

Mineral Filler Reinforcement for Commingled Recycled-Plastic Materials

H. D. Putra,¹ Y. Ngothai,¹ T. Ozbakkaloglu,² R. Seracino³

¹School of Chemical Engineering, The University of Adelaide, Adelaide, South Australia 5005, Australia

²School of Civil, Environmental and Mining Engineering, The University of Adelaide, Adelaide, South Australia 5005, Australia

³Department of Civil, Construction and Environmental Engineering, North Carolina State University, Raleigh, North Carolina 27695-7533

Received 26 December 2007; accepted 6 December 2008

DOI 10.1002/app.29861

Published online 6 March 2009 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Three types of mineral fillers, gypsum, wollastonite, and talc, were investigated for their ability to modify the mechanical properties of commingled recycled-plastic composites containing 0.07–0.26 v/v of filler. Mechanical test results showed that the talc reinforced composites were significantly better in mechanical properties when compared with the gypsum and wollastonite composites. Scanning electron microscopy (SEM) showed that gypsum formed large agglomerates in the matrix. Interfacial adhesion between filler and matrix was evaluated using simple empirical models. To enhance the adhesion, talc, and wollastonite were pretreated with silane coupling agents, 3-methacryloxypropyltrimethoxy silane

(γ -MPS) and 3-aminopropyltriethoxy silane (γ -APS). This did not result in any significant improvement to the material properties. The γ -APS treatment, however, increased the tensile properties of the composites by $\sim 5\%$ when compared with the γ -MPS treatment. The SEM investigations showed that the γ -APS treatment provided better adhesion of filler particles and hence voids were less likely to form in the matrix when compared with the γ -MPS composites. © 2009 Wiley Periodicals, Inc. *J Appl Polym Sci* 112: 3470–3481, 2009

Key words: commingled recycled plastic; mineral fillers; silanes

INTRODUCTION

The high consumption of plastics generates large volumes of plastic waste. In 2006, Australia consumed at least 1.5 million tonnes of plastic, of which only 16% was recycled.¹ Because plastics seem to be almost irreplaceable in some applications, for example, their use as packaging material, then recycling is the best way to reduce postconsumer plastics from disposal into landfill.

There are many ways to recycle postconsumer plastics by the manufacture of products such as vineyard and fence posts, decking boards and outdoor furniture. This type of industry which uses the recycled plastic in a load bearing role has shown a significant growth in Australia. Even more so with the prohibition of CCA preserved timber for domes-

tic use in Australia which has led to increased markets for the recycled plastic products.

However, consistent sourcing of feedstock is a major challenge due to the “commingled” nature of the postconsumer waste stream, which often varies in composition and quality. Waste segregation is considered to be a costly and time consuming procedure; therefore, the direct recycling of commingled plastics is still a cost-effective practice.^{2,3}

The majority of the “raw” material is predominantly sourced from polyolefins.^{4–6} Because of the inferior mechanical characteristics of polyolefins, fillers are often added to the plastic waste to enhance stiffness and strength of the final products. Common fillers used in plastic recycling include mineral particulates (e.g., talc, calcium carbonate, gypsum, and wollastonite) and fibrous fillers (e.g., glass fiber and saw dust).

Despite the excellent reinforcement provided by glass fiber for plastic composites, recyclers often avoid this reinforcement agent due to its high cost. Instead, they turn to lower cost alternatives, such as mineral fillers. It has been reported that mineral fillers can enhance the strength and stiffness of polymers.^{7–9} When used to reinforce polymers, the performance of a filler is influenced by particle shape,

Correspondence to: Y. Ngothai (yngothai@chemeng.adelaide.edu.au).

Contract grant sponsor: The ARC Research Linkage; contract grant number: LP 0455409.

Contract grant sponsor: Advanced Plastic Recycling of South Australia (APR) of South Australia.

particle size, and/or particle size distribution as well as adhesion at the filler-matrix interface.

