

Compatibilisation of Polypropylene/Ethylene Propylene Diene Terpolymer/Kaolin Composites: The Effect of Maleic Anhydride-Grafted-Polypropylene

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Abstract: *The tensile properties and morphology of polypropylene (PP)/ethylene propylene diene terpolymer (EPDM)/Kaolin composites were investigated. Maleic anhydride-grafted-polypropylene (MAPP) was used as a compatibiliser to improve the compatibility of kaolin filled PP/EPDM composites. Results show that incorporation of MAPP increased the tensile strength and Young's modulus, but reduced the elongation at break. Scanning electron microscopy (SEM) of the tensile fracture surfaces of composites indicates that the MAPP improved the interfacial interaction between kaolin and PP/EPDM matrix.*

Keywords: polypropylene, ethylene propylene diene terpolymer, kaolin, compatibiliser, composites

1. INTRODUCTION

The incorporation of mineral fillers into thermoplastics' elastomers has been widely practiced in industry to extend the elastomers and to enhance certain properties. Fillers often increase the performance of polymeric products. The degree of improvement depends on the choice of filler origin, particle size and shape, the fraction of filler, and the surface treatment promoting interaction between the polymer matrix and filler.¹ The addition of fillers to polymers is a fast and cheap method to modify the properties of the base materials. For this reason, particulate filled polymers have been, and continue to be, the subject of increasing interest in both industry and research. In this way, strength, stiffness, electrical and thermal conductivity, hardness and dimensional stability, among other properties can be tailored to the required values.²

The addition of filler to polyolefins seeks to reduce production costs with subsequent change in tensile and impact properties. Research by Han et al.,³⁻⁴ Tabtiang et al.,⁵ and Qiu et al.⁶ have reported that the addition of a filler to polymer systems results in a deterioration of the breaking and impact resistance; this behaviour has been attributed to weakness in the structure of these two-phase systems caused by stress concentration or discontinuity in stress transfer at the

narrow portions of the matrix at the dispersed phase and the matrix. This poor interaction between both components has given rise to the formation of large filler agglomerates in the polymer matrix, markedly influencing the mechanical response of the finished material.

Kaolin commonly has highly polar hydrophilic surfaces, whereas the polymers (e.g., polypropylene) into which they are introduced are often non-polar and hydrophobic. Consequently, poor adhesion occurred between the filler surface and the matrix. In addition, achieving uniform dispersion of the fillers tended to be difficult.⁷ Filler coating improved filler dispersion, which resulted in enhanced mechanical properties and easier processability.

The main problem in preparing kaolin-thermoplastic elastomeric composites is the incompatibility of hydrophilic kaolin and hydrophobic PP/EPDM matrix. However, applying compatibiliser and coupling agents on the surface of the mineral filler can promote filler-polymer interaction, which in turn improves the tensile and impact properties of the composite, as well as its processing capability. In fact, MAPP is commonly used as a compatibiliser, because it can efficiently improve the filler-matrix bonding due the formation of covalent linkages and hydrogen bonds between the maleic anhydride and the hydroxyl group of the fibre.⁸⁻¹⁴

This article reports the result of an investigation on the effect of MAPP as a compatibiliser on the mechanical properties and morphology of PP/EPDM/Kaolin composites.

2. EXPERIMENTAL

2.1 Materials

The PP homopolymer used in this study was of injection molding grade, from Titan PP Polymers (M) Sdn. Bhd., Johor, Malaysia (code 6331) with an MFI value of $14.0 \text{ g } 10 \text{ min}^{-1}$ at 230°C . EPDM, grade "950", was obtained from Luxchem Trading Sdn. Bhd., Selangor, Malaysia. MAPP was obtained from the Aldrich Chemical Company. Kaolin was obtained from Ipoh Ceramic Sdn. Bhd, Malaysia with an average size of $9.7 \text{ }\mu\text{m}$ (density, 2.2 g cm^{-3}). The formulation of PP/EPDM/Kaolin composites used in this study is shown in Table 1.

Table 1: The formulation of PP/ EPDM/Kaolin composites with and without MAPP.

