Effect of Chemical Treatments on the Mechanical, Flow, and Morphological Properties of Talc- and Kaolin-filled Polypropylene Hybrid Composites

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ABSTRACT: This study was performed with commercially available phenyl trimethoxysilane (PTMS) and neoalkoxytitanate [i.e., neopentyl(diallyl)oxytri(dioctyl)phosphato titanate (LICA 12)] as coupling agents. PTMS and LICA 12 were used to treat talc and kaolin to compare their effects with untreated fillers upon incorporation into polypropylene (PP). Single-filler PP composites (containing either talc or kaolin) and hybrid-filler composites (containing a mix of both talc and kaolin) were compounded in a twin-screw extruder and subsequently injection-molded into dumbbells. The incorporation of PTMS and LICA 12 slightly decreased the tensile and flexural properties in terms of modulus and strength but increased the elongation at break for both single-filler and hybrid-filler composites. There was also a sig-

INTRODUCTION

Polypropylene (PP) is a commodity thermoplastic that has been widely used as a matrix material in many industrial applications because of its versatility for accepting numerous types of fillers and reinforcements, wide variety of physical properties, good mechanical property balance, good processing properties, and low cost. The use of PP composites in automotive and mechanical engineering has been increased mainly because of its excellent stiffness, which enables it to be substituted for conventional materials in demanding engineering applications.¹

The incorporation of fillers into thermoplastics is a common practice in the plastics industry for reducing the production costs of molded products. Fillers are also used to improve thermoplastic working properties, such as strength, rigidity, durability, and nificant improvement in the impact strength of the composites, particularly those treated with LICA 12. The hybrid composites, through the synergistic coalescence of positive characteristics from talc and kaolin with the aid from chemical treatment provided an economically advantageous material with mechanical properties comparable to those of the single-filler-filled PP composites. Further investigations on flow and morphological properties were also done to correlate the mechanical properties of the single- and hybrid-filler-filled PP composites. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 110: 2770–2779, 2008

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hardness.² Among particulate fillers for PP, talc has been used in the largest quantity in various applications.^{3–6} In the automotive sector, talc-filled PP has found application in the form of bumpers, heater housings, door pockets, timing belt covers, and claddings.⁷ Other fillers that are also being widely used are kaolin, mica, and calcium carbonate (CaCO₃).^{8–10}

The performance of particulate-filled polymers is generally affected by the filler size, interfacial adhesion, and surface characteristics. In a highly filled polymer system, a major problem in the nonuniformity of properties due to poor dispersion of the filler in the matrix is frequently reported.¹¹ Therefore, the application of some coupling agents and compatibilizers in particulate-filled polymers has generally been directed toward overcoming the dispersion problem and enhancing the mechanical properties of the composites through the improvement of adhesion across the interface and thus upgrading the performance of the composites.^{6,11} There are several types of coupling agent that have been used in many applications. However, the three most commonly used are silane, titanate, and zirconate.¹²

The use of silane and titanate coupling agents has been widely reported by many researchers. Demjen¹³ reported the treatment of CaCO₃ with as many as

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Filler Specifications						
Filler	Density (g/cm ³)	Hardness (Moh's scale)	Mean particle diameter (µm)			
Talc Kaolin	2.8 2.6	1 2	6.3 1.7			

eight types of silane coupling agents to determine their effect on $PP/CaCO_3$ composites. The incorporation of aminofunctional silane coupling agents resulted in the reactive coupling of PP and CaCO₃, in which both were inactive components, which led to increased strength and decreased deformability.

Ai Wah et al.¹¹ studied the effect of a titanate coupling agent on the rheological and dispersion characteristics and mechanical properties of a talc-filled PP composite. They reported improved melt flow and filler particle dispersion with the treatment of PP pellets with neopentyl(diallyl)oxytri(dioctyl)phosphato titanate (LICA 12), a type of titanate coupling agent. The chain scission of PP molecules and the plasticizing and lubricating effects of the coupling agent were cited as the main causes of the decrease in melt viscosity. The tensile and flexural properties of the composites were also reported to decrease, whereas the toughness properties improved with the incorporation of the coupling agent.

