

Structure and Properties of Talc-Filled Polypropylene: Effect of Phosphate Coating

Z. LIU and M. GILBERT*

Institute of Polymer Technology and Materials Engineering, Loughborough University of Technology, Loughborough, Leicestershire LE11 3TU, United Kingdom

SYNOPSIS

Talc surface treatment was carried out using a series of phosphate coating levels. The coated talc was characterized using quantitative diffuse reflectance FTIR analysis, which revealed a saturation of surface coverage at around 6 wt % phosphate. Coated and uncoated talc had a nucleation effect; and the degree of crystallinity of composites increased initially, then gradually decreased with increasing phosphate coating level. The talc dispersion was improved as coating level increased; however, the platelet alignment was reduced. The addition of 0.5 wt % phosphate to talc resulted in a maximum increase in tensile strength and a significant drop in flexural modulus. Improvement of falling weight impact properties of the composites were achieved at high coating levels. © 1996 John Wiley & Sons, Inc.

INTRODUCTION

Talc-filled polypropylene is widely used in automobile and appliance applications. These composites possess good surface quality, lower shrinkage, and higher heat deflection temperature than unfilled polypropylene. In addition, talc imparts greater stiffness, tensile strength, and creep resistance to the composites than other particulate fillers because of its plateletlike structure. Its use, however, is limited by a reduction in some mechanical and processing properties due to the poor affinity of talc with the polymer matrix.¹ Quite a lot of attention has been paid to minimizing the negative effects by means of talc surface treatment.²⁻⁶ However, in contrast to other silicate fillers, the work up to now has not produced appreciable improvement in the mechanical properties of the composites in question.

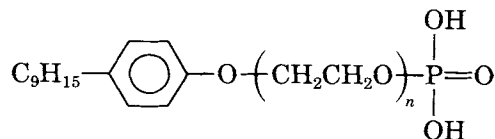
As a coupling agent, phosphate has been reported to be useful for calcium carbonate surface treatment.⁷ Fu et al. found that the toughness of high-density polyethylene/calcium carbonate (HDPE/CaCO₃) compounds was considerably enhanced by treating calcium carbonate with phosphate due to the improved

wettability of filler particle by the polymer, in turn reducing the defects in the compounds.⁸ A series of phosphates were investigated in our present work with the objective of improving talc dispersion and modifying the polypropylene-talc interface, hence enhancing the mechanical properties of the composite. Investigating the relationship between phosphate coating level and structure and mechanical properties was particularly interesting.

EXPERIMENTAL

Materials

Polypropylene was in powder form (Isplen PP.070) with a melt flow index (MFI) of 12 g/10 min (ASTM D1238). Talc (Talc de Luzenac, Lembalel 2SG 891/1), with a surface area of 6.74 m²/g, measured using the Brunauer-Emmett-Teller (BET) method, was used as filler. The phosphate used (Phospholan PNP9, Lankro Chemicals) is a water soluble liquid with a pH value of 2.5 (1% aqueous solution). Its molecular structure is



* To whom correspondence should be addressed.

Table I Apparatus and Test Conditions for Mechanical Measurement

Mechanical Test	Apparatus	Test Specimen Dimension	Test Condition
Tensile test	Lloyd Instruments tensile tester 1000, 10 kN load cell	50 × 12.65 × 3.2 mm	Crosshead speed 50 mm/min, temp. 22°C
Flexural test	Lloyd Instruments tensile tester 2000R, 5 kN load cell	50 × 12.65 × 3.2 mm	Crosshead speed 10 mm/min, temp. 22°C
Falling weight impact test	Rosand Instruments falling weight impact tester, type 5	Diameter 63 × 1.6 mm	Mass 25 kg, speed 3.0 m/s, temp. 22°C

The polypropylene powder and talc were dried in an oven at 90°C for 3 h prior to compounding to drive off moisture. The phosphate was used as received.

Talc Surface Treatment

A solution coating method was employed for talc surface treatment. To optimize coating conditions, a series of coating experiments were carried out using a phosphate/water solution. Temperature (30–85°C), time, and phosphate concentration were varied according to an orthogonal design method. The coated talc was then characterized by using quantitative diffuse reflectance Fourier transform infrared spectrophotometry (DRIFT) analysis (see following section). The results revealed that the coating on the talc surface, although rather limited, increased with increasing coating temperature. It was decided therefore to posttreat the coated filler at elevated temperatures. The DRIFT analysis results of postheated coated talc showed that postheating could significantly increase the phosphate coating on the talc surface as the temperature was increased from 100 to 125°C. A further increase in temperature did not result in any obvious change. Moreover, the amount of coating was reduced at 155°C because of decomposition of the phosphate. It was also found that 1-h postheating time was beneficial to talc surface coating. Based on these experimental results, the following talc surface treatment process was identified.

The phosphate was dissolved in water at 85°C, then talc was added and the slurry was agitated for 1 h. After being dried at 95°C, the coated talc was then postheated at 125°C for one more hour to allow the reaction to occur. The percentage of the phosphate ranged from 0.5 to 8.0% based on talc weight.

To investigate whether the phosphate had been chemisorbed on the talc surface, the coated and postheated talc was washed with warm water repeatedly to eliminate unreacted phosphate, then dried in a vacuum oven, and finally characterized

by DRIFT analysis. The results were compared with those of unwashed samples.

Compounding and Injection Molding

Polypropylene powder and talc were mixed in an 8-L high-speed Fielder mixer (T. K. Fielder, TRV8) at a rotation speed of 1500 rpm for 5 min. The mixtures were compounded using an APV (MP2000) corotating twin screw extruder and calibrated feeder maintaining the screw rotation speed at 100 rpm and the temperature of zones from the feeder to the die at 100, 200, 220, and 220°C. The extrudates were cooled, pelletized, and then dried in an oven at 80°C to drive off residual water left over from the cooling process. A Negri Bossi NB55 injection molding machine was used to produce unfilled and filled polypropylene mechanical test specimens under identical conditions (injection pressure 140 bar, injection temperature 230°C, and mold temperature 60°C). To investigate the influence of phosphate coating level on composite properties, a constant talc load of 40% based on polypropylene weight was used. Three samples containing phosphate alone at levels corresponding to a talc coating of 0.4, 0.8, and 1.2% were also produced.

