

The Effect of Organo-Montmorillonite on the Compatibility and Properties of Nylon 66/Polypropylene Blend

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ABSTRACT: Nylon 66/polypropylene (PP) blends containing maleated polypropylene and organo-montmorillonite (OMMT) have been prepared by melt compounding using a well-dispersed master-batch of nylon 6/OMMT nanocomposite as the source of OMMT. The effects of OMMT platelets on the compatibility and properties of the blends have been investigated. The blend morphology has been observed by the use of field emission scanning electron microscopy, showing a sharp decrease in domain size. The dispersion and location of OMMT have been investigated by X-ray diffraction and transmission electron microscopy. A high-density

occupation of OMMT at the interface (i.e. Nylon-g-PP inter-phase region) is revealed by Fourier transform infrared spectroscopy and thermogravimetric analysis of the extraction residue after the removal of nylon phase by formic acid. A significant gain in stiffness is realized with the use of OMMT while the toughness of the material is maintained. The possible mechanism of compatibilization also is discussed. © 2009 Wiley Periodicals, Inc. *J Appl Polym Sci* 115: 3697–3704, 2010

Key words: nylon 66; polypropylene; organo-montmorillonite; nanocomposites; compatibilization

INTRODUCTION

Polymer blending is one of the common methods to modify the properties of single polymers and produce new materials with enhanced properties. However, most polymer blends are thermodynamically immiscible because of the large unfavorable enthalpy and thus, phase separation often occurs, resulting in poor mechanical properties. Physical compatibilizers routinely are added to the immiscible blend systems to improve interfacial adhesion and reduce phase separation.

The recent trend is to explore organoclay as a compatibilizer for immiscible polymer blends. The compatibilization effect of organoclay, apart from being just a nano-filler, has been observed in various immiscible polymer blends such as nylon 6/PP,^{1–10} nylon 6/POE,^{11,12} PS/PP,^{13,14} and PS/PMMA.^{15–21} The size of the dispersed phase is dramatically reduced. Reduced interfacial tension, single T_g , and enhanced mechanical properties also are reported. Organoclay usually is located in one phase or at the interface of the blends, depending on the strength of the interaction with the two blend polymers. It seems that if a strong interaction exists between organoclay and one

of the polymers, then the organoclay would be preferably located in that polymer phase, no matter which phase (sea or island) it is. Otherwise, organoclay will be located at the interface. For example, in most nylon 6-based blends, organoclay is located in nylon 6 phase because of a strong affinity to nylon 6, although often a great density of organoclay is observed at the interface.^{1,2} In PS/PMMA and PC/SAN systems,¹⁶ where the organoclay–polymer interaction is favorable, but not very strong, for only one (the former) or both (the latter) of the components, organoclay is located at the interface.

As to the mechanism of the compatibilization brought by organoclay, there are mainly two viewpoints. One considers organoclay as physical barrier,^{11,22} which prevents coalescence of the dispersed phases during mixing. The other proposes the formation of in situ (polymer) grafts during mixing,^{16,23} which are unstable in either of the phases and thus localized at the interface of the blends. The grafts can act the same way as block copolymers, effectively decreasing the interfacial tension and reducing the domain size.

Nylon 66/PP is a typical example of immiscible polymer blend and also an important blend in both industry and academia. Although the compatibilization effect of organoclay on nylon 6/PP blend has been extensively investigated,^{1–10} there is no report involving effect of organoclay on the compatibility

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and properties of nylon 66/PP blend. Both nylon 6 and nylon 66 are versatile engineering plastics. It is commonly considered that clay is more difficult to get exfoliated in nylon 66 than in nylon 6 because of the structural difference.²⁴ González et al.²⁵ have used nylon 6/OMMT nanocomposite as a master-batch to produce exfoliated nylon 66/OMMT nanocomposites.

In this work, the role of organoclay as a compatibilizer for nylon 66/PP blend is investigated. The melt compounding process is used to prepare the nylon 66/PP/OMMT nanocomposites. A master-batch of nylon 6/OMMT is taken as the source of OMMT to facilitate dispersion of OMMT. Traditional compatibilizer MAH-g-PP (MAPP) is added to assist the compatibilization. Melt flow index (MFI) and formation of MAPP were 2.1 g/10 min and 1.2 wt % MAH. The phase morphology is observed by field emission scanning electron microscopy (FESEM). The dispersion and location of OMMT are studied by the use of X-ray diffraction (XRD) and transmission electron microscopy (TEM). High-density location of OMMT at the interface is further confirmed by dissolution of the nylon phase with formic acid, followed by analyzing the residue with Fourier transform infrared spectroscopy (FTIR) and thermogravimetric analysis (TGA). The mechanical properties of the nanocomposites are investigated. A direct melt compounding process, where a mixture of nylon 66, PP, OMMT, and MAPP, are melted mixed with the use of methyl methacrylate (MMA), as an intercalation agent,^{26,27} is also used for the purpose of comparison.