Over the years, the development of hybrid materials has been oriented toward composites because they are more attractive systems used to achieve synergistic effects with particular improvement in the mechanical characteristics and functional properties at a reduced cost. Nevertheless, like other materials, the improvement in mechanical properties is still limited in demand applications without the use of any coupling agent or compatibilizer.

This study focused on the effects of silane and titanate coupling agents on the mechanical, flow, and morphological properties of talc- and kaolin-filled PP hybrid composites. Previously, various workers have only targeted the effects of filler treatments on a single-filler PP composite. Here, the extent of filler–matrix interaction and filler–filler interaction between two different fillers when incorporated into a single matrix, before and after surface treatment of the fillers, is discussed. In this study, a hybrid composite with a formulation of 20 wt % talc and 10 wt % kaolin (PPT20K10) was chosen as the final hybrid formulation because it produced a material with mechanical properties comparable to those of talc and kaolin single-filler-filled PP composites.^{14,15}

EXPERIMENTAL

Materials

The PP copolymer used (Pro-Fax SM240, Titan PP Polymers Sendirian Berhad, Johor, Malaysia) had a melt index of 25 g/10 min. The talc (supplied by Chung Chemicals Sdn. Bhd., Selangor, Malaysia) and kaolin (supplied by Finn Chemicals Sdn. Bhd., Kuala Lumpur, Malaysia) had average particle diameters of 6.3 and 1.7 µm, respectively. Table I shows the filler specifications. The silane coupling agent used was phenyl trimethoxysilane (PTMS) grade Z-6124 supplied by Dow-Corning Midland, MI. A commercial grade of titanate (LICA 12), supplied by Kenrich Petrochemicals, Inc. (Bayonne, NJ), was also used as a coupling agent. Table II shows the chemical formulas of the coupling agents. Other additives, including an antioxidant (Irganox 1010, CIBA, Switzerland) and an ultraviolet stabilizer (Tinuvin 770 DF, CIBA, Switzerland), were used to counter the effects of oxidation and ultraviolet degradation on the PP matrix during processing and storage.

Filler treatment

The surface treatment of the filler was carried out in solution. The silane coupling agent was diluted in ethanol solution (a 95% ethanol/5% water solution adjusted to pH 4.5–5.5 with acetic acid) to yield a 2% final concentration. The 2% silane/ethanol solution was left for 5 min to prehydrolyze the silane and activate the silane group. After treatment, the fillers were then dried in an oven at 110°C for 24 h.

	List of Coupling Agents for the Chemical	Treatment
Coupling agent	Chemical formula	Producer
PTMS	(CH ₃ O) ₃ Si 🔘	Dow Corning USA
LICA 12	CH2-CH-CH2O-C H2 O CH3-CH2- C -CH2-O-Ti-[O-P(OCH	Kenrich Petrochemicals, Inc.
	CH2=CH-CH2O-CH2	

TABLE II List of Coupling Agents for the Chemical Treatment

Sample formulation	PP (wt %)	Talc (wt %)	Kaolin (wt %)	Silane (wt %) ^a	LICA 12 (wt %) ^a			
PP	100	0	0	0	0			
PPT30	70	30	0	0	0			
PPT30S	70	30	0	0.5	0			
PPT30L	70	30	0	0	0.5			
PPK30	70	0	30	0	0			
PPK30S	70	0	30	0.5	0			
PPK30L	70	0	30	0	0.5			
PPT20K10	70	20	10	0	0			
PPT20K10S	70	20	10	0.5	0			
PPT20K10L	70	20	10	0	0.5			

 TABLE III

 Compositions of the Compounded Formulations Containing Untreated and Treated Fillers

^a 0.5 wt % with respect to the weight percentage of the fillers.

The dried fillers were then rinsed again with ethanol to drive away any residual silane before they were dried again at the same temperature and for the same drying period.

According to the reference manual supplied with the LICA 12^{16} the recommended dosage of LICA 12 is either 0.5 wt % total filler or 0.2 wt % resin, whichever provides the greater absolute amount of titanate. Because all of the composites in this study were filled with 30 wt % filler, the incorporation of 0.5 wt % titanate was chosen for both talc and kaolin. Toluene was used as a solvent to reduce the viscosity of the titanate. The ratio of solvent to toluene was fixed at 3 : 1. The solvent was added to a vessel, and the titanate was blended mechanically into the solvent for approximately 30 min. The filler was then added to the vessel, and the slurry was mixed until a good dispersion was achieved. The slurry was then poured into a tray and dried in an oven at 80°C for 5 h. The fillers were washed again with toluene to drive off excess titanate and dried again for the same period of time at the same temperature.