Mechanical Testing

Tensile, flexural, and falling weight impact tests were carried out to obtain mechanical properties. Test conditions are listed in Table I. At least five specimens were used for the tensile and the flexural tests, and 10 for the falling weight impact test. Average values and standard deviations were recorded.

DRIFT Characterization

The coated talc was characterized by DRIFT. An FTIR spectrophotometer (Mattson 3000) and a DRIFT accessory (PIN 19900 Series) were used at a resolution of 2 cm⁻¹, and 64 scans were accumu-

lated. The samples were dried in a vacuum oven at 80°C overnight prior to DRIFT characterization. KBr was used as standard and diluent.

Thermal Analysis

Differential scanning calorimetry (DSC) measurements were performed using a Du Pont 2000 Thermal Analyzer fitted with a DSC cell. Samples taken from the waisted section of injection molded tensile test specimens were heated up to 205°C, kept at this temperature for 5 min, and then cooled down to 40°C at a constant rate of 10°C/min under N₂ atmosphere. Typical thermograms of the compound with 0.5 wt % phosphate coated talc and that of pure polypropylene are shown in Figure 1.

Scanning Electron Microscopy (SEM) Observation

Injection molded tensile testing specimens of unfilled and filled polypropylene were cooled with liquid nitrogen and then fractured. The fracture surfaces were observed with a scanning electron microscope (Cambridge Instruments Steroscan 360) to obtain information about filler dispersion and interfacial morphology.

RESULTS

Coating on Talc Surface

Figure 2 shows the DRIFT spectra from uncoated and coated talc. An absorbance peak around 2900

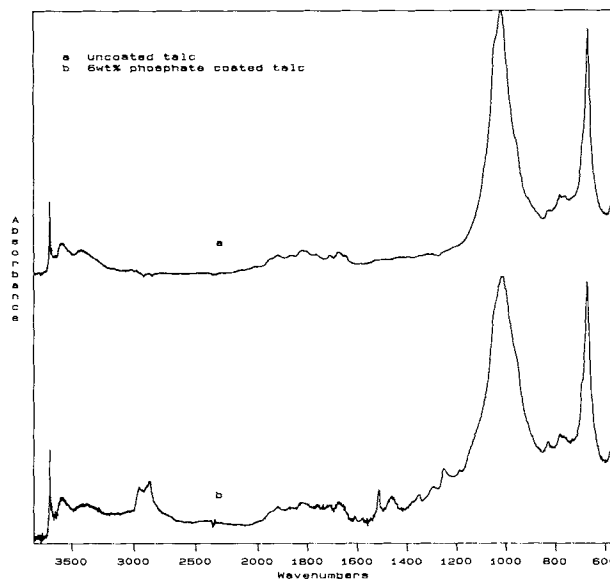


Figure 2 DRIFT spectra of (a) uncoated and (b) coated talc.

cm⁻¹, due to C—H stretching absorbance arising from the alkyl chain of the phosphate molecule, can be clearly observed for the coated sample [Fig. 2(b)]. The peak is still very clear for washed coated talc [Fig. 3(b)], although it is smaller than that of the unwashed sample [Fig. 3(a)]. This suggested that a reaction had taken place between the phosphate and talc surface. To investigate the effect of phosphate concentration, an internal standard method was employed.⁹ From the DRIFT spectra of

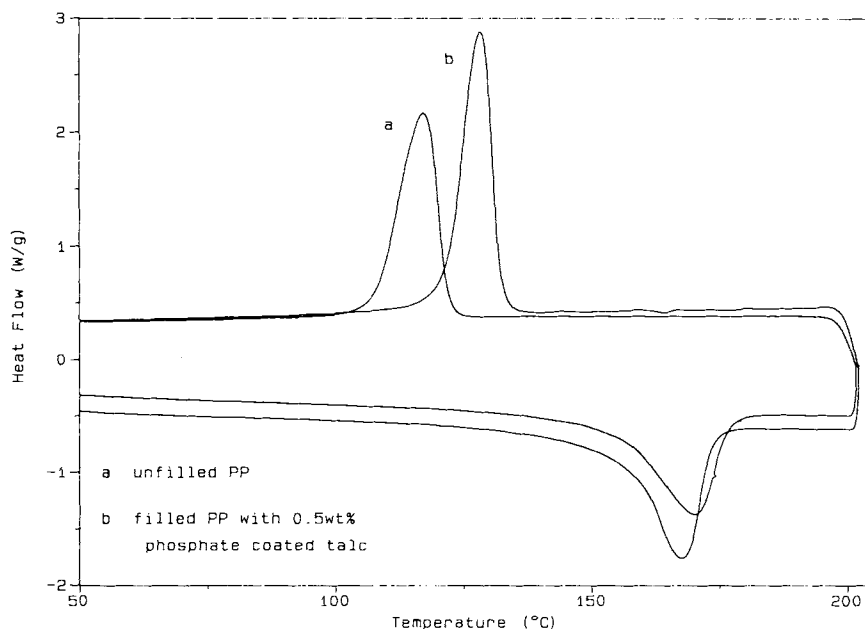


Figure 1 DSC thermograms of (a) unfilled and (b) filled polypropylene.

