POLYAMIDE 6/POLYPROPYLENE/ORGANOCLAY NANOCOMPOSITES: INFLUENCE OF ORGANOCLAY AND COMPATIBILISER ON THE MORHOLOGICAL AND TENSILE PROPERTIES

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Abstract. Polyamide 6/polypropylene (PA6/PP) nanocomposite was prepared by twin screw extruder and injection molding machine. The effect of organoclay loading from 2 wt% up to 10 wt% on the tensile properties was studied. 4 wt% organoclay has been observed to be the optimum loading for PA6/PP blends. Maleic anhydride grafted polypropylene (MAH-g-PP) at 5 wt% was used as a compatibiliser for 4 wt% organoclay filled PA6/PP nanocomposites. Failure mechanisms of PA6/PP/ organoclay nanocomposites, assessed by fracture surface studies in a scanning electron microscopy (SEM) are discussed. The incorporation of MAH-g-PP increased mechanical properties of PA6/PP/ organoclay nanocomposites. A coarse and brittle morphology was observed for uncompatibilised PA6/PP nanocomposites. On the contrary, MAH-g-PP compatibilised PA6/PP nanocomposites showed a more fibrilous dominant morphology, which indicates that MAH-g-PP is capable of promoting a better adhesion and interaction between PA6, PP and organoclay.

Keywords: Polyamide 6/polypropylene (PA6/PP) nanocomposit, organoclay, maleic anhydride grafted polypropylene (MAH-g-PP), tensile properties, morphology

Abstrak. Nanokomposit poliamida 6/polipropilena (PA6/PP) telah disediakan dengan menggunakan pengekstrud skru kembar and mesin pengacuanan suntikan. Kesan penambahan organo-tanah liat daripada 2% mengikut berat hingga 10% mengikut berat ke atas sifat-sifat tensil bagi komposit PA6/PP telah dikaji. Daripada kajian yang telah dijalankan, didapati komposit yang mengandungi organo-tanah liat sebanyak 4% mengikut berat telah menunjukkan penambahan yang optimum bagi komposit PA6/PP. 5% mengikut berat MAH-g-PP telah ditambahkan sebagai agen penyerasian dalam nanokomposit PA6/PP yang mengandungi 4% mengikut berat organo-tanah liat. Permukaan perekahan selepas ujian tensil bagi nanokomposit PA6/PP telah dikaji dengan menggunakan mikroskop penskanan elektron (SEM). Penambahan MAH-g-PP telah meningkatkan sifat-sifat mekanikal bagi nanokomposit PA6/PP/organo-tanah liat. Morfologi yang kasar dan rapuh telah diperhatikan bagi nanokomposit PA6/PP/organo-tanah liat tanpa MAH-g-PP. Manakala, bagi nanokomposit PA6/PP/organo-tanah liat yang mengandungi MAH-g-PP, morfologi yang menunjukkan dominasi fibril telah diperhatikan. Ini menunjukkan bahawa MAH-g-PP berkebolehan untuk meningkatkan pelekatan antara muka dan interaksi antara PA6, PP dengan organo-tanah liat.

Kata kunci: Nanokomposit poliamida 6/poli propilena (PA6/PP), organo-tanah liat, MAH-g-PP, sifat-sifat tensil, morfologi

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1.0 INTRODUCTION

In recent years, polymer nanocomposites have attracted great interest due to their superior properties such as enhanced stiffness and strength, reduced gas permeability, and improved flame retardancy. Layered silicates dispersed as a reinforcing filler in a polymer matrix are one of the forms of hybrid organic-inorganic nanocomposites [1]. The incorporation of nanometer scale reinforcement in polymeric materials could dramatically improve the performance of the polymer. Direct polymer melt intercalation is most attractive because of its low cost, high productivity and compatibility with current polymer processing techniques. For direct polymer melt intercalation process, the polymer chain spread from the molten mass into the silicate galleries to form either intercalated or delaminated hybrids [2].