EXPERIMENTAL

Materials

Nylon 66 is Zytel[®] 101L (Dupont, USA) and Nylon 6 is B3S (BASF, Germany). PP was purchased from Yan shan Petroleum, China (trade mark: 1300) with a melt flow index of 2.5 g/10 min and a density of 0.9 g/cm³. MAPP was prepared in our laboratory with a content of maleic anhydride of 1.2 wt % and miscible with PP. The OMMT was purchased from Zhejiang Huate Group, China (NB-901). It is a montmorillonite modified with tallow quaternary ammonium (30 wt %) to increase the interlayer spacing. The cation exchange capacity (CEC) of the OMMT is 95 mequiv/100 g. MMA is of analytical grade purchased from Beijing Yilishiji Corp. (China).

Preparation of nylon 66/PP/OMMT nanocomposites

A master-batch of nylon 6/OMMT having 10 wt % of OMMT was prepared by melt compounding with MMA as an intercalation agent. After a preparatory mixing process, PP (20 wt %), MAPP (10 wt %), nylon 66, and nylon 6/OMMT master-batch were

melt compounded by the use of a TE-34 twin-screw extruder (Keya Co., China), with a screw speed of 120 rpm. The twin-screw extruder had a screw diameter of 34 mm and an *L/D* value of 28. The blending temperature profile was set to 250–260–260–265–265–240°C. The amounts of nylon 66 and nylon 6/OMMT master-batch were adjusted so that total amount of nylons were 70 wt % and the OMMT content was 1, 3, and 5 wt % (relative to nylons). The nanocomposites were coded as NP1, NP3, and NP5, respectively. For comparison, nylon 66/PP/MAPP/OMMT nanocomposite, which had the same proportion of nylon/PP/MAPP (7 : 2 : 1) and 5 wt % of OMMT (relative to nylon), was prepared by direct melt mixing of all the components with MMA as an intercalation agent, and the sample was coded as DM5. Nylon 66/PP/MAPP (7 : 2 : 1) blend also was prepared under similar conditions and named as NP0.

Characterization

XRD patterns were obtained on injection-molded Izod bars by the use of a BRUKER D8-Advance diffractometer connected to a computer. The diffraction scans were collected at 1.5°–10° by the use of a scanning speed of 1°/min. TEM was performed on a Cambridge S250MK3 with an accelerating voltage of 200 kV. For TEM examinations, ultra-thin specimens with a thickness of about 50 nm were cut from the middle section of the injection-molded bars. Cutting operations were conducted by the use of a Reichert Ultracut S microtome under cryogenic conditions, and the film was stained with RuO₄ vapor then retrieved onto copper grids. FESEM was performed on a JEOL model JSM-7401 apparatus with an operating voltage of 1 kV. The samples were fractured by impact test and observed. The average particle sizes were obtained by analyzing FESEM photos by the use of an electronic microscope photo processor.²⁸

FTIR and TGA were used to confirm the existence of OMMT in the interfacial region by analyzing the residue after removal of the nylon phase, because OMMT do not exist in the PP phase as evidenced by TEM. Thus, the nylon 66/PP/OMMT nanocomposite NP5 was treated with formic acid for 12 h at room temperature. Nylon phase was removed by repetitive washing with formic acid. The residue after drying was subjected to FTIR and TGA measurements. FTIR analysis was conducted on a NICOLET 560 instrument with compression-molded films (thickness was ~ 0.1 mm). TGA was conducted on a TGA 2050 at a heating rate of 10°C/min under nitrogen atmosphere, from room temperature to 900°C.

Tensile and flexural tests were performed with a GT-TCS 2000 machine (Gotech), according to ASTM

D638 and ASTM D790 at a crosshead speed of 50 mm/min and 5 mm/min, respectively. Izod impact tests were conducted on notched specimens with a GT-7045-I Izod impact tester (Gotech) according to ASTM D256. Before mechanical tests, all sample bars were dried in a vacuum oven at 110°C for dry-testing and were placed at a humidistat of 60 RH at 23°C for wet-testing.

