollution. Use of these residues as filler/fiber in polymeric materials may solve the problem. It will also add value to the agribusiness supply chain system. Fiber-

reinforced polymer composites have recently been applied in the construction industry to replace steel and concrete. Natural fibers are replacing synthetic fibers as reinforcement in polymer composites due to their non-toxicity, non-corrosiveness, high strength, low density, low cost, renewability, biodegradability and, therefore, positive impact on the environment (Khalil *et al.* 2008; Azwa *et al.* 2013). The use of biodegradable sources in polymer products may reduce carbon emissions during the burning of plastics (Sahari *et al.* 2013). In spite of the attractiveness, natural fiber-based composites suffer from lower modulus, lower strength, limited durability, and relatively poor moisture resistance compared to synthetic fiber-reinforced composites. In order to solve these problems, natural fibers can be mixed with a stronger synthetic or natural fiber in the polymer matrix. This will produce hybrid composites that take full advantage of the best properties of the constituents and thereby an optimal, superior, and economical composite can be obtained.

Natural fiber-reinforced hybrid polymer composites provide designers the freedom of tailoring composites at a low cost that cannot be attained in binary systems having one fiber/filler dispersed in the matrix (Thakur *et al.* 2014). Natural cellulosic fibers are renewable, non-abrasive, exhibit excellent mechanical properties and are environmentally friendly, which make them very popular in engineering markets such as the automotive and construction industries (Saw and Datta 2009; Thakur and Thakur 2014; Thakur *et al.* 2014). Among all natural fibers, wood and coir have the highest potential as hybrid polymer reinforcements all over the world. Wood fiber has gained enormous acceptance as a filler in polymers due to its low density, high stiffness, biodegradation, easy availability, renewability, and relatively low cost. Various types of wood-reinforced polymers have already been manufactured and commercialized (Hetzer and Kee 2008). The addition of wood fiber has improved polymer properties by enhancing its rigidity, stiffness, strength, hardness, and heat resistance (Boopalan *et al.* 2013). On the other hand, coir, which is extracted from the outer shell of coconuts, has the highest toughness amongst all natural fibers. Coir fiber is abundantly available in Malaysia, especially in the coastal areas, since the coir tree needs sandy soil for its growth. Coir fiber is moth-proof, resistant to fungi and rot, flame-retardant, tough, durable, and unaffected by moisture and dampness. It also has a high lignin content and low cellulose content and high microfibrillar angle that yield lower tensile strength and higher elongation compared to other lignocellulosic fibers (Satyanarayana *et al.* 1986). This chemical composition limits its reaction with polymer radicals produced during processing and the service life of the product (Thakur *et al.* 2014).

Nanotechnology is one of the promising approaches towards the development of advanced materials for future engineering applications. Polymer nanocomposites possess better properties as compared to the neat polymer without scarifying density, toughness, and processibility (Peter *et al.* 1999; Ray and Okamoto 2003). Nanocomposites are composed of particles having at least one dimension in the nanometer range (*i.e.,* 100 nm or less), which allows the particles to act more effectively as fillers in a matrix. Nanoclays are nanoparticles that consist of layered mineral silicates. The use of nanoclay as a reinforcing agent in a polymer depends strongly on its shape, particle size, aggregate size, surface characteristics, and degree of dispersion (Kumar and Singh 2007). Nanoclay has a high surface energy and large specific surface area that result in strong interfacial interaction between the polymer and the nanofiller and significantly enhance the polymer properties (Peter *et al.* 1999). Polymer nanocomposites reinforced with organic and inorganic nanoparticles have attracted much attention due to their outstanding

mechanical, optical, electrical, and flame retardancy properties (Dong *et al.* 1992; Kim *et al.* 2003).

Nanocomposites comprising of biopolymer/biopolymers and one or more inorganic nanomaterials is an emerging group of nanostructured hybrid materials. They represent the frontier between nanotechnology, material science, and life science (Fernandes *et al.* 2013). Efforts have been made for the development of nanocomposites with enhanced thermal, mechanical, and functional properties (Mohanty *et al.* 2000). In addition, biopolymer-based materials manufactured using green technology has shown biodegradability and biocompatibility in pharmaceutical, food packaging, and agriculture technologies (Majeed *et al.* 2013). A number of studies have been conducted on natural fiber-reinforced polymer composites hybridized with synthetic fiber (Samal *et al.* 2009; Rahman *et al.* 2012). Very little research, however, has been conducted on natural fiber hybrid polymer nanocomposites and their hybridization combined with natural fibers and montmorillonite nanoclay. Therefore, the main objective of the present research was to evaluate the effects of combining of fiber types and the further addition of montmorillonite (MMT) nanoclay on the physical, mechanical, and biodegradable properties of wood/coir/MMT hybrid polypropylene biocomposites.

MATERIALS AND METHODS

Materials

Wood fiber, coir fiber, and MMT nanoclay were used as reinforcement filler in the composite system. Wood fiber was processed and collected from the middle bark of Brown Rubber wood in a wood factory in Kelantan, Malaysia. Coir fiber was processed and collected from a farm in East Malaysia. Sodium hydroxide and polypropylene (PP) were obtained from Nature Works LLC (USA). The MMT nanomaterial (1.30TC) was purchased from Nanocor Inc. (Arlington Heights, IL, USA). It was organically modified with approximately 30 wt% octadecylamine, had a dry particle size of 16 to 22 mm, and was used as received.

Methods

Alkaline treatment of fiber

The dried raw wood and coir fibers (20 g fiber in 250 mL solution) were immersed in the 2% (w/v) NaOH solution in a beaker. The fibers were left in the treatment for about 5 h at room temperature without any stirring. The treated coir and wood fibers were thoroughly washed with distilled water until the fibers became neutral (pH = 7.0). Finally, the washed and treated fibers were air dried under direct sunlight for 7 d.