Numerous researchers described polymer-clay nanocomposites based on single polymer matrix, for example polypropylene nanocomposites [3-8], and polyamide nanocomposites [9-13]. The present paper is a preliminary study on the thermoplastic nanocomposites based on blends of PA6/PP. It is expected that the combination of PA6, PP and organoclay could bring some synergistic effect due to the ease of processability and insensitivity to moisture of PP, good mechanical and good barrier against oxygen of PA6 and high aspect ratio of organoclay. However, PA6 and PP are known to be incompatible and a compatibiliser is required to promote a better adhesion and interaction between PA6 and PP. In the present study, MAH-g-PP was used as a compatibiliser. MAH-g-PP was used by Ide and Hasegawa to compatibilize the binary blends of PA6 and PP, and achieving an almost homogeneous morphology [14]. The effect of organoclay and maleic anhydride grafted polypropylene (MAH-g-PP) on the mechanical properties and morphology of PA6/PP nanocomposites is discussed.

2.0 EXPERIMENTAL PROCEDURE

The PA6 used in this study was a commercial product from Toray Nylon Resin, Japan. MFI and density of PA6 is 35 g/10 min at 230°C and 1.14 g/cm^3 respectively. PP was supplied by Titan Himont Polymer (M) Sdn. Bhd. MFI and density of PP is 25 g/10 min. at 230°C and 0.9 g/cm^3 respectively. Organoclay was a commercial product from Nanocor, Inc, USA. Melt compounding of the PA6/PP (70/30) blends and nanocomposites were done on counter-rotating twin screw extruder. The extrusion zone temperature ranged from 220-230°C. The extrudates were pelletized with the pelletizer. The pellets were injection molded into standard tensile bar using an injection molding machine. Injection molding temperature ranged from 225-240°C. Tensile test was carried out according to ASTM D638. Fracture surface morphology studies of selected PA6/PP blends nanocomposites were done by using scanning electron microscopy (SEM, model S 360 of Leica Cambrige Ltd.). The fracture surface was gold coated to avoid electrostatic charging during examination.

3.0   RESULTS AND DISCUSSION

Figure 1 shows the effect of organoclay loading on the Young's modulus of PA6/PP blend. Young's modulus increased steadily with increasing organoclay content. In all cases, the modulus of organoclay composite is higher than the modulus of PA6/PP blend. This may be attributed to the stiffness and high aspect ratio of the organoclay. Reichert *et al.*, [4] also showed that the Young's modulus increased as a function of organophilic layered silicates content.