RESULTS AND DISCUSSION

Phase morphology of nylon 66/PP blends containing OMMT

Figure 1 shows FESEM pictures of the fractured surfaces of the nylon 66/PP blends containing different amounts of OMMT. Nylon 66/PP blend is known as an immiscible polymer blend having typical sea-

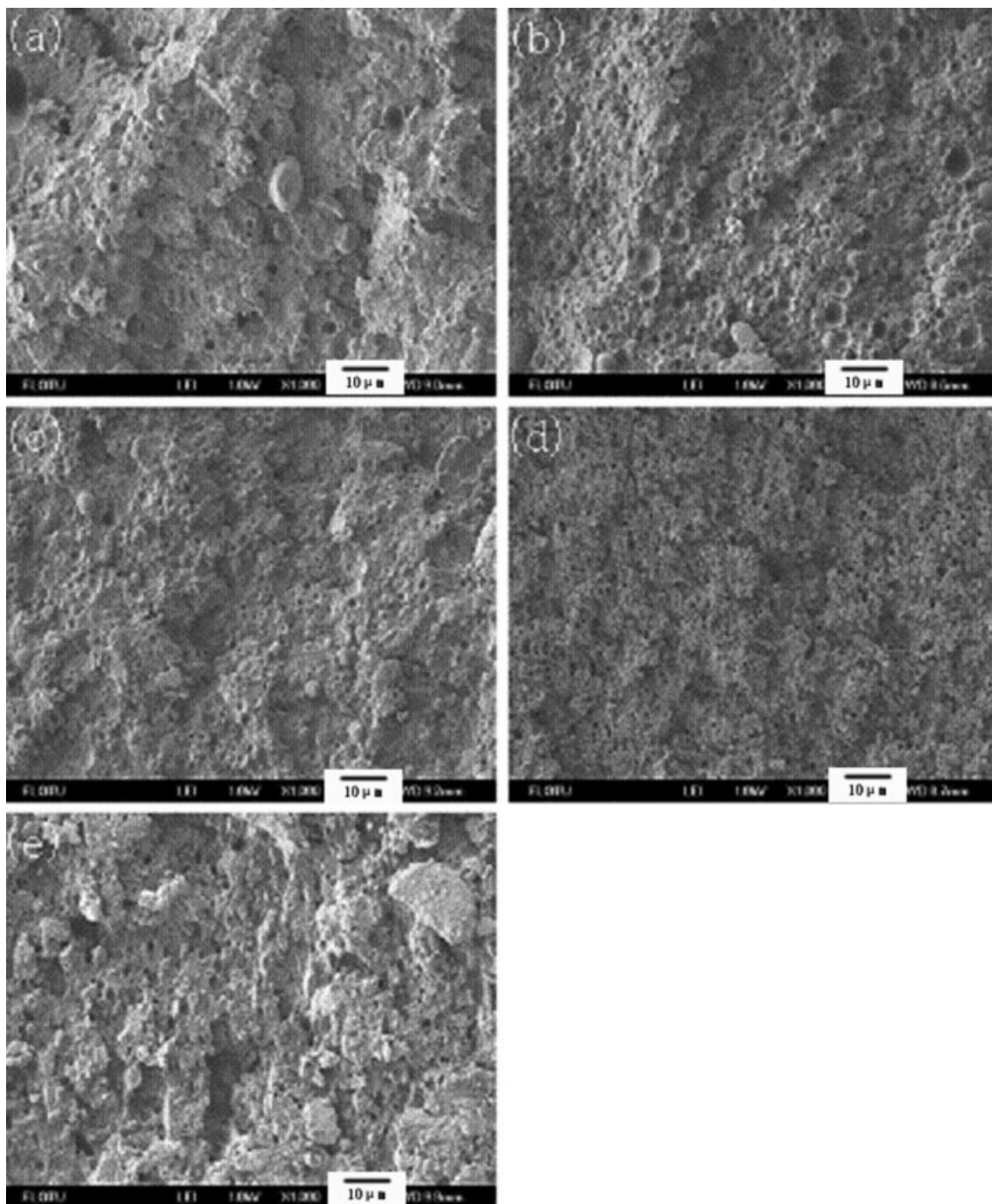


Figure 1 FESEM micrographs of the fracture surfaces of nylon 66/PP/OMMT nanocomposites: (a) NP0; (b) NP1; (c) NP3; (d) NP5; and (e) DM5.

island morphology. The average size of the dispersed PP phase is big ($3.9\ \mu\text{m}$), and the size distribution is wide [Fig. 1(a)], even in the presence of the classical compatibilizer MAPP. When a small amount of OMMT (1 wt %) is added to the blend, the particle size does not change much [Fig. 1(b)]. However, the average particle size decreases significantly, to 2.9 and $2.0\ \mu\text{m}$, respectively, when OMMT content is increased to 3 and 5 wt % [Fig. 1(c,d)]. The more OMMT is added, the smaller the particle size becomes. The reduction in particle size is related to reduced coalescence and thus is an indication of improved compatibility between the two blend polymers attributed to the presence of OMMT. As for DM5 [Fig. 1(e)], the average particle size is $2.9\ \mu\text{m}$, which is larger than in NP5. This difference indicates that the compatibilization effect of OMMT is more pronounced in master-batch approach than in direct mixing approach.