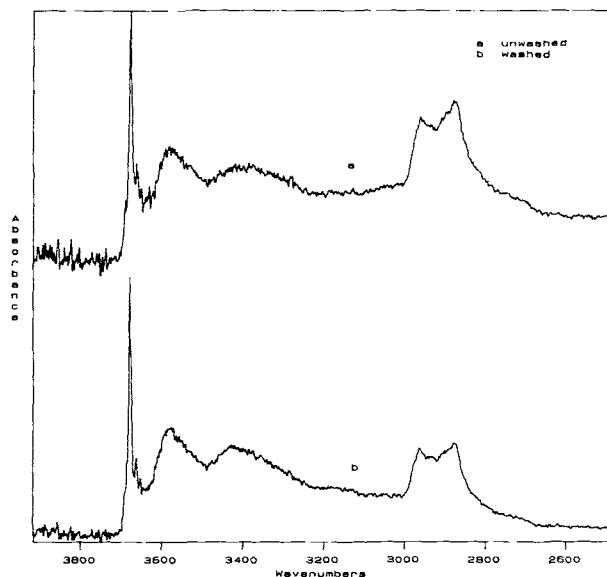


Figure 3 DRIFT spectra of 6% phosphate coated talc.

talc coated with various percentages of phosphate, the integrated area of the CH group peak and that of the OH group peak at 3676 cm^{-1} from talc were calculated; and the ratioed areas of CH/OH peaks representing the coating of phosphate on the talc surface are plotted against the phosphate coating level in Figure 4.

The coating on the talc surface increased with increasing phosphate concentration up to around 6 wt %, where a plateau in the curve begins due to surface coating saturation.

Crystallization Behavior

From DSC thermograms the enthalpies attributed to melting of polypropylene were determined and divided by enthalpy of a 100% crystalline sample of polypropylene to give the degree of crystallinity of samples.¹⁰ For 100% crystalline polypropylene, the enthalpy value of 163 J/g was used.¹¹ The degree of crystallinity of filled polypropylene samples as a function of phosphate coating level is plotted in Figure 5. The peak temperature (T_p) and onset of recrystallization temperature (T_o), which give an indication of the nucleation effect of the talc, were also obtained from the DSC traces and are shown in Table II.

By inspection of these recrystallization data and the thermograms in Figure 1, it is clear that the addition of talc, both coated and uncoated, resulted in a significant increase in temperature of crystallization, demonstrating the nucleating effect of talc.

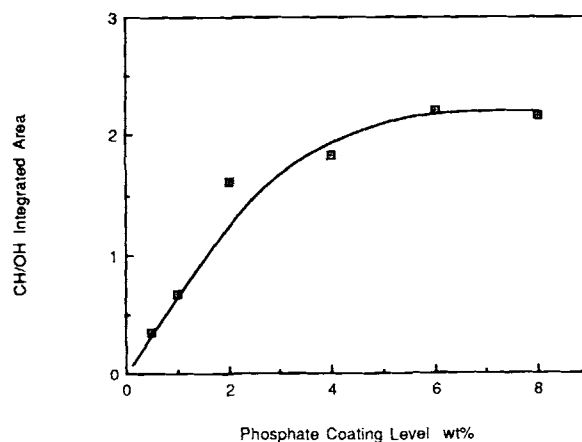


Figure 4 Variation of CH/OH integrated area with coating level.

However, the influence of phosphate coating level on the temperature of crystallization was not very large, although there was a maximum value at around 0.5 wt %. Beyond 2 wt % phosphate the temperatures became independent of coating level. When phosphate was used without filler no significant nucleation was observed. The curve of crystallinity vs. coating level for talc-filled samples peaked at 0.5 wt % and then gradually dropped down with increasing coating level to values even lower than that of virgin polypropylene (Fig. 5). An increase in crystallinity was also observed when phosphate was used alone (Fig. 6), although in this case the plateau, or maximum, occurred at a higher phosphate concentration.

Fracture Surfaces

The fracture surface of polypropylene and its composites with coated and uncoated talc were studied

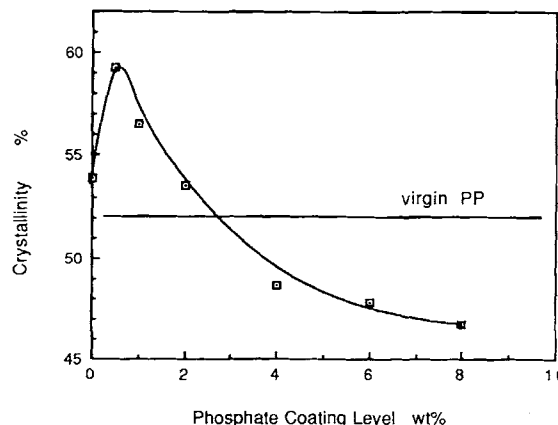


Figure 5 Effect of coating level on crystallinity.

using SEM in the core zone of the injection molded tensile specimens. Typical micrographs are shown in Figure 7. The fracture surface of uncoated talc filled polypropylene clearly showed some talc agglomerates and long narrow holes that resulted from plateletlike talc pull-out [Fig. 7(a)]. For the samples with coated talc, the agglomerates of talc and the holes were still observable at low coating level. However, the size of talc agglomerates and of the holes became smaller as coating level increased, and the agglomerates and holes were hardly seen for the sample with 8 wt % phosphate coated talc [Fig. 7(e)]. The coating process itself did not seem to have any effect on the talc particles (Fig. 8). These photographs implied the breakdown of talc agglomerates during processing when coated talc was used, hence the improvement of filler dispersion in the polypropylene composites. A close examination of coated talc particles on the fracture surface did not reveal any adherence of the matrix material (Fig. 9), which suggested that the interfacial adhesion between phosphate coated talc and polypropylene was not very good.

Mechanical Properties

The tensile and flexural properties of virgin and filled polypropylene are presented in Table III and Figures 10 and 11. Tensile yield strength appears higher than break strength because values were not corrected for reduction in cross-sectional areas. Corrected values showed similar trends. The addition of talc produced a stronger but more brittle material. The introduction of about 0.5 wt % of phosphate to the talc resulted in the maximum improvement in tensile strength. With further increase in coating level, the tensile strength gradually decreased and reached a plateau around 6 wt % of coating level (Fig. 10). Phosphate alone appeared to produce a similar result together with corresponding changes in elongation at break (Fig. 12). The addition of uncoated talc to polypropylene produced a significant drop in elongation at break. It showed some improvement as phosphate was ap-