Preparation of hybrid nanocomposites

Hybrid nanocomposites were fabricated using a hot press machine. Dried fibers were chopped to an approximate length of 2 mm and mixed homogeneously with PP granules and MMT using a Brabender mixer machine (Brabender, Germany). The fibers and PP matrix were mixed at different weight fractions, as summarized in Table 1. The mixtures were heated to 170 °C for 15 min inside the hot press machine. The same steps were repeated with the addition of 2 parts per hundred (phr) MMT. The composites were cooled in open air for several minutes in order to prevent any shrinkage that might form

during the extraction of the composite. The design of mold fabrication and desired composite thickness were in accordance with the ASTM D638 (2007) and ASTM D790 (2007) standards.

Table 1. Formulation of Hybrid Nanocomposites

Types of Samples	Polypropylene (wt.%)	Coir fiber (wt.%)	Wood fiber (wt.%)	MMT (phr)
Wood/PP composite	70	0	30	0
Coir/PP composite	70	30	0	0
Wood/PP/MMT composite	70	0	30	2
Coir/PP/MMT composite	70	30	0	2
Wood/Coir/PP hybrid composite	70	15	15	0
Wood/Coir/PP/MMT hybrid nanocomposite	70	15	15	2

Tensile test

Mechanical properties were tested using an Instron universal testing machine (Model 4032, USA) following ASTM D638 (2007). The sample was clamped at both ends with one end fixed to the machine and the other end at the removable clamp. Loads were applied until the samples were broken. Five tests were conducted for each type of specimen and the average values are reported.

Fourier transform infrared (FTIR) spectroscopy

In order to investigate the interaction between the polymer and the fibers, ground powder samples were tested by FTIR (Shimadzu 81001 spectrophotometer; Japan). The FTIR spectra of the composites were recorded in the 4000 to 400 cm^{-1} region.

Scanning electron microscopy (SEM)

Surface morphology of the manufactured hybrid nanocomposites was studied under a scanning electron microscope (JSM-5510, JEOL Ltd.; Japan) operating at an accelerating voltage of 5 kV. Tensile fracture surface micrographs were taken to study the fracture mechanisms and interface adhesion of the composites. The composite samples were sputter-coated with platinum before SEM observation.

Water absorption test

Rectangular specimens with a dimension of 39 mm (L) x 10 mm (W) x 4.1 mm (T) were prepared from each set of composites. The specimens were dried for 1 h in an oven maintained at 80 °C and left in open air for cooling to a constant weight. The composite specimens were then immersed in distilled water for 10 d according to ASTM D570 (1999). The specimens were taken out of the water each day, wiped with tissue paper, reweighed, re-measured and immediately put back into the water. Water absorption values were calculated according to the given formula,

$$\text{Water absorption (\%)} = [(W_2 - W_1)/W_1] \times 100\% \qquad (1)$$

where W_2 is the specimen weight after soaking and W_1 is the specimen weight before soaking. Five tests were conducted for each type of specimen, and the average values were reported.

Biodegradability test

A soil burial test was employed on a laboratory scale to examine the biodegradability of the hybrid nanocomposites. Rectangular specimens with a dimension of 100 mm (L) × 150 mm (W) × 1 mm (T) were buried in the dark inside the Rengam series soil randomly at a depth of one foot from the top. The soil and samples were incubated for a total of three months at 26 °C. The moisture content was maintained at an optimal condition for microbial activity that is 40 to 50% of the maximum water holding capacity of the soil (Chandra and Renurustgi 1998). The pots were covered with plastic film for avoiding water evaporation from the soil surface. Biodegradation was estimated by monitoring weight changes as a function of burial time. The composites were removed from the soil after each 30 d, and the debris on the specimens was removed by water washing. The composite samples were dried in an oven at 100 to 105 °C for 24 h and weighed using an electronic balance.

RESULTS AND DISCUSSION

Fourier Transform Infrared (FTIR) Spectroscopy

In order to reduce the hydrophilicity of the fibers and to increase the compatibility between the fibers and PP matrix, the raw fibers were chemically treated with a 2% NaOH solution. The effect of alkali treatment on the fibers was analysed using FTIR testing. Figure 1 shows the FTIR spectra of the untreated and treated fibers. For the untreated fiber, there are some characteristic absorption bands in the 800 to 3400 cm^{-1} region, which may be due to O-H, C=O, C=C, and CH$_2$ stretching vibrations (Islam *et al.* 2012). On the other hand, all these characteristic peak intensities shifted towards higher or lower wave numbers after alkali treatment (Fig. 1b). It can also be observed that the intensity of -OH peaks was reduced after the alkali treatment, indicating that some of the hydroxyl groups were removed from the fiber. The OH groups were more prominent on the untreated fibers compared to the treated fibers, which are in the region of 3344 to 3342 cm^{-1}. It clear from Fig. 1 that carbonyl groups had disappeared from the peak around 1700 cm^{-1} after alkali treatment, and there was a reduction in the amide peak around 1500 cm^{-1}. The alkali treatment resulted in the decrease of the OH stretching vibration. The sodium hydroxide reacted with accessible OH groups of the fiber by the following reaction (Islam *et al.* 2012):

$$\text{Fiber-OH} + \text{NaOH} \longrightarrow \text{Fiber-} O^- Na^+ + H_2O + \text{impurities} \qquad (2)$$