Dispersion and location of OMMT

The dispersion of OMMT is evaluated by a combination of XRD and TEM. The latter also can provide information about the location of OMMT. Figure 2 shows XRD patterns of the three nanocomposites prepared by master-batch approach with different OMMT contents (NP1, NP3, and NP5) and that obtained from direct melt mixing approach (DM5). XRD patterns of OMMT and NP0 also are shown for comparison. The XRD pattern of commercial OMMT (curve a) shows a broad peak centered at $2\theta = 4.3^\circ$, corresponding to a basal spacing of 2 nm as calculated from Bragg equation. No obvious peak was observed for NP0 and NP1, suggesting possible exfoliation of OMMT in NP1. However, a weak broad "peak" at 2° – 6° was observed for NP3 and NP5, indicating that a portion of OMMT is only intercalated with various degrees of intercalation when OMMT content is increased to 3 and 5 wt %. As for DM5, a weak broad peak at 4° – 6° along with

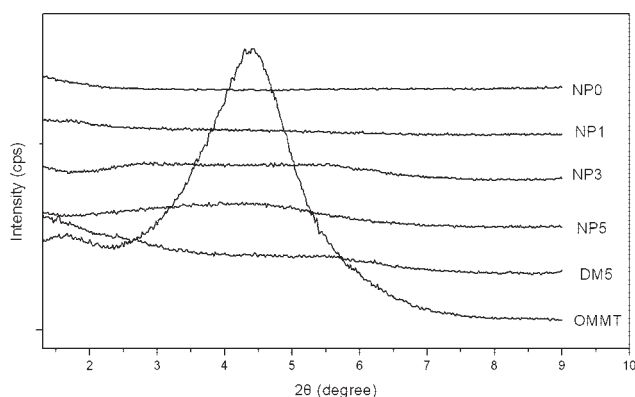


Figure 2 XRD patterns of OMMT, NP0, and nylon 66/PP/OMMT nanocomposites NP1, NP3, NP5, and DM5.

the strong declining tendency of the spectrum at 1° – 4° suggests intercalated OMMT structure with various degrees of intercalation.

Figure 3 gives TEM photos of nylon 66/PP blends having 5 wt % of OMMT prepared by either master-batch or direct mixing approach (sample NP5 and DM5) at different magnifications. Figure 3(c,d) corresponds to Figure 3(a,b), respectively. Under low magnification [Fig. 3(a,b)], phase separation is clearly observed. The white particles are PP phase, and the grey background is the nylon phase. The particle size with the master-batch approach [0.2 – $1\ \mu\text{m}$, Fig. 3(a)] is smaller than that with the direct mixing approach [0.5 – $2\ \mu\text{m}$, Fig. 3(b)], which is a finding similar to our FESEM observation. Under higher magnification [Fig. 3(c,d)], individual platelets or stacks containing a few platelets are observed as dark lines in grey region and at the interface for both samples, indicating that OMMT is in partial exfoliated and partial intercalated state and either in nylon phase or at the interface. No OMMT platelets are found in PP phase.

FTIR and TGA of the extraction residue

To evaluate the amount of OMMT located at the interface, the nanocomposite having 5 wt % of OMMT (NP5) and the control sample nylon 66/PP/MAPP (7/2/1) blend without OMMT (NP0) were extracted with formic acid to remove the nylon phase. The residues are coded as NP5-ext and NP0-ext, respectively. Because OMMT is in nanoscale dispersion and has a good interaction with nylon, OMMT located in nylon phase should be removed along with nylon by extraction with formic acid, although OMMT alone does not dissolve in formic acid. To demonstrate this point, a sample of nylon 66/OMMT nanocomposite with nanoscale OMMT dispersion was dissolved in formic acid. The resulting homogeneous stable sol can completely pass through a sintered filter or filter paper, and no OMMT residue was observed. Because there is no OMMT in PP phase as observed by TEM, the OMMT remained in the residue will be that located in the interphase region. Nylon-g-PP can not dissolve in formic acid and thus not be removed by extraction with formic acid.

