

Influence of Stearic Acid Treatment of Filler Particles on the Structure and Properties of Ternary-Phase Polypropylene Composites

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ABSTRACT: In this study, ternary-phase polypropylene (PP) composites containing an ethylene–octene copolymer (EOR) and calcium carbonate (CaCO_3) were investigated. Particular consideration was given to the influence of stearic acid treatment of the filler on the phase morphology and mechanical properties of the composites. In composites containing an uncoated filler, a separate dispersion of the elastomer and filler particles in the PP matrix was observed. The use of filler treated with stearic acid had no effect either on the dispersion or the interaction of the filler and the polymer components. However, the surface-treated filler was found to promote the β -hexagonal crystallization of PP and gave a composite with lower $T_{c \text{ onset}}$ and T_c values. As a consequence, differences in mechanical properties, in particular, impact strength, were exhibited in which calcium carbonate with stearic acid treatment was apparently more effective in increasing the impact strength of the composites in comparison with the composites containing the uncoated filler. © 1999 John Wiley & Sons, Inc. *J Appl Polym Sci* 74: 3445–3454, 1999

Key words: polypropylene; ethylene–octene copolymer; calcium carbonate; ternary composite; morphology; mechanical properties

INTRODUCTION

Ternary-phase polymer composites containing a soft elastomer and a rigid filler have both theoretical and practical significance due to the possibility of producing composites with increased impact strength and stiffness. In ternary polypropylene (PP) composites, various elastomers and fillers have been used.^{1–8} In these composites, two extremes in the phase structure may occur, where either elastomer and filler particles are dispersed separately in the PP matrix^{5,9} or the elastomer encapsulates filler particles, resulting in a low

modulus interlayer between the matrix and filler.^{1,10} The relative extent to which these structures develop depends on several factors, including the rheology and surface energies of the constituents, mixing conditions, and geometry of the rigid fillers.

Extensive analysis of the mechanical properties of ternary-phase composites was carried out.^{11–14} Most studies have focused on the influence of the phase morphology. Ternary composites containing an encapsulated filler have been reported to have somewhat higher tensile impact strength but lower modulus than those with a separation structure, because the effect of the incorporated elastomer is extended by the filler.³ However, composites having a separation structure yielded a marked increase in the composite modulus.³ Although some experimental data on the mechanical properties of such ternary com-

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posites has been published in the literature, contradictory results on the influence of the filler on the microstructure and crystallization of the ternary composites were reported.^{11–14}

In the present study, the relationship between the structure and mechanical properties of ternary-phase PP composites, containing an ethylene–octene copolymer and calcium carbonate, was investigated. Particular attention was also given to the influence of surface-treated filler on the microstructure and crystallization behavior of the composites.

EXPERIMENTAL

Materials

Composites were prepared from a PP homopolymer (P400S, Thai Polyethylene Co., Thailand), with a melt flow rate of 4 g/10 min (230°C/2.16 kg), together with an ethylene–octene copolymer (EOR) (Engage 8150, DuPont Dow Elastomer Co.) containing a 25% wt octene monomer with a melt-flow rate of 0.5 g/10 min (190°C/2.16 kg). The calcium carbonate (CaCO₃) fillers used in this study were supplied by the Lime Quality Co. (Thailand) in an uncoated form (Microcal) and surface-coated with 2% wt stearic acid (Turbo-plex). The particle size and surface area of the two fillers were determined by a Mastersizer S Ver. 2.11 (Malvern Instruments) using isopropanol as a dispersing agent. The uncoated carbonate had an average particle diameter of 5.3 μm and specific surface area of 1.8 m²/g, while the surface-coated carbonate had an average particle diameter of 4.3 μm with a specific surface area of 2.2 m²/g. According to the literature,^{15,16} 1 g of stearic acid would cover around 445 m² of the available mineral surface. A 2% wt stearic acid treatment, therefore, would ensure 100% surface coverage of the calcium carbonate used in this study.

Compounding and Sample Preparation

PP/elastomer/filler composites were prepared by melt-mixing all the components in a one-step procedure using a corotating twin-screw extruder (Prism TSE16), under conditions which ensured dispersion of both coated and uncoated fillers. All composites contained the same amount of PP at 60% vol. The elastomer and filler contents were varied in the range of 0–40% vol EOR and 0–30% vol filler while maintaining the total amount of EOR and filler in each composite at 40% vol. The

barrel temperature profile was set between 160 and 200°C (from feed zone to die). The screw speed used was 220 rpm, giving a throughput rate of 3 kg/h. Test specimens for tensile and impact tests were prepared by injection molding, using a barrel temperature of 210°C.

Mechanical Testing and Structural Analysis

Tensile properties were measured in accordance with ASTM D638-89, using an Instron Model 4301 tensile-testing machine with a crosshead speed of 50 mm/min. Izod impact strength was obtained from notched specimens, using a pneumatic impact tester (Radmana ITR-2000). An impact velocity of 3.4 m/s was used. Fifteen specimens were analyzed for each composite. All mechanical testing was undertaken at 23°C.