Several researchers have reported that alkali-sensitive hydroxyl (OH) groups present among the molecules are broken down and that they react with water molecules (H-OH); byproducts move out from the fibre structure, thus decreasing the proportion of hemicelluloses, lignin, pectin, wax, and oil covering materials (Ray *et al.* 2001; Li *et al.* 2007; Mwaikambo *et al.* 2007; Kabir *et al.* 2012). It was noticed that hydrophilic

hydroxyl groups are reduced by alkali treatment and that such treatment increases the fibers moisture resistance property (John and Anandjiwala 2008).
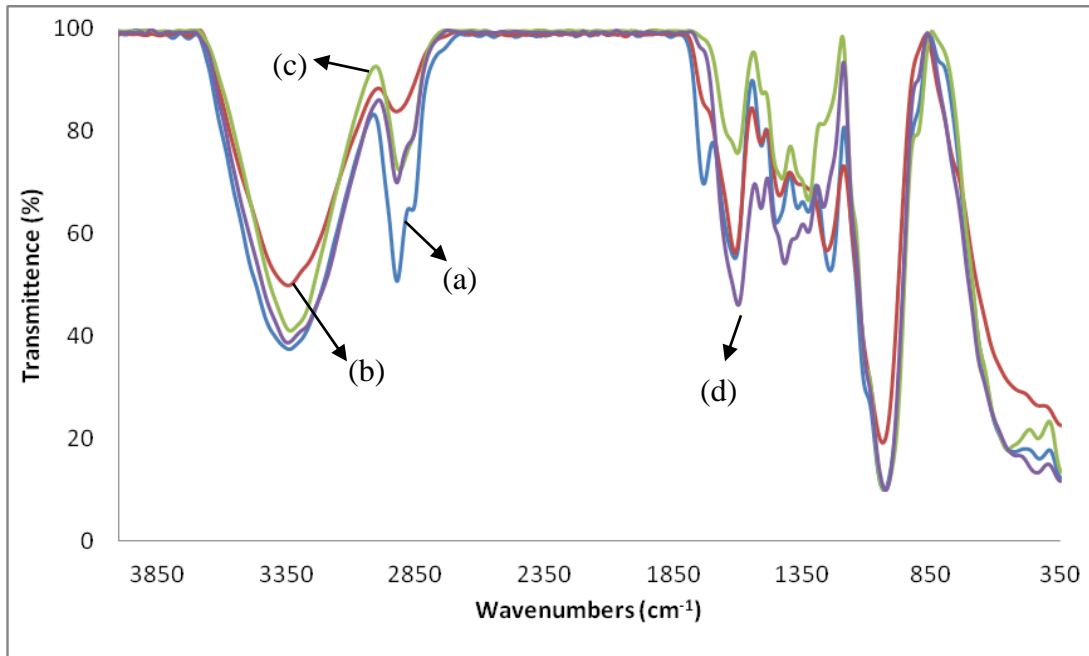
Fig. 1. FTIR spectrum of (a) untreated wood fiber, (b) untreated coir fiber, (c) treated wood fiber, and (d) treated coir fiber

Typical FTIR spectra of coir/PP, wood/PP, and coir/woood/PP composites are represented in Fig. 2.
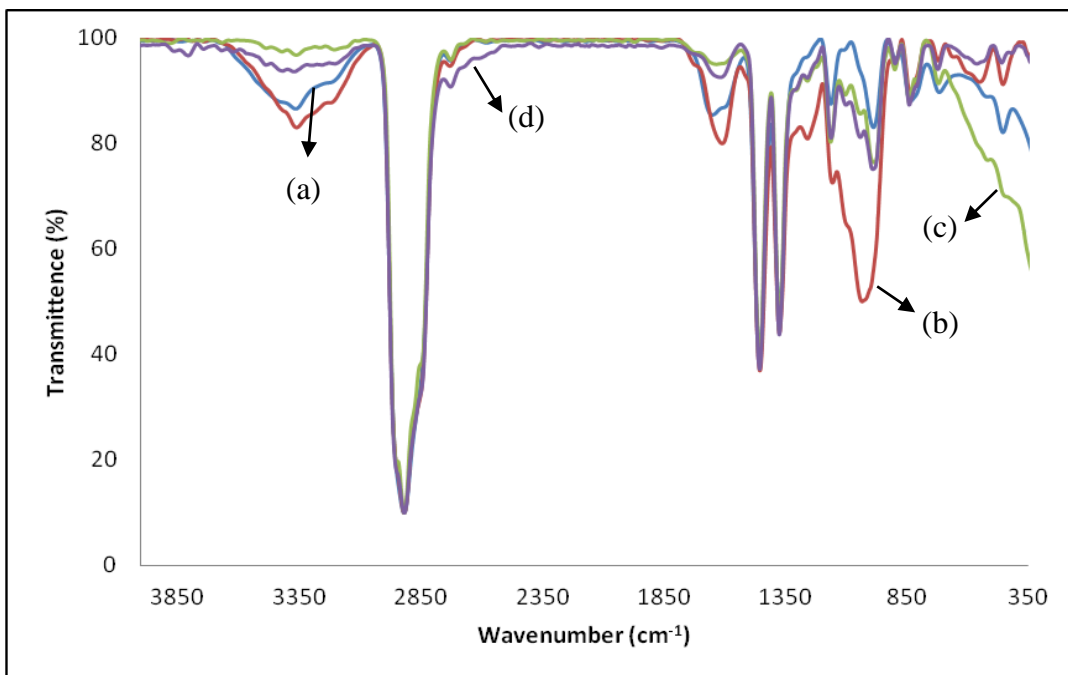
Fig. 2. FTIR Spectrum of (a) Pure PP, (b) Coir/ PP, (c) Wood/PP, and (d) Coir/ Wood/ PP

From the spectra, it can be seen that some new peaks appeared in the composites, which corresponds to the PP content. These peaks are due to CH, CH_2, and CH_3 and are found at 2916 cm^{-1} and 1454 cm^{-1} wavenumbers. All the peaks showed a similar variation of intensity because the PP content was the same for all composites. In the composites, -OH group peaks appeared in the region of 3400 cm^{-1}, which was due to the fiber content; however, the peaks of the wood fiber-reinforced PP composite had higher intensity as compared to the coir fiber reinforced PP composite. The increase in intensity of peaks was related to the hemicellulose content in the composite, as there was a higher content of particles, which were more hydrophilic compared to the other components of the composite. From Fig. 2, it is observed that all peaks of the hybrid composite (curve c) shifted toward the left, having higher wave numbers compared to their composites. Additionally, one new peak was apparent in the spectra at 3804 cm^{-1}. This peak may be due to the combining of fibers.