Sample preparation

Compounding was carried out with a twin-screw extruder (model Haake Rheomex CTW 100, Haake, Karlsruhe, Germany). The barrel temperatures of the four zones were 160, 170, 180, and 190°C from the feeding to the die zones. The compounds were blended at a screw speed of 25 rpm. The extrudates were cooled in a water bath, pulled, and pelletized. Test specimens were injection-molded in a tensile mold with a 35-ton Battenfeld BA 350 CD machine (Battenfeld, Bielefeld, Germany) with a Unilog 4000 control system. The injection-molding temperature ranged from 190 to 230°C, whereas the back pressure was 60 psi. Table III lists the compounded formulations containing untreated and treated fillers.

Mechanical testing

The tensile and flexural properties were measured with an Instron 5580 100-kN electromechanical tensile testing machine (Instron, Canton, MA) with a series IX control system in accordance with ASTM D 638 and ASTM D 790-86, respectively, with type-1 test specimen dimensions. For tensile tests, a crosshead speed of 5 mm/min was used, and the average value of at least five specimens was taken for each sample. Tensile modulus, tensile strength, and elongation at break were evaluated from the stress–strain data

For flexural tests, a three-point loading system was used, and the support span length was adjusted to 50 mm. The crosshead speed was 3 mm/min, and the average value of at least five specimens was taken for each sample. The flexural modulus and flexural strength were determined.

Impact testing (unnotched) was done with a Ray-Ran impact pendulum tester (Ray-Ran, Warwickshire, UK) at an impact energy of 7.5 kJ according to ASTM D 256-88. The hammer velocity was set at 2.88 m/s, whereas the weight of the hammer was 1.811 kg. The Izod impact strength was measured for this test. The average value of at least eight specimens was taken for each sample. All tests were carried out in an air-conditioned room (27° C).

Mode of failure analysis

The mode of fracture was studied by the analysis of the fracture surfaces of the tensile samples. This was done with a scanning electron microscopy (SEM) machine (Cambridge S-360, Leica, Cambridge, UK); the fracture surfaces of the tensile test specimens were coated with a thin gold–palladium layer to prevent electrical charge accumulation during the examination.

and kaolin fillers treated with silane and LICA 12. The appearance of the SiO group (~ 1300 cm⁻¹) and CO (~ 1720 cm⁻¹) were used to confirm the presence of the silane coupling agent. As shown in Figure 1, the treatment of silane on talc was detected, which thereby suggested that the condensation of silane on the surface of the filler took place during the drying process. Apart from the SiO group, the bands at 1298–1652 cm⁻¹ showed the presence of Si–N, NH(C₂H₃)–OCH₃, and CH₃–NH–COCH₃,¹⁷ all of which are the characteristic groups of the aminosilane used when it is hydrolyzed in ethanol. LICA 12 treatment, on the other hand, affected the spectrum of talc at approximately 2200–2500 cm⁻¹, which signified the presence of phosphate groups.

As shown in Figure 2, the presence of the silane coupling agent was still detected with the broad band in the range 1415–1570 cm⁻¹, although the appearance of the SiO group peak was weak. The drying treatment of the silane coupling agent on the filler surface, which could have resulted in the evaporation of a fairly large amount of the agent, as was proven by Nakatsuka et al.,¹⁸ could have

with silane and LICA 12 coupling agents. Fourier transform infrared (FTIR) spectroscopy

FTIR analyses were done for fillers treated by silane and LICA 12 to determine the chemical structure before and after the filler treatments. About 5 mg of filler was mixed with about 95 mg of potassium bromide (KBr) before it was compacted into thin pellets with a hydraulic press with 8 tons of force and maintained for about 3 min. The pellets were then inserted into a Nicolet Avatar 360 FTIR spectrophotometer (Nicolet Avatas, Karlsruhe, Germany) to be scanned with OMNIC software.