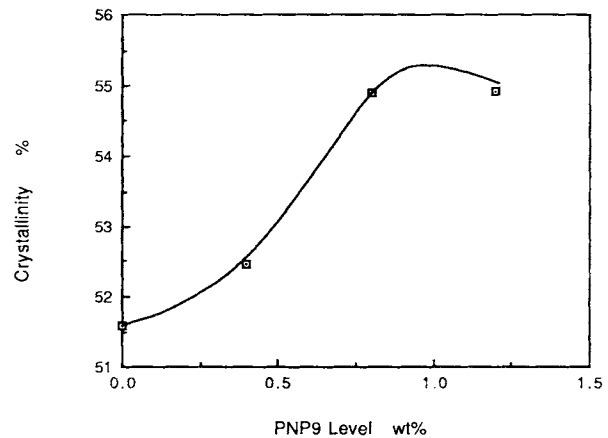


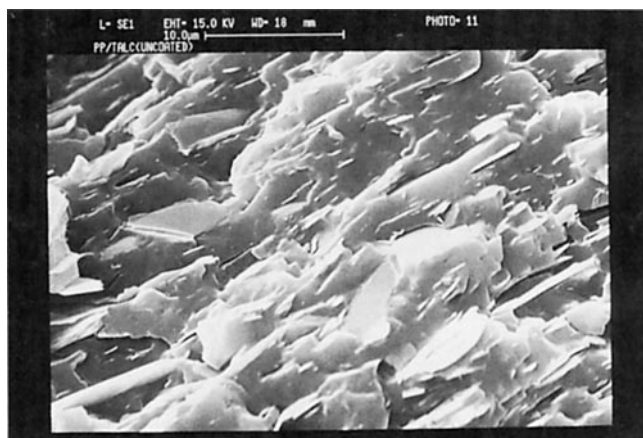
Figure 6 Effect of phosphate concentration on crystallinity.

plied, although remaining lower than that of unfilled polypropylene. Figure 11 clearly shows that the flexural modulus of the composites decreased with increasing coating level up to about 6 wt %. An addition of as little as 0.5 wt % phosphate caused the largest reduction. However, the modulus values of the composites with both coated and uncoated talc were much higher than that of pure polypropylene, as would be expected. Phosphate used alone produced a small increase in modulus when used at a concentration up to the equivalent of 1.2% on talc (Fig. 13). (Property results for this series are presented as a ratio to the property obtained for unfilled polypropylene.)

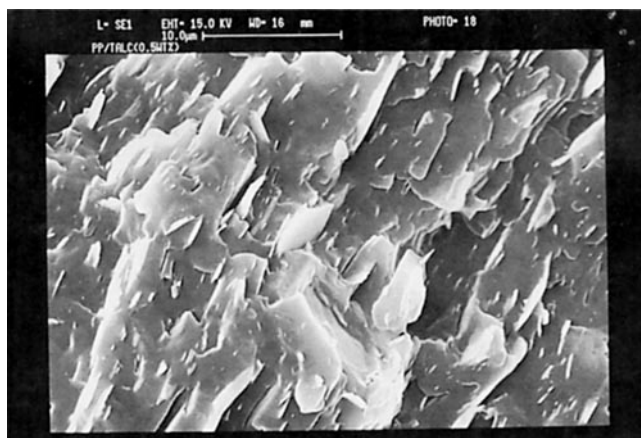
The effect of coating level on falling weight impact energy is shown in Figure 14. It seems that the increase of coating level did not give rise to a major change in impact strength up to 4 wt %, but some improvement was achieved at higher coating levels. The values for failure energy and also failure deflection of composites were even higher than that of unfilled polypropylene when more than 6 wt % phosphate was used. Low levels of phosphate used alone produced very small changes in impact properties, approximately in line with those produced by similar levels of phosphate coating.

Table II Recrystallization Temperatures

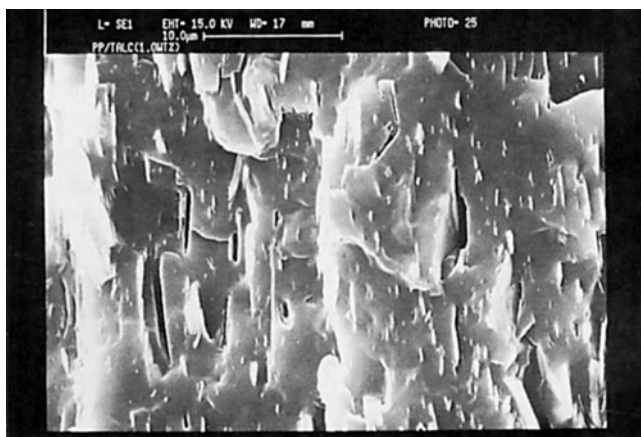
	Coating Level (wt %)							
	PP	0.0	0.5	1.0	2.0	4.0	6.0	8.0
T_p (°C)	117	127	128	127	127	126	126	126
T_o (°C)	121	132	132	131	130	131	131	131



(a)



(b)



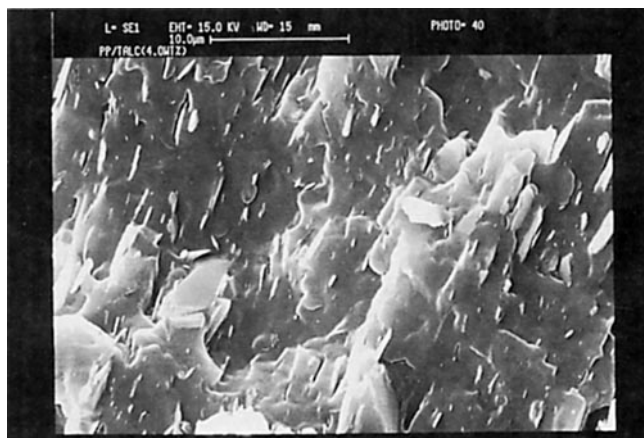
(c)

Figure 7 SEM photographs of fracture surfaces: (a) with uncoated talc; (b–e), with 0.5, 1.0, 4.0, and 8.0% phosphate coated talc, respectively; and (f) pure polypropylene.

