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SYNTHESIS AND CHARACTERISATION OF MALEATED POLYPROPYLENE/*LALANG* FIBER/POLYPROPELYNE COMPOSITES: OPTIMISATION PREPARATION CONDITIONS AND THEIR PROPERTIES

(Penyediaan dan Pencirian Komposit Polipropelena Termaleik/Gentian Lalang/Polipropilena: Keadaan Penyediaan Optimum dan Sifatnya)

Sharil Fadli Mohamad Zamri[1,2*], Hasratul Nadiah Mohd Rashid[1,2], Nurul' Ain Jamion[1,2] and Rahmah Mohamed[2]

[1]International Education College (INTEC),
UiTM Campus Section 17, 40200 Shah Alam, Selangor
[2]Faculty of Applied Science,
Universiti Teknology MARA, 40450 Shah Alam, Selangor

**Corresponding author: sharil7240@salam.uitm.edu.my*

Abstract

Polypropylene (PP) blend with natural *lalang* fiber (LF) were investigated in this study. LF was chosen in preparation of polymer composites because it is low cost, eco-friendly, abundance, renewable, biodegradable and shows excellent mechanical properties. Hence, LF has highly potential as a natural fiber filler in polymer composites. LF was prepared by drying the *lalang* leaves under the sun light and were crushed using hammer mill before they were soaked in 5% sodium hydroxide (NaOH) solution. The fiber was then washed with distilled water until neutral. LF was dried in an oven then grinded and sieved into specific sizes. LF/PP and maleated polypropylene/LF/PP (MAPP/LF/PP) composites were prepared by extrusion method using a twin-screw extruder between LF, PP and MAPP at different compositions. The LF/PP and MAPP/LF/PP composites were shaped into 3 mm sheet samples using an electric hydraulic hot press at the temperature of 180 ˚C. Several studies were carried out to investigate the optimum preparation conditions including the effect of size and percentage of LF, as well as the percentage of MAPP on their mechanical properties using tensile tester and izod impact tester. Meanwhile, the thermal properties of PP, 15% M size LF/PP and 1% MAPP/15% M size LF/PP composites were analyzed using thermogravimety analysis (TGA) and the characteristic of LF on the cross section of 15% M size LF/PP and 1% MAPP/15% M size LF/PP composites were investigated using a digital microscope.

Keywords: Lalang, Polypropylene, Extrusion, Maleated polypropylene, Polymer composite

Abstrak

Polipropilena (PP) diadun bersama gentian lalang semulajadi (LF) dikaji dalam kajian ini. LF dipilih dalam penyediaan polimer komposit kerana ianya rendah kos, mesra alam, tersedia dalam kuantiti yang banyak, boleh diperbaharui, boleh terhurai secara biologi dan menunjukkan sifat mekanikal yang baik. Justeru, LF mempunyai potensi yang tinggi sebagai pengisi gentian semulajadi dalam komposit polimer. LF disediakan dengan mengeringkan daun lalang di bawah cahaya matahari dan dihancurkan menggunakan penghancur tukul sebelum ia direndam di dalam larutan 5% natrium hidroksida (NaOH). Gentian tersebut kemudiannya dibasuh dengan air suling sehingga neutral. LF dikeringkan di dalam oven kemudian dikisar dan diayak kepada saiz tertentu. Komposit LF/PP dan polipropilena termaleik/LF/PP (MAPP/LF/PP) disediakan dengan kaedah pengekstrudan menggunakan pengekstrud skru-berkembar di antara LF, PP dan MAPP pada komposisi berlainan. Komposit LF/PP dan MAPP/LF/PP dibentuk kepada kepingan sempel 3 mm menggunakan hidrolik elektrik tekan panas pada suhu 180 ˚C. Beberapa kajian telah dijalankan untuk mengkaji keadaan penyediaan optimum termasuk kesan saiz dan peratusan LF, dan juga peratusan MAPP terhadap sifat mekanikalnya menggunakan penguji tensil dan penguji hentaman izod. Sementara itu, sifat haba PP, komposit 15% saiz M LF/PP dan 1% MAPP/15% saiz M LF/PP dianalisis menggunakan analisis termogravimetri (TGA) dan ciri-ciri LF pada keratan rentas komposit 15% saiz M LF/PP dan 1% MAPP/15% saiz LF/PP disiasat menggunakan mikroskop digital.