Figure 4 shows FTIR spectra of NP5-ext and NP0-ext and also that of PP for comparison. The spectrum of NP0-ext resembles that of PP, except that a small amount of nylon is observed at 1550 , 1640 , and $3300\ \text{cm}^{-1}$ because of incomplete extraction.²⁹ Compared with NP0-ext, NP5-ext has three extra peaks at 470 , 525 , and $1000\ \text{cm}^{-1}$, attributable to the vibrations of Si–O–Al and Al–O–H bonds, indicating the presence of OMMT in the extraction residue.³⁰ FTIR spectrum of NP5 demonstrates

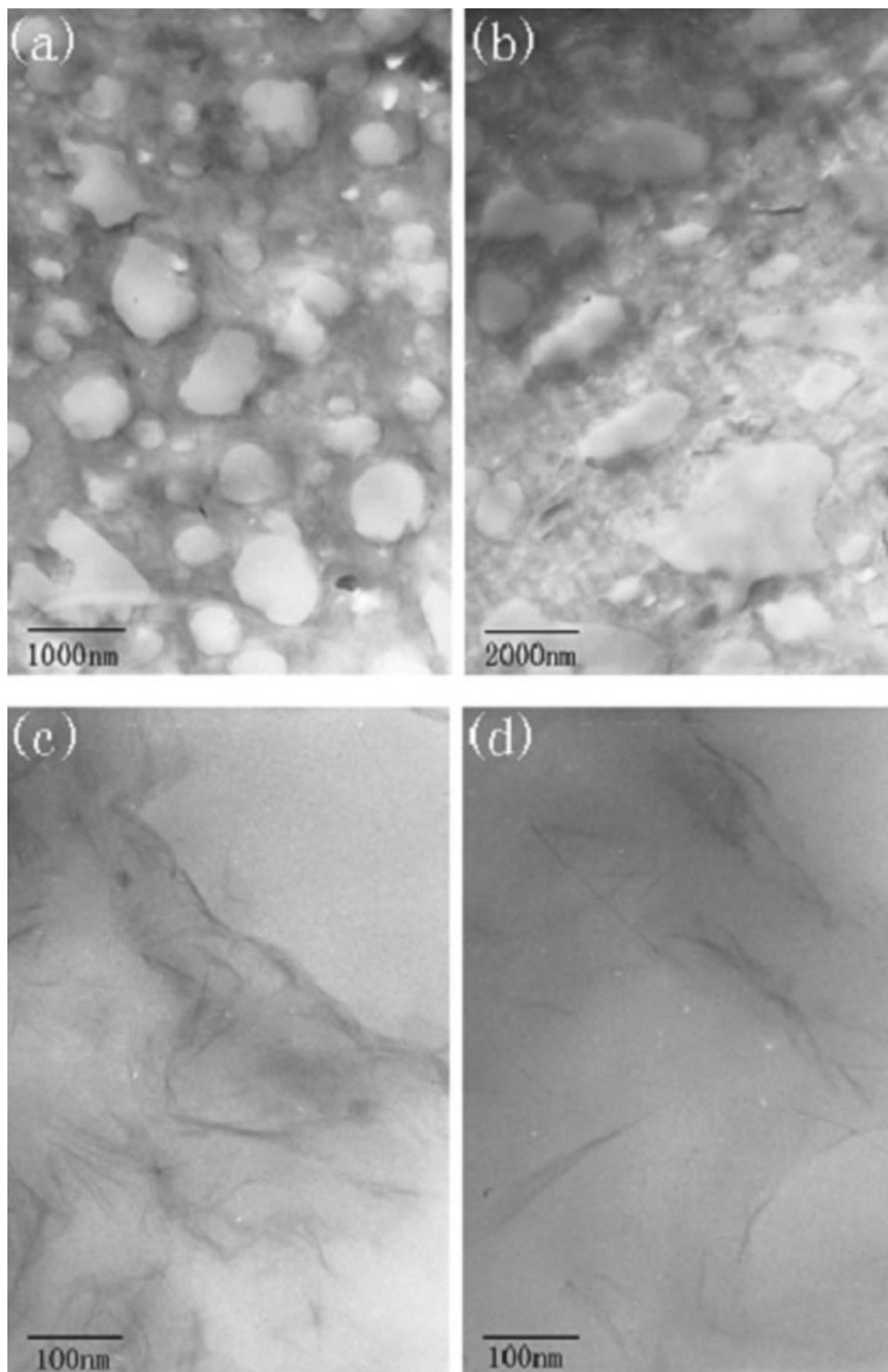


Figure 3 TEM micrographs of nylon 66/PP/OMMT nanocomposite: (a) low magnification of NP5; (b) low magnification of DM5; (c) high magnification of NP5; (d) high magnification of DM5.

qualitatively the presence of OMMT in the extraction residue.