The dynamic mechanical properties of selected composites were determined using a Polymer Laboratories dynamic mechanical thermal analyzer. Testing was carried out in the bending mode over a temperature range of –100 to 120°C at a frequency of 6.28 rad/s (1 Hz).

The melting and crystallization behavior of the composites was studied using a Perkin–Elmer DSC-7 differential scanning calorimeter (DSC). Samples were first heated from 50 to 230°C at a scan rate of 10°C/min and then maintained at 230°C for 5 min before cooling to 50°C at the same rate. The percentage crystallinity of the composites was calculated from the heat of fusion using a ΔH° value of 189 J/g.¹⁷ Some compositions were also characterized on a JEOL JDX-3530 X-ray diffractometer (30 kV, 30 mA) using Ni-filtered CuK α radiation.

The phase structures of the composites were examined by scanning electron microscopy (SEM). Specimens were prepared by immersing test pieces in liquid nitrogen, before breaking them using an impact testing machine. To improve the contrast between the PP and EOR phases, the fractured surfaces were exposed to hot heptane vapor for 20 s to remove the EOR particles from the PP matrix. The surfaces prepared in this way were platinum/palladium sputter-coated and examined under a Hitachi S2500 scanning electron microscope.

RESULTS AND DISCUSSION

Composite Microstructure

The effect of stearic acid coating on the filler dispersion and phase morphology of the ternary PP

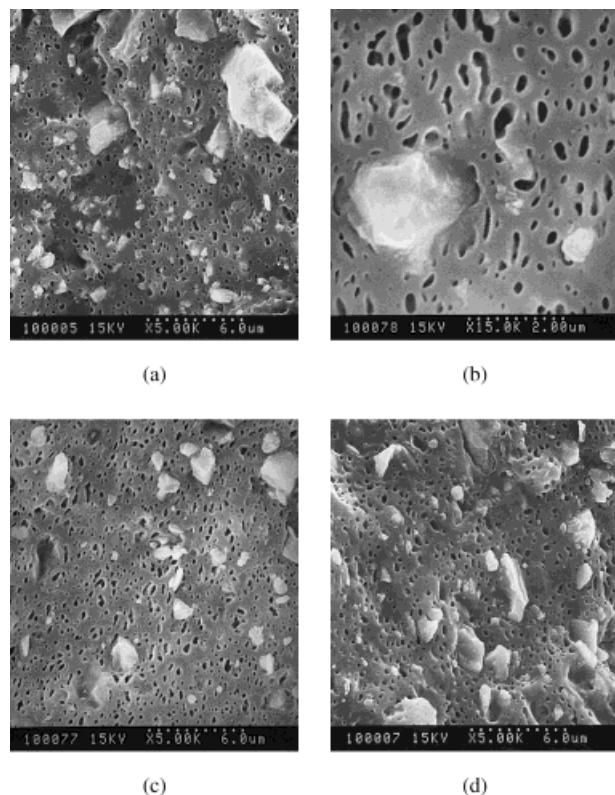


Figure 1 Cryogenic fractured and etched surfaces of ternary-phase PP composites: (a) PP/EOR/CaCO₃ (60/20/20); (b) higher magnification of (a); (c) PP/EOR/CaCO₃ (60/30/10); (d) PP/EOR/coat CaCO₃ (60/20/20).

composites was studied using SEM and dynamic mechanical analysis (DMA) techniques. Figure 1(a) shows the microstructure of the PP/EOR/CaCO₃ composite. Calcium carbonate particles are seen to be well dispersed without agglomeration within the PP matrix. Complete filler encapsulation by the EOR was not apparent; however, some filler particles were found to be partially wetted by the EOR as seen in Figure 1(b). This structure remained similar when the concentration of EOR in the composites was increased [Fig. 1(c)]. In compositions where the calcium carbonate filler had been treated with stearic acid (PP/EOR/coat CaCO₃), a separate dispersion structure was again observed [Fig. 1(d)].

Figure 2 shows the temperature dependencies of $\tan \delta$ at 1 Hz for various PP composites. In binary PP/EOR blends, two $\tan \delta$ peaks were observed at 15 and -37°C , corresponding to the glass transition temperatures (T_g) of PP and EOR, respectively. In both PP/EOR/CaCO₃ and PP/EOR/coat CaCO₃ composites, the location of T_g peaks for PP were the same at 15°C . A T_g value for EOR in the PP/EOR/coat CaCO₃ composite

was seen at -36°C , which is similar to that observed in the binary PP/EOR blend, indicating that coated CaCO₃ had no influence on the mobility of the EOR chains. Thus, the filler and rubber in this system were independently dispersed in the PP matrix. This result confirmed the SEM observations described above, in which the structure of the PP/EOR/coat CaCO₃ composites was a phase-separated dispersion. In the ternary composites containing uncoated CaCO₃, some shift in the T_g peak of EOR was observed. In such composites, the EOR peak shifted from -37°C (in the binary blend) to -33°C (in the ternary blend), indicating immobilization of some rubber chains on the filler surface. This is attributed to the higher surface energy and roughness of the uncoated surface.