Melt flow index (MFI)

The processability of the filled PP composites was determined with a Ray-Ran melt flow indexer according to ASTM D 1238-90b. A load of 2.16 kg at 230°C was used for the measurement.

RESULTS AND DISCUSSION

FTIR spectroscopy analysis

Figures 1 and 2 show the 4000-500-cm⁻¹ wave-number range of the difference in the spectra of the talc

Figure 2 FTIR difference spectra of kaolin fillers treated with silane and LICA 12 coupling agents.

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contributed to the weak peak of the silanes. More phosphate groups were seen in LICA 12 treated kaolin with the presence of peaks at 2976 cm^{-1} .

MFI

Figure 3 shows the effect of the silane and titanate coupling agents on the melt flow behavior of the single-filler and hybrid-filler composites. Normally, the addition of particulate fillers to PP restricts molecular motion in the matrix and thus imposes resistance to flow. As a result, it gives a lower MFI value as the filler content increases. The MFI value of kaolinfilled PP was significantly lower than those of the unfilled PP and talc-filled PP composites. The incorporation of very fine particle size (refer Table I) fillers and, hence, a higher surface area like kaolin hindered plastic flow and increased the viscosity of the polymer melt. Thus, a decrease in MFI value was expected. For the hybrid talc/kaolin-filled PP composites, it was expected that the MFI value of the hybrid composite would be intermediate between the MFI values of talc and kaolin because 20 wt % talc and 10 wt % kaolin were used in the final hybrid formulation (Table III).

As shown in Figure 3, the MFI values for both PP single and hybrid composites filled with LICA 12 and silane-treated fillers significantly increased compared to those of the untreated fillers. This observation was in agreement with the results of other related studies reported by several workers.^{11,16,19} This increase in melt flow may have been due to the lubricating/plasticizing effect induced by the coupling agent. According to Han et al.,¹⁹ the reduction in melt viscosity with the presence of a coupling agent may result from the surface modification of the filler particles. Thus, under shear stresses, there could be far less frictional resistance to flow with treated filler particles. This, however, implies that



Figure 3 MFI for single-filler and hybrid-filler composites with and without the filler surface treatment.



Figure 4 Tensile modulus for single-filler and hybrid-filler composites with and without the filler surface treatment.

there is no true coupling (in the chemical sense) between the coupling agent and the polymer matrix. There is also the possibility that the coupling agent might have diffused into the polymer matrix, thus acting as an internal plasticizer, which would have increased the melt flow of the polymer phase.

Silane has been proven to enhance the particle dispersion of fillers by acting as a wetting agent.¹² Good wetting of a filler is often evidenced by the reduced melt viscosity of the polymer composite because there is an improvement in terms of filler dispersion in the polymer matrix. Because the reduction in agglomerated particles reduces hindrance to polymer melt flow, an enhancement of the MFI value can be expected. According to Boaira and Chaffrey,²⁰ who studied the effect of silane treatment on the mechanical and rheological properties of mica-reinforced PP, a melt viscosity reduction of up to 50% could be achieved, which was attributed to a reduction in the surface energy of the mica and a reduction in mica flake interaction.



Figure 5 Elongation at break for single-filler and hybrid-filler composites with and without the filler surface treatment.



Figure 6 Tensile strength for single-filler and hybrid-filler composites with and without the filler surface treatment.

Mechanical properties

The tensile, flexural, and impact properties of the talc- and kaolin-filled single and hybrid PP composites are shown in Figures 4-9. As expected, the incorporation of 30 wt % filler, that is, talc or kaolin, into the PP matrix resulted in a significant increase in the both tensile and flexural moduli of the PP composites. However, the presence of chemical treatment, such as by PTMS or LICA 12, did not have a significant influence on the moduli of the composites. According to Pukanszky,6 in particulate-filled polymer composites, the polymer itself solely contributes to the elasticity of the composite. Therefore, the modulus cannot be used to characterize the strength of filler-matrix interactions; thus, the value of the modulus should not be affected even if there is the existence of strong filler-matrix interactions due to filler surface modification. On the contrary, the observed changes in toughness (indicated by the impact strength values) and flow (as indicated by the MFI values) properties with both PTMS and



Figure 7 Flexural modulus for single-filler and hybrid-filler composites with and without the filler surface treatment.