Figure 2 shows the effect of organoclay loading on the tensile strength of PA6/PP blends. The tensile strength increased until an optimum loading of organoclay, is attained at 4 wt%. Further incorporation of organoclay reduced the strength gradually. The slight increment of the tensile strength with the incorporation of organoclay up to 4 wt% may be due to the interaction of organoclay with PA6. According to LeBaron *et al.*, [1], the coupling between the tremendous surface area of the clay and the polymer matrix, facilitates stress transfer to the reinforcement phase, and allowing for tensile improvements. Another possible reason is the high aspect ratio of organoclay and ease of delamination of organoclay up to 4 wt%. However, a mixture of the delaminated, intercalated and aggregated tactoids may exist in the PA6/PP nanocomposites. Above an optimum loading (*i.e.* > 4 wt%), the ease of delamination of the organoclay is more restricted. Above a critical clay concentration, the clay aggregates persist [15]. Although the high aspect ratio of silicate nanolayers is ideal for reinforcement, the nanolayers are not easily dispersed in most polymers due to their preferred face-to-face stacking in agglomerated tactoids [1].
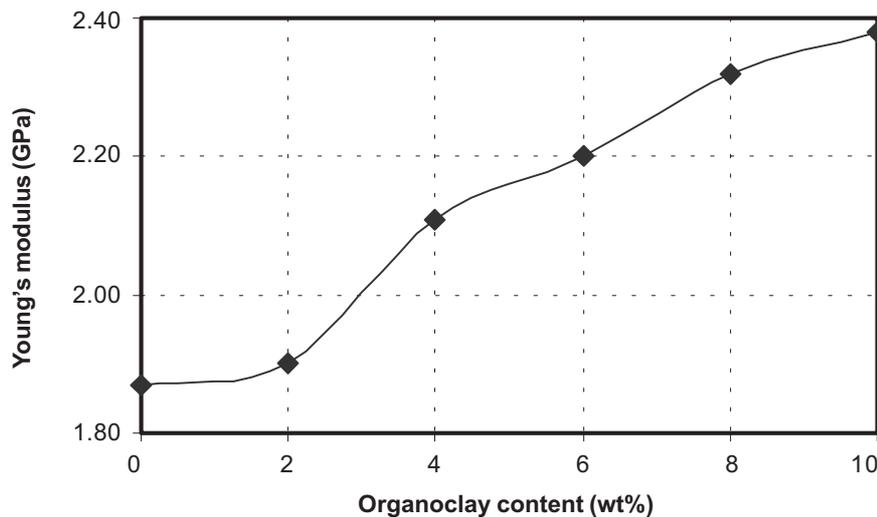
Figure 1   The effect of organoclay on the Young's modulus of PA6/PP blends
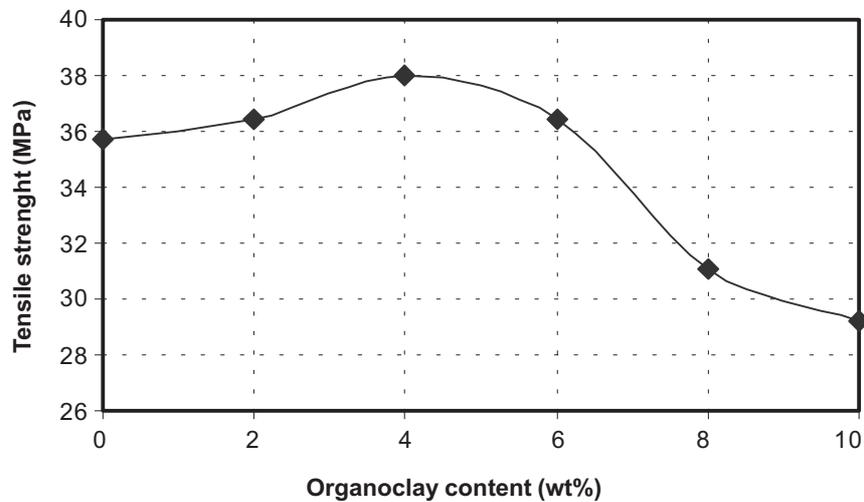
Figure 2   The effect of organoclay on the tensile strength of PA6/PP blends

The effect of organoclay loading on the elongation at break of PA6/PP composites is shown in Figure 3. The elongation at break drastically decreased with the incorporation of up to 4 wt% organoclay loading. A similar result also reported by Cho and Paul [10] and Reichert *et al.,* [4] for PA6 and PP nanocomposites. The brittleness of this material can be associated with the disappearance of plastic deformation of the polymer matrix as observed in the scanning electron microscopy. This may be due to the aggregated nanolayers tactoids which leads to the sacrifice of the elongation at break [1].

Figure 4 shows the effect of MAH-g-PP on the tensile modulus and yield stress of PA 6/PP blends and PA6/PP/organoclay nanocomposites. In this study, 4 wt% of organoclay filled PA6/PP nanocomposites was studied. The organoclay is able to act as a reinforcing filler, due to its high aspect ratio and platelet structure. The addition of organoclay increases the Young's modulus of the uncompatibilised PA6/PP blends. The enhancement in the Young's modulus of PA6/PP nanocomposites became more significant with the incorporation of the MAH-g-PP.

The incorporation of the MAH-g-PP has also increased the yield stress of the PA6/PP blends. When MAH-g-PP is added to PA6/PP blends, the anhydride group of MAH will react with the terminal amino group of PA6 during melt compounding. The PP group in the MAH-g-PP has the same affinity with the PP minor phase in the blends. PA6-g-PP copolymer will be formed. The improvement of the interfacial adhesion of the PA6 and PP will subsequently increase the yield stress of the PA6/PP blends.

