

# Mechanical Properties and Morphologies of PP/mPE/Filler Composites

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**ABSTRACT:** Because of the poor impact behavior of polypropylene (PP) at low temperatures, the blending of PP with metallocene-polymerized polyethylene (mPE) elastomers was investigated in this study. However, a reduced modulus of the overall blend was inevitable because of the addition to elastomers. To obtain a balance of the properties, we introduced rigid inorganic fillers to PP/mPE blends. The performance of the composites was characterized with tensile and Charpy notched impact tests, and the fracture morphology was examined with scanning electron microscopy. The results showed that the effects of fillers in a brittle matrix and in a ductile matrix were quantitatively different. For PP/mPE/filler ternary composites, the dependence of Young's modulus and yield strength on CaCO<sub>3</sub> content was not significant compared with that of PP/filler binary composites, whereas the elongation at break and tensile toughness at room temperature for PP/mPE/filler systems were more improved. The impact strength of the PP/mPE blends filled with untreated glass beads and CaCO<sub>3</sub> at a low temperature was lowered because of the weak interfacial bond. However, the values of the impact strength of the PP/mPE/filler composites at a low temperature remained at a high level compared with that of pure PP. In particular, a PP/mPE blend filled with surface-treated kaolin had a higher low-temperature impact toughness than the unfilled blend. © 2002 John Wiley & Sons, Inc. *J Appl Polym Sci* 83: 3029–3035, 2002; DOI 10.1002/app.2333

**Key words:** poly(propylene) (PP); metallocene polyethylene (mPE); toughening modification; fillers; composites

## INTRODUCTION

It is generally known that polypropylene (PP) has a rather low fracture toughness at low temperatures and a high notch sensitivity at room temperature. Compounding PP with a dispersed elas-

tomeric phase [e.g., ethylene–propylene–diene rubber (EPDM)] is widely practiced<sup>1–5</sup> because the rubber can increase the overall toughness of the PP matrix.<sup>6</sup> However, the addition of elastomers often results in damage to some properties of PP, such as stiffness and hardness.

The development of metallocene catalysts has led to numerous new polyolefinic materials, among which polyolefin elastomers are extremely attractive for both the rubber and plastics industries. Metallocene-polymerized polyethylene (mPE) elastomers polymerized with octene as a comono-

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mer possess a very homogeneous copolymer distribution and a narrow molecular mass distribution.<sup>7</sup> In comparison with conventional EPDM, mPE can accomplish a higher knit-line impact strength as a modifier of PP.<sup>8</sup> In addition, previous work has shown an improved fracture behavior at low temperatures and better dynamic properties with an mPE modifier.<sup>9</sup> Although mPE provides many advantages with respect to the toughening effect and processability in PP modification, it also unavoidably reduces the stiffness of PP like most conventional elastomers.

For engineering plastics, high stiffness and toughness are often required, characterizing the performances of structural materials. Through modification, general-purpose PP can serve as an engineering plastic that is widely used in automobile and electrical equipment and transportation. The toughness of PP can be improved dramatically through the addition of elastomers, but the stiffness and strength are often significantly reduced. Therefore, the optimum overall properties cannot be achieved only with changes in the PP/elastomer blending ratio. Fibers can be used for enhancing both the stiffness and strength properties of the blend,<sup>10,11</sup> but fiber-reinforced composites often display an anisotropy of properties. Another interesting way is the addition of rigid inorganic particles into the PP/elastomer blend.<sup>12–17</sup> This article describes the observed mechanical properties and morphology of PP/mPE blends filled with different fillers.