Kata kunci: Lalang, Polipropilena, Pengekstrudan, Polipropilena termaleik, Polimer komposit

Introduction

Several studies were conducted by researchers to investigate the potential application of natural fibers in biodegradable polymer composites in order to minimise the dependency of artificial fillers such as fiberglass and carbon fiber for polymer composites [1-5]. The use of natural fillers in polymer composites has been reported by researchers including abaca [6], bamboo fiber [7], cotton [8] and wood fiber [9]. Natural fibers have been given a big attention due to the ability to promote several advantages over artificial fibers including greater value added, sustainability, renewability, biodegradability, non-toxicity, excellent mechanical properties and lower costs. These fibers are able to use in the preparation of natural fiber polymer composites as engineering materials especially in the automotive and construction industries [10-12].

Lalang plant or long-bladed grass (*Imperata arundinacea*) is the most abundant green short plant which grows wildly in the rain forests of tropical countries including Malaysia. The plant does not usually have any economical value and become one of the major problems to agriculture industries. This is because planters need to shed a lot of effort especially allocation of money, energy and time to control its population in order to eliminate the competition for nutrients, water and light with agro-economical plant [13].

As a natural fiber, lalang plant consists of several components including cellulose, hemicelluloses, lignin, pectins and extractives [14]. Cellulose which is the main component became one of ingredients for preparation of natural fiber polymer composites. The impurities such as hemicelluloses, lignin, pectins need to be eliminated from the natural fiber before it can be used in the preparation of natural fiber polymer composites trough a chemical treatment. The chemical treatment using NaOH hydroxide has been reported by Bessadok et al. (2003) and Bachtiar et al. (2008). The treatment will result some advantages including breaking down the bundle fibers into individual filament fibers and increasing contact area of the fiber with their matrix hence enhance the properties of the polymer composites [2;15].

Cellulose of natural fibers contains polar groups in its structure and gives a low compatibility with non-polar structure of polymeric matrices. Thus, the prepared natural fiber polymer composites show a dramatical decrease in mechanical properties of natural fiber polymer composites. Fiber surface treatments were reported to be able to improve these weaknesses [6]. Unfortunately, the modification of these natural fibers techniques such as graft copolymerization [16] and silane treatment [17] bring other implication such as high production cost, generate toxic waste and inflexible processes which may cut the potential of these fibers for a mass production scale. "*In situ*" surface modification via blending natural fiber with polymeric matrices with the presence of maleated polyolefin such as maleated polypropylene as MAPP and maleated polyethylene (MAPE) as the compatibiliser is able to avoid mentioned disadvantages [18].

PP is a thermoplastic has been used widely including packaging, plastic parts and various types of reusable containers of various types, loudspeakers, automotive components, and polymer banknotes [19] due to their lightweight property, inexpensive, inert against most of chemicals, high softening temperature, better thermal stability and flexible processability [20]. PP has lack of wetting characteristic against natural fiber filler. Thus, MAPP was used as compatibiliser. It has been proven that this blend is able to improve fiber–matrix bonding, resulting in improvements in the physical properties of the natural fiber polymer composites [21].

In this study, investigation to optimise the preparation conditions including the effect of size and percentage of LF, as well as the percentage of MAPP on their mechanical properties using tensile and izod impact tester were conducted. Meanwhile, the thermal stability of PP, 15% M size LF/PP and 1% MAPP/15% M size LF/PP composites were analyzed using TGA and the characteristic of LF on the cross section of 15% M size LF/PP and 1% MAPP/15% M size LF/PP composites were investigated using a digital microscope.

Materials and Methods

Materials

Lalang plants which grew wildly were collected form a rural area in Kampung Tekah Baru, Kampung Kepayang, Perak, Malaysia. Industrial grade PP was supplied from Titan (M) Sdn. Bhd. NaOH was purchased from Merck, German and MAPP was obtained from SIRIM, Shah Alam, Selangor, Malaysia.

