Mechanical Properties of Green Recycled Polypropylene Composites: Effect of Maleic Anhydride Grafted Polypropylene (MAPP) Coupling Agent

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Abstract: Recycled polypropylene/microcrystalline cellulose (rPP/MCC) composites were prepared by adding different loadings of maleic anhydride grafted polypropylene (MAPP) coupling agent. The tensile, impact and morphological properties of the composites were investigated. The obtained results show that the tensile and impact strengths of the composites were significantly enhanced with the addition of MAPP loading from 2 to 5 wt%, as compared with unfilled rPP/MCC composites. However, it was found that at low filler content, different amounts of MAPP resulted in no appreciable change in the tensile strength and modulus. Moreover, dynamic mechanical analysis (DMA) results indicated that, increasing the amount of MAPP loading from 2 to 5 wt% in rPP/MCC provide better stiffness of the composite compared to those neat rPP and neat PP. Field emission scanning microscopy (FESEM) has shown that the composite, with MAPP loading, promotes better fiber-matrix interaction.

Introduction

Polypropylene (PP) is one of the most versatile thermoplastic polymers and widely used in many applications such as in automotive parts, household equipments, telecommunication and electronic devices and food packaging due to its superior characteristics such as low in cost, high thermal stability, low density and recyclability [1]. Recycling of polypropylene waste is the most acceptable way of disposal. It offers many benefits; not only the recycling can reduce the environmental problem but it is also cost-effective. Through recycling, PP can be converted to valuable products for various applications and at the same time it can reduce the use of virgin PP. However, the recycled polypropylene usually faces reduction in its thermal and mechanical properties. This reduction was caused by the high temperature and shearing during processing [2].

Cellulose fibers extensively used as reinforcement fillers in polymer composites because of its low density, good mechanical properties, biodegradability, recyclable [3], relatively inexpensive [4] abundantly available [5]. Cellulose contains both amorphous and crystalline regions. Crystalline cellulose is much stronger and stiffer than amorphous cellulose and the cellulose itself. As a result, the crystalline cellulose is better as reinforcing filler than cellulose [6]. However, cellulose fibers are generally hydrophilic and are inherent incompatible with hydrophobic PP, in which this condition can reduce the mechanical properties of the composite materials. Consequently, the use of coupling agent is normally required to improve the interfacial bonding between the cellulose fibers and the polymer matrix [7]. Maleic anhydride grafted polypropylene (MAPP) has been shown to be very efficient in improving the interfacial adhesion between the fibers and the PP matrix [8]. In this present study, to improve the mechanical properties of rPP/MCC composites, MAPP has been used as a coupling agent to enhance the interfacial adhesion of the composites. The effects of the MAPP on mechanical behavior were evaluated using tensile testing, impact testing and dynamic mechanical analysis testing. The morphology of the composites was also evaluated by Field Emission Scanning Electron Microscopy (FESEM).

Experimental

Materials. Recycled polypropylene (rPP) resin with melt flow of 1.34 g/10 min at 230 °C and density of 1.07 g/cm³ was supplied by Top Flow Industry Sdn. Bhd. Commercial MCC (average diameter, 20 μ m) with density of 0.6 g/cm³. MAPP with density of 0.934 g/cm³ were obtained from Sigma-Aldrich Company. Both MCC and MAPP were added in the matrix as reinforcing filler and coupling agent respectively.

Preparation of the composites. RPP/MCC-MAPP were extruded in a twin screw extruder with a screw speed and temperature of 100 rpm and 180 °C respectively. The composites were prepared with different loading of MAPP(2%, 3%, 5% w/w) and the MCC content was kept constant at 4% w/w. The extruded material was palletized followed by injection molding using Battenfeld HM 600/210 injection molding machine. The temperature profile was kept at 180 °C to 190 °C and 200 °C in the nozzle. The samples were prepared into dumbbell type IV and rectangular shape according to D638 and D790 standard.

Characterization of Composites. Tensile and impact tests were carried out according to ASTM standard. Tensile tests were conducted according to ASTM D638 using a Universal Testing Machine (LLOYD Instruments) at a cross speed of 50 mm/min. Notched Charpy impact strength tests were conducted according to ASTM D 256 using Universal Impact Testing Machine (Dynisco Polymer Test). Dynamic mechanical analysis was perform on a Pyris Diamond DMA machine. The frequency was set at 1 Hz and static force of 0.01 N. The temperature range was set from -100 to 120 °C with a 0.5 °C mm/min heating rate. The fracture surfaces morphology of the composites were observed by using field emission scanning microscopy (JEOL-JSM 5600) at 10 kV. The sample surfaces were sputter coated with gold coating before analysis to eliminate electron charging effect.