The effect of MMT on the hybrid nanocomposite was also investigated through FTIR testing, and the spectra are shown in Fig. 3. A peak at around 470 cm^{-1} was observed after blending with MMT. This peak is the characteristic peak of clay and is due to Si-O-Si stretch vibration (Ray and Okamoto 2003). The OH group was slightly shifted after MMT addition. These results indicate that the MMT strongly interated and bonded between the polymer and fiber in the hybrid nanocomposite (Majeed *et al.* 2013).
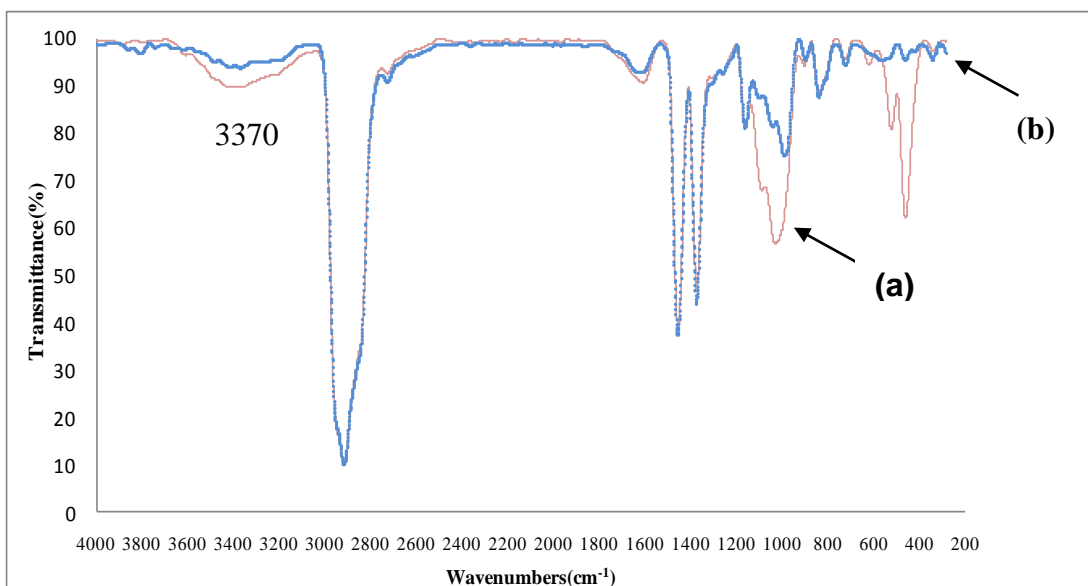

Fig. 3. FTIR spectra of the(a) coir/wood/PP/MMT hybrid nanocomposite and (b) coir/wood/PP hybrid composite

Scanning Electron Microscopy (SEM)

The surface morphology of the composite samples was investigated using scanning electron microscopy (SEM). Figure 4 shows the tensile fracture surfaces of the various composites and hybrid nanocomposites. From Figs. 4a and 4b, it can be seen that the wood/PP composite and coir/PP composites had the roughest surfaces among all the manufactured composites, resulting in low mechanical properites. The surface of the wood/coir/PP hybrid composite was also similar to the wood/PP and coir/PP composites, as shown in Fig. 4c. Thus, the interfacial adhesion between the fibers and matrix was not

strong enough to resist fiber pull-out during impact, as the energy consumed was small (Rosoff 2001). It can be observed from Fig. 4d that the hybrid nanocomposite exhibited a smoother surface than the single fiber composite. The addition of nanoclay in the hybrid composite resulted in high interficial adhesion and surface smoothness, which increased the mechanical properties of the hybrid nanocomposite (Smart *et al.* 2008).
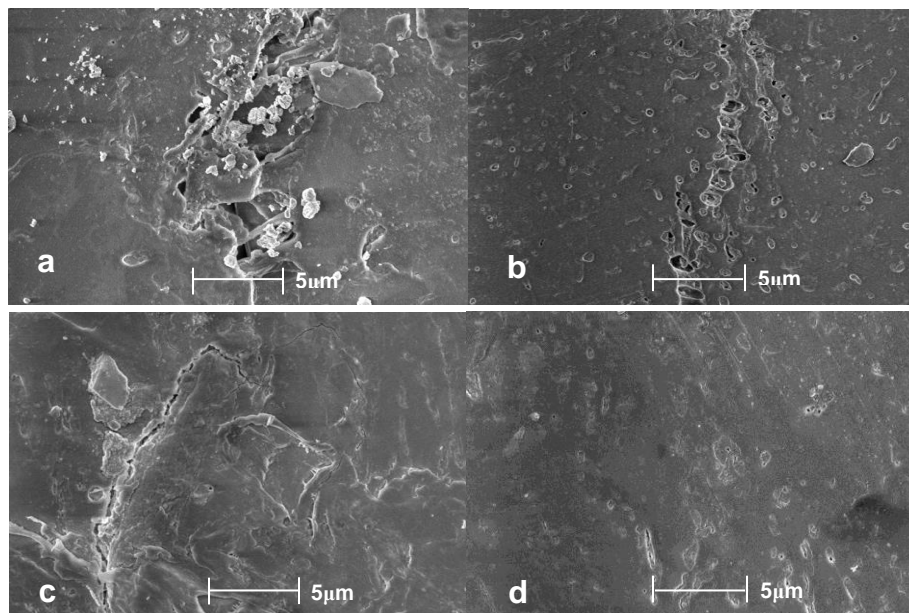

Fig. 4. SEM micrographs of the tensile fracture surfaces of the (a) wood/PP composite, (b) coir/PP composite, (c) wood/coir/PP hybrid composite, and (d) wood/coir/PP/MMT hybrid nanocomposite