TGA is used to quantify the amount of OMMT in the residue. Figure 5 shows the TGA curves of NP5-ext and NP0-ext. PP and nylon decompose at 300–500°C. The decomposition residue at 600°C for NP0-ext is nearly 0 as predicted, and the small amount of

residue (0.7 wt %) is caused by incomplete combustion, whereas that for NP5-ext is 4.6 wt %, which can be taken as MMT content in the extraction residue because almost all the organic components should be burned up. Accordingly, the amount of MMT at the interface is estimated to be 1.4 wt % ($4.6 \text{ wt } \% \times 30\%$) to total resin, corresponding roughly to 60 wt % of the

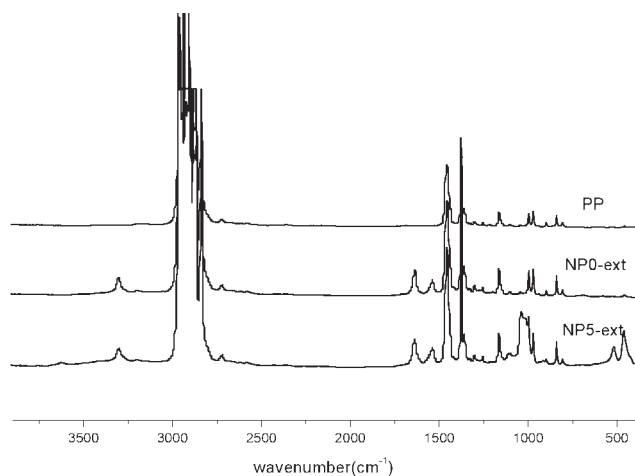


Figure 4 FTIR spectra of PP, NP0-ext, and NP5-ext.

total MMT used in the preparation (note that OMMT content is 5 wt % to nylon, i.e., 3.5 wt % to total resin, the real MMT content is 2.3 wt % after deduction of the organic intercalation agent). This clearly indicates a much higher density occupation of OMMT at the interface than in inner nylon phase. This finding is somehow in agreement with the case of nylon 6/PP/MAPP/OMMT nanocomposites reported by Chow et al., where OMMT is located in nylon 6 phase and nylon 6-g-PP interphase region (i.e. the interface)¹ and likely to have a preference to localize in the interphase region.³¹ The location and the compatibilization mechanism of OMMT in the current blend system may be explained by a combination of the two main existing viewpoints mentioned in the introduction part. First, OMMT has a much higher affinity to nylon than to PP, and it is more likely to be in nylon phase and play a role of physical barrier to prevent coalescence of PP phases. Second, because the nylon-g-PP copolymer is known to form at the interface by reaction between the anhydride group of MAPP and the amine (primary and secondly) of nylon during melt mixing, intercalation of nylon-g-PP, apart from intercalation of nylon, is likely to happen as a result of the

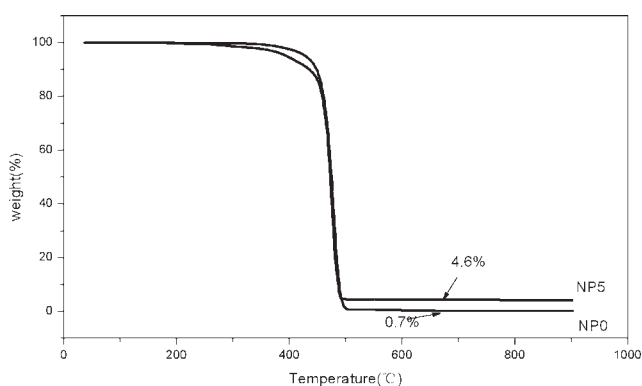


Figure 5 TGA patterns of NP0-ext and NP5-ext.

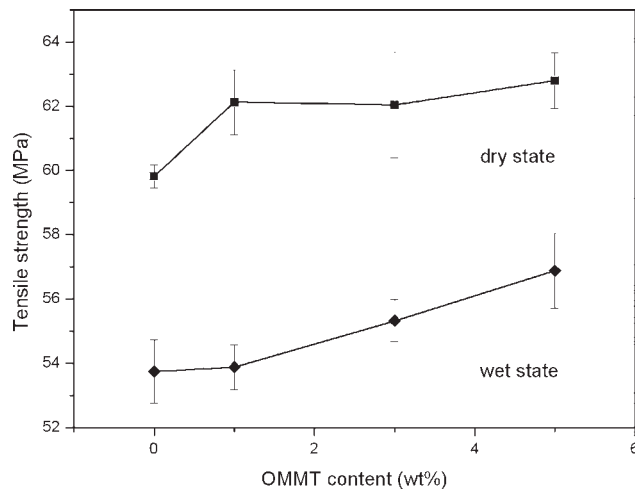


Figure 6 Effect of OMMT content on tensile strength of nylon 66/PP/OMMT nanocomposites.

interaction between the amide group of nylon and the ammonium group of the intercalation agent or the hydroxyl group of MMT through hydrogen bonding. Furthermore, intercalation of MAPP is also likely to happen because of the interaction between the anhydride group of MAPP and the hydroxyl group of MMT through hydrogen bonding. The OMMT intercalated simultaneously by nylon, nylon-g-PP, or MAPP molecules are unstable in either PP or nylon phase and thus localizes in nylon-g-PP interphase region, and acts the same way as block copolymers, effectively playing the role of compatibilizers.