The incorporation of 4 wt% organoclay in PA6/PP blends could not enhance the yield stress of the composites significantly. However, in the presence of MAH-g-PP, a more pronounced improvement in yield stress of the PA6/PP/organoclay
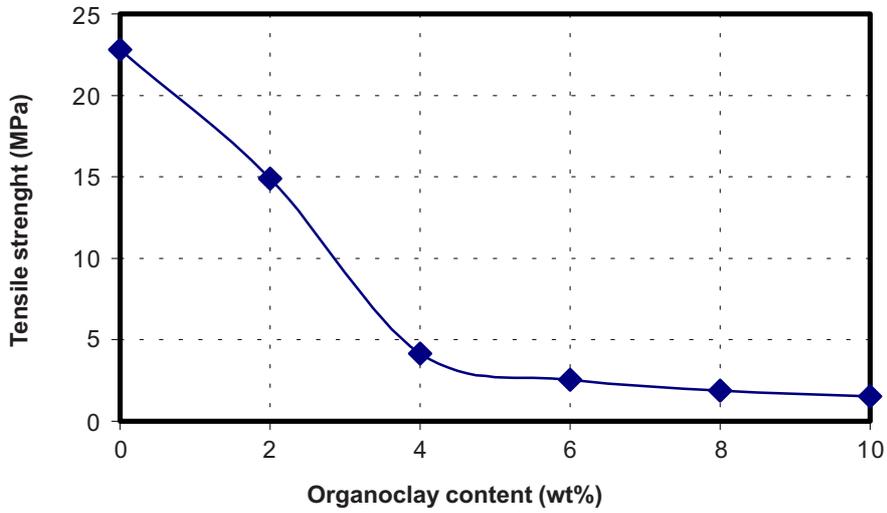
Figure 3 The effect of organoclay on the elongation at break of PA6/PP blends
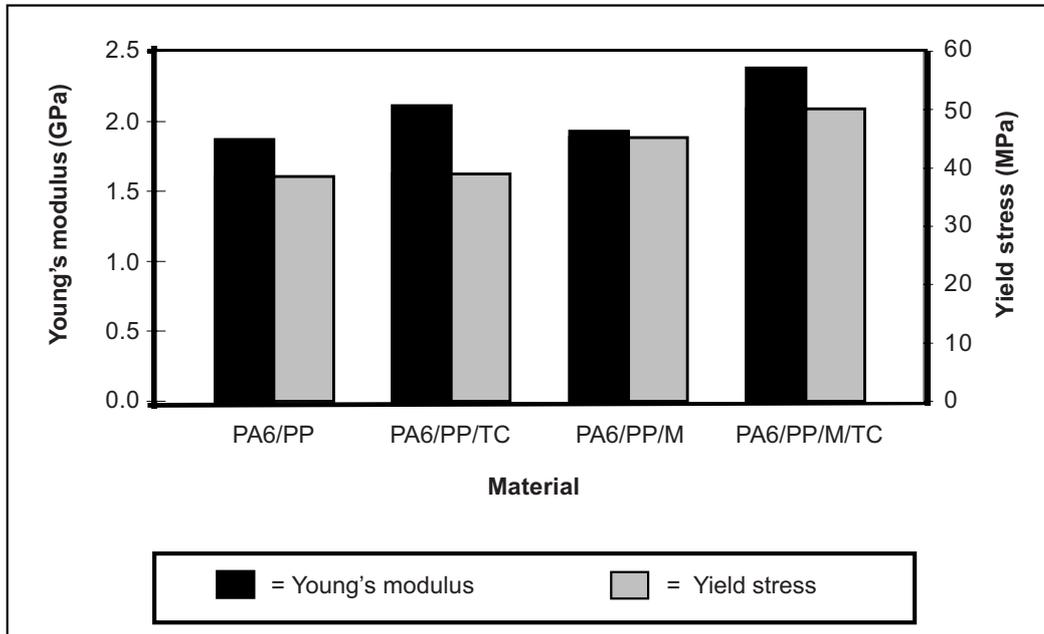
Figure 4 The effect of MAH-g-PP on the Young's modulus and yield stress of PA6/PP blends and PA6/PP/organoclay nanocomposites. (M = MAH-g-PP; TC = organoclay)

nanocomposites was observed. This may be attributed to the possible interaction between organoclay and MAH-g-PP. The mechanism of interaction is reported elsewhere [16].

