Chapter 11

COMMINGLED COMPOSITES FROM COMMINGLED GRANULES

Abstract

Polypropylene/banana fiber commingled composites has been prepared from commingled PP/banana fiber granules by injection moulding method with special reference to the effect of maleic anhydride modified polypropylene (MAH-PP) concentration. The mechanical properties of the composites has been found to depend on the concentration of MAH-PP. The tensile and flexural properties of the composites increased with the addition of MAH-PP up to 2 wt%. After 2 wt% addition of MAH-PP, these properties tend to be stabilised. On the other hand the unmodified composites showed the maximum impact strength. Fourier transform infrared spectroscopic analysis of the MAH-PP modified composites showed the evidence of a chemical bridge between the hydroxyl group of the banana fiber and maleic anhydride of the MAH-PP through esterification reaction. The feature peak of the esterification occurred in the range ~1743cm⁻¹. The tensile fracture surfaces of the untreated and MAH-PP modified PP/banana fiber composites were studied by scanning electron microscopy. An improvement in adhesion between the fiber and the matrix was observed in the case of MAH-PP treateded composites.

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11.1 Introduction

PP/natural fiber commingled granules can be fabricated from commingled slivers of natural fiber and PP fiber. The basic principle for the manufacture of commingled granules is the use of slivers. These slivers are made by mixing of the reinforcing fiber and PP fiber by textile equipments. The fiber content of the composite can be adjusted by the mixing ratio. The slivers are twisted like in flyer yarn production but at the same time are heated up and are compacted (Pull-drill-process) [1]. By twisting of the sliver the reinforcing fibers are compacted and bonded together with the molten matrix material. A compact material strand is formed that can transfer high tensile forces and can be taken off continuously. Afterwards the rope is quenched in a cooling zone and consolidated. The solid strand is cut in order to pelletize the material. The process of commingled granule manufacturing (Pull-drill-process) is shown in Fig.1.14. Commingled granules consisted of 30% by weight of banana fiber and 70% by weight of PP. The length of the granule is 10 mm. These granules can be easily converted into composites by heating the material above the melting point of the matrix either by compression moulding or injection moulding. They offer significant environmental advantage including total recyclability. The commingled granules provide an even distribution of fiber and the matrix. The homogeneous distribution of the reinforcement and the matrix require low shear energy during the processing steps. Because of the premixing of the matrix and the reinforcement fibers in an earlier stage, the molten matrix will be gluing the outer reinforcing fibers, but inside the granules, most of the matrix materials will be in the fiber form. As a result natural fibers are only subjected to low thermal stress. The commingled granules have a helical fiber arrangement as it is made by twisting of the sliver [1]. The fiber content in the granules can be varied from 20-60% and the cutting length from 5-30 mm. Another characteristic feature of the granules
is the skin-core structure. In addition such a textile based granule manufacturing process is energetically more favourable than the conventional granule production process like pultrusion or extrusion [1]. But the properties of the composites made from these granules depend on the fiber content, their interfacial adhesion as well as the properties of the individual components. The inherent incompatibility of the hydrophilic natural fibers with hydrophobic polypropylene matrix leads to a poor adhesion between the natural fiber and the polypropylene matrix in the composite [2-4].

This chapter deals with the effect of maleic anhydride grafted polypropylene on the mechanical properties of composites prepared from polypropylene/banana fiber granules. This study is important as no studies have reported in literature where commingled granules were prepared from polypropylene fiber and banana fiber.

11.2 Results and Discussion

11.2.1 Effect of varying MAH-PP concentration

Preparation of the commingled granules and various formulations used to prepare the composites are described in Chapter 2, Section 2.10. 2. Fig. 11.1 shows the tensile stress-strain behaviour of the MAH-PP treated PP/BF composites. The stress is found to increase linearly with strain at low elongation followed by a non-linear behaviour due to plastic deformation. The stress value is found to be the highest for 2 wt% MAH-PP treated composites and the value then decreases on increasing the concentration of MAH-PP. At 2 and 3 wt% addition of MAH-PP, the stress-strain curve seems to overlap emphasizing the maximum allowable MAH-PP concentration.
Fig. 11.1  Tensile strain-stress behaviour of the MAH-PP treated PP/banana fiber composites. (1) untreated (2) 1% MAH-PP (3) 2% MAH-PP (4) 3% MAH-PP (banana fiber loading 30%)

Fig. 11.2  Effect of wt % MAH-PP on tensile strength of PP/banana fiber composites (banana fiber loading 30%)
The influence of MAH-PP concentration on the tensile strength of PP/BF composites is shown in Fig.11.2. The tensile strength increases up to 2 wt% concentration of the MAH-PP and after that the values tend to level off. The addition of 1 wt% MAH-PP cause an increase of the tensile strength to about 5%, while the addition of 2 and 3 wt% of MAH-PP enhanced the tensile strength to about 13.5 and 14.5% respectively.