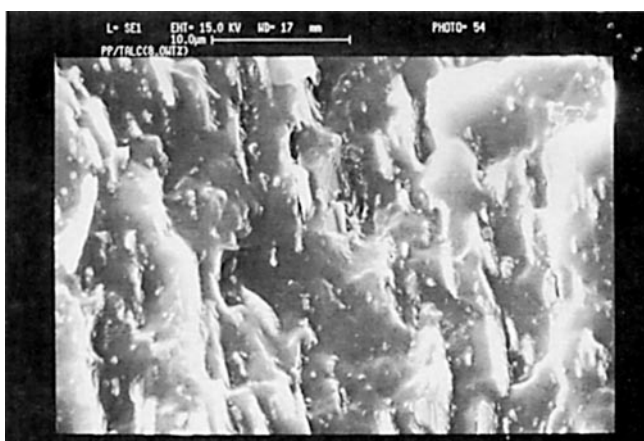
DISCUSSION

It is beneficial if a coating is chemically bonded to the filler by reaction between a functional group and a group on the filler surface. In earlier work using a

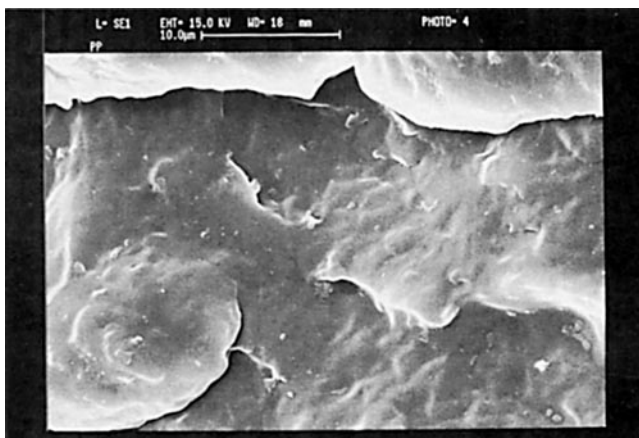
phosphate coating, Nakatsuka⁷ showed that calcium carbonate reacted with dihydrogen phosphates, and proposed a reaction scheme to explain the reaction products. In the present work it was anticipated that the hydroxyl groups from the phosphate would react



(d)



(e)



(f)

Figure 7 (Continued from the previous page)

with the Si—OH groups present at the edges of the talc particles with the elimination of water.¹² The DRIFT technique demonstrated that phosphate coating of talc occurred, although the reaction described produced no unique chemical groups, so it

cannot be confirmed. The washing experiment showed that a proportion of the coating was water soluble, suggesting that it was partly physisorbed and partly chemisorbed. If CH/OH ratios are calculated for the traces in Figure 3 for the sample with

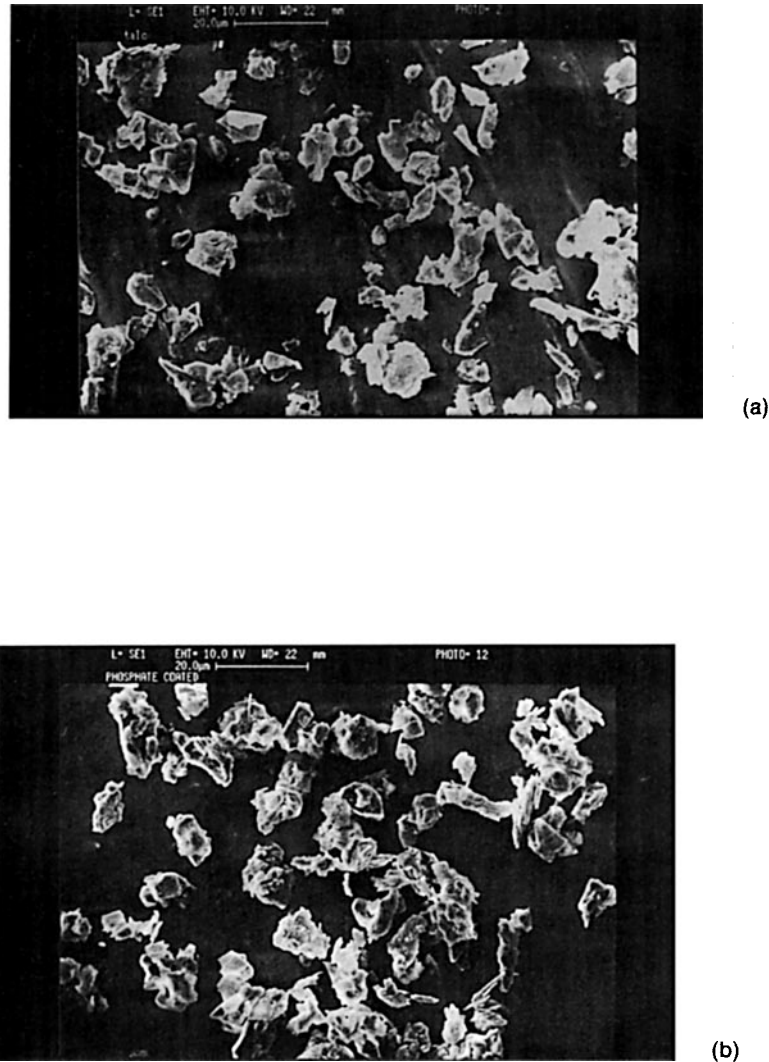


Figure 8 SEM photographs of (a) uncoated and (b) coated talc.

6% phosphate coating, and compared with the data in Figure 4, it is found that the coating concentration was reduced to about 4.8% by washing, suggesting that 80% of the coating had reacted with the filler.

Fillers used for the compounding experiments were not washed, so both chemisorbed and physisorbed phosphate would be present. Scanning electron micrographs (Fig. 7) suggested that the phosphate coating improved wettability of the filler, hence facilitating its dispersion and reducing the size of the platelets. Dispersion of the talc in the polypropylene matrix increased with phosphate concentration. As described previously,^{13,14} thermal analysis results suggested that talc acted as a nucleating agent for the polypropylene. The addition of talc increased both crystallization temperature and crystallinity of the matrix polymer. Both increased

further with 0.5% phosphate coating, then decreased to a plateau as the level of phosphate coating was increased. When the phosphate coating level was 4% and above, the crystallinity was lower than unfilled polypropylene, although the nucleation effect still existed. Low levels of phosphate used alone appeared to affect crystallinity in a similar manner, suggesting that reacted phosphate initially behaves as a plasticizer. It appeared that higher phosphate levels restricted the level of crystallinity, while crystallization rate was unaffected. It is likely that there is more physisorbed phosphate present at higher phosphate concentrations; it is possible that this could account for the observed crystallinity decrease by hindering crystallite growth.