Tensile Properties

　　　　Figures 5 and 6 show the tensile strength and tensile modulus of the various manufactured composites, respectively. The tensile strength and tensile modulus of the coir/PP composite and wood/PP composites were almost same, as is clear from Fig. 1. This may be due to similar cellulosic content in wood and coir fibres (Jawaid and Khalil 2011). Coir fiber is more hydrophilic than wood fiber due to higher hemi-cellulose content, whereas polypropylene is hydrophobic in nature. Thus, the hydrophilic coir/wood did not interact well with the hydrophobic polypropylene, resulting in lower tensile strength and tensile modulus. When combining coir fiber with wood fiber in PP and further addidition of MMT was carried out, then hybrid composites (coir/wood/PP and coir/wood/MMT/PP) displayed better tensile strength and modulus than wood/PP and coir/PP composites. It assumed that hybrid composites with coir/wood display better tensile properties due to strong fiber/matrix bonding compared with other formulations, leading to even and effective distribution of stress among fibers. Researchers have reported tensile properties of coir/oil palm/PP hybrid composites, and the results indicated that hybrid composites display better tensile properties than coir/PP and oil palm/PP composites (Zainudin *et al*. 2014). Furthermore, the hybrid nanocomposite exhibited the highest values of tensile strength and modulus after addition of MMT. The nanoclay enhanced interfacial interaction and adhesion between the fiber and the polymer matrix, thus improving the mechanical properties of the composites. Similar results were also reported by a previous work (Islam *et al.* 2011).
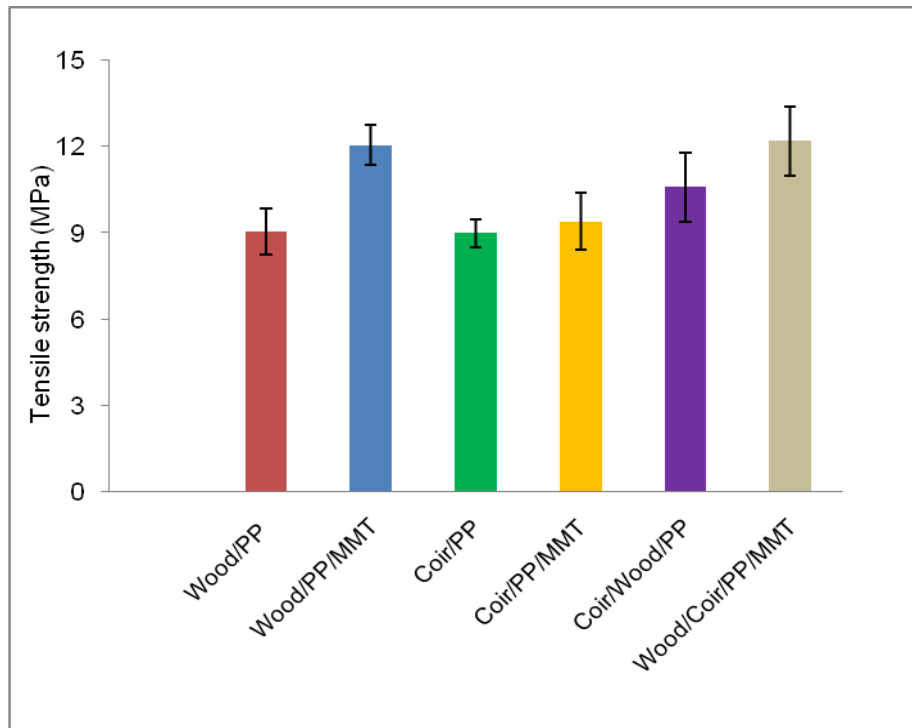
Fig. 5. Tensile strength of the various manufactured composites. Data reported as the average ± standard deviation
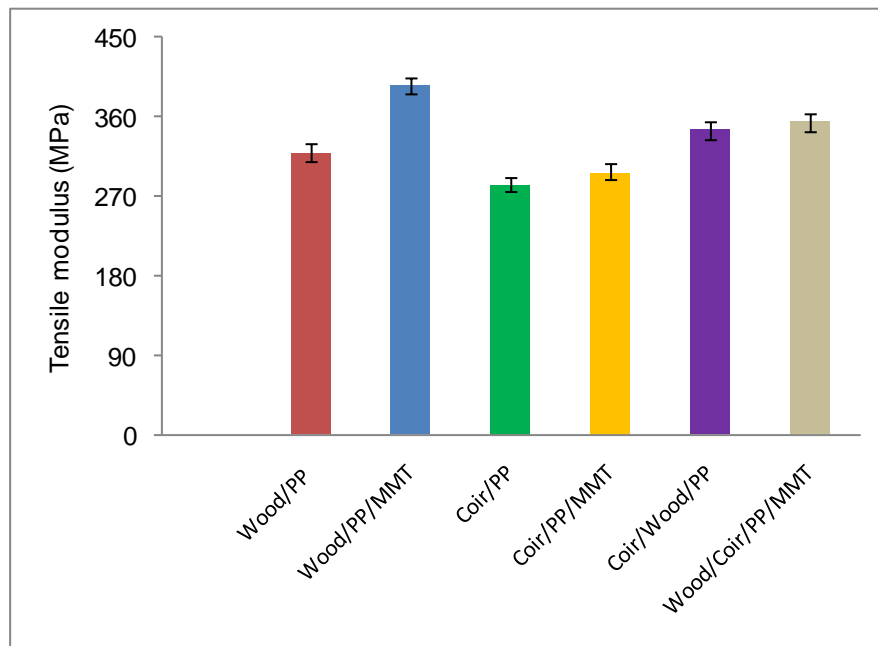
Fig. 6. Tensile modulus of the various manufactured composites. Data reported as the average ± standard deviation