It has been reported that stearic acid coating decreases the filler surface energy, leading to the promotion of a separate dispersion structure.³ Kolarik et al.³ found that the phase structure of PP/EPDM/CaCO₃ composites could be controlled by several factors, the surface treatment of the filler being the most important factor. The authors found that calcium carbonates without any treatment were extensively encapsulated by the incorporated elastomer. If the filler surface was treated with stearic acid, its surface energy was reduced and the percentage of the encapsulated filler diminished.

To analyze the occurrence of differing composite structures, the surface energies of each component were compared. Table I shows published

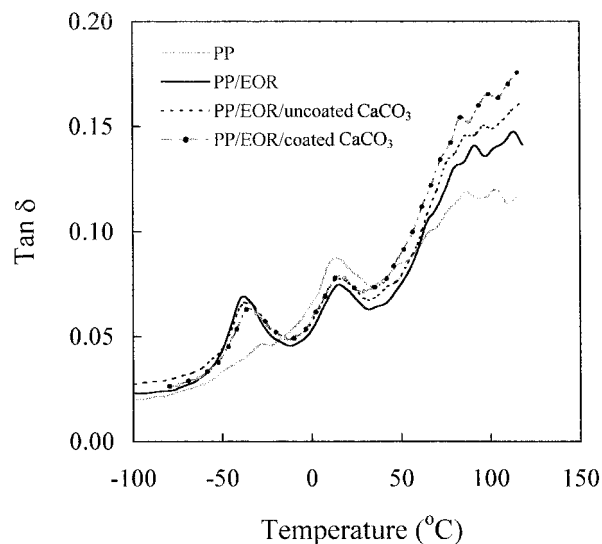


Figure 2 Temperature dependency of $\tan \delta$ for PP, PP/EOR, PP/EOR/CaCO₃, and PP/EOR/coat CaCO₃.

Table I Dispersion (γ^d) and Polar (γ^p) Component of the Surface Energy (γ) of Polymers and Fillers Used in the Ternary Composites

Component	Surface Energy (mJ/m ²)		
	γ^d	γ^p	γ
PP ¹⁸	26.0	4.0	30.0
EOR ¹⁸	26.4	3.9	30.3
EPDM ¹⁹	33.1	3.1	36.2
CaCO ₃ (uncoated) ¹⁹	54.4	153.4	207.9
CaCO ₃ (coated with stearic acid) ²⁰	23.4	18.0	41.4

surface energy values for PP, EOR, EPDM, and CaCO₃. In comparison with calcium carbonate fillers, all polymers are low surface-energy materials. Therefore, calcium carbonates might be expected to be wetted by polymer melts rather easily. Due to the higher surface energy of EPDM, in the system of PP/EPDM/CaCO₃, the ability of EPDM to encapsulate calcium carbonate particles is thus greater than that of PP. This explains the observation of an encapsulation structure in such systems.³ In the present study, PP and EOR have similar surface-tension values. Thus, both components would be expected to wet filler particles to a similar degree. However, due to the lower viscosity and higher concentration of PP in the compositions, calcium carbonate particles were favorably coated by PP than by EOR, resulting in the separate dispersion structure observed. In the case of the composite containing coated CaCO₃, the lower surface energy of the filler makes it more amenable to wet-out by the polymer melt.

Crystal Structure and Crystallization Behavior

Wide-angle X-ray diffractograms (WAXD) for the unmodified PP and its composites are shown in Figure 3. In the given range of the scattering angle, unmodified PP shows four maxima at 2θ of 14.0°, 16.9°, 18.5°, 21.0°, and 21.8°, corresponding to the (110), (040), (130), and overlapping (131) and (111) reflections which are characteristic of the monoclinic α -phase. With the incorporation of EOR, the dominating α -monoclinic modification remained unchanged. The incorporation of CaCO₃ into the PP/EOR blends also gave the same structure. However, differences were observed in composites containing coated CaCO₃ in which the (300) reflection of the β -hexagonal-

phase PP was clearly seen at 2θ of 15.7°. These results were corroborated by the DSC thermograms recorded during the melt process. Figure 4(a) shows the DSC thermograms of various PP/EOR/CaCO₃ composites in which a single-peak characteristic of the melting of α -PP was observed at $\approx 166^\circ\text{C}$. Unlike the composites containing coated CaCO₃, no evidence of β -PP was observed. At the filler concentration from 10% vol, a shoulder at 145°C, characteristic of the melting temperature of β -PP, was apparent in the PP/EOR/coat CaCO₃ composites [Fig. (4b)].