The tensile modulus of the composites Fig. 11.3 is also found to increase, but a decrease in elongation at break Fig. 11.4 is observed with increasing MAH-PP concentration. The modulus, which denotes the stiffness of the material, reaches a maximum value at 2 wt% MAH-PP content and then level off as observed in the case of tensile strength. Elongation at break of a material is a measure of the ductility of the material. The decrease in elongation at break is expected, as the composites with MAH-PP are stiffer and have higher strength. Thus the untreated composites are more ductile and the ductility of the composites decreases after the MAH-PP addition.

Flexural strength is a measure of how well a material resists bending. The deformation under flexural load is a combination of compressive and tensile stress.
Fig. 11.3  Effect of wt% of MAH-PP on Young’s modulus of PP/banana fiber composites (banana fiber loading 30%)

Fig. 11.4  Effect of wt% of MAH-PP on elongation at break of PP/banana fiber composites (banana fiber loading 30%)
Fig. 11.5 shows the flexural stress-strain of the MAH-PP treated PP/BF composites. The slope of the curve increases and the plastic deformation occurs at a higher stress level for MAH-PP treated composites and it is higher for 2 wt% MAH-PP treated composites. Figs 11.6 and 11.7 show the result of varying the concentration of the MAH-PP on the flexural strength and flexural modulus of the PP/BF composites. From the figure we can see that the flexural strength and flexural modulus increase with the increase in wt% addition of MAH-PP. The flexural properties increase to a maximum value at 2 wt% addition of MAH-PP. After this, the flexural properties decrease. The addition of 1 and 2 wt% of MAH-PP to the PP/BF granules increases the flexural strength of the composites to 14.2 and 21.4% while the flexural modulus increases to 9.3 and 15.6% respectively.
Fig. 11.6  Effect of wt % MAH-PP on flexural strength of PP/banana fiber composites (banana fiber loading 30%)

Fig. 11.7  Effect of wt% MAH-PP on flexural modulus of PP/banana fiber composites (banana fiber loading 30%)
The Fig. 11.8 shows the impact strength of the composites with different wt% of MAH-PP. The nature of the interface region is of extreme importance in determining the toughness of the composite. It therefore could be expected that the impact strength of the untreated composites should be higher than the MAH-PP treated composites. The measured impact strength of the untreated composites is high due to the separation of the fibers from the matrix, helping to create rough new surfaces during the fracture and thus consuming more energy. But in the case of MAH-PP treated composites, the interfacial adhesion is more and hence less energy is consumed in the process of creating new surfaces. The impact strength decreases in the order untreated >1wt% MAH-PP treated >2wt% MAH-PP treated >3wt% MAH-PP treated fiber composites. This explanation can be further substantiated by considering the FTIR studies and SEM observations.

**Fig. 11.8** Effect of wt% of MAH-PP on impact strength of PP/banana fiber composites (banana fiber loading 30%)
The FTIR spectra of the PP/BF composites are shown in Figure 11.9a. The spectra shows an absorbance band at ~3427 cm⁻¹ which is attributed to the hydroxyl group stretching vibrations and another at ~2900 cm⁻¹, which is associated with the -C-H stretching vibrations. A band near ~1734 cm⁻¹ is attributed to the -C=O stretching vibrations due to the presence of hemicellulose [5]. The peak at ~1122 cm⁻¹ is likely due to C-O stretching vibrations and C-C stretching vibrations from components of cellulose in the BF [6,7]. The emergence of bands near ~2840 cm⁻¹ is the characteristic of the IR signals of the PP back bone.