Results and Discussion

Tensile Properties. Fig. 1 shows the tensile strength and modulus of the composites prepared with various MAPP loadings. For unfilled rPP/MCC composites, the tensile strength and modulus is lower compared to that of neat rPP. The result implied that the interfacial adhesion of MCC fiber and rPP matrix is poor due to hydrophilic nature of cellulose fiber and hydrophobic of rPP matrix. However, both the tensile strength and tensile modulus increased by incorporating MAPP into rPP/MCC composites. It is presumed that the better performance of rPP/MCC composites added with MAPP is due to improvement in the interfacial bond strength between maleic anhydride groups and cellulose fibers. The presence of MAPP also enhanced the compatibility between the rPP and the PP segments of MAPP. Nevertheless, it is also noticed that various content of MAPP (2 to 5 wt %) resulted in insignificant changes in the tensile strength. Although a slight increase in the tensile strength of rPP/MCC composite was observed at 3 wt % MAPP, but these differences are only minor. This result indicated that only a certain amount of MAPP loading could provide an effective interaction between the rPP matrix and MCC fiber.



Fig. 1: (a) Tensile strength and (b) modulus of rPP/MCC composite with 4 wt% of MCC with different MAPP loading.

Impact Properties. Fig. 2 shows the notched Charpy impact strengths of the rPP/MCC composites added with different MAPP loadings. In contrast to the tensile strength result, the impact strength of unfilled rPP/MCC composites is higher than that of the neat rPP. Since the MCC fibers are stiffer than the rPP matrix, thus their presence would enhance the stiffness of the rPP composites and led to the improvement in the impact strength. Apparently, greater impact strength was observed in rPP/MCC composites added with MAPP than unmodified rPP/MCC composites. This result is consistent with our findings in the tensile strength. Therefore, the good bonding between the rPP matrix and MCC fibers improved the stress transfer and absorbed energy in the composites. However when the MAPP loadings were varied from 2 to 5 wt %, the impact strength decreased. This phenomena could be due to too small amount of MCC loadings caused the free volume of the composites to increase and consequently led to segmental movements and restricted the stress transfer in the composites when subjected to an impact force [8].



Fig. 2: Impact strength of rPP/MCC composites with 4 wt% of MCC at different MAPP loadings.

Dynamic Mechanical Analysis. Fig. 3 is the result of dynamic modulus and tan delta of rPP/MCC composites. At different MAPP loadings, the storage modulus of the rPP/MCC composites increased with an increased in MAPP loadings, indicating an enhanced stiffness due to strong interaction between the rPP matrix and MCC fibers. From Fig. 3(a), rPP composites have shown improvement in its storage modulus to about 47.1% by addition of only 4 wt% MCC fibers and 3 wt% MAPP coupling agent.



Fig.3: DMA curves for rPP/MCC with different MAPP loading: (a) dynamic modulus, and (b) Tan delta.

It is also observed from Fig. 3(b) that the incorporation of MAPP from 2 to 5 wt % caused shifting in the peak value of tan delta. It is known that these peak values are assigned to the glass transition temperature, T_g . From Fig. 3(b) different MAPP loadings give different T_g values; the T_g values of rPP composites increased from 78.4 to 91.2 °C as the MAPP loadings were increased from 2 to 3 wt% respectively. The increased in T_g value indicated that MAPP coupling agent improved the compatibility between rPP matrix and MCC fibers. It is hypothesized that improvements in the compatibility had increased the interface bonding force. As a result, the thermal motion of polymer molecules decreased and thus reduced the friction thermal loss between the interface of the matrix and cellulose fibers [9]. However, further addition of MAPP at 5 wt% tends to lower the T_g value of rPP composites. This result suggests that there is a critical amount of MAPP at which MAPP exhibits optimum interaction between the matrix and cellulose fibers.

Morphology Analysis. The tensile fracture surface morphology of rPP/MCC composites were studied with FESEM. Neat rPP (Fig. 4(a)) shows the appearance of rough and coarse fibrous structure. At 3 wt% of MAPP loading (Fig. 4(c)), the composite shows much more smooth surface and the MCC fibers are well distributed compared to the composite at 2 wt% of MAPP loading (Fig. 4(b)). A good adhesion of the MCC fiber with the rPP matrix also can be seen from Fig.4 (b) and (c); the MCC fibers were still intact with the matrix. This shows that the MAPP provides a strong interfacial adhesion between the rPP matrix and MCC fibers which is consistent with our tensile results as discussed in the previous section.



Fig. 4: FESEM micrograph of (a) rPP, (b) 2 wt %, and (c) 3 wt% of MAPP loading.

Conclusion

Recycled polypropylene/microcrystalline cellulose (rPP/MCC) composites with different loadings of maleated polypropylene (MAPP) were fabricated and characterized. The tests results indicated that tensile and impact properties of the composites made with MCC and MAPP were superior to those of neat rPP, due to the stronger interfacial bonding between the MCC fiber and the rPP matrix. Nevertheless, various content of MAPP does not give significant changes for both tensile and modulus strength in the composite. DMA results shows that the MAPP loading provide better stiffness for rPP/MCC composites and increased the glass transition of the composite. FESEM morphology demonstrates that the interfacial adhesion between MCC and the rPP was improved. From this study, it can be concluded that the presence of 3 wt% of MAPP loading in rPP/MCC composites improved the mechanical properties of rPP/MCC composite at higher MCC content.

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