Materials	Composites without MAPP	Composites with MAPP
PP (php)	50	50
EPDM (php)	50	50
Kaolin (php)	0, 15, 30, 45, 60	0, 15, 30, 45, 60
MAPP (php)*	-	3

php = part per-hundred polymer

* 3 php from weight PP

2.2 Mixing Procedure

Composites were prepared in a Haake Reomix PolyDrive. Mixing was done at 180°C and 50 rpm. The EPDM was first charged to start the melt mixing. After 3 min, filler and MAPP were added. After 5 min total time, PP was added. Mixing was continued for another 5 min. At the end of 10 min, the composites were taken out and sheeted through a laboratory mill at a 2.0 mm nip setting. Samples of the composites were compression molded in an electrically heated hydraulic press. Hot-press procedures involved preheating at 180°C for 6 min, compressing for 4 min at the same temperature and then cooling under pressure for 4 min. Table 2 shows a semi-quantitative analysis of the kaolin used in this study.

Table 2: Semi quantitative analysis of kaolin using X- Ray Fluorescence Spectrometer Rigaku RIX 3000.

Components	Wt. (%)
MgO	0.33
Al ₂ O ₃	30
SiO ₂	63
P ₂ O ₅	0.065
SO ₃	0.030
K ₂ O	1.5
CaO	0.042
TiO ₂	0.025
Fe ₂ O	0.84
NiO	0.012
Br ₂ O	0.59
Rb ₂ O	0.044
ZrO ₂	0.018
LOI	3.5

Note: LOI – Loss of Ignition

2.3 Measurement of Tensile Properties

Tensile tests were carried out according to the ASTM D-412 standard on an Instron 3366. One mm thick dumb bell specimens were cut from the molded sheets with a Wallace die cutter. A cross head speed of 50 mm min^{-1} was used and the test was performed at $25^\circ\text{C} \pm 3^\circ\text{C}$.

2.4 Morphology Study

Studies on the morphology of the tensile fracture surface of the composites were carried out using a SEM, model Leica Cambridge S-360. The fracture ends of the specimens were mounted on aluminium stubs and sputter coated with a thin layer of gold to avoid electrostatic charging during examination.

3. RESULTS AND DISCUSSION

Figure 1 shows the effect of filler loading on the tensile strength of kaolin filled PP/EPDM composites, with and without compatibiliser (MAPP). It can be seen that the tensile strength of all PP/EPDM composites decreases with increasing filler loading. For irregular shape fillers, the tensile strength of the composites decreases due to the inability of the filler to support stresses transferred from matrix. However, at similar filler loading, kaolin filled PP/EPDM composites with MAPP have higher tensile strength than similar composites without MAPP. The better tensile strength of PP/EPDM/ CaCO_3 with the presence of MAPP can be attributed to the better dispersion and adhesion of calcium carbonate and PP/EPDM matrix.

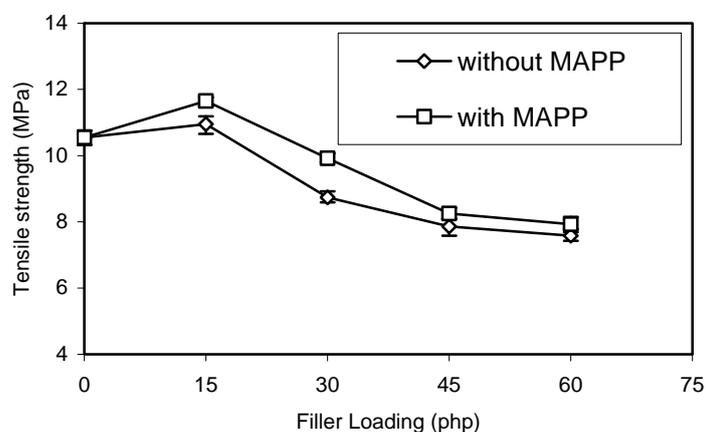


Figure 1: The effect of filler loading on the tensile strength of PP/EPDM/Kaolin composites, with and without MAPP.