Preparation of LF

The LF was prepared by drying the lalang leaves under the sun light then they were crushed using a hammer mill before they were soaked in 5% sodium hydroxide solution for 48 hours to remove their hemi-cellulose, lignin and pectins components. After that, LF was washed using distilled water until a solution reached pH 7 and LF was dried again in an oven at the temperature of 60 ˚C until it reached constant weight. Finally, the LF was grinded using domestic electrical grinder and sieved into specific of sizes.

Preparation of LF/PP and MAPP/LF/PP composites

The MAPP/LF/PP composites were prepared by extrusion method using a twin-screw extruder between LF, PP and MAPP at different compositions. The MAPP/LF/PP composites were extruded twice to improve the distribution of filler in their matrix. The LF/PP and MAPP/LF/PP composites were shaped into 3 mm sheet samples using an electric hydraulic hot press at the temperature of 180 ˚C.

The weight percentage of MAPP/LF/PP composites was calculated by following formula:

$$\text{Percentage of LF} = \frac{\text{weight of LF}}{(\text{weight of PP} + \text{weight of LF} + \text{weight of MAPP})} \times 100 \qquad [22]$$

Results and Discussion

Table 1 shows the mechanical properties of LF/PP composites at various LF sizes prepared by extruding PP with LF at 20% of LF loading. The tensile strength of LF/PP composites are not influenced by the size of LF. The same phenomenon was observed by Ruksakulpiwat et al. [13]. Meanwhile, Young's modulus of LF/PP composites shows significant decreases with the increasing of LF size. The impact strength of LF/PP composites increased as the size of LF gets bigger. This may due to the increases of LF increasing the adhere area of PP on individual LF's filaments.

Table 1: Tensile strength, Young's modulus and impact strength of 20% LF/PP composites at various LF sizes loaded.

LF size	Tensile strength (MPa)	Young's modulus (N/mm^2)	Impact strength (kJ/m^2)
S	20.6	1434.2	10.6
M	20.5	1111.2	10.8
L	20.3	792.9	14.3

S: <1 mm; M: 1-2 mm; L: 2-3 mm

The variation of tensile strength, Young's modulus and impact strength of LF/PP composites using M size LF as a function of percentage of LF loading is represented in Table 2. All tensile strength, Young's modulus and impact strength of LF/PP composites show the similar trends where when the percentage of LF increases, it leads to the decreasing of their mechanical properties from 0 to 10 of LF loaded. The decreases in these properties are due to the lack of affinity between the LF and PP. As a natural fiber, LF has a poor compatibility demonstrated between the fiber and the matrix. This will cause a non-homogeneous dispersion of fiber in their matrix and decreases in the mechanical properties of the polymer composites [23]. However, the results show there are no significant effects after 15% of LF has been loaded into the polymer composites.

Table 2: Tensile strength, Young's modulus and impact strength of M size LF/PP composites at various percentage of LF loaded.

Percentage of LF loaded	Tensile strength (MPa)	Young's modulus (N/mm^2)	Impact strength (kJ/m^2)
0	31.1	1834.1	37.1
5	28.0	1586.1	13.0
10	24.1	1463.6	12.0
15	20.6	1442.1	10.0
20	20.5	1431.2	10.8
25	20.3	1449.4	10.2

The effect of MAPP as a compatibilizer for MAPP/LF/PP composites on their tensile strength, Young's modulus and impact strength are shown in Table 3. It clearly revealed that by the introduction of 1% of MAPP to the MAPP/LF/PP composites, it increases the tensile strength, Young's modulus and impact strength of the polymer composites by 18, 42 and 48%, respectively. As a natural fiber, LF naturally is hydrophilic. This will cause incompatibility between LF with hydrophobic PP and leads to a decrease in mechanical properties of the polymer composites. Good filler-matrix interface interaction is the main key for properties of polymer composites. The incorporation of MAPP in the polymer composites may improve filler-matrix adhesion strength [24]. Compatibility of the polymer composites may improve by the formation of hydrogen bond between maleic anhydride of MAPP with hydroxyl group of LF and the presence of PP segment as shown in Figure 1 [25].