Figure 5 shows the effect of MAH-g-PP on the tensile strength and elongation at break of PA6/PP blends and PA6/PP/organoclay nanocomposites. Tensile strength of the uncompatibilised PA6/PP blends increased slightly in the presence of 4 wt% organoclay. The incorporation of MAH-g-PP increases the tensile strength of PA6/PP/ organoclay nanocomposites significantly.

It is quite clear that improvements of tensile strength are dependent on the synergistic effect of organoclay and MAH-g-PP. This may be due to the extra chemical and/or physical interaction of silicate layers of organoclay that is distributed in the compatibilised interfacial layer of the PA6-g-PP copolymer. Necking and partially fibrous fracture was observed for the 5 wt% MAH-g-PP compatibilised PA6/PP blends. The elongation at break of the PA6/PP blends has increased in the presence of MAH-g-PP. This indicates the formation of PA6-g-PP copolymer which induces the interfacial bonding between PA6 and PP in the presence of MAH-g-PP. Subsequently, the compatibilised PA6/PP blends could withstand the tensile deformation to a higher elongation at break. When the compatibilised PA6/PP blends were subjected to the tensile deformation, the interfacial adhesion between PA6 and PP are still strong enough to withstand the deformation until necking zone. The addition of organoclay decreased
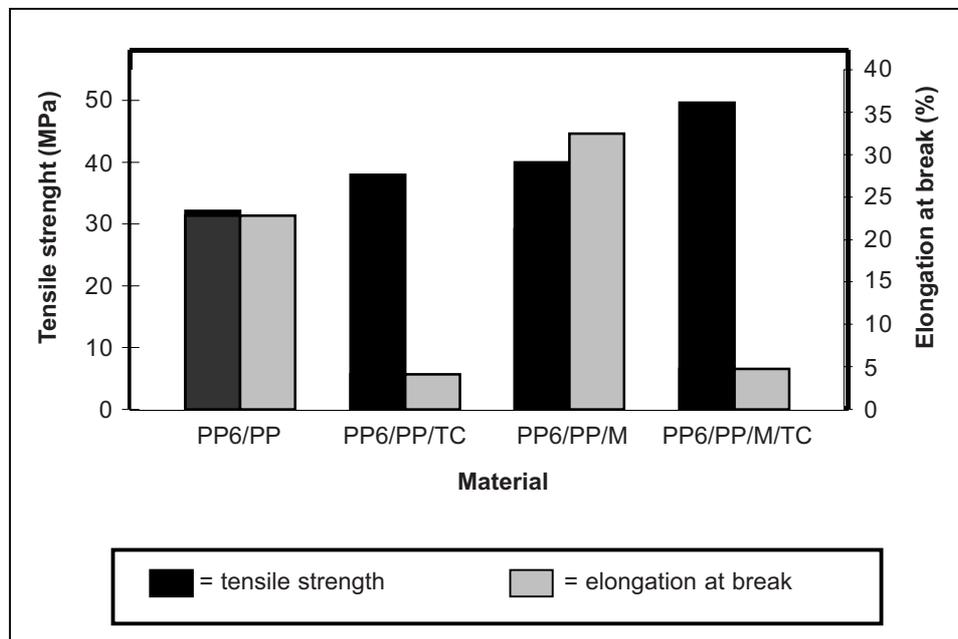
Figure 5   The effect of MAH-g-PP on the tensile strength and elongation at break of PA6/PP blends and PA6/PP/organoclay nanocomposites. (M = MAH-g-PP; TC = organoclay)

the elongation at break of PA6/PP blends. However, the presence of MAH-g-PP in the PA6/PP blends nanocomposites has a limited influence on the elongation at break.