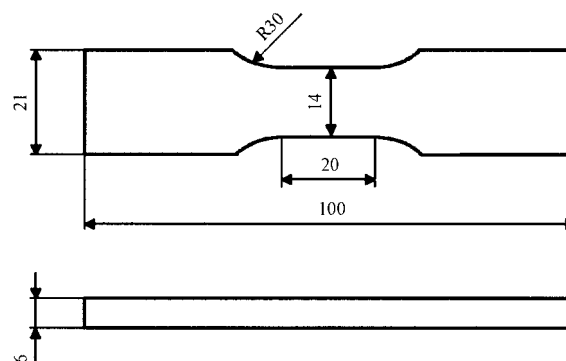
## EXPERIMENTAL

### Materials

The PP used was a commercial block copolymer [Vestolen P9500;  $d = 0.901 \text{ g/cm}^3$ , MFI (190/5) = 0.5 g/10 min] supplied by Vestolen/DSM (Gelsenkirchen, Germany). The mPE sample used was Engage EG8842 [ $d = 0.857 \text{ g/cm}^3$ , MFI (190/5) = 1.5 g/10 min] produced by DuPont/Dow Elastomers SA (Geneva). Three kinds of inorganic fillers were used: CaCO<sub>3</sub> (Millicarb;  $d_{50} = 5.5 \mu\text{m}$ ; OMYA GmbH), kaolin (Translink, HF-900; 1.8  $\mu\text{m}$ , surface-treated; Engelhard Co.), and glass beads (3M, K20; 11–125  $\mu\text{m}$ ; Scotchlite Glass Bubbles).

### Blend Preparation

Blend samples were prepared by the compounding of elastomers, PP, and fillers in a corotating



**Figure 1** Sample geometry (dimensions in millimeters).

twin-screw extruder (Leistritz LSM 30/34 GL; mass temperature = 200°C, screw speed = 100 min<sup>-1</sup>). The extruder was equipped with two feeding hoppers that could quantitatively feed the materials to the extruder. PP and mPE were first mechanically mixed and then put into one hopper, whereas the filler was put into another hopper. The extruder was cooled in a water bath at room temperature, cut into small granules, and then dried for 2 h at 70°C.

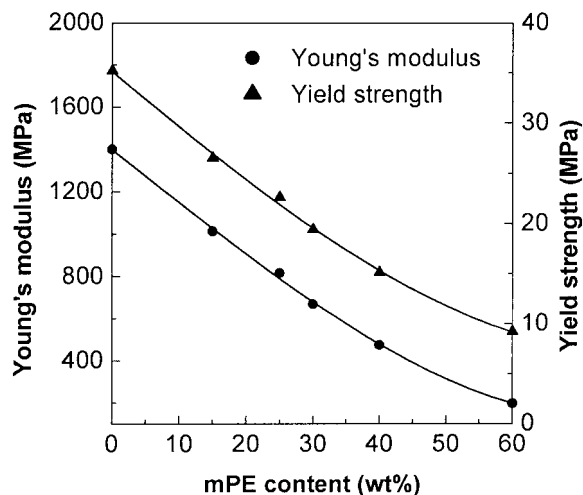
The samples for tensile tests were made with injection molding (Arburg Allrounder 370V), with a mass temperature of 200°C and a mold temperature of 40°C. The specimen geometry is shown in Figure 1.

### Testing Equipment and Procedures

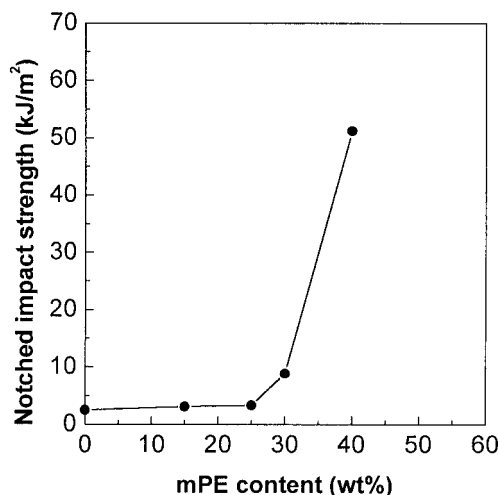
A Zwick 1465 universal testing machine was used in the tensile tests. Measurements were carried out at room temperature and 50 % relative humidity, with a crosshead speed of 200 mm/min. The reported tensile properties were the mean values of six specimens.

To determine the response of the materials under high-speed loading, we measured the Charpy notched impact strength at -30°C for the evaluation of the effectiveness of the different components with increasing fracture toughness of PP. The bars (10 mm × 6 mm × 80 mm) were notched in a V form for the measurements. The remaining depth was 8 mm, and the notch-tip radius was 0.25 mm.

The morphology of the blends was determined with scanning electron microscopy (SEM; Cam Scan type CS 24). Fracture surfaces taken from the impact fracture section were examined. The specimens were coated with gold/palladium before SEM observations.