Water Absorption Characteristics

The water absorption values of the various composites were determined and are presented in Fig. 7. It was observed that the initial water absorption rate for all the composites was rapid, whereas this rate became almost constant and slower in the final stage. It was also found that the water absorption for all the composites increased with time. The initial water absorption (evaluated after 1 day) was 7 to 13%, and it increased up to 15-22% for various manufactured composites. Among all the composites, the hybrid nanocomposites (wood/coir/PP/MMT) prepared with nanoclay exhibited the highest rate of water absorption. This may be due to the increased hydrophilic nature of the hybrid nanocomposite upon fiber mixing and addition of MMT. The coir/PP composite displayed the highest water absorption values compared to the other composites. This is due to the large number of –OH groups present in coir fiber surfaces. The number of hydroxyl groups and micro-voids increased in the coir/PP composites; thus the amount of water absorption increased a lot. On the other hand, the hybrid composite of wood and coir had the lowest amount of water absorption. The hybrid nanocomposite, however, had higher water absorption as compared to the hybrid composite due to the presence of hydrophilic MMT in the composite (Nadir *et al.* 2011).
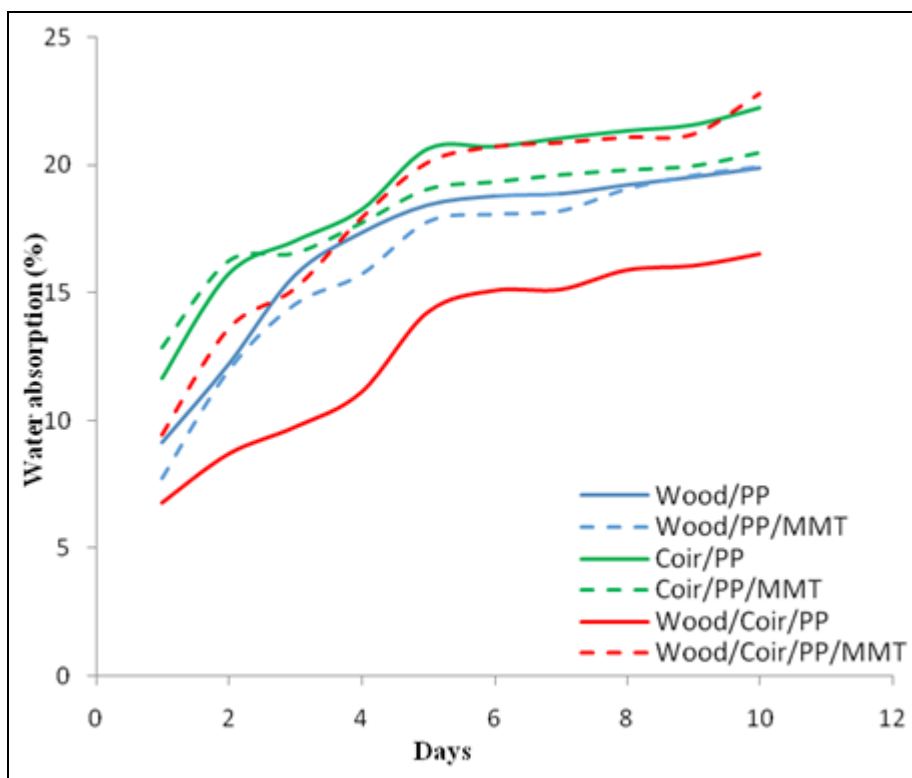

Fig. 7. Water absorption of the various manufactured composites

Biodegradability

Biodegradation of the composites caused by microorganisms was determined by weight loss after burying them in soil, and the results are shown in Fig. 8. The weight loss increased with an increase in degradation time (Wang *et al.* 2014). However, the weight

loss decreased when MMT was incorporated in the composites. The MMT had the ability to create better interaction and adhesion between the fiber and PP matrix, thus reducing the biodegradibilty of the hybrid nanocomposites. Furthermore, MMT nanoparticles were agglomerated due to the attractive force between the PP matrix and MMT, thus decreasing the biodegradation rate. The hybrid composite (wood/coir/PP) had a higher weight loss as compared to thesingle fiber/PP composite. The weight loss of the hybrid composite decreased, however,when MMT wasincorporated into the composite. This was expected and due to the differences in structure, the number of active hydroxyl groups and nanoparticles arrangment (Bledzki and Gassan 1999). Thus combining of fibers can increase the biodegradable properties of composites; however, MMT had the opposite effect on the same.
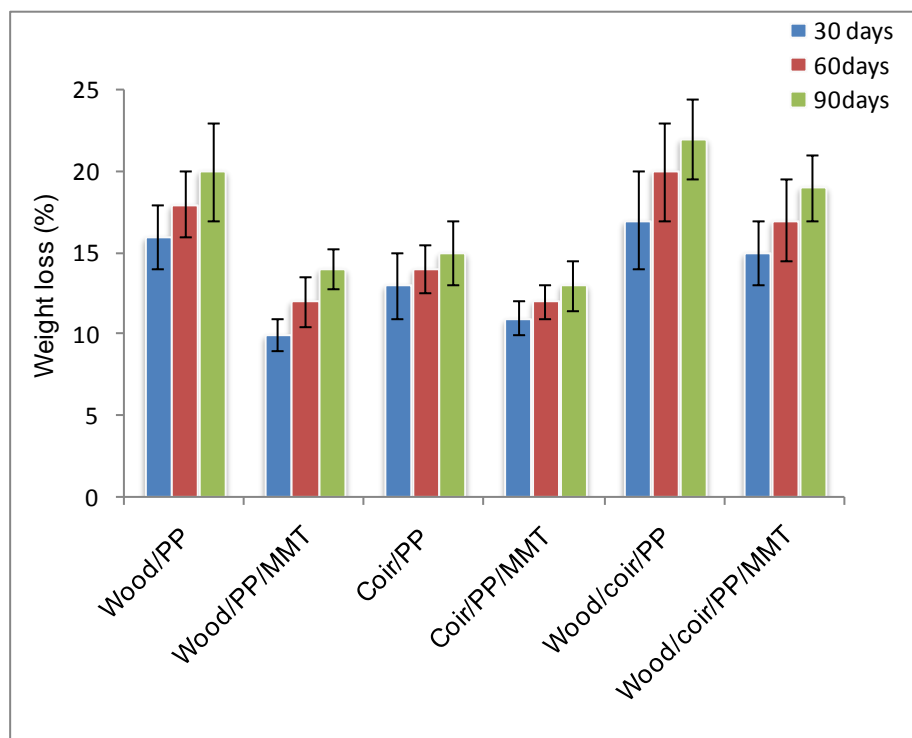
Fig. 8. Biodegradability of thevarious manufacturedcomposites. Data reported as the average ± standard deviation