Stearic acid coating of the filler was also found to influence the crystallization behavior of PP. The incorporation of uncoated CaCO₃ to PP/EOR blends led to an increase in the $T_{c \text{ onset}}$ and T_c of the composites [Fig. 5(a,b)]. An increase in $T_{c \text{ onset}}$ clearly indicated that the crystallization started earlier in the filled composites than in the unfilled PP. In other words, the CaCO₃ in this system acted as nucleating agents, promoting the crystallization process. As the concentration of the filler was increased, the $T_{c \text{ onset}}$ and T_c of PP increased. This effect was more pronounced in composites containing up to 10% vol of the filler. With

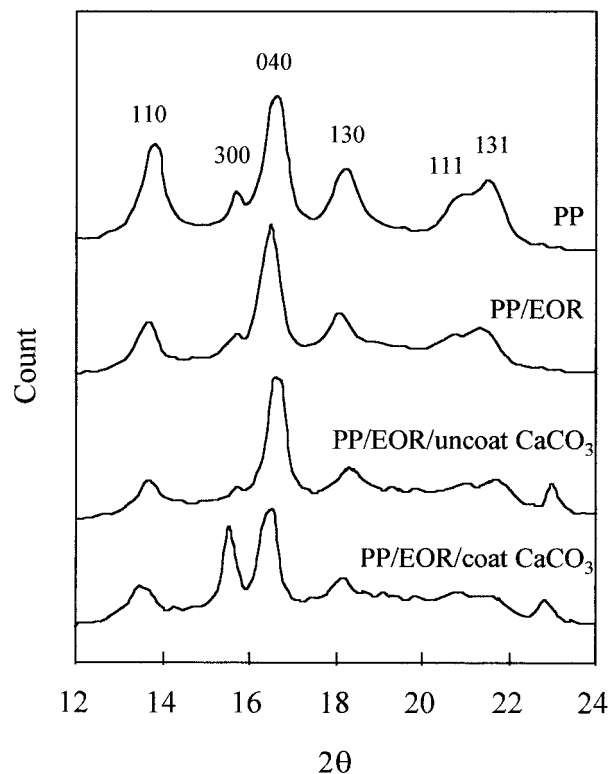
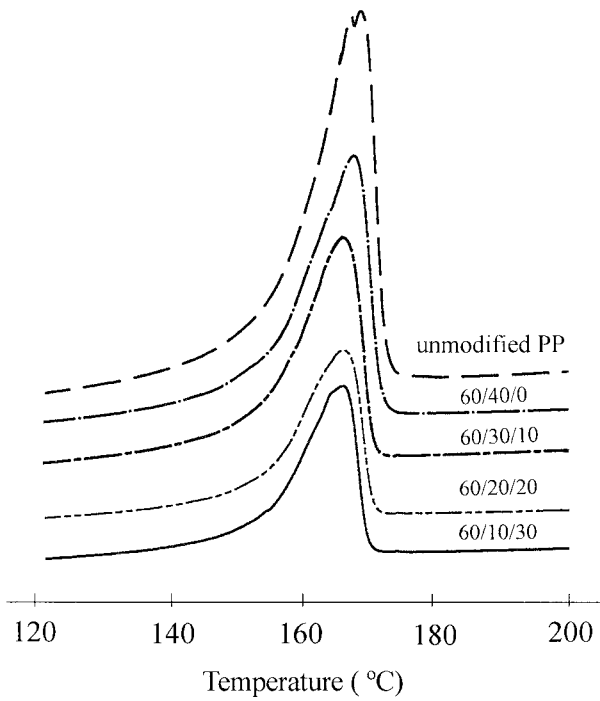
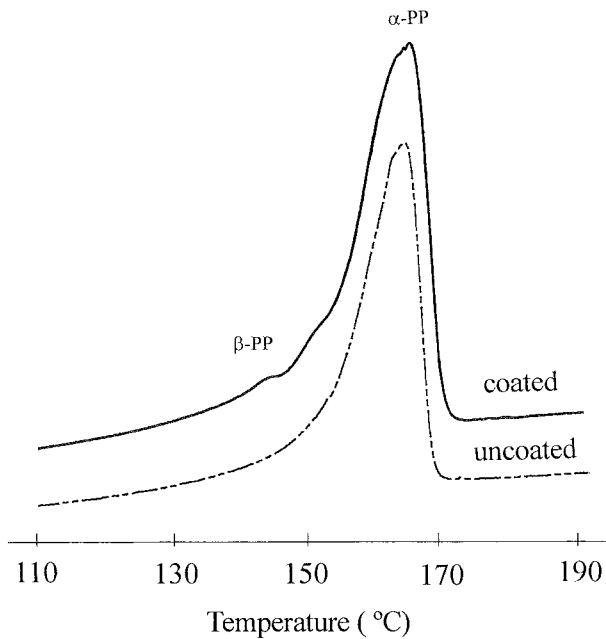


Figure 3 X-ray diffractograms of PP, PP/EOR (60/40), PP/EOR/CaCO₃ (60/30/10), and PP/EOR/CaCO₃ (60/30/10).