**Figure 2** Young's modulus and yield strength of PP/mPE blends as a function of elastomer content.



**Figure 3** Charpy notched impact strength ( $-30^{\circ}\text{C}$ ) of PP/mPE blends as a function of elastomer content.

## RESULTS AND DISCUSSION

### PP/mPE Binary Blends

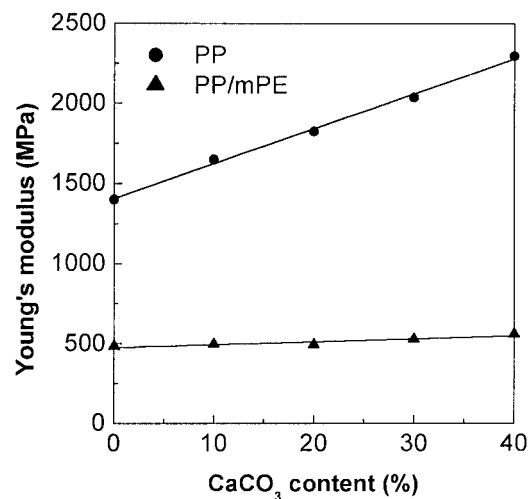
In Figure 2, the yield strength and Young's modulus, as measured in static tensile tests, are shown as dependent on the content of the elastomer component. As expected, with an increase in the elastomer content, both Young's modulus and the yield strength decreased significantly.

mPE with 20–35 wt % octene has a very low glass-transition temperature ( $-40^{\circ}\text{C}$ ) and low crystallinity, so it can be expected to serve as an effective modifier for increasing the toughness of PP at low temperatures. The Charpy notched impact strength determined at  $-30^{\circ}\text{C}$  is shown in Figure 3. The blend with 40 wt % mPE showed excellent low-temperature impact behavior, and with 60 wt % mPE, the blend could not be broken at all. Below a content of 30 wt %, mPE could not improve the low-temperature impact strength of PP. For a significant enhancement of the low-temperature impact toughness, the blends must contain at least 40 wt % mPE. However, the yield strength and Young's modulus of the blends with 40 wt % mPE were lowered 50% more compared with those of pure PP.

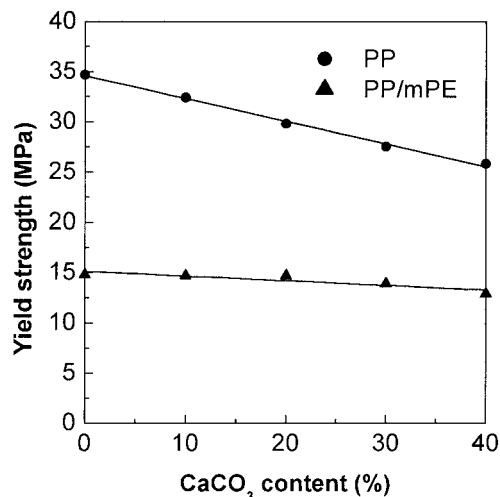
### PP/Filler and PP/mPE/Filler Composites

A PP/mPE blend containing 40 wt % mPE displayed good low-temperature toughness, whereas pure PP often showed brittle fracture behavior at a low temperature. We used untreated  $\text{CaCO}_3$  as

a filler to increase the stiffness; however, in brittle and ductile matrices, the fillers functioned rather differently. Figure 4 describes the relationship between Young's modulus and the filler content in both PP and PP/mPE blend matrices. Young's modulus is an important parameter, characterizing the stiffness of a material. For the PP brittle matrix, with an increase in the filler content, Young's modulus increased significantly in a linear form. This means that the addition of an inorganic filler can greatly enhance the stiffness of PP. For a PP/mPE blend ductile matrix in which the mPE content was fixed at 40 wt %,