There seem to be diverse results from the various studies that have been performed to determine how particle size affects the mechanical properties of virgin polymers.^{10–12} In general, the studies show that smaller particle size provides greater improvement in the tensile modulus, but Bose and Mahanwar¹³ and Texeira et al.¹⁴ have reported that fillers with larger particle size provide higher moduli. Still other researchers noted that mechanical properties are independent of particle size.^{15–17}

Fillers are often coated with coupling agents to enhance chemical bonding between filler particles and the polymer matrix. The selection of an appropriate coupling agent involves identification of a suitable group of agents that is compatible with the targeted polymers. Although there is no fundamental rule regarding this relationship, researchers have reported notable improvements in stiffness and yield strength of polyolefin composites with the use of methacrylate and amino silanes.^{14,18–20} In particular, two kinds of silanes, 3-methacryloxypropyltrimethoxy silane (γ -MPS) and 3-aminopropyltriethoxy silane (γ -APS), are the most commonly used coupling agents.

In the manufactures of recycled plastic profiles, many attempts have been made to improve the mechanical properties of the composite materials. Little research has been carried out on the effectiveness of mineral fillers as reinforcement for commingled recycled-plastic waste. Therefore, this study examined the effects of three kinds of mineral fillers (i.e., gypsum, wollastonite, and talc) on the mechanical properties of commingled recycled plastics. In addition, an investigation into the effects of pretreatment of some fillers with two commercial silanes, γ -MPS and γ -APS on the properties of the resulting composites was undertaken. The results

give some insights into the fillers that can be used in the production of the composites.

EXPERIMENTAL

Materials

Commingled plastics flake was supplied by Advanced Plastic Recycling (APR) Pty of South Australia. The material was obtained from postconsumer household and industrial waste. The waste contained mainly packaging materials (e.g., plastic bags, shrink wrap containers, drink bottles, and beverage crates), as well as factory off-cuts and polystyrene foams. The composition of the mixture was determined by the sink-float method to be 70–80 wt % polyolefins (polyethylene and polypropylene) and expandable polystyrene (EPS) foam beads. The remaining fraction was made up of other polymers, such as polycarbonate, nylon, polystyrene (PS), and polyethylene terephthalate (PET) which were removed before compaction into pellets.

Three commercial mineral fillers were studied: talc, wollastonite, and anhydride gypsum. Glass fiber was also used to enable comparisons with the mineral fillers. The two silanes, γ -MPS and γ -APS, were blended with the fillers and plastic waste. Properties of these materials are summarized in Table I. The plastics flake was compacted into pellets to ensure good blending with the fillers. The pellets were compounded with the filler and then this compound was injection molded.

Compaction of plastic waste

The plastic flakes were fed through a John's single-screw extruder (the screw length and diameter ($L/$

TABLE I
Material Properties

Material	Particle shape	Hardness (Mohs)	Specific gravity	Descriptions ^a
APR plastic waste ^b	–	–	0.95	MFI: 0.811 g/10 min
Anhydride gypsum (CaSO ₄)	Sphere	2	2.90	$d_{10} = 1 \mu\text{m}$ $d_{50} = 4.8 \mu\text{m}$ $d_{90} = 45.9 \mu\text{m}$
Wollastonite (CaSiO ₃)	Acicular	5	2.96	$d_{10} = 3.5 \mu\text{m}$ $d_{50} = 20.8 \mu\text{m}$ $d_{90} = 59.5 \mu\text{m}$
Talc (Mg ₃ Si ₄ O ₁₀ (OH) ₂)	Plate	1	2.76	$d_{10} = 3.3 \mu\text{m}$ $d_{50} = 14.5 \mu\text{m}$ $d_{90} = 42.4 \mu\text{m}$
Chopped strand glass fiber				Average length = 3 mm Average diameter = 10 μm
3-Methacryloxypropyltrimethoxy silane	–	–	1.045	–
3-Aminopropyltriethoxy silane	–	–	0.95	–

^a d_{10} , d_{50} , and d_{90} are mean diameters where 10, 50, or 90% of the particles are below the corresponding size.

^b After separation from nonpolyolefin polymers.

D) ratio was 15) located at Plastic Granulating Services (PGS) of South Australia. The pellets were produced at the rate of 5 kg/h with the optimum screw rotation being 110 RPM, whereas the temperatures of zones 1, 2, 3, and die were set at 210°C, 210°C, 220°C, and 220°C, respectively. The extruded strands were chopped into small pellets having diameters of 2–3 mm.