(a)



(b)

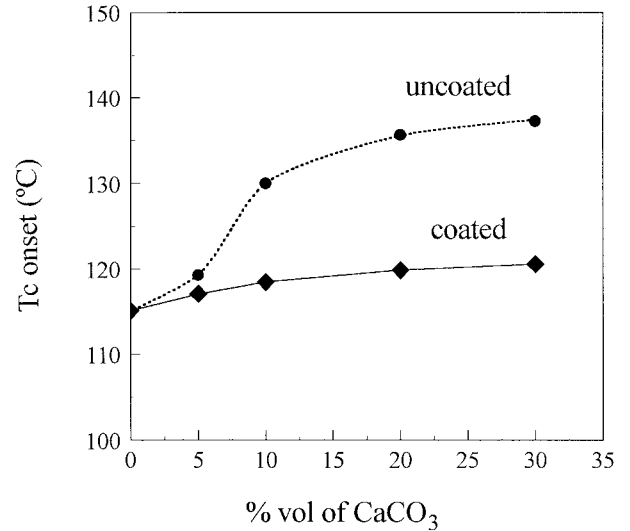
Figure 4 DSC thermograms of PP and its ternary-phase composites: (a) DSC thermograms of PP, PP/EOR/CaCO₃ (60/40/0), (60/30/10), (60/20/20), and (60/10/30). (b) DSC thermograms of PP/EOR/CaCO₃ (60/30/10) and PP/EOR/coat CaCO₃ (60/30/10).

further increasing of the level of the filler (from 10 to 30% vol), $T_{c\ onset}$ and T_c still increased but less sharply. In comparison to the uncoated CaCO₃,

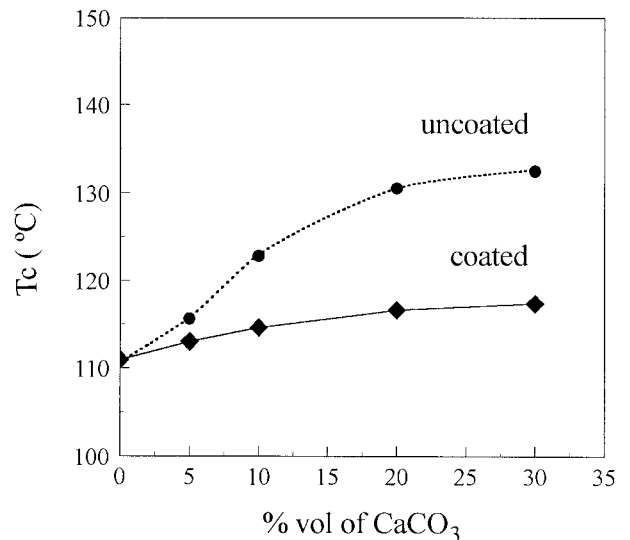
the stearic acid-coated filler had less influence on the crystallization behavior of PP in the composites.

Tensile Properties

The effect of EOR and CaCO₃ concentration on the composite modulus is shown in Table II. It can be seen that incorporation of 40% vol of EOR



(a)



(b)

Figure 5 Effect of stearic acid treatment of filler on the crystallization behavior of PP: (a) effect on the onset temperature of crystallization ($T_{c\ onset}$); (b) effect on the crystallization temperature (T_c). (●) composites containing uncoated CaCO₃; (◆) composites containing coated CaCO₃.

Table II Tensile Yield Stress, Tensile Modulus, and Impact Strength of Various PP Composites

PP/EOR/CaCO ₃ (vol %)	Tensile Stress (MPa)		Tensile Modulus (GPa)		Impact Strength (J/m)	
	Uncoated	Coated	Uncoated	Coated	Uncoated	Coated
100/0/0	34.35		1.62		32.11	
60/40/0	18.78		0.87		777.16	
60/35/5	18.02	17.45	0.98	0.99	795.99	841.97
60/30/10	17.59	17.32	1.22	1.28	749.84	866.80
60/20/20	17.06	16.53	1.91	1.93	235.43	615.70
60/10/30	17.07	16.51	2.99	2.99	49.24	83.54

(60/40/0 composition) markedly reduced the composite modulus, whereas the modulus of ternary composites noticeably increased with increasing calcium carbonate content. Composites having filler contents greater than 20% showed higher modulus values than those of pure PP. No significant difference in the modulus value was observed between the use of uncoated and coated CaCO₃.

Figure 6 also shows comparison data from the current study and theoretical data. The theoretical composite modulus was determined from the Einstein,²¹ Kerner,²² and Nielsen²³ expressions for polymers filled with rigid spheres:

$$E_R = \frac{E_c}{E_m} = (1 + 2.5\nu_f) \quad (\text{i})$$

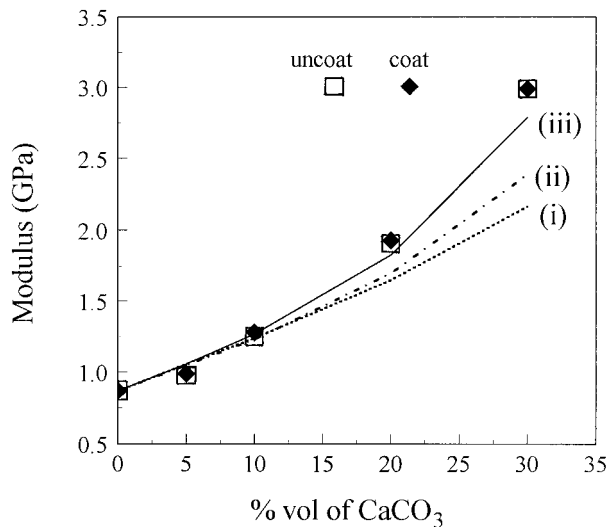


Figure 6 Tensile modulus of ternary-phase PP composites. Curves (i), (ii), and (iii) were calculated using the Einstein, Kerner, and Nielsen equations, respectively. (□) Composites containing uncoated CaCO₃; (◆) composites containing coated CaCO₃.