Mechanical properties are modified by phosphate coating of the talc. Such changes can be caused by

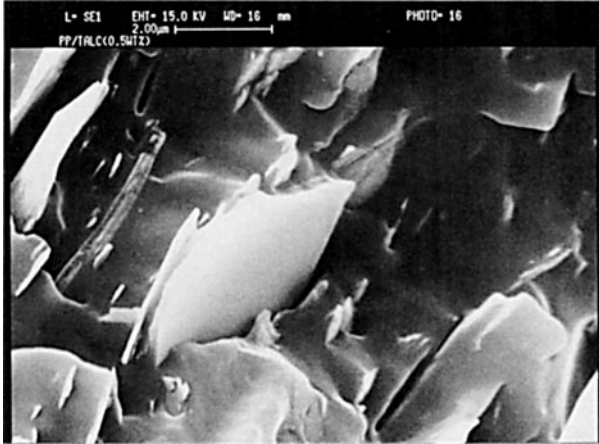


Figure 9 SEM photograph of fracture surface of composite with 0.5% phosphate coated talc.

a number of different factors. Furthermore, these factors can interact, making the understanding and prediction of properties difficult. For the system studied here, the observed changes in tensile properties can be attributed to crystallinity. Both yield and tensile strength mirror the changes in crystallinity with coating level (Figs. 5, 10). The same is true of the yield properties obtained without filler (Fig. 12). Extension at break also reflected crystallinity changes, showing higher values at lower crystallinities, and vice versa.

Flexural test results showed similar trends. Yield strength appeared to show a maximum as coating level increased, although the variation in flexural yield stress was small. Elongation at yield results showed some scatter, but were not inconsistent with

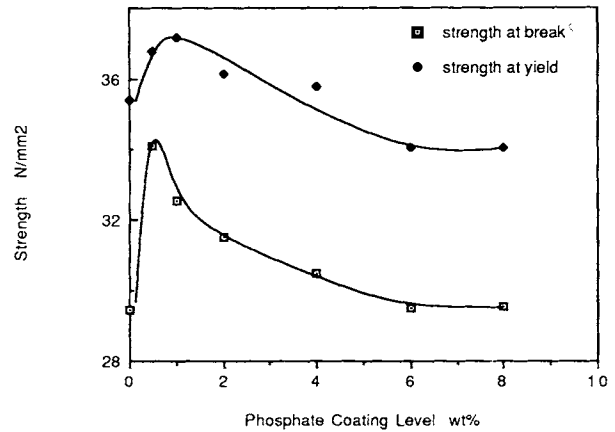


Figure 10 Effect of coating level on tensile properties.

results discussed so far. Flexural modulus was increased significantly by the introduction of filler, as expected. Coating the filler resulted in a steady and significant decrease in modulus as coating level increased. However, phosphate alone produced a small increase in modulus. Although the decreased crystallinity at higher coating levels would be expected to decrease the modulus, these results cannot be explained by crystallinity changes alone because no maximum is observed at low coating concentrations. However, the substantial alignment observed for the large uncoated platelets [Fig. 7(a)] may account for the high modulus of this sample. These large aligned platelets disappear as coating level increases, and could account for the reduction in modulus (see below). It is known¹⁵ that the exposed basal plates on the larger (upper and lower) surfaces of talc particles contain no hydroxyl groups; these are found

Table III Tensile and Flexural Properties

	Coating Level (wt %)							
	PP ^a	0.0	0.5	1.0	2.0	4.0	6.0	8.0
Tensile break strength (N/mm ²)	23.7 (3.9)	29.5 (0.9)	34.1 (1.9)	32.5 (0.9)	31.5 (0.8)	30.5 (0.6)	29.5 (1.3)	29.6 (1.0)
Tensile yield strength (N/mm ²)	35.9 (0.4)	35.4 (0.2)	36.8 (0.9)	37.2 (0.2)	36.2 (0.2)	35.8 (0.1)	34.1 (0.2)	34.1 (0.1)
Tensile elongation (%)	25.3 (5.6)	8.9 (1.8)	6.5 (1.7)	8.8 (1.2)	11.3 (2.1)	11.1 (1.0)	10.6 (1.9)	10.8 (2.4)
Flexural modulus (N/mm ²)	1626 (52.6)	3782 (95.8)	3276 (51.2)	3261 (46.5)	2883 (100.9)	2981 (76.8)	2856 (103.6)	2763 (56.2)
Flexural yield strength (N/mm ²)	61.8 (0.9)	60.6 (1.0)	61.7 (0.5)	63.2 (0.7)	63.6 (0.7)	62.3 (0.3)	61.0 (0.5)	61.5 (0.2)
Flexural yield strain (%)	16.7 (1.2)	10.7 (0.4)	11.2 (1.1)	12.2 (0.2)	11.7 (1.3)	12.3 (0.8)	12.3 (0.5)	12.4 (0.5)

Standard deviations shown in parentheses.

^a Unfilled polypropylene.

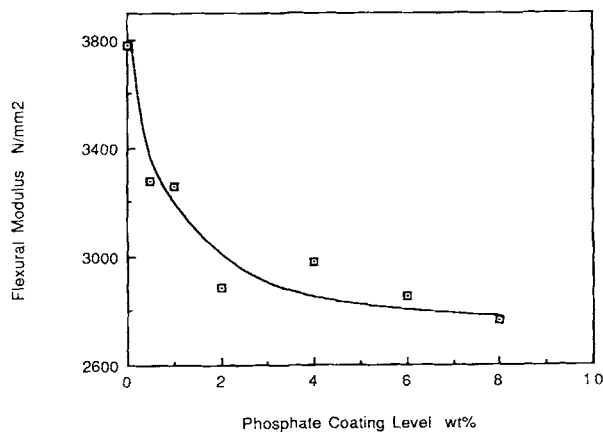


Figure 11 Effect of coating level on flexural modulus.

at the broken edge of the silicate planes, and initial reaction with the phosphate coating would occur here. However, talc has a layered structure with layers linked by internal hydroxyl groups. The phosphate coating agent used in this work is strongly polar [calculated solubility parameter approximately $10.3 \text{ (MJ/m}^3)^{1/2}$]. It may be speculated that some interaction could occur from the edges of the talc particles with internal hydroxyl groups, resulting after processing in a reduction in talc agglomerate size.