Mechanical properties of nylon 66/PP/OMMT nanocomposites

Figures 6–11 provide the mechanical properties of nylon 66/PP/OMMT nanocomposites having different amounts of OMMT in terms of tensile strength, tensile modulus, elongation at break, flexural

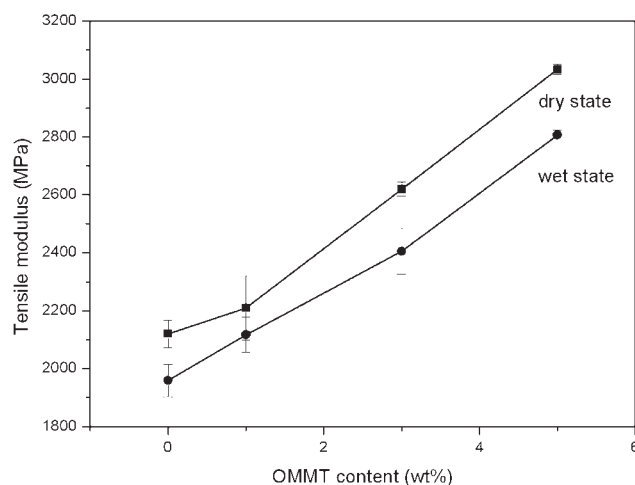


Figure 7 Effect of OMMT content on tensile modulus of nylon 66/PP/OMMT nanocomposites.

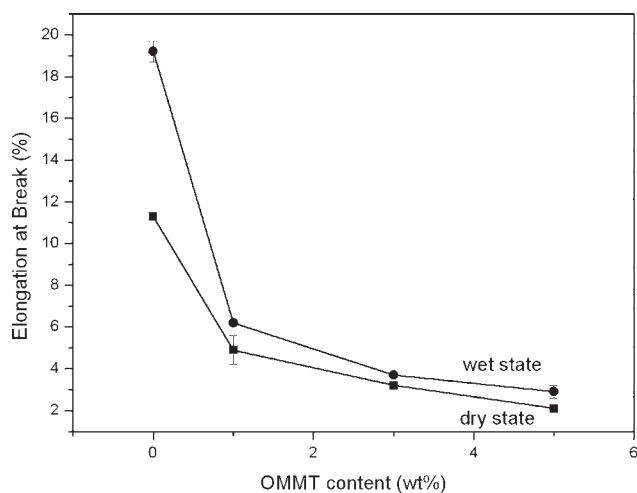


Figure 8 Effect of OMMT content on elongation at break of nylon 66/PP/OMMT nanocomposites.

strength, flexural modulus, and impact strength measured at both dry and wet states. The tensile strength increases slightly, tensile modulus increases significantly and elongation at break decreases sharply with the increasing OMMT content at both dry and wet states. The flexural strength measured at dry state increases significantly with increasing OMMT content, whereas at wet state decreases with a small addition of OMMT (1 wt %), and then begins to raise and reaches a much higher value with 5 wt %. The flexural modulus behaves similarly to the flexural strength. A significant gain in flexural modulus is observed at both dry and wet states with 5 wt % of OMMT. This seems reasonable because a significant improvement in stiffness is always expected when a hard inorganic filler is added to a polymer. The impact strength measured at both dry and wet states behaves similarly with the addition of OMMT. With a small OMMT content (1 wt %), it decreases dramati-

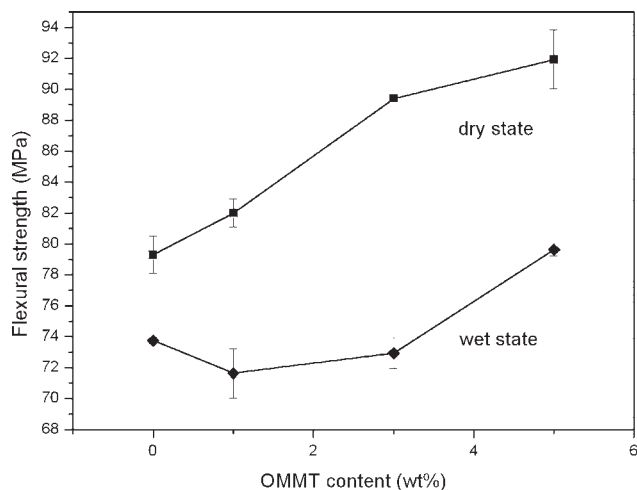


Figure 9 Effect of OMMT content on flexural strength of nylon 66/PP/OMMT nanocomposite.