$$E_R = \frac{E_c}{E_m} = 1 + \frac{15(1 - \gamma_p)}{8 - 10\gamma_p} \frac{\nu_f}{1 - \nu_f} \quad (\text{ii})$$

$$E_R = \frac{E_c}{E_m} = \frac{1 + AB\nu_f}{1 - B\psi\nu_f} \quad (\text{iii})$$

where

$$B = \frac{(E_f/E_m) - 1}{(E_f/E_m) + A} \quad \text{and} \quad \psi = 1 + \left(\frac{1 - \phi_m}{\phi_m^2} \right) \nu_f$$

E_R is the relative modulus; E_c , the modulus of the composite; E_m , the modulus of the matrix polymer; E_f , the modulus of the filler; ν_f , the volume fraction of filler; γ_p , Poisson's ratio of the polymer; and ϕ_m , the maximum volume fraction of the filler. Experimental data measured from the two-phase (PP/EOR) of the same compositions were taken as the matrix values for the ternary-phase systems

From Figure 6, it is evident that the modulus of the ternary composites containing both uncoated and coated fillers agreed reasonably well with Nielsen's model. This would imply that the presence of EOR in the composites did not produce an encapsulation structure. If the incorporated filler is encapsulated by rubber, suppression of the reinforcing efficiency of CaCO₃ by EOR would have been observed and the experimental modulus would have been lower than that predicted.

The effect of EOR and CaCO₃ on composite yield stress is shown also in Table II. The tensile stress of an unmodified PP was determined as 34.35 MPa. With addition of either EOR or CaCO₃, the tensile yield stress decreased to about 17 MPa, representing a reduction of 50% and indicating that both EOR and CaCO₃ have similar load-bearing capacities in the composites. The effect of stearic acid treatment on the tensile yield stress of various composites is shown in Figure

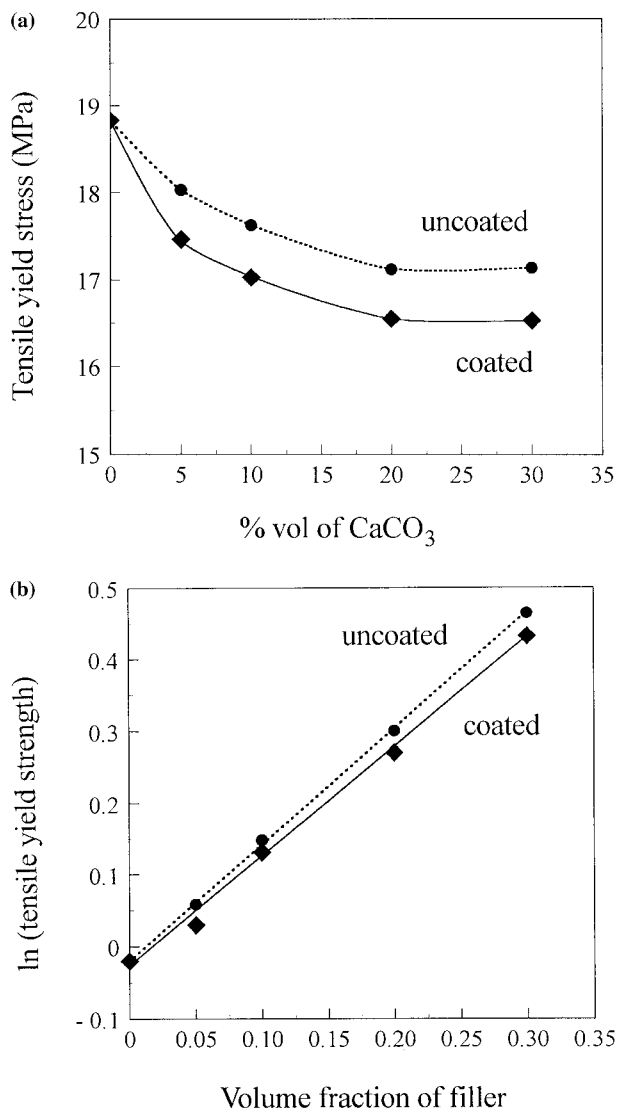


Figure 7 Tensile yield stress of ternary-phase PP composites: (a) tensile yield stress of ternary-phase PP composites as a function of filler concentration; (b) a plot of \ln (tensile yield strength) as a function of filler concentration: (●) composites containing uncoated CaCO₃; (◆) composites containing coated CaCO₃.

7(a). Composites with the coated filler exhibited a lower tensile yield stress. A reduction in tensile properties after introducing the coated filler may be due to poor stress transfer between the filler and the polymer matrix brought about by the surface coating. To analyze the interaction between the polymer and filler in the composites, some of the most commonly used theoretical predictive models were considered.