Impact results also suggest that property changes cannot be attributed to crystallinity alone. Although the least crystalline samples do have the highest impact strength, effects of coating level are different. Suetsugu¹⁶ reported that improvement of filler dispersion drastically improves the falling weight impact failure energy of calcium carbonate filled polypropylene. The improved dispersion and reduced filler alignment observed here with increased phosphate coating level may be having a similar effect.

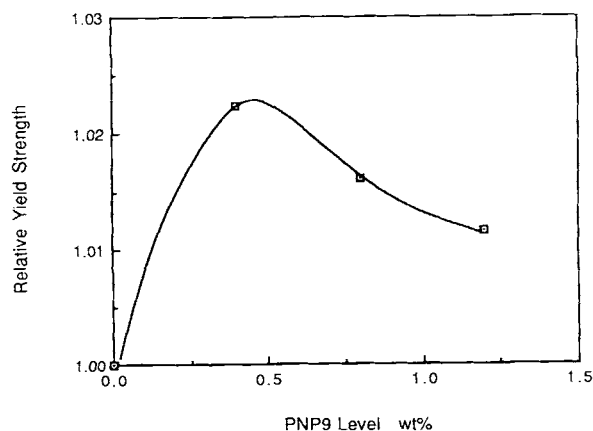


Figure 12 Effect of phosphate concentration on tensile yield properties.

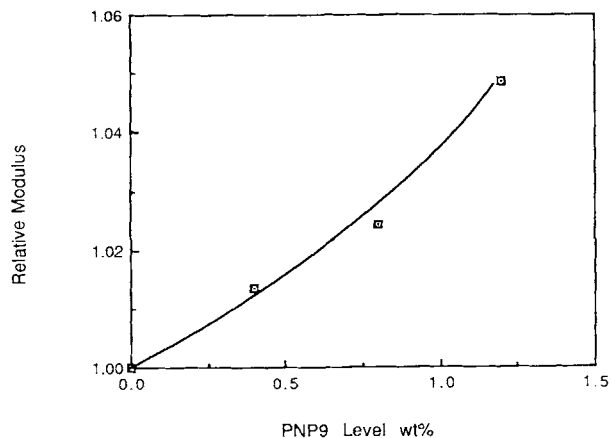


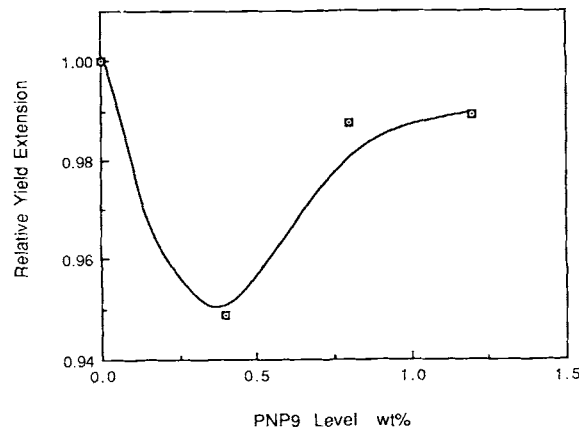
Figure 13 Effect of phosphate concentration on flexural modulus.

Also, highest impact strengths are observed for samples containing greater than saturation levels of phosphate coating, so there may be some matrix plasticization at the filler interface.

The effect of fillers on the mechanical properties of thermoplastics has been studied extensively, and theoretical equations to predict properties have been proposed. Nielsen¹⁷ derived the following equation for the change in elongation at break with filler content for systems where there is perfect adhesion between the filler and the matrix.

$$\epsilon'/\epsilon = 1/(1 - \phi_F^{1/3}) \quad (1)$$

where ϵ is the elongation at break for the polymer, ϵ' is the elongation at break for the filled compound, and ϕ_F is the volume fraction of the filler. In the present work the volume fraction of filler was maintained constant at 0.176 (calculated from a polypropylene density of 890 kg/m^3 and a talc density of



2780 kg/m³). From eq. (1) the ratio ϵ'/ϵ would therefore be 0.44 for perfect adhesion. Actual values for compounds studied in this work are shown in Table IV. It should be noted however, that this equation assumes that the elongation at break of the matrix remains unchanged. This will not apply because of changes in crystallinity. Figure 5 shows that the sample with 3% phosphate coating does have a similar crystallinity to the unfilled polypropylene, and ϵ'/ϵ for samples with 2 and 4% coating are very close to 0.44. Values of σ'_y/σ_y where σ_y is the yield strength of the polymer and σ'_y is the yield strength of the compound can also be considered. In the absence of any adhesion, this ratio would decrease in the presence of filler because the effective cross-sectional area of the polymer would decrease. Data in Table IV, particularly for the samples containing 2 and 4% phosphate coating on the filler, show virtually no change in yield stress due to the presence of the filler. Attempts have been made to predict yield stresses for filled polymers, but are complicated for nonspherical particles that may be present at a variety of orientations. A number of assumptions have to be made, and comparison between theory and experimental results would require that data are available for a range of volume fractions. Nevertheless, the fact that σ'_y/σ_y remains virtually unchanged suggests that there must be significant adhesion between the talc and polypropylene. This is consistent with the observed nucleating effect, which is irrespective of talc coating (Table II). Clearly, some crystallization originates on the talc surface, although it has been shown¹⁸ that only a small proportion of particles are nucleation active. Despite the conclusions reached above, Figure 9 does show that good adhesion does not always occur. However,