Figure 8 Flexural strength for single-filler and hybrid-filler composites with and without the filler surface treatment.

LICA 12 treatment were expected because their role as lubricants/plasticizers in the filled polymers.^{16,21}

Both the silane- and titanate-treated PP composites experienced a slight decrease in both the tensile and flexural strengths compared to the untreated PP composites. The decrease in the strength of the LICA 12 treated PP composites was in coherence with the results obtained by Ai Wah et al.¹¹ This may have again been due to the role of the titanate coupling agent in acting as a plasticizer in the filled system.^{16,19}

For composites filled with silane-treated fillers previously, decreases in both the tensile and flexural strengths were rather unexpected. Previous workers have reported positive coupling effects of silane in particulate-filled PP composites.^{6,10,19} This contradiction is still unclear; perhaps it is worth mentioning that the recommended dosage of the silane might not be correct for the effective treatment of the fillers to produce an adequate coupling ability. Underdosage of the silane could lead to inhomogeneity and inadequate coupling between the fillers and matrix,



Figure 9 Impact strength for single-filler and hybrid-filler composites with and without the filler surface treatment.

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whereas overdosage could cause severe particle agglomeration.²² This will be the subject of a future publication.

The presence of both PTMS and LICA 12 significantly improved the elongation at break and impact strength of the PP composites, regardless of whether they were single-filler or hybrid-filler composites. As mentioned earlier, in this investigation, both PTMS and LICA 12 did play a role as plasticizers/lubricating agents. This significant improvement in deformability seemed to suggest that the coupling between the filler and matrix was not strong enough because the fillers failed to hinder the plastic deformation of the polymer matrix. The impact strengths of the kaolin- and hybrid talc/kaolin-filled PP treated with both silane and LICA 12 showed better improvement compared to those of the talc-filled PP single-filler composites. The use of kaolin with a very fine particle size provided a positive contribution toward the improvement of the impact strength of the PP composites. Because silane and titanate could have acted as good wetting or dispersion agents, they could have contributed to the ease of particle dispersion of talc and kaolin in the PP matrix. Better filler dispersion would have obviously reduced the stress concentration sites, which are very sensitive to impact loading. The plasticizing effects of both chemical treatments operated in conjunction with interfacial adhesion between the filler and matrix to yield an increase in the toughness values of the composites. Thus, if the smaller filler particles such as kaolin were dispersed well in the polymer matrix, an improvement of the toughness would not be possible.

Morphology

Figure 10 shows the SEM micrographs of the tensile fractured surfaces of the talc-filled PP composites. A



Figure 11 SEM micrograph of the fractured surface of the silane-treated, single-filler PPT30S composite at $1000 \times$ magnification. The circles show talc particles.

preferred orientation of talc particles was clearly seen, with their basal sheet planes mostly parallel to the surface of the injection molding. This unique organization of particles was the consequence of the platelike structure of talc and its motion in a viscous medium during the injection-molding process. Most of the talc particles were clearly visible and well dispersed and tended to be embedded inside the PP matrix. Thus, it was not surprising that the talc-filled PP (PPT30) composites exhibited better strength and stiffness (ca. Figs. 4–8) even without any form of filler treatment.

Figure 11 shows the more ductile fracture surface morphology of the silane-treated, talc-filled PP composite (PPT30S). The presence of plastic deformation in the form of matrix fibrillation on the fracture surface is believed to be responsible for the improvement in the elongation at break and impact strength of the PPT30S composite.

A similar surface morphology was also displayed by the LICA 12 treated, talc-filled PP composite



Figure 10 SEM micrograph of the fractured surface of the untreated, single-filler PPT30 composite at $1000 \times$ magnification. The circle shows dispersed talc particles.



Figure 12 SEM micrograph of the fractured surface of the titanate-treated, single-filler PPT30L composite at $1000 \times$ magnification. The circle shows dispersed talc particles.



Figure 13 SEM micrograph of the fractured surface of the untreated, single-filler PPK30 composite at $1000 \times$ magnification. The circle shows agglomerated kaolin particles. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

(PPT30L) in Figure 12. There was no severe agglomeration of particles, which suggested that LICA 12 helped in filler dispersion. There was billowy or ground swell formation, which proved that the PP matrix most certainly retained its ductility when it was incorporated with LICA 12-treated talc. As suggested by Monte,¹⁶ LICA 12 did play a role as plasticizer/lubricant, which improved the toughness properties, such as elongation at break and impact strength, of the composites.