Equation (iv), proposed by Nielsen,²⁴ enables the strength of a filled polymer (σ_c) to be calculated from

$$\sigma_c = \sigma_m(1 - \nu_f)S \quad (\text{iv})$$

where σ_c and σ_m are the tensile strengths of the composite and polymer matrix, respectively, and ν_f is the volume fraction of filler in the composite. A parameter S accounts for the weakness in the structure resulting from a discontinuity in stress transfer and the generation of stress concentrations at the filler-polymer interface. The maximum value of S is unity, where stress concentration is absent. The lower the value of S , the greater the stress concentration or the poorer the adhesion.

Nicolais and Nicodemo²⁵ expressed the strength of a filled composite as

$$\sigma_c = \sigma_m(1 - a\nu_f^b) \quad (\text{v})$$

where a and b are constants. The value of a is related to the stress concentration or to the quality of adhesion between the matrix and filler and b is related to the geometry of the filler. In spherical fillers uniformly distributed with no adhesion, a becomes 1.21. Where there is some adhesion, a becomes smaller than 1.21. The constant b is equal to 1 if the material fails by planar fracture and 2/3 if it fails by random fracture.

Pukanszky²⁶ proposed that the tensile yield strength of composites could be calculated from

$$\sigma_c = \frac{1 - \nu_f}{1 + 2.5\nu_f} \sigma_m \exp(B\nu_f) \quad (\text{vi})$$

where the parameter B reflects the interfacial adhesion between the filler and polymer and may be estimated by the plot of $\ln(\sigma_c)$ versus ν_f [Fig. 7(b)]. In this study, the calculated B values of 1.61 and 1.53 were obtained for composites containing an uncoated and stearic acid coated filler, respectively. These results together with the S and a values determined from eqs. (iv) and (v) and shown in Table III suggest little difference in the interfacial adhesion between uncoated and coated fillers and the polymer matrix.

Impact Properties

Notched Izod impact strengths for the uncoated and coated composites are shown in Table II and Figure 8. In the PP/EOR blends without filler addition (60/40/0), EOR significantly increased the impact resistance of PP. Addition of filler into the blends led to a gradual decrease in the impact strength of the ternary composites. However, it

Table III Calculated S and α Values from Eqs. (iv) and (v)

% Vol of Filler	Calculated S Values		Calculated α Values	
	CaCO ₃	Coated CaCO ₃	CaCO ₃	Coated CaCO ₃
5	0.94	0.91	0.77	0.97
10	0.92	0.91	0.78	0.84
20	0.90	0.87	0.81	0.88
30	0.91	0.88	0.81	0.86

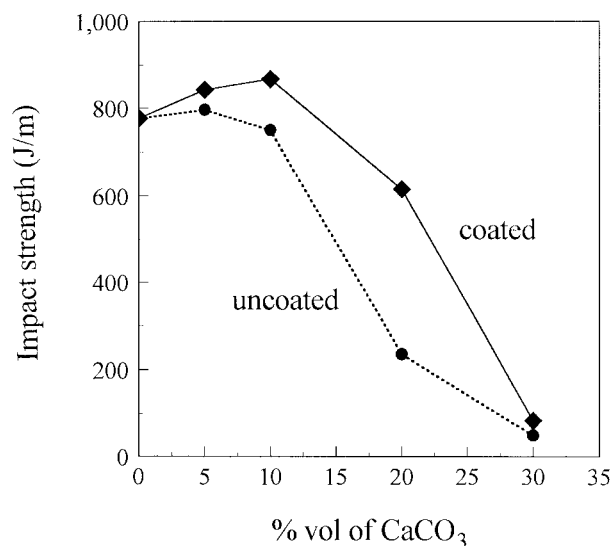
can clearly be seen that all ternary composites prepared in this work exhibited improved impact properties in comparison with pure PP. Incorporation of calcium carbonate with stearic acid treatment was even more effective in this respect. This effect was particularly notable at 20% vol filler where material containing the coated filler achieved impact strengths nearly three times that of materials containing the uncoated filler and 20 times that of the unfilled PP.

The origin of the difference between coated and uncoated fillers deserves further discussion. Riley et al.²⁷ studied the effect of filler coating. The authors reported that for PP/CaCO₃ composites the coating aids the dispersion of the filler, so making available a large number of particles to retard crack propagation. The electron microscopy undertaken in the present work [Fig. 1(a)–(d)] did not appear to support this theory; filler dispersion was not dependent on the surface treatment. Failure due to the presence of large agglomerates does not seem likely. In both filled composites, “filler pull-out” seemed to be the mechanism that resulted in improvement of the impact properties observed here. Figure 9(a)–(c) shows the impact-fractured surfaces of the unfilled PP and its ternary composites containing uncoated and coated calcium carbonate, respectively. Under impact loadings, the unfilled PP exhibited a brittle failure characteristic at room temperature. Extensive dewetting occurred in its ternary composite. In ternary composites containing the coated filler, the extent of plastic deformation increased. This is in accordance with data published in the literature,^{28–30} where it has been reported that plastic deformation initiated by a filler is responsible for increase in the composite toughness.