**Figure 4** Young's modulus as a function of filler content.



**Figure 5** Yield strength as a function of filler content.

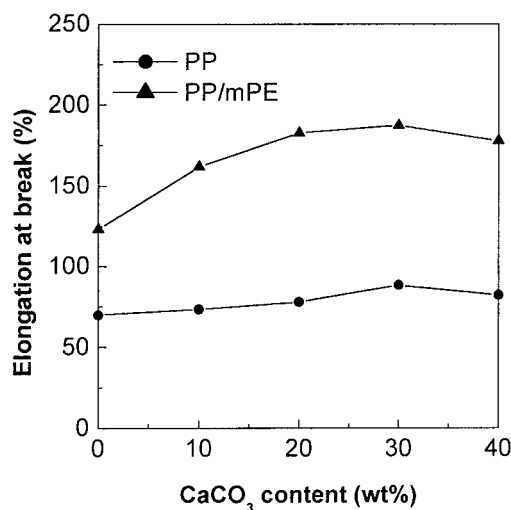
Young's modulus increased slightly. It is assumed that the stiffness and strength of a polymer blend are mainly provided by its continuous phase; however, in the PP/mPE blends, a large amount of filler was dispersed in the mPE phase. Compared with that of the pure PP matrix, the filler content in the PP phase of the PP/mPE blend was relatively low, so the stiffness of the PP/mPE/filler composites showed only a little improvement.

Among the mechanical properties, the yield strength of composites is of primary importance, determining the maximum allowable load without considerable plastic deformation. The depen-

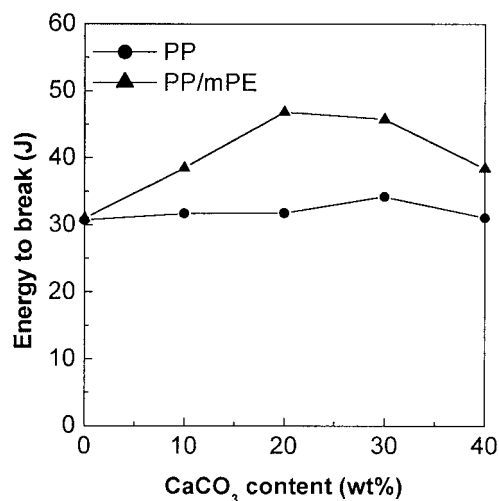
dence of the yield strength on the filler content is shown in Figure 5. With an increase in the filler content, the yield strength of the PP/filler composites decreased significantly, whereas the yield strength of the PP/mPE/filler composites did not decrease until to a filler content of 20 wt %.

The elongation at break generally reflects the ductility of materials, whereas the energy to break, the tensile fracture energy, is a very important parameter that is often used to characterize the tensile toughness of materials. The dependence of the elongation at break and energy to break are shown in Figures 6 and 7, respectively. The results show that with an increase in the filler content, the elongation at break and energy to break of the PP/filler composites slowly increased. For PP/mPE/filler composites, an increase in the filler content led to a significant increase in the elongation at break and tensile toughness described by the tensile fracture energy. This indicates that ductility and tensile toughness can be improved by the addition of an inorganic filler. It suggests that under tensile stress, the matrix layer around rigid particles produces an obvious plastic deformation to absorb a great deal of energy because of the stress concentration. Above a filler content of 30 wt %, the energy to break will be reduced because of the decrease in the elongation at break and tensile stress.

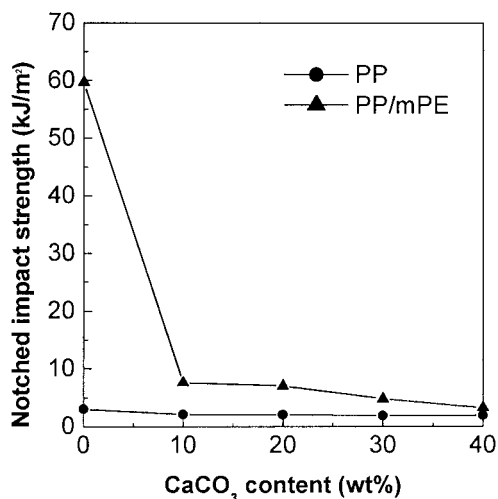
The influence of the filler content on the notched impact strength at  $-30^{\circ}\text{C}$  is shown in Figure 8. In both PP and PP/mPE blend matrices,



**Figure 6** Elongation at break as a function of filler content.



**Figure 7** Energy to break as a function of filler content.



**Figure 8** Charpy notched impact strength ( $-30^{\circ}\text{C}$ ) as a function of filler content.

small amounts of filler caused a sudden decrease in the low-temperature impact toughness, and then it decreased only a little with an increase in the filler content. When filler was added to the PP/mPE blend, the reduced impact strength led to a complete break of the specimen. However, the impact strength at a low temperature still remained above the value for the unfilled blends with less than 30 wt % mPE.