The FTIR spectra of MAH-PP treated PP/BF composites (Fig.11.9b) also shows an absorption band at ~3408 cm⁻¹, but the intensity of the peak is decreased compared to the PP/BF composites, indicating that there are less –OH groups on the surface of the MAH-PP treated samples [6,7]. This is expected based on the esterification reaction between the BF hydroxyl groups and anhydride functionality of MAH-PP as shown in Chapter 6, Fig.6.21.

![FTIR spectra of (a) PP/banana fiber composites (b) MAH-PP treated PP/banana fiber composites](image)

**Fig. 11.9** FTIR spectra of (a) PP/banana fiber composites (b) MAH-PP treated PP/banana fiber composites
The esterification reaction expected to occur between the vegetable fiber with MAH-PP has been discussed in other previous publications [7,8]. A band ~1734 cm⁻¹ in the untreated sample associated with (−C=O) stretching exhibits a shift and appears at ~1743 cm⁻¹ in the MAH-PP modified composites is also an indication of the esterification reaction. The increased intensity of the peak at ~1462 cm⁻¹ and ~1372 cm⁻¹ in MAH-PP modified composites is contributed to the grafting reaction, suggesting more -C-H character in the modified samples [7,8].

The evidence supporting the chemical bonding of MAH-PP with BF obtained by FTIR was further proved by SEM. Figs. 11.10 a and b show the SEM of the fracture surface of the PP/BF composites. SEM indicates extensive debonding due to the poor fiber/matrix adhesion. One can see clean BF and some voids on the fracture surface. The voids are created by the debonding of BF during the tensile fracture indicating that the fiber and the matrix are not totally compatible with each other. But for MAH-PP treated composites (Fig. 11.11 a and b), a significant improvement in the fiber/matrix adhesion is observed as shown by the absence of holes and debonding of the fibers. In MAH-PP treated composites, the peeling or tearing of the fiber from the matrix and the fiber breakage can also be observed due to the strong fiber/matrix adhesion. The improved fiber/matrix adhesion is due to the formation of bonds existing between the BF and the matrix provided by the reaction between BF hydroxyl group and maleic anhydride of MAH-PP. Furthermore, Fig. 11.11 b clearly shows the presence of polypropylene coated on the BF surface due to the high interfacial adhesion for the MAH-PP modified composites, which can be explained as follows.
Fig. 11.10 SEM of the fracture surface of untreated PP/banana fiber composites at different magnifications

Fig. 11.11 SEM of the fracture surface of the MAH-PP treated PP/banana fiber composites at different magnifications
The increase in tensile and flexural properties of the MAH-PP treated composite is associated with the ester linkages formed by the chemical reaction of the acidic anhydride group of the MAH-PP and the hydroxyl group of the BF [7,8] and the physical entanglement of PP molecules from the MAH-PP coupling agent and the PP matrix. This entanglement causes a reduction in the interfacial tension and an increase in interfacial adhesion between PP and the BF. Felix and Gatenholm [9] showed that in MAH-PP modified cellulose fiber composites the treatment improves the wetting resulting in better PP/BF adhesion in comparison with the untreated system. Fig. 11.12 shows the schematic representation of the interface of MAH-PP treated PP/BF composites.

It is also important to add that the levelling of the tensile and flexural properties after 2 wt% concentration of MAH-PP is due to the interfacial saturation attained by the system and the excess concentration of MAH-PP (higher than 2 wt%) did not contribute further in the interfacial adhesion between the PP/BF in the composites.
11.3 Conclusions

Commingled PP/banana fiber granules were fabricated from commingled slivers of polypropylene fiber and banana fiber by pull-drill-process. PP/banana composites were prepared from commingled PP/banana granules by injection moulding method with special reference to the effect of MAH-PP concentration. The effect of incorporating different wt% of maleic anhydride grafted polypropylene on the mechanical properties of composites was evaluated. It was found that the tensile and flexural properties of the composites increase with addition of MAH-PP up to 2 wt% followed by levelling off due to the interfacial saturation. The unmodified composites showed the maximum impact strength. The FTIR analysis of the unmodified and MAH-PP modified composites indicated the evidence of a chemical bridge between the banana fiber and polypropylene through esterification. The succinic and half succinic ester units were the two primary covalent linkages to bond the banana fibers and the polypropylene matrix. The scanning electron micrographs of the fracture surface of the composites showed better compatibility between the fiber and the matrix for MAH-PP modified composites.

References