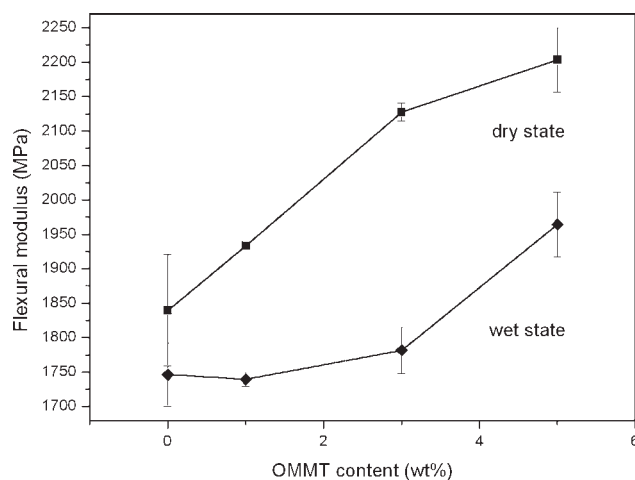


Figure 10 Effect of OMMT content on tensile modulus of nylon 66/PP/OMMT nanocomposite.

cally. When the OMMT content increases to 3 and 5 wt %, the impact strength increases significantly and approaches the value of the control blend sample when measured at wet state. Because OMMT is playing the role of nanofiller and compatibilizer at the same time, the values of impact strength should be considered as a consequence of the combined effects. When OMMT content is small (1 wt %), the compatibilizing effect is modest as observed by FESEM, and an addition of a hard filler to polymer matrices often brings a decrease in toughness; therefore, the impact strength of the blend composite decreases dramatically. When OMMT content is bigger (3 and 5 wt %), the compatibilization effect is obvious. The smaller PP particle size increases the interfacial area between PP and nylon, allowing easier absorption of impact energy. As a result, the impact strength increases significantly and approaches that

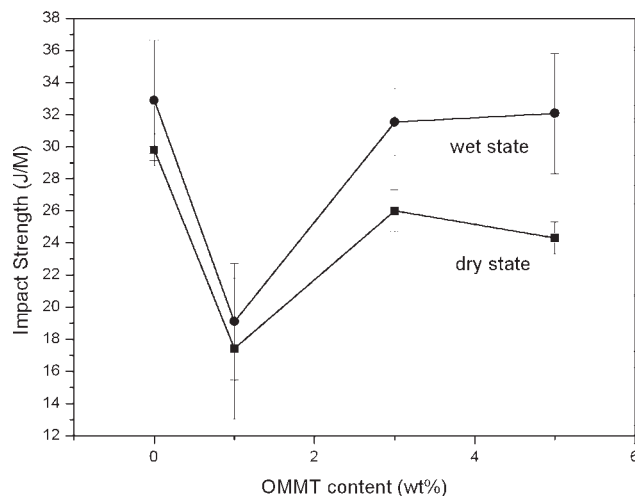


Figure 11 Effect of OMMT content on impact strength of nylon 66/PP/OMMT nanocomposite.

of the control sample without OMMT. The retention of toughness with OMMT content of 3 and 5 wt % may also be related to more addition of the tougher nylon 6 which is added to the system along with OMMT.

The mechanical properties of the nanocomposite prepared by direct melt mixing approach (DM5) are similar to those of NP5 discussed previously in this work except that there is a sharp decrease in impact strength (19.7 J/M compared with 32.1 J/M of NP5), which could be understood by comparing the degree of compatibilization efficiency of OMMT in the two samples. The compatibilization efficiency is lower in DM5 than in NP5, as observed from FESEM. It is common sense that a higher degree of compatibilization favors toughness of the blend material. Therefore, the toughness of NP5 is much better than that of DM5 because of higher compatibilization efficiency brought by OMMT.

CONCLUSIONS

OMMT can play the role of compatibilizer in nylon 66/PP blend prepared by melt compounding using nylon 6/OMMT master-batch as the source of OMMT. The size of the dispersed PP phase is dramatically reduced with only 3–5 wt % of OMMT, which is in nanoscale dispersion in nylon phase and at the interface. About 60 wt % of OMMT is localized at the interface. The possible reasons for the location and compatibilization mechanism of OMMT are discussed based on a combination of the two main existing viewpoints. Interaction of OMMT with nylon, Nylon-g-PP and MAPP through hydrogen bonding is thought to be the most important factor for intercalation of those macromolecules into OMMT galleries, which is decisive for the location and compatibilization effect of OMMT. The increased compatibility brought by OMMT is also reflected by a significant increase in stiffness without sacrifice of toughness.

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