CONCLUSIONS

1. Hybrid nanocomposites were successfully fabricated from wood fiber, coir fiber, PP, and MMT through a hot press method.

2. The FTIR and SEM results revealed that the structure and morphology of the composites were significantly changed after nanoclay incorporation. However, these results were not much affected when two fibers were used in combination.

3. The tensile strength and modulus were also found to be improved for hybrid nanocomposites. These improvements in composite properties can be ascribed to

good interaction between the fiber, polymer, and nanoclay and their support to the composites.

4. Nanoclay strongly improved the interfacial adhesion between the fibers and PP matrix, as observed by SEM analysis.

5. The biodegradability and water absorption characteristics of the hybrid nanocomposites were improved by the combined use of wood and coir fibers, while MMT had an opposite effect on those properties.

REFERENCES CITED

ASTM D570. (1999). "Standard test method for water absorption of plastics," *ASTM International*, West Conshohocken, PA.

ASTM D638. (2007). "Standard test method for tensile properties of plastics," *ASTM International*, West Conshohocken, PA.

ASTM D790. (2007). "Standard test method for flexural properties of unreinforced and reinforced plastics and electrical insulating materials," *ASTM International*, West Conshohocken, PA.

Azwa, Z. N., Yousif, B. F., Manalo, A. C., and Karunasena, A. (2013). "A review on the degradability of polymeric composites based on natural fibres," *Mater Design* 47, 424-442. DOI: 10.1016/j.matdes.2012.11.025

Bledzki, A. K, and Gassan, J. (1999). "Composites reinforced with cellulose-based fiber," *Progress Polym. Sci.* 24(2), 221-274. DOI: 10.1016/S0079-6700(98)00018-5

Boopalan, M., Niranjanaa, M., and Umapathy, M. J. (2013). "Study on the mechanical properties and thermal properties of jute and banana fiber-reinforced epoxy hybrid composites," *Compos B* 51, 54-57. DOI: 10.1016/j.compositesb.2013.02.033

Chandra, R., and Renurustgi. (1998). "Biodegradable polymers," *Progress Polym. Sci.* 23(7), 1273-1335. DOI: 10.1016/S0079-6700(97)00039-7

Dong, S., Sapieha, S., and Schreiber, H. P. (1992). "Rheological properties of corona modified cellulose/polyethylene composites," *Polym. Eng. Sci.* 32(22), 1734-1739. DOI: 10.1002/pen.760322212

Fernandes, E. M., Pires, R. A., Mano, J. F., and Reis, R. L. (2013). "Bionanocomposites from lignocellulosic resources: Properties, applications and future trends for their use in the biomedical field," *Progress Polym. Sci*.38(10-11), 1415-1441.

Hetzer, M., and Kee, D. D. (2008). "Wood/polymer/nanoclay composites, environmentally friendly sustainable technology: A review," *Chem. Eng. Res. Design* 86(10), 1083-1093. DOI: 10.1016/j.cherd.2008.05.003

Islam, M. S., Hamdan, S., Jusoh, I., Rahman, M. R., and Talib, Z. A. (2011). "Dimensional stability and dynamic Young's Modulus of tropical light hardwood chemically treated with MMA in combination with HMDIC crosslinker," *Ind. Eng. Chem. Res.* 50(7), 3900-3906. DOI: 10.1021/ie1021859

Islam, M. S., Hamdan, S., Jusoh, I., Rahman, M. R., and Ahmed, A. S. (2012). "The effect of alkali pretreatment on mechanical and morphological properties of tropical wood polymer composites," *Mater. Design.* 33, 419-424. DOI:10.1016/j.matdes.2011.04.044

Jawaid, M., and Khalil, A. H. P. S. (2011). "Cellulosic/synthetic fibre reinforced polymer hybrid composites: A review." *Carbohyd. Polym.*86, 1-18. DOI:10.1016/j.carbpol.2011.04.043.

John, M. J., and Anandjiwala, R. D. (2008). "Recent developments in chemical modification and characterization of natural fibre-reinforced composites," *Polym. Compos.* 29 (2), 187-207. DOI: 10.1002/pc.20461.

Kabir, M. M., Wang, H., Lau, K. T., and Cardona, F. (2012). "Chemical treatments on plant-based natural fibre reinforced polymer composites: An overview," *Compos B* 47(3), 2883-2892. DOI:10.1016/j.compositesb.2012.04.053

Khalil, A. H. P. S., Firdaus, M. Y. N., Anis, M., and Ridzuan, R. (2008). "The effect of storage time and humidity on mechanical and physical properties of medium density fiberboard (MDF) from oil palm empty fruit bunch and rubberwood," *Polym. Plast. Technol. Eng.* 47(10), 1046-1053. DOI: 10.1080/03602550802355644

Kim, T. H., Lim, S. T., Lee, C. H., Choi, H. J., and Jhon, M. S. (2003). "Preparation and rheological characterization of intercalated polystyrene/organophilic montmorillonite nanocomposite," *J. Appl. Polym. Sci.* 87(13), 2106-2112.DOI: 10.1002/app.11481

Kumar, A. P., and Singh, R. P. (2007). "Novel hybrid of clay, cellulose, and thermoplastics. I. Preparation and characterization of composites of ethylene-propylene copolymer," *J. Appl. Polym. Sci.* 104(4), 2672-2682.DOI: 10.1002/app.25659

Li, X., Tabil, L. G., and Panigrahi, S. (2007). "Chemical treatment of natural fibre for use in natural fibre-reinforced composites: A review," *J. Polym. Environ*. 15 (1), 25-33. DOI 10.1007/s10924-006-0042-3.