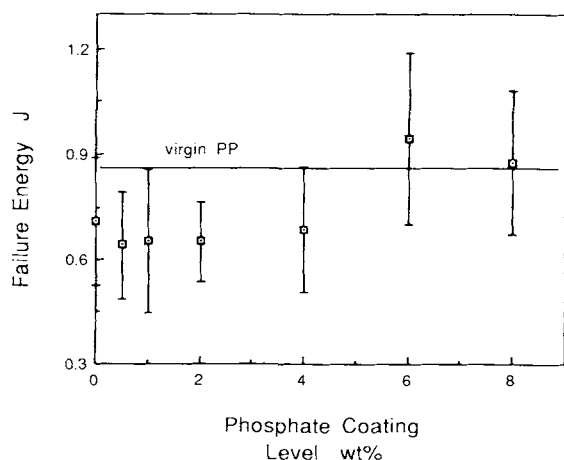


Figure 14 Effect of coating level on falling weight impact failure energy.

Table IV Relative Tensile Properties

Coating Level (wt %)	ϵ'/ϵ	σ'_y/σ_y	E'/E
0	0.35	0.99	2.33
0.5	0.26	1.02	2.01
1	0.35	1.04	2.00
2	0.44	1.01	1.78
4	0.44	1.00	1.83
6	0.42	0.95	1.76
8	0.43	0.95	1.70

Figure 7 suggests that it may increase with coating level, because fewer holes are observed on the fracture surface. It must be realized that these systems are complex, and further information about crystallite orientation and morphology would be required to explain them fully.

Modulus values of filled polymers are not affected by adhesion, but depend on filler and polymer modulus, and for nonspherical particles depend on filler aspect ratio and filler alignment. Radosta^{19,20} and Riley et al.²¹ obtained mechanical property data for talc filled polypropylene and demonstrated the high level of modulus enhancement achievable through the addition of talc. Radosta¹⁹ showed that flexural modulus could be enhanced by a factor of 2.6 with a filler level equivalent to that used in this work; data in Riley's article give a value of about 3.1. Table IV shows an enhancement of 2.3 (ϵ'/ϵ) for the uncoated talc used here; this is significantly reduced by filler coating. Radosta used the Halpin-Tsai equation, modified for platy fillers, and was able to obtain a good match between experimental and predicted data. The modulus measured in this work for the compound filled with uncoated talc also fits with predicted data, but the prediction does include an empirical scaling factor.

CONCLUSIONS

The phosphate coating used reacted with talc filler particles. A proportion of the coating appeared to be physisorbed onto the filler surface. Saturation of the surface occurred at a phosphate concentration of about 6%. Changes in coating significantly affected the morphology and mechanical properties of the polypropylene based composites. Increased coating level improved the dispersion of talc in the polymer matrix. Crystallinity increased on coating, then decreased gradually as phosphate concentra-

tion increased. At the highest coating level, talc still acted as nucleating agent, even though the crystallinity of the polypropylene was lower than the unfilled polymer. Tensile properties appeared to mirror the crystallinity changes exactly. Impact properties appeared to be attributable to crystallinity changes and improved filler dispersion. Flexural modulus can be explained in terms of the size and alignment of filler platelets. Property measurements imply some filler/matrix adhesion at a coating level of about 3%; photomicrographs suggest that this may be less at lower coating levels.

The authors are grateful to Cabot Plastics Limited for the support of the project, to the Chinese State Educational Commission and British Council for financial support for Z. L., and to Mr. R. A. Venables for his help in sample preparation.

REFERENCES

1. R. Gachter, *Plastics Additives Handbook*, 3rd ed., Hanser, New York, 1990.
2. S. N. Maiti and K. K. Sharma, *J. Mater. Sci.*, **27**, 4605 (1992).
3. V. Khunoua, M. Sain, and Z. Brunowska, *Polym. Plast. Technol. Eng.*, **32**, 311 (1993).
4. M. Ramos, M. S. Berna, and J. Matheu, *Polym. Eng. Sci.*, **31**, 245 (1991).
5. J. E. Stamhuis, *Polym. Comp.*, **9**, 72 (1988).
6. J. E. Stamhuis, *Polym. Comp.*, **5**, 202 (1984).
7. T. Nakatsuka, *Polym. Sci. Technol.*, **27**, 51 (1985).
8. Qiang Fu, Jiusi Shen, and Guiheng Wang, *Polym. Mater. Sci. Eng.*, **1**, 107 (1992).
9. M. Gilbert, I. Sutherland, and A. Guest, *Conference Proceedings, Filplas 92*, Manchester, United Kingdom, 1992.
10. J. V. Dawkins, *Development in Polymer Characterization-1*, Applied Science Publishes, Ltd, London, 1978.
11. J. P. Runt, in *Encyclopaedia of Polymer Science and Engineering*, 2nd ed, Vol. 4, H. F. Mark, N. M. B. Kales, C. G. Overberger, and G. Menges, Eds., Wiley-Interscience, New York, 1985, p. 482.
12. A. C. D. Newman, *Chemistry of Clays and Clay Minerals*, Longman, Scientific and Technical, Harlow, 1987, p. 412.
13. J. Menczel and J. Varga, *J. Thermal Anal.*, **28**, 161 (1983).
14. M. A. Ramos and J. P. V. Matheu, *Polym. Comp.*, **9**, 105 (1988).
15. T. P. Bragg, *Plast. Eng.*, **September**, 31 (1973).
16. Y. Suetsugu, *Int. Polym. Proc.*, **3**, 184 (1990).
17. L. E. Nielsen, *J. Appl. Polym. Sci.*, **10**, 97 (1966).
18. F. Rybniker, *J. Appl. Polym. Sci.*, **27**, 1479 (1982).
19. J. A. Radosta, 33rd SPE-Antec Conference, May 1975.
20. J. A. Radosta, 34th SPE-Antec Conference, April 1976.
21. A. M. Riley, C. D. Puynter, P. M. McGenitz, and J. M. Adams, *Plast. Rubber Proc. Appl.*, **14**, 85 (1990).

Received April 11, 1995

Accepted July 25, 1995