Compounding of fillers and plastic pellets

Fillers and plastic pellets were compounded using a Brabender corotating, intermeshing, twin-screw extruder (RMIT University, Victoria) having an L/D ratio of 20. The barrel temperatures of the extruder were set at 170°C, 195°C, and 195°C. The mixture strands were chopped into small pellets (2–4 mm diameter) for injection molding. The fillers were blended at 20, 30, 40, and 50% of the total composite weight. The weight compositions correspond to volume fractions of 0.08, 0.12, 0.18, and 0.25 of gypsum; 0.07, 0.12, 0.18, and 0.24 of wollastonite; 0.08, 0.13, 0.19, and 0.26 of talc; 0.08, 0.13, 0.19, and 0.26 of glass fiber.

Injection molding

Pellets from the compaction stage were injection molded into tensile and flexural specimens using a Battenfeld injection molder (PGS, South Australia). The injection pressure was adjusted to 137 bar for all samples. The front, centre, and rear temperatures across the barrel were set at 240°C, 235°C, and 180°C, respectively.

Silane treatment of mineral fillers

Two commercial silanes, 3-methacryloxypropyltrimethoxy (γ -MPS) and 3-aminopropyltriethoxy silane (γ -APS) were applied to the fillers at 0.5, 1, and 2% with respect to the total weight of the filler, from water solutions. These are equivalent to 0.01, 0.03, and 0.05 of the volume of the filler. The fillers were added to the aqueous solutions and the slurry was continuously stirred for 30 min. The silane-treated fillers were then removed from water, dried, and held at 80°C for 24 h.

Mechanical testing

Specimens were subjected to tensile and flexural tests. Both tests were performed using an INSTRON 5543 universal tester with a maximum load capacity of 1 kN. Five samples were tested for each composite formulation and their average results were reported. For the tensile test, dog-bone specimens similar to Type II specimens from ASTM D 638 were

tested with a constant strain rate of 0.1 mm/min. Axial displacement was recorded using a clip-on extensometer with a 50-mm gauge length.

In the flexural tests, small strips of 127 mm \times 12.7 mm \times 3 mm size were subjected to a three-point bending test and rested on a 50.8 mm support span, according to ASTM D793. A cross-head rate of 1.23 mm/min was applied to each strip. The test was terminated when the strain in the specimens reached 5%.

Scanning electron microscopy

A Philips XL-20 scanning electron microscope was used to observe the inner surface of the tensile-tested samples. Small portions adjacent to the fracture area of the samples were cut and coated with carbon before the microscopy observation.

RESULTS AND DISCUSSION

This study investigated how various mineral fillers affect the mechanical properties of recycled-plastic composites. It was also evaluated how those properties change when silane coupling agents were added to the fillers. The compatibility of the polymers in the matrix phase was not investigated in this study. In some research, the use of compatibilizers was considered for multiple polymer blends.^{21,22} This study was the first stage of a program of research looking at various variables and the use of compatibilizer will be considered in the subsequent stages. There appeared to be homogenous mixing of the polymer components so the compatibility question was not addressed in this stage.

“Effect of mineral fillers on mechanical properties” section describes the response of mechanical properties to additions of mineral fillers and compares the results with those observed when glass fiber is used as filler. In addition, some empirical models frequently used in polymer-filler interaction are used to quantify the amount of interfacial adhesion provided by mineral fillers. “Effect of silane treatments on behavior of mineral fillers” section describes how mechanical properties respond when silane coupling agents are included with mineral fillers.

Effect of mineral fillers on mechanical properties

This part of the study discussed the tensile and flexural testing of different filler-plastic composites.