The increase in Young's modulus with increasing filler loading (Fig. 2) is expected since the addition of filler increases the stiffness of the composites, which in turn decreases the elongation at break (Fig. 3). The reduction in elongation at break with increasing filler loading might be due to the decreased deformability of a rigid interface between the filler and PP/EPDM matrix. At similar filler loading, composites with MAPP indicate lower elongation at break, than composites without MAPP. Modification with MAPP as the compatibiliser has increased the tensile strength of composites, with an enhancement in the rigidity and reduction of the ductility of composites, which consequently lowered the elongation at break of PP/EPDM/Kaolin composites. Again at a similar filler loading, the Young's moduli of kaolin filled PP/EPDM composites with MAPP exhibited higher compared composites without MAPP. The application of compatibiliser in polymer composites was used to overcome the dispersion problem and to enhance the mechanical strength of composites by improving adhesion across the interface.

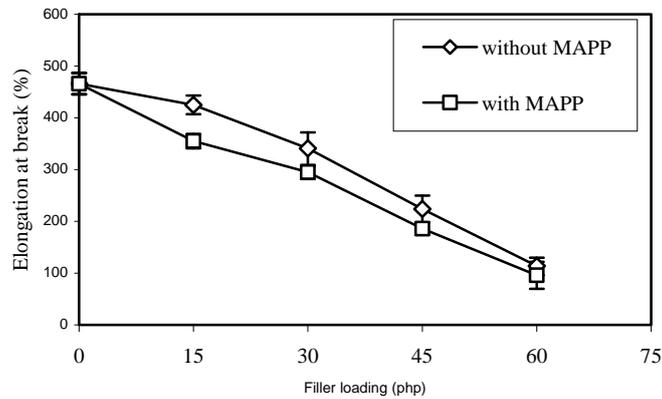


Figure 2: The effect of filler loading on the elongation at break of PP/EPDM/Kaolin composites, with and without MAPP.

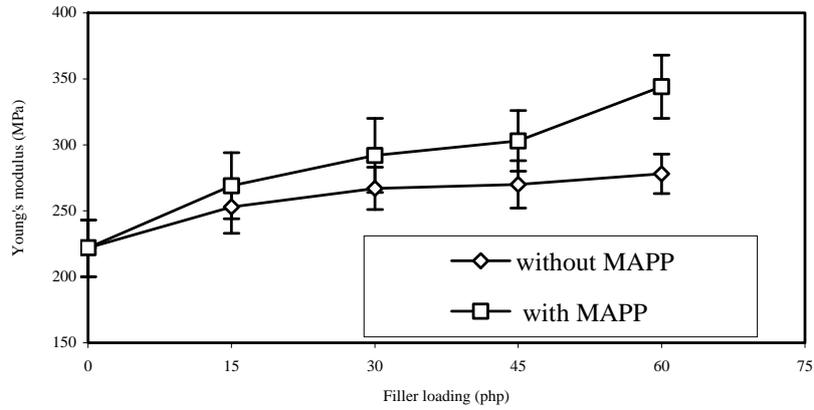


Figure 3: The effect of filler loading on the Young's modulus of PP/EPDM/Kaolin composites, with and without MAPP.

SEM was used to compare the tensile fracture surfaces of kaolin filled PP/EPDM composites containing 30 wt. % and 60 wt. % of kaolin. SEM micrographs of the fracture surfaces of PP/EPDM/Kaolin composites without MAPP are shown in Figures 4 and 5. The micrographs of the composites without MAPP exhibit poor wetting of kaolin by the PP/EPDM matrix. It can be seen that the fracture occurred at the interface of the kaolin and the PP/EPDM matrix and the kaolin was pulled out because of insufficient adhesion between the kaolin and the PP/EPDM matrix. However, for composites with MAPP (Figs. 6 and 7), the fracture occurred in the matrix material and the kaolin was covered by layers. There was less pull of the PP/EPDM matrix with MAPP present, and there is evidence of improvement in the interfacial bonding between the kaolin and PP/EPDM composites.