Table 3: Tensile strength, Young's modulus and impact strength of MAPP/15% M size LF/PP composites at various percentage of MAPP loaded.

Percentage of MAPP loaded	Tensile strength (MPa)	Young's modulus (N/mm^2)	Impact strength (kJ/m^2)
0	20.6	1442.1	10.0
1	24.3	2047.9	14.8
3	23.0	1609.0	12.7
5	21.6	1468.3	11.1

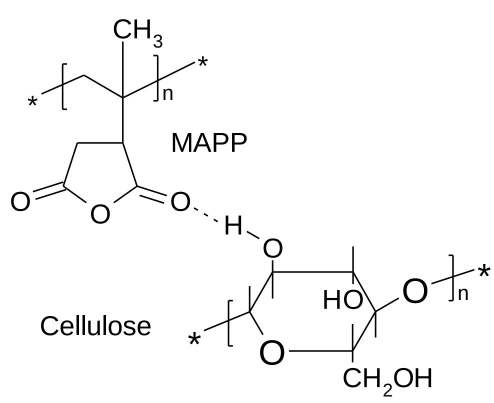

Fig. 1: Formation of hydrogen bonding between maleic anhydride in MAPP and hydroxyl group in cellulose of LF.

Thermal stability of polymer composites has been improved by the introduction MAPP as shown in Figure 2. The figure shows that the thermal stability of 1% MAPP/15% M size LF/PP composite is in between the thermal stability of 15% M size LF/PP composite and PP. Table 4 confirms that the decomposition temperature of polymer composites measured at 5 and 50% mass loss has shifted to the higher temperature for 1% MAPP/15% M size LF/PP composite compared to 15% M size LF/PP composites. Furthermore, the different in mass loss measured at the temperature of 250 to 350 °C for 1% MAPP/15% M size LF/PP composite is lesser compared to the 15% M size LF/PP composite. MAPP improved the compatibility of the polymer composites by improving the interfacial adhesion between LF and PP. The formation of hydrogen bond in between LF and MAPP gives additional improvement toward polymer composite's properties [24-25].

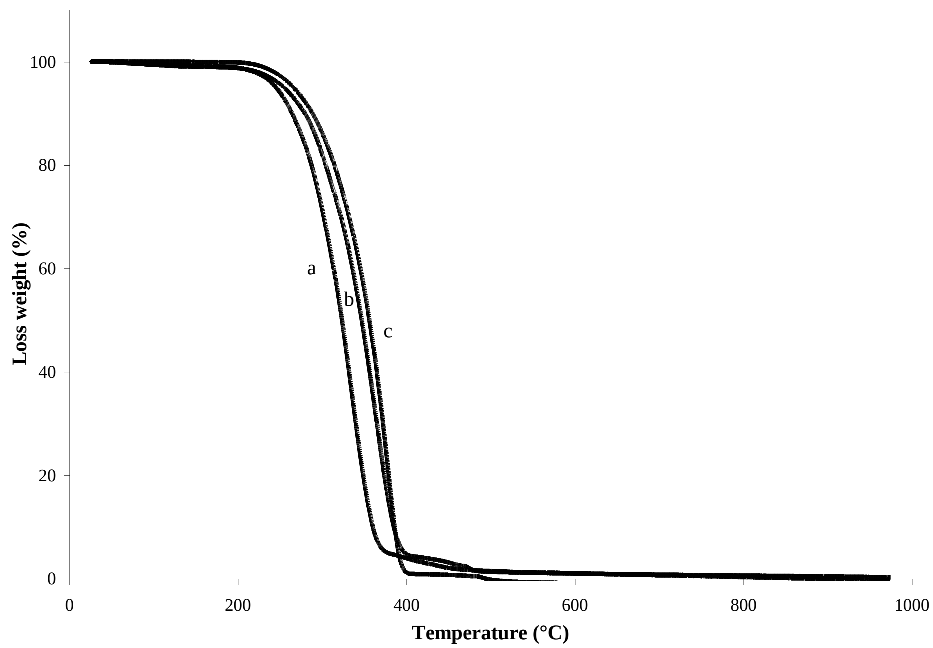

Figure 2: TG thermograms of (a) 15% M size LF/PP composite, (b) 1% MAPP/15% M size LF/PP composite and (c) PP.