Tensile and flexural properties

Plots are presented in Figure 1 for the effects of filler type and volume fraction of filler on the tensile properties of mixed recycled-plastic blends. The

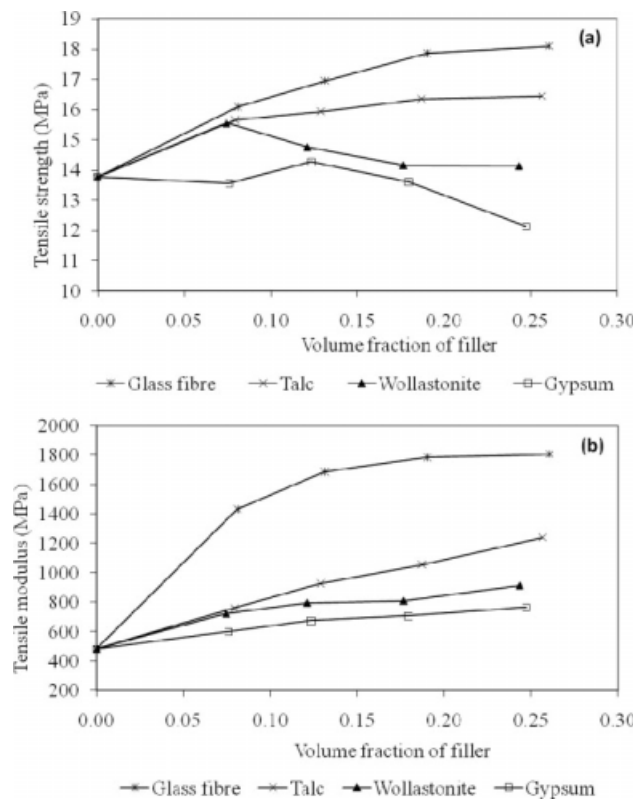


Figure 1 Tensile strength (a) and tensile modulus (b) of filler-recycled plastic composites.

tensile strength was determined at the break point of the specimen, whereas the modulus was calculated as the slope of the stress–strain curve at 1% strain (i.e., secant modulus).

As expected, Figure 1 clearly shows the addition of glass fiber results in the largest improvement in the tensile properties, especially in tensile modulus. Increases of 300% in tensile modulus and 30% in the strength of unfilled samples were achieved with 0.26 v/v glass fiber content. Within the mineral filler group, talc was found to be the most effective where the composites having 0.26 v/v talc showing increases of almost 170% in the modulus and 20% in strength. It has been previously reported that the tensile strength of filled polymers may increase or decrease with increased filler content.^{10,11,13,15,17,23}

Figure 1 shows that the tensile strength provided by talc increases with increasing filler content, whereas that provided by wollastonite and gypsum decreases once the filler content reaches a certain amount. In the gypsum case, the reduction in strength may be caused by agglomeration of gypsum particles, which increases at high filler loadings. This will be further discussed in the morphology section below. As for wollastonite, the large wollastonite particles possibly disrupt matrix continuity, thereby decreasing the effective load-bearing cross-sectional area. Figure 1(b) shows that the tensile

modulus increases with increasing content of fillers, which is a common finding.^{10,11,13,23}

Figure 2 shows the flexural strength and moduli of the plastic composites. The flexural strength was determined at the maximum stress of the stress–strain curve, whereas the modulus was calculated as the slope of the stress–strain curve at 1% strain (i.e., secant modulus).

The addition of glass fiber at 0.26 v/v resulted in increases of nearly 200% in the flexural modulus and 30% in the strength of the base plastic. For the mineral fillers, talc still dominates both the flexural strength and modulus, although the performance differences between the mineral fillers are not as significant as those observed for tensile properties. At 0.26 v/v content, talc increases the flexural modulus by as much as 65% over that of the unfilled polymer and increases the strength by as much as 40%. Overall, the flexural strength of the composites increases with increasing filler content.

Generally, the flexural moduli of the composites are lower than the tensile moduli. This has also been reported by other researchers and might be because in flexural testing the maximum stress occurs at the surface, where filler concentration is relatively low when compared with that in the core section.^{7–9} In injection-

