Structural characterization and mechanical properties of polypropylene reinforced natural fibers

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Abstract. Recently the development of natural fiber composite instead of synthetics fiber has lead to eco-friendly product manufacturing to meet various applications in the field of automotive, construction and manufacturing. The use of natural fibers offer an alternative to the reinforcing fibers because of their good mechanical properties, low density, renewability, and biodegradability. In this present research, the effects of maleic anhydride polypropylene (MAPP) on the mechanical properties and material characterization behaviour of kenaf fiber and coir fiber reinforced polypropylene were investigated. Different fractions of composites with 10wt%, 20wt% and 30wt% fiber content were prepared by using brabender mixer at 190 °C. The 3wt% MAPP was added during the mixing. The composites were subsequently molded with injection molding to prepare the test specimens. The mechanical properties of the samples were investigated according to ISO 527 to determine the tensile strength and modulus. These results were also confirmed by the SEM machine observations of fracture surface of composites and FTIR analysis of the chemical structure. As the results, the presence of MAPP helps increasing the mechanical properties of both fibers and 30wt% kenaf fiber with 3wt% MAPP gives the best result compare to others.

1. Introduction
Over the last few decades, there is a growing trend in using natural fibers as reinforcement for polymeric composites due to their attractive properties such as lightweight, renewability, low density, high specific strength, non-abrasively, combustibility, non toxicity, low cost and biodegradability [1]. Natural fibers such as abaca, jute, kenaf, coir, ijuk have all been proved to be good reinforcement in thermoset and thermoplastics matrices [2-5]. These days, the use of natural fibers reinforced composites is well known in various applications such as automotives components, building materials, and the aerospace industry due to the economical and environmental concerns over traditional
inorganic reinforcements and fillers [6,7]. Figure 1 presents the different components made of lignocellulosic reinforced polymer composite in a Mercedes Benz sedan [8].

Nowadays, the global plastic production and consumption is rising progressively. The increased demand shows that the usage is important and also an integral part of our life [9]. A vigorous effort to explore more environmentally friendly, sustainable materials to change the current glass fiber and carbon fiber composites are required, hence able to reduce the reliance on petroleum based fuel and products [10]. Presently, due to high stiffness and strength properties of synthetic fibers like glass, carbon and aramid, thus it is being used extensively in polymer-based composites. Nevertheless, these fibers have serious downsides in terms of their biodegradability, initial processing costs, recyclability, energy consumption, machine abrasion, health hazards, and others [11]. Therefore, the interest has recently moved to the fabrication and properties of natural fiber reinforced materials.

![Image](image.jpg)

**Figure 1.** Automobile components made of natural fiber reinforced composites [8].

Despite of the advantage of natural fibers, the use of natural fibers as reinforcement agents, particularly in hydrophobic polymeric matrices, also have several drawbacks such as poor wettability, incompatibility with some polymeric matrices and high moisture absorption by the fibers because of the hydrophilic nature of natural fibers [12]. Recently, there are several chemical treatment methods have been studied to improve the interfacial bonding such as acetylation, mercerization, isocyanate treatment, acrylation, silent treatment and others. Among these methods, mercerization or alkaline treatment may be considered to be the most economical technique [13]. The surface of fibers becomes rough and enhances mechanical interlocking with resin after the chemical treatments which lead to decrease in moisture absorption as well as improving the wettability of fibers by matrix. This results in imparting better mechanical properties to the composite [14].

The compatibility between the fibers and matrix also can be enhanced by modifying the matrix with a compatibilizers that adheres well to both the fibers and matrix. One of the most suitable compatibilizers available for use in natural fiber reinforced polypropylene composites is Maleic anhydride polypropylene (MAPP). The presence of MAPP in natural fibers reinforced PP composites acts as a bridge between the non-polar polypropylene matrix and the polar fibres by chemically bonding with the cellulose fibers through the MA groups, and bonding to the matrix by means of polymer chain entanglement as shown in Figures 2 [15,16].

In this investigation, a study was performed to determine the effect of Maleic Anhydride Polypropylene (MAPP) on the mechanical properties and material characterization behavior of kenaf fiber/coir fiber composites. Composites were developed from polypropylene, kenaf fiber (KF) and coir fiber (CF) (which was treated with NaOH), and MAPP by using injection molding process.
2. Experimental

2.1 Material
Thermoplastic polypropylene (PP) was obtained from Lotte Chemical Titan (M) Sdn Bhd (Malaysia) in the form of copolymer pellets as the matrix material. For the reinforcement, kenaf fiber (KF) was obtained from Institut Penyelidikan dan Kemajuan Pertanian Malaysia (MARDI) in the form of coarse fibers from their core and the coconut coir fiber (CF) was received from Nature At Work Sdn Bhd. The maleic anhydride polypropylene, MAPP (Orevac CA100) as a compatibilizer was supplied from Syarikat Saintifik Bersatu (M) Sdn Bhd.