Table 4: Thermogram data of PP, 15% M size LF/PP and 1% MAPP/15% M size LF/PP composites.

	T_{-5} (°C)	T_{-50} (°C)	Different Mass Loss (250-350 °C) (%)
PP	266.138	356.079	41.21495
15% M size LF/PP	245.564	323.515	75.32318
1% MAPP/15% M size LF/PP	254.755	346.942	48.86728

Note: T_{-5}: Decomposition temperature at 5% loss weight, T_{-50}: Decomposition temperature at 50% loss weight

Figure 3 shows pictures of cross section samples of (a) 15% M size LF/PP and (b) 1% MAPP/15% M size LF/PP composites taken using a digital microscope. The samples were initially tested for tensile strength then the picture of the broken area resulting from the test was taken in order to investigate the relationship between pulled LF filaments with their tensile strength, Young's modulus and impact strength of the composites. The figure clearly shows that the LF filament was pulled out individually resulting from the force generated from the tensile test for uncompatibilized 15% M size LF/PP. Contrary, the compatibilized 1% MAPP/15% M size LF/PP composites picture shows that the pulled out LF filaments are covered with PP as its polymeric matrix. By introduction of 1% MAPP may improve the adhering property of MAPP/LF/PP composites. This agrees with the tensile strength, Young's modulus and impact strength of MAPP/LF composites.

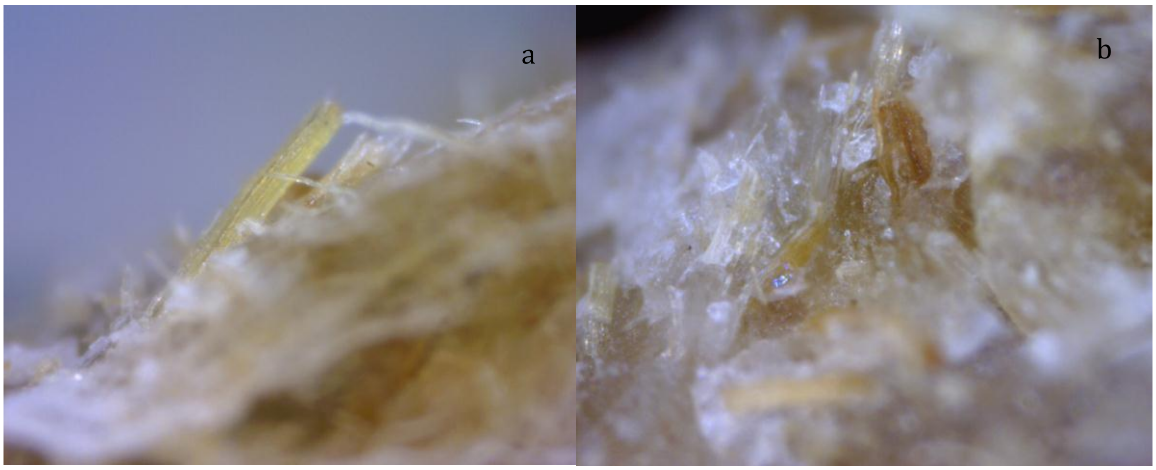

Figure 3: Digital microscopic pictures of (a) 15% M size LF/PP (b) 1% MAPP/15% M size LF/PP composites.

Conclusion

LF/PP and MAPP/LF/PP composites were successfully prepared. The mechanical properties of LF/PP composites were varied according to size and percentage LF loaded. Bad wetting characteristic between LF and PP leads to decreasing in mechanical properties of LF/PP composites. By introduction of 1% MAPP give the higher mechanical and thermal properties to MAPP/LF/PP composites. This due to ability of MAPP to improve the compatibility characteristic between LF and PP. Digital microscopic pictures of cross section samples revealed that MAPP enhanced adhering properties between LF and PP.

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