Table I illustrates the mechanical properties of a PP/mPE (60/40) blend filled with 10 wt % fillers of different kinds. With respect to the unfilled blend, the addition of fillers could enhance the elongation at break and energy to break. Among three filled composites, the PP/mPE/CaCO<sub>3</sub> system showed the largest elongation and tensile fracture energy, but its impact toughness at a low temperature was the lowest. Kaolin and glass beads could increase the tensile strength of the

blends and keep the impact strength at a high level. It is significant that the blend filled with kaolin showed a better low-temperature impact toughness than the unfilled blend; this happened because the surface-treated kaolin particles had good interfacial adhesion with the matrix. The results also show that, unlike in the pure PP matrix, for PP/mPE blends, a small amount of fillers could not significantly improve the stiffness of the blends.

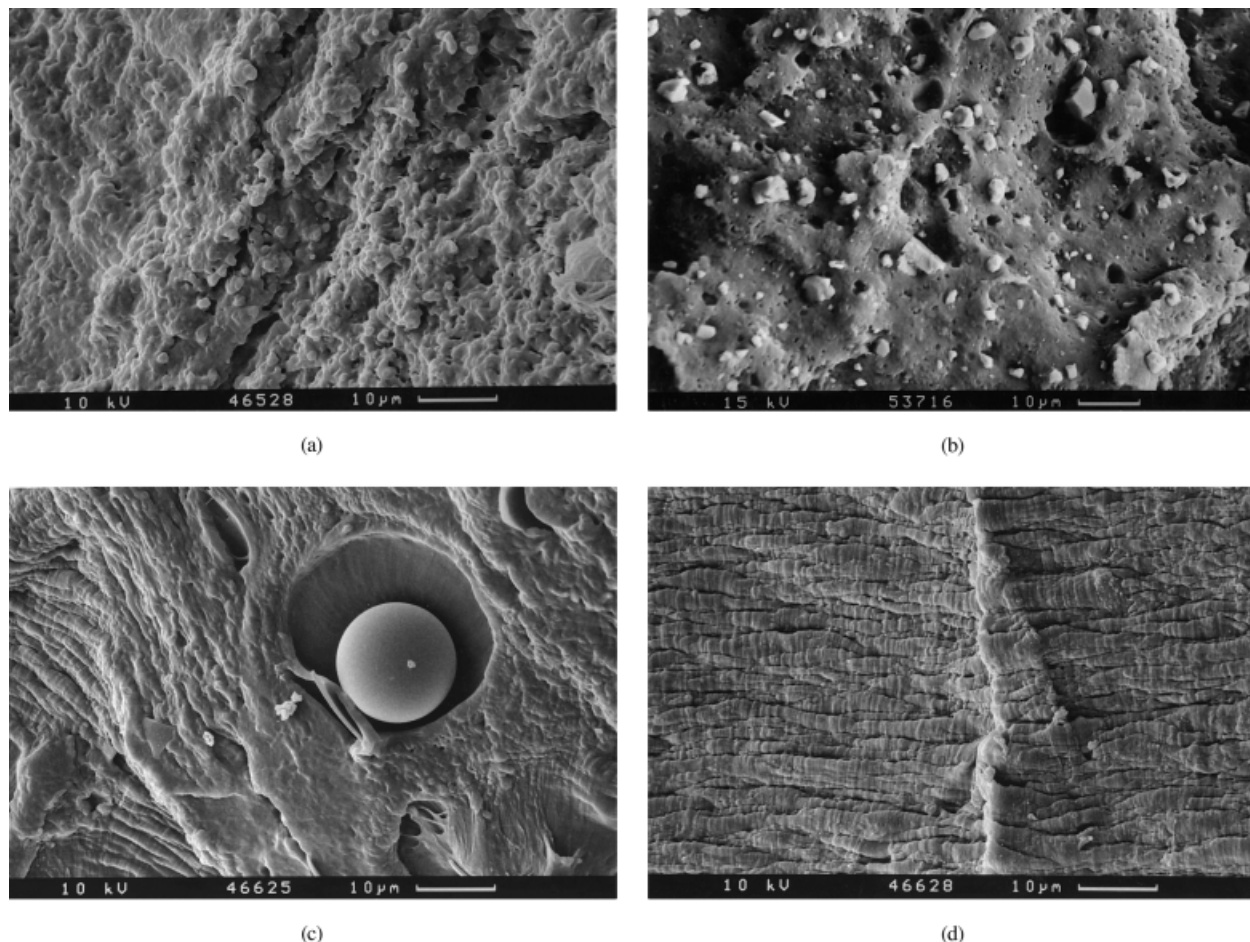
#### Morphology of PP/mPE Blends

It is well known that the properties of materials greatly depend on their morphological structure. For polymer blends or composites, the dispersion of the components is extremely important. Figure 9(a) shows an SEM micrograph of the fracture surface of a PP/mPE blend (60/40) fractured at  $-30^{\circ}\text{C}$ . The rough surface is a typical feature of ductile fracture. Figure 9(b) shows the low-temperature fracture surface of a PP/mPE (60/40) blend filled with 10 wt % CaCO<sub>3</sub>. The untreated CaCO<sub>3</sub>-filled PP/mPE blend had a relatively smooth fracture surface, and the coagulation of CaCO<sub>3</sub> and the debonding of filler particles from the matrix can be seen. This resulted in the poor impact strength of the PP/mPE/CaCO<sub>3</sub> composites. In Figure 9(c), although the untreated glass beads also displayed poor interfacial bonding with the matrix, the plastic deformation around the glass beads appears to be relative large, so the composites maintained a high level of impact strength. Figure 9(d) reveals that surface-treated kaolin had a very good filler-matrix interfacial adhesion, and kaolin particles were dispersed in lamellar form in the PP/mPE matrix. This may be the reason the kaolin-filled PP/mPE blend exhibited better impact performance at a low temperature.

**Table I** Mechanical Properties of PP/mPE Blends (60/40) Filled with Different Fillers

Filler in the Blends	Young's Modulus (MPa)	Yield Strength (MPa)	Elongation at Break (%)	Energy to Break (J)	Impact Strength (kJ/m <sup>2</sup> )