The interaction between the polymer and filler is one of the factors influencing the impact strength in filled composites. According to Kendall and Sherliker,³¹ the role of a filler as a reinforcement is influenced by the surface interaction

between the polymer and filler, resulting in a layer of polymer adhering to the filler surface. This polymer interphase causes energy dissipation and toughness in a filled material. As discussed above, little difference in the interfacial adhesion was found between composites containing uncoated and coated calcium carbonate. In this study, dispersion and interfacial adhesion between the filler and polymer matrix, therefore, was not a determining factor in the difference of impact strength between the composites containing coated and uncoated filler. Surface treatment of CaCO₃ with stearic acid had virtually no effect on filler dispersion or matrix/filler adhesion. Thus, it is proposed that improvement in the impact strength by the use of stearic acid coating was due to other effects.

It is possible that the effect on the polymer microstructure which might result from using a nucleating (uncoated) or nonnucleating (stearic acid-coated) filler may be responsible for improvement of the impact properties. Although the predominating crystal structure in both systems is α -PP, stearic acid coating of the filler tended to promote the β -modification of PP and also gave a composite with lower $T_{c\text{ onset}}$ and T_c values. Hultley and Darlington^{32,33} observed a correlation between $T_{c\text{ onset}}$ and T_c in carbonated-filled PP, when it was cooled from the melt, and impact strength. Our studies have, to some extent, confirmed this finding, where, within the same system, high onset temperatures were observed for composites of poor impact strength.

**Figure 8** Effect of stearic acid treatment of filler on impact strength of ternary-phase PP composites.

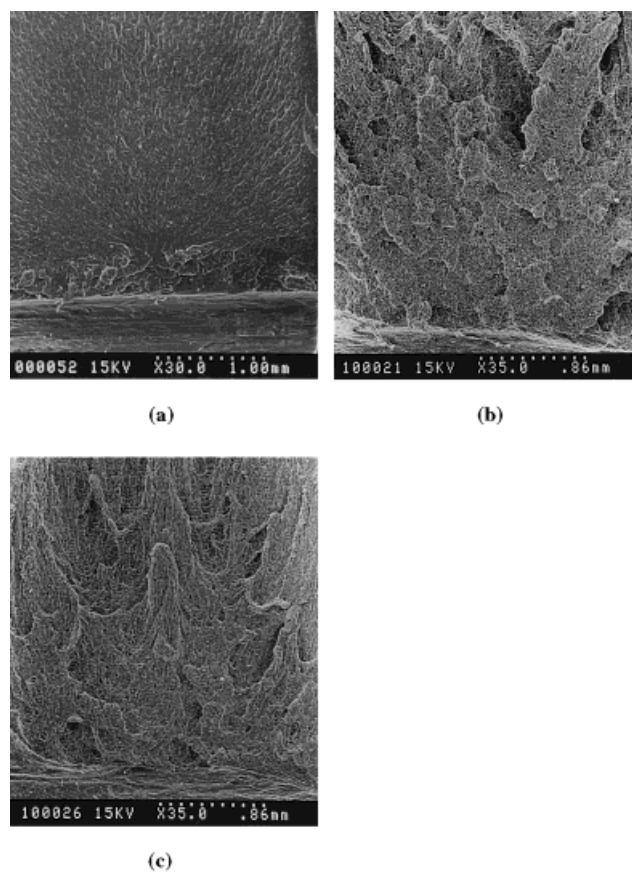


Figure 9 Impact-fractured surfaces of PP and its ternary-phase composites: (a) unfilled PP; (b) PP/EOR/CaCO₃ (60/30/10); (c) PP/EOR/coat CaCO₃ (60/30/10).

There is also disagreement between the results of the current study and published behavior with respect to β -PP. Murphy et al.³⁴ found that in unfilled PP a high β -phase content correlated with poor impact performance. Contrary to this, Tjong et al.^{35–37} recently studied the static tensile and impact behavior of high-purity β -form PP and found that the β -form of PP exhibited lower yield strength but higher impact strength than that of the α -PP material. In addition, microfibrils and microcrazes were formed extensively in the impact-fractured surfaces of the β -form PP specimens. The ease of microfibril formation in the β -form PP is attributed to its spherulites showing a sheaflike structure, leading to greater energy dissipation during impact tests.^{35,36} Such a suggestion is in accordance with the data presented here.

CONCLUSIONS

The influence of stearic acid treatment on the microstructure and mechanical properties of ter-

nary-phase PP composites containing EOR and calcium carbonate was investigated. A study of the phase morphology by SEM and DMA revealed a separate dispersion of elastomer and filler particles in the PP matrix. Stearic acid treatment of filler particles showed no significant influence on the dispersion and interaction of the filler and polymer components.

All ternary composites studied exhibited a reduction in tensile yield stress but showed great improvement in modulus and impact strength, relative to unfilled PP. Extensive dewetting and plastic deformation around the filler particles were the important mechanisms in the failure process. Calcium carbonate with stearic acid treatment was more effective in increasing the impact strength of the composites in comparison with composites containing the uncoated filler. The benefit of surface treatment is attributed principally to a reduction in filler nucleation effects, and, consequently, crystallization is induced in a temperature region which favors the presence of β -modifications of the PP structure.

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