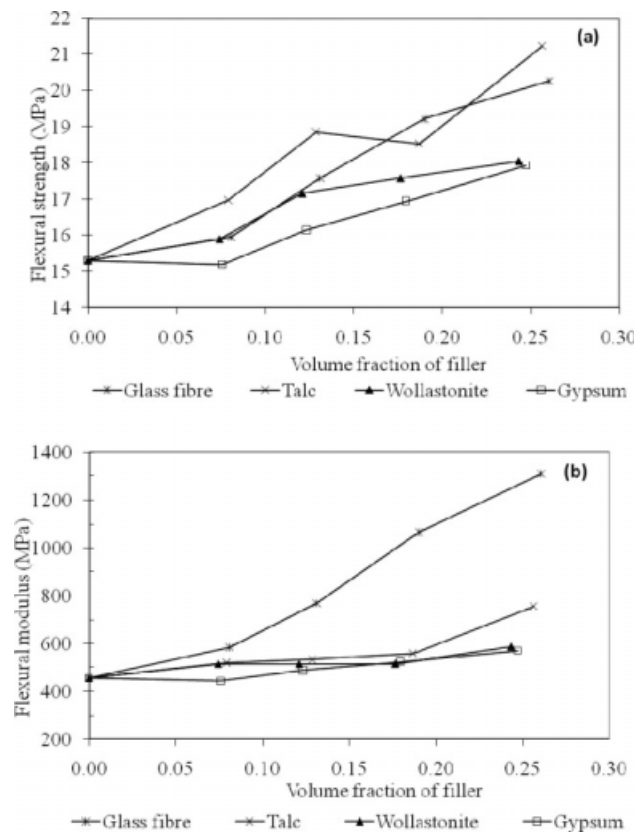


Figure 2 Flexural strength (a) and flexural modulus (b) of filler-recycled plastic composites.

TABLE II
Values of the Constant B in the Turcsanyi Model
[eq. (2)] for Mineral Fillers

Filler	B (Tensile strength)	ΔR^a (%)	B (Flexural strength)	ΔR^a (%)
Gypsum	3.01	5.96	3.58	1.19
Wollastonite	3.38	6.78	3.66	1.89
Talc	4.01	5.50	4.37	3.18

^a Mean value of relative differences between experimental and computed (using the Turcsanyi model) tensile/flexural strengths of composites.

molded composites, filler particles tend to accumulate in the core rather than on the surface, because as the blend passes the molding gate, particles move from the gate (i.e., the front surface) to the interior.³ Furthermore, during cooling of molded specimens, filler particles tend to migrate from cold regions (outer skin) to the melt or core (inner section).⁹

Gypsum was not found to improve tensile and flexural properties as much as talc and wollastonite. This might be because the particle size of gypsum is smaller than those of the other two minerals, and when no surface coating is present, small particles tend to agglomerate.⁹ This will be discussed in the morphology section below.

Particle shape might also influence filler performance. The plate-like structure of talc and the fiber-like (acicular) structure of wollastonite exhibit high aspect ratios, which improve wetting with the polymer matrix and hence reduce the number of micro voids between the filler and matrix.⁹ In addition, talc is known to have high degree of lubricity or dispersion due to much lower Van der Waals forces holding together its crystal structure.⁹

Interfacial adhesion: predictive models of tensile and flexural strength

Various models have been proposed to relate tensile strength to filler content in composites.^{8,17,19} These models often contain a parameter that characterizes the extent of interfacial adhesion between filler particles and the matrix. An example is the Nicolais-Narkis model,^{9,24,17}

$$\sigma_c = \sigma_p \left(1 - 1.21\phi_f^{2/3}\right) \quad (1)$$

Here, σ_c is the tensile strength of the composite, σ_p is the tensile strength of the polymer, and ϕ_f is the volume fraction of filler. This model assumes no adhesion at the interphase between the filler and the matrix; hence, load is sustained only by the matrix. As a result eq. (1) predicts that the tensile strength of polymer composites decreases as more filler is added. The increase in tensile strength observed in this study (Fig. 2) is opposite to that predicted by

the Nicolais-Narkis equation. This suggests that good adhesion was established at the interphase.