Figure 13 shows the fracture surface morphology of the untreated kaolin filled PP composite (PPK30). The very fine particle size of kaolin increased the particle–particle interaction and resulted in the agglomeration of kaolin particles. Filler agglomeration could have acted as a stress concentration point or weak point, which could have reduced the me-



Figure 14 SEM micrograph of the fractured surface of the silane-treated, single-filler PPK30S composite at $1000 \times$ magnification. The arrow shows kaolin particles, whereas the circle shows long matrix fibrils. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]



Figure 15 SEM micrograph of the fractured surface of the titanate-treated, single-filler PPK30L composite at $1000 \times$ magnification. Talc particles are depicted by arrows. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

chanical properties, especially elongation at break and impact strength (Figs. 5 and 9).

Figure 14 shows the fracture surface morphology of the silane-treated kaolin filled PP composite (PPK30S). The extensive matrix fibrillation suggested the occurrence of a plastic deformation process. This, coupled with the fact that the silane could act as a wetting or dispersion agent for the kaolin and particles, would have consequently led to an increase in the toughness of the composites.

Figure 15 shows the fracture surface morphology of the LICA 12 treated kaolin-filled PP composite (PPK30L). The good matrix yielding on the fracture surface represented a seemingly ductile failure mode, which suggested that the plastic deformations could have occurred between these polymer layers. There might have been a lubricating/plasticizing effect induced by the incorporation of the LICA 12 coupling agent, which improved the toughness properties, such as elongation at break and impact strength, of PPT30L.

Figure 16 shows the fracture surface morphology of the untreated talc/kaolin-filled PP hybrid composite (PPT20K10). The absence of long fibrils and billowy or ground swell formation indicated that PPT20K10 exhibited a brittle failure mode. The talc particles could hardly be spotted on the fracture surface, which suggested that they could have been embedded deep in the matrix, whereas kaolin particles seemed to have the tendency to form aggregations or agglomerations.

Figure 17 shows the fracture surface morphology of the silane-treated talc/kaolin-filled PP hybrid composite (PPT20K10S). As a good wetting or dispersing agent, silane could have contributed to easing the particle dispersion in the polymer matrix



Figure 16 SEM micrograph of the fractured surface of the untreated, hybrid-filler PPT20K10 composite at $1000 \times$ magnification. The arrows point to kaolin particles, whereas the circles show talc particles.

and, consequently, reduced the stress concentration sites on the composites. With minimal presence of stress concentrators, higher stresses would have been needed to initiate a crack, whereas the impact energy could have been absorbed more efficiently by the higher plastic deformation imparted by the matrix. This led to an improvement in the impact strength and elongation at break of the hybrid composite (Figs. 5 and 9).

Figure 18 shows the fracture surface morphology of the LICA 12 treated talc/kaolin-filled PP hybrid composite (PPT20K10L). The presence of long matrix fibrils on the fracture surface represented a ductile failure mode, which suggested that plastic deformations could have occurred between the matrix layers. This indicated the role of LICA 12 as plasticizer/ lubrication, which consequently led to an improvement of both elongation at break and impact strength of PPT20K10L.



Figure 17 SEM micrograph of the fractured surface of the silane-treated, hybrid-filler PPT20K10S composite at $1000 \times$ magnification. The arrows point to kaolin particles, whereas the circles show talc particles.



Figure 18 SEM micrograph of the fractured surface of the titanate-treated, hybrid-filler PPT20K10L composite at $1000 \times$ magnification. The arrows show kaolin particles, whereas the circle shows talc particles. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

CONCLUSIONS

The incorporation of silane and titanate coupling agents into filled PP notably increased the processing properties (MFI), elongation at break, and impact strength and improved the filler dispersion of PP single-filler and hybrid-filler composites. Nevertheless, the incorporation of both coupling agents did not improve the strength and stiffness properties because of their plasticizing or lubricating effects on the PP matrix. The morphological studies demonstrated the major role of both silane and titanate as wetting or lubricating agents in both the single-filler and hybrid-filler PP composites.

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