Figures 6 and 7 show the scanning electron micrograph of uncompatibilised and MAH-g-PP compatibilised PA6/PP blends nanocomposites respectively. Whilst two phase morphology and phase separation was observed in the former, a fibrilous dominant morphology was observed in the later. This indicates that a good interfacial interaction was formed between PA6, PP, MAH-g-PP and organoclay. In Figure 6, the presence of PP droplets that were detached from the PA6 matrix is very obvious on the fracture plane. The coarse morphology of uncompatibilised PA6/PP/organoclay nanocomposites indicates the brittleness of the material. This explains the low elongation at break displayed by PA6/PP nanocomposites at 4 wt% and above organoclay loading (Figure 3). Figure 7 shows some morphological changes which can be associated to the incorporation of MAH-g-PP. Extensive fibril structure morphology on the fracture plane is very obvious. This indicates that MAH-g-PP is capable of increasing the compatibility between PA6, PP and organoclay. The extensive plastic deformation also explains the slight increment in the ductility of the MAH-g-PP compatibilised nanocomposites as compared with the uncompatibilised counter part. Further confirmation of this compatibilising effect will be derived from the impact test. This will be the subject of our future publication.

4.0   CONCLUSION

4 wt% organoclay has been observed to be the optimum loading for PA6/PP blends. The filler-filler interaction of organoclay and the formation of organoclay agglomerate which acted as stress concentration point is believed to be responsible for the
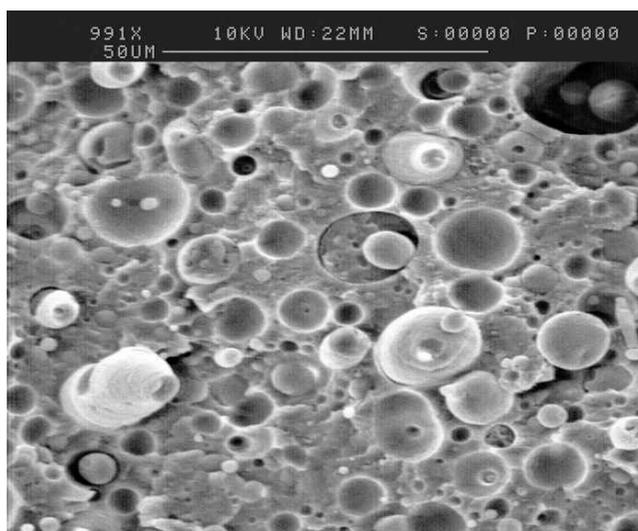
Figure 6   SEM micrograph showing the fracture surface of uncompatibilised PA6/PP/organoclay
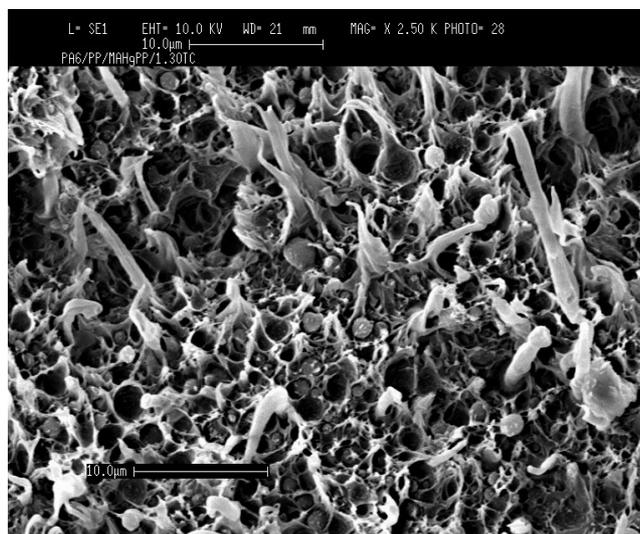
Figure 7   SEM micrograph showing the fracture surface of MAH-g-PP compatibilised PA6/PP/ organoclay nanocomposites

embrittlement of the composites and also for the deterioration in the mechanical properties above 4 wt% loading. Interestingly, the incorporation of MAH-g-PP increased the tensile properties significantly. The synergistic effect of organoclay and MAH-g-PP improved the strength and stiffness of PA6/PP nanocomposites remarkably.

ACKNOWLEDGEMENT

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