2.2 Fiber Preparation
The natural fiber of KF and CF were immersed in 5% of NaOH water solution at room temperature for 24 h before washing it with running water until water used for washing the fiber no longer gave any alkalinity reaction as shown in Figures 3a [17]. The washed fibers were then dried in an open air for one day before drying it finally in an oven for another 24 h at 80°C as shown in Figures 3b [18,19]. The treated fibers were then milled by rotor mill machine to get fine sizes random fibers by using ring sieves 1 mm, 0.5 mm and 0.2 mm at 12 rpm as shown in Figures 3c. The milled fibers were sieved for 30 min using a sieve with 200 μm mesh size as shown in Figures 3d [20].

![Figure 3. Fiber preparation process for kenaf fiber and coir fiber.](image_url)
2.3 Composite Fabrication
Mixing of fibers and polymer were prepared with few sets of different composition of 0 %, 10 %, 20 %, and 30 % of the fibers in the composites. The MAPP (3wt%) was added during mixing as a compatibilizer as shown as in Table 1. The fibers and polypropylene were mixed by using twin screw Brabender Plastograph machine at 190 °C melt temperature setting and at a speed of 20 rpm. Then, the compound was crushed by using granulator machine to make a form of pellet shape. The test samples were prepared by using horizontal screw type injection molding (Nissei Plastic Industrial Co. Ltd, model NP7-1F).

| Sample      | Composite | Polypropylene | Fibers | MAPP  |
|-------------|-----------|---------------|--------|-------|
| Pure        | 1         | 100           | 0      | 0     |
| Unmodified  | 2         | 90            | 10     | 0     |
|             | 3         | 80            | 20     | 0     |
|             | 4         | 70            | 30     | 0     |
| Modified    | 5         | 87            | 10     | 3     |
|             | 6         | 77            | 20     | 3     |
|             | 7         | 67            | 30     | 3     |

2.4 Characterization of composites

2.4.1 Scanning electron microscope
The morphology of the KF and CF reinforced PP composites with and without MAPP were investigated by using scanning electron microscope SEM (Hitachi, Japan) at 10 kV. The fractured surfaces of tensile test samples were studied with SEM after being sputter coated with gold.

2.4.2 Fourier Transform Infrared Spectroscopy
The FTIR spectra of composites with and without MAPP were recorded in the range of 4000 to 600 cm⁻¹ at a resolution of 4 cm⁻¹ with 32 scans using an FTIR machine Model Perkin-Elmer Spectrum 100 FT-IR Spectrometer (Perkin-Elmer, Norwalk, CT, USA).

2.4.3 Mechanical Properties
Tensile test was conducted according to ISO 527 at a test speed 5 mm/min by using Shimadzu Universal Testing Machine (Model AG-1 10kN, Japan) for both natural fiber composites with and without compatibilizer [21]. An average of five tests was reported.

3. Result and Discussion

3.1 Morphology Analysis
The morphology of composites were studied the effect of the MAPP on the interface between coir fibers and kenaf fibers reinforced polypropylene with different composition were investigated by referring the fracture surface of the tensile specimens. For unmodified composition, it shows there were poor fiber matrix adhesion which is justified by the presence of gaps between both of the fibers and the matrix and fiber pull-outs as shown in Figures 4. Meanwhile, the presence of the MAPP compatibilizer gives a better improvement of the wettability of both fibers surface by the polymer. The gaps were less distinctly for both composites which is indicating good adhesion and better wettability between the fibers and matrix as shown in Figures 5. The good dispersion during blending avoids the unoccupied area; hence it was reflected by giving the good mechanical properties compared to composites without compatibilizer [9].
Figure 4. Image of fractured surface of composites with (a) unmodified 10% coir fiber (b) unmodified 20% coir fiber (c) unmodified 30% coir fiber (d) unmodified 10% kenaf fiber (e) unmodified 20% kenaf fiber, and (f) unmodified 30% kenaf fiber.

Figure 5. Image of fractured surface of composites with (a) modified 10% coir fiber (b) modified 20% coir fiber (c) modified 30% coir fiber (d) modified 10% kenaf fiber (e) modified 20% kenaf fiber, and (f) modified 30% kenaf fiber.