Another frequently used model, proposed by Turcsanyi et al.,⁸ takes into account both active and inactive effects of fillers on tensile strength. This model is given by

$$\sigma_c = \sigma_p \frac{(1 - \phi_f)}{1 + A\phi_f} \exp(B\phi_f) \quad (2)$$

Here, the constant B is an adhesion parameter for the filler where large values of B indicate better adhesion or activity of the filler. The term $(-\phi_f)/(1 + A\phi_f)$ measures the extent to which filler reduces the effective load-bearing cross-section of the matrix. The quantity A is a packing factor of the filler; for particulate fillers, $A \approx 2.5$.⁸

From the test data, values of B were determined for each mineral filler as shown in Table II. It can be seen that values of B derived for tensile strength are similar to those obtained for flexural strength. The B value for tensile strength of talc is similar to that estimated by Turcsanyi et al.⁸ for talc-HDPE composites (i.e., 3.926). These B values demonstrate that talc exhibits the strongest interaction or adhesion

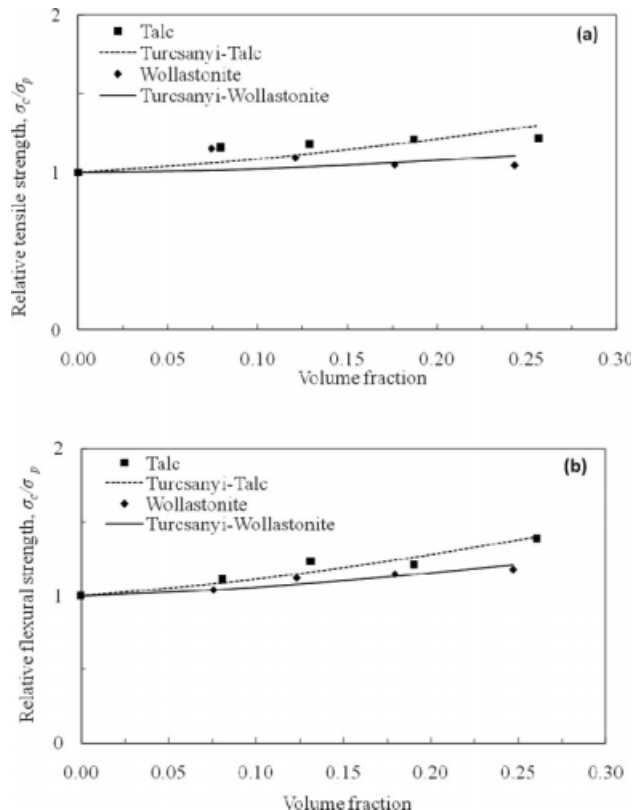


Figure 3 Composites with talc and wollastonite fillers: tensile strength (a) and flexural strength (b) as functions of filler volume fraction. (Lines are drawn using eq. (2) with B values of 4.01 and 3.38 for talc and wollastonite, respectively).

with the polymers in the matrix; gypsum exhibits the weakest interactions, and wollastonite falls in between. These findings are consistent with the behavior described earlier for the mechanical properties.

Figure 3(a,b) compare experimental and predicted values for the tensile and flexural strengths of talc and wollastonite. To give a clear presentation, the gypsum data and their predicted values are excluded from the figure. It can be observed from Figure 3(a) that the measured tensile strengths deviate slightly from the correlations given by eq. (2), especially at low filler concentrations. This might have been caused by the fillers establishing different interactions with the various polymers at the interphase of the plastic waste. However, the measured flexural strengths are in good agreement with the correlation [Fig. 3(b)] and this is reflected by the smaller errors shown in Table II.

Interfacial adhesion: predictive models of tensile and flexural modulus

Attempts were also made to fit the tensile and flexural modulus data to theoretical models. One simple model is the Einstein equation,⁹ which predicts the

tensile behavior of particulate-filled polymers and has the form:

$$E_c = E_p(1 + a\phi_f) \tag{3}$$

Here, E_c and E_p are the tensile moduli of the composite and matrix, respectively. The constant a is equal to 2.5 for good adhesion between the filler and polymer. A modification to the Einstein model gives the Guth-Smallwood model, which has been frequently used by others,^{19,25,26}

$$E_c = E_p \left(1 + 2.5\phi_f + 14.1\phi_f^2 \right) \tag{4}$$

For rigid spherical particles, the Kerner equation is commonly used,

$$E_c = E_p \left[1 + \frac{15\phi_f(1 - \tau_p)}{(1 - \phi_f)(8 - 10\tau_p)} \right] \tag{5}$$

where τ_p is the Poisson's ratio of the matrix. For high-density polyethylene, τ_p has been determined to be 0.35.²