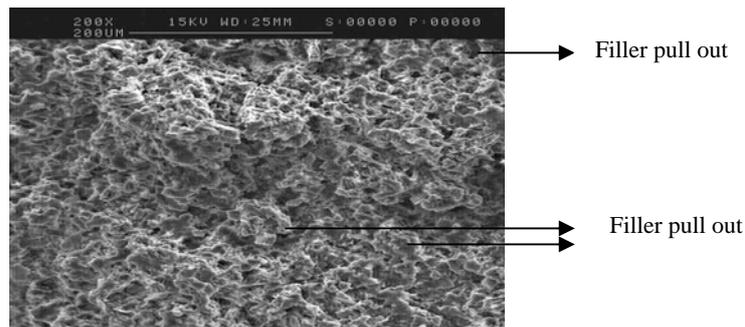


Figure 4: A scanning electron micrograph of the tensile fracture surface of PP/EPDM/Kaolin composites without MAPP (30 wt. %) at a magnification of 200X.

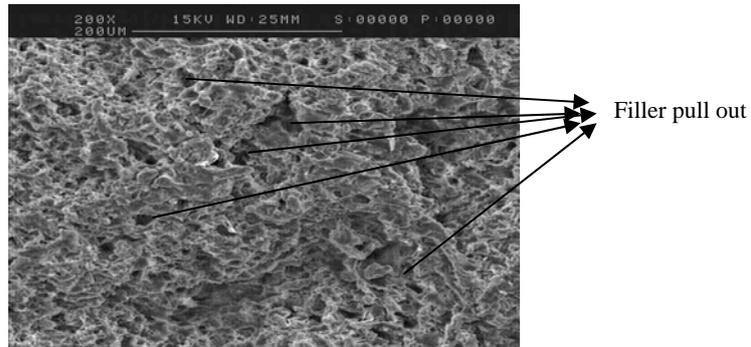


Figure 5: A scanning electron micrograph of the tensile fracture surface of PP/EPDM/Kaolin composites without MAPP (60 wt. %) at a magnification of 200X.

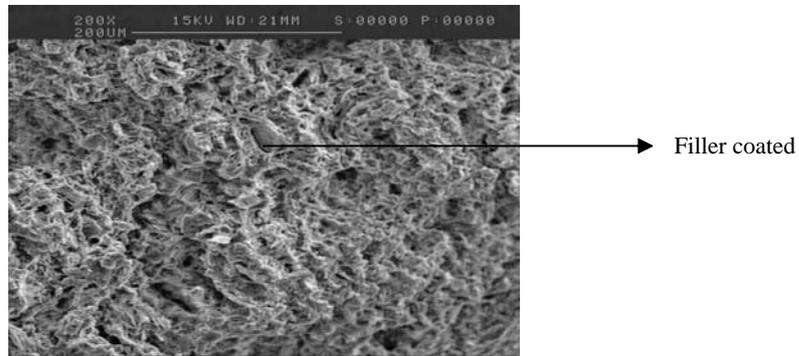


Figure 6: A scanning electron micrograph of the tensile fracture surface of PP/EPDM/Kaolin composites with MAPP (30 wt. %) at a magnification of 200X.

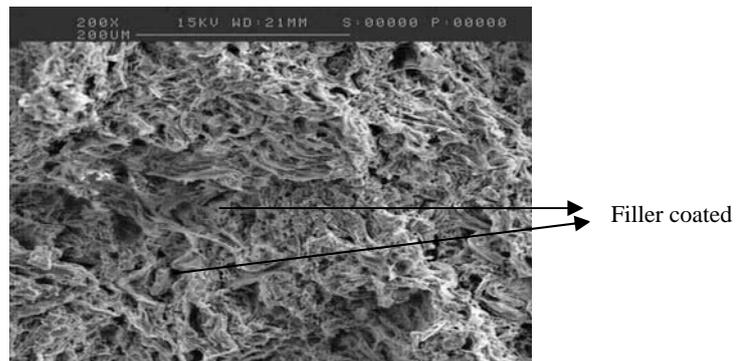


Figure 7: A scanning electron micrograph of the tensile fracture surface of PP/EPDM/Kaolin composites with MAPP (60 wt. %) at a magnification of 200X.

4. CONCLUSION

The compatibility between kaolin and PP/EPDM matrix is significantly improved by the addition of MAPP as a compatibiliser. Consequently the tensile strength, and Young's modulus of kaolin filled PP/EPDM composites with MAPP is better than those of untreated composites. SEM studies indicate that the interfacial adhesion between kaolin and PP/EPDM matrix is improved with the presence of MAPP.

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