3.2 FTIR Analysis
The FTIR spectra of unmodified and modified CF and KF were shown in Figure 6 and Figure 7. In Figure 6, the spectrum of the CF fibers are dominated by the peak at 2918 cm\(^{-1}\). The peak at around 2900 cm\(^{-1}\) is due to the C-H asymmetric and symmetric stretching from aliphatic saturated compounds which is corresponding to the aliphatic moieties in cellulose and hemicellulose [22]. On the other hand, the peaks in the region 1455 cm\(^{-1}\) are due to C-H groups from various lignin. The intensity of the
peak at 1376 cm$^{-1}$ is sharply reduced after the presence of MAPP which is indicating C-H groups in methyl and phenolic alcohol and results from bending vibration in the molecule. Apart from that, the peaks found at 1261 cm$^{-1}$, 1100 cm$^{-1}$ and 807 cm$^{-1}$ for unmodified CF seems disappear at peak for modified CF. Based on the FTIR spectra, a new peak was found at 1729 cm$^{-1}$ for modified CF. This peak shows the evidence of the presence of ester linkage (C–O) between CF and MAPP.

Figure 7 shows the FTIR spectra of unmodified and modified KF. The peaks at 2917 cm$^{-1}$ is assigned to C-H stretching vibrations. Also, the absorption band of 1455 cm$^{-1}$ and 1376 cm$^{-1}$ were represented to –CH2 and –CH3 bending vibration. A new peak also have been found at 1737 cm$^{-1}$ for modified KF. This result expressed that the MA functional groups of MAPP can be reacted with the hydroxyl groups of the kenaf fiber to produce covalent bonding and esterification reaction [23].

It can be summarized that FTIR only observed the possibility of interaction, functional group change on rigidity imposed by composite formation. Moreover, the treatments with alkali and the addition of MAPP had removed most of the lignin and hemicelluloses components from the fiber surface [24]. Furthermore, the H-bonding shift and differences in the intensities of the absorptions be the main factor that contribute to the mechanical properties of the natural fiber composites.

3.3 Mechanical Properties Analysis
The mechanical properties of natural fiber reinforced polymer composites are one of major importance for all applications. Figure 8 shows the effect of MAPP on the tensile strength and tensile modulus of PP/CF composites. Obviously, the tensile strength of the CF composites modified with MAPP increased as the fiber content increased compared to unmodified CF composites, in which were found decrease with the increased fiber content. The incorporations of MAPP was enhanced the tensile strength of CF composites, which may be due to a good compatibility at interfacial regions between natural fiber and matrix as shown in Figure 8a [25].

The tensile modulus of PP/CF composites were illustrated in Figure 8b. It were observed that tensile modulus of PP/CF modified with MAPP for 30% fiber content found to be 12 % and 94 % better compared to unmodified CF at 30% fiber content and neat PP. The results indicate that the MAPP was able to enhance the stiffness of the composites. It may be due to the anhydride groups can react with the hydroxyl groups of CF and form ester bonds, which improves the interfacial adhesion between the matrix and fillers [26].
Figure 8. (a) Tensile strength of PP/Unmodified CF and PP/Modified CF (b) Tensile modulus of PP/Unmodified CF and PP/Modified CF.

Figure 9a shows the influence of MAPP on the tensile strength of PP/KF composites. The tensile strength of PP/KF composites modified with MAPP increased with the increased of fiber fraction compared to unmodified PP/KF. The improvement in tensile strength may be attributed to the hydrophilic anhydride groups of MAPP, which was promoting the dispersion and surface wetting of KF in the matrix. The tensile modulus of PP/KF with modified MAPP also gave the good improvement compare to the unmodified PP/KF as shown in Figure 9b. For the 30% fiber content, tensile modulus of modified PP/KF composites increased by 14.2 % and 96 % compared to unmodified PP/KF and pure PP. This is a common behaviour which is natural lignocellulosic fillers have been found as having elastic modulus higher than PP [27].

Figure 9. a) Tensile strength of PP/Unmodified KF and PP/Modified KF (b) Tensile modulus of PP/Unmodified KF and PP/Modified KF.

As the result, these compatibilizers act on both fiber and PP matrix to achieve superior fiber matrix adhesion. It formed carbon-carbon covalent bond with the matrix and reinforced with hydroxyl groups of the fiber, hence providing effective interlocking. Compatibilizer also diminished fibers fractures and enhanced the tensile strength. The reaction of anhydride of compatibilizer with the hydroxyl groups of fibers also lessen the compatibility problems between the fiber and matrix [28].
4. Conclusion
In the present study, the effect of different fiber fraction of PP/CF and PP/KF composites, as well as the effect of the addition of MAPP as a compatibilizer on the microstructure and mechanical properties were investigated. Based on the finding results, it can be concluded that the presence of the MAPP as a compatibilizer showed good compatibility for KF and CF between the matrices. The 30 % KF reinforced PP with 3 % of MAPP was found to be good results compared to unmodified PP/KF, unmodified PP/CF and modified PP/CF. Furthermore, the SEM and FTIR results were clearly revealed that the interfacial adhesion of the KF and CF and PP matrix was significantly improved due the addition of MAPP.

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