Without filler	484	14.8	123	29.5	59
CaCO <sub>3</sub> <sup>a</sup>	497	14.7	161	38.5	8
Glass beads <sup>a</sup>	507	15.4	126	31.1	19
Kaolin	492	15.5	129	32.5	65

<sup>a</sup> CaCO<sub>3</sub> and glass beads were untreated.



**Figure 9** SEM micrographs of PP/mPE blends (60/40) fractured at  $-30^{\circ}\text{C}$  by the impact test: (a) unfilled blend, (b) blend filled with 10 wt %  $\text{CaCO}_3$ , (c) blend filled with 10 wt % glass beads, and (d) blend filled with 10 wt % kaolin.

## CONCLUSION

The impact toughness of the PP exhibited an increase at low temperatures when compounded with mPE elastomers. The significant enhancement of the low-temperature impact strength was achieved only with a relatively high elastomer content. However, it inevitably led to a decrease in stiffness and strength in the polymer blend. With the synergistic effect of elastomers and fillers, a balance of mechanical properties was expected. For the rigid particle-filled polymer composites, the mechanical properties depended on the features of the matrix and filler, the filler content, and the interfacial morphology. The results showed that the inorganic fillers performed differently in the brittle matrix and in the ductile matrix.  $\text{CaCO}_3$  as a filler effectively enhanced the

stiffness of PP, but in the PP/mPE blend matrix, its reinforcing effect was not as significant.  $\text{CaCO}_3$  enhanced the tensile toughness both for pure PP and for the PP/mPE blend, whereas a small amount of  $\text{CaCO}_3$  resulted in a great decrease in the low-temperature impact toughness. However, at a high filler content, the impact strength of filled PP/mPE blends at low temperatures still remained higher than that of unfilled blends with less than 30 wt % mPE. The performances of different fillers in the PP/mPE blend matrix were not the same. PP/mPE/ $\text{CaCO}_3$  composites had a large elongation at break and low-temperature impact strength. PP/mPE/kaolin composites exhibited a better low-temperature impact toughness than the unfilled blend because of the surface treatment of kaolin particles. An SEM examination showed that unfilled PP/mPE

exhibited a ductile fracture. The fracture morphologies and interfacial structures of the PP/mPE/filler composites with different fillers were quite different.

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