Majeed, K., Jawaid, M., Hassan, A., Bakar, A.A., Khalil, A. H. P. S., Salema, A.A., and Inuwa, I. (2013). "Potential materials for food packaging from nanoclay/natural fibres filled hybrid composites," *Mater Design* 46,391-410. DOI:10.1016/j.matdes.2012.10.044

Mohanty, A. K., Misra, M., and Hinrichsen, G. (2000). "Biofibres, biodegradable polymers and biocomposites: An overview," *Macromol. Mater. Eng.* 276-277(1), 1-24.DOI: 10.1002.

Mwaikambo, L.Y., Tucker, N., Clark, A. J. (2007). "Mechanical properties of hemp fibre reinforced euphorbia composites," *Macromol. Mater. Eng.* 292 (9), 993-1000. DOI: 10.1002/mame.200700092.

Nadir, A., Jarusombuti, S, Fueangvivat, V., Bauchongkol, P., and White, R. H. (2011). "Coir fiber reinforced polypropylene composite panel for automotive interior applications," *Fibers Polym.* 12(7), 919-926. DOI: 0.1007/s12221-011-0919-1

Peter, C., Baron, L., Wang, Z., and Pinnavaia, T. J. (1999). "Polymer-layered silicate nanocomposites: An overview," *Appl. Clay Sci.* 15(1-2), 11-29. DOI: 10.1016/S0169-1317(99)00017-4

Rahman, N. A., Hassan, A., Yahya, R., Araga, R. A. L., and Hornsby, P. R. (2012). "Polypropylene/glass fiber/nanoclay hybrid composites: Morphological, thermal, dynamic mechanical and impact behaviors," *J. Reinf. Plast. Compos.* 31(18), 1247-1257. DOI: 10.1177/0731684412456445

Ray, S. S., and Okamoto, M. (2003). "Polymer/layered silicate nano-composites: A review from preparation to processing," *Progress Polym. Sci.* 28(11), 1539-1641. DOI:10.1016/j.progpolymsci.2003.08.002.

Ray, D., Sarkar, B. K., Rana, A. K., and Bose, N. R. (2001). "Effect of alkali treated jute fibres on composite properties," *Bull. Mater. Sci*. 24 (2), 129-135.

Rosoff, M. (2001). *Nano-Surface Chemistry*, Marcel Dekker Inc., New York.

Sahari, J., Sapuan, S. M., Zainudin, E. S., and Maleque, M. A. (2013). "Mechanical and thermal properties of environmentally friendly composites derived from sugar palm tree," *Mater Design* 49, 285-289. DOI: 10.1016/j.matdes.2013.01.048

Samal, S. K., Mohanty, S., and Nayak, S. K. (2009). "Polypropylene-bamboo/glass fiber hybrid composites: Fabrication and analysis of mechanical, morphological, thermal, and dynamic mechanical behavior," *J. Plast. Compos.* 28(22), 2729-2747. DOI: 10.1177/0731684408093451

Satyanarayana, K. G., Sukumaran, K., Kulkarni, A. G., Pillai, S. G. K., and Rohatgi, P. K. (1986). "Fabrication and properties of natural fibre-reinforced polyester composites," *Compos.* 17(4), 329-333. DOI: 10.1016/0010-4361(86)90750-0

Thakur, V. K., and Thakur, M. K. (2014). "Processing and characterization of natural cellulose fibers/thermoset polymer composites," *Carbohyd. Polym.* 109, 102-117. DOI:10.1016/j.carbpol.2014.03.039

Thakur, V. K., Thakur, M. K., and Gupta, R. K. (2014). "Review: Raw natural fiber-based polymer composites," *Int. J. Polym. Analysis Characterization* 19(3), 256-271. DOI: 10.1080/1023666X.2014.880016

Thakur, V. K., Thakur, M. K., Raghavan, P., and Kessler, M. R. (2014). "Progress in green polymer composites from lignin for multifunctional applications: A review," *ACS Sustainable Chem. Eng.* 2(5), 1072-1092. DOI: 10.1021/sc500087z

Saw, S. K., and Datta, C. (2009). "Thermomechanical properties of jute/bagasse hybrid fibre reinforced epoxy thermoset composites," *BioResources* 4(4), 1455-1476. DOI: 10.15376/biores.4.4.1455-1475

Smart, G., Kandola, B. K., Horrocks, A. R., Nazare, S., and Marney, D. (2008). "Polypropylene fibers containing dispersed clays having improved fire performance. Part II: Characterization of fibers and fabrics from PP-nanoclay blends," *Polym. Adv. Technol.* 19(6), 658-670. DOI: 10.1002/pat.1137

Wang, X., Sun, H., Bai, H., and Zhang, L. P. (2014). "Thermal, mechanical, and degradation properties of nanocomposites prepared using lignin-cellulose nanofibers and poly(lactic acid)," *BioResources* 9(2), 3211-3224. DOI:10.15376/biores.9.2.3211-3224

Zainudin, E. S., Yan, L. H., Haniffah, W. H., Jawaid, M., and Alothman, O. Y. (2014). "Effect of coir fibre loading on mechanical and morphological properties of oil palm fibres reinforced polypropylene composites," *Polym. Compos.* 35(7), 1418-1425. DOI: 10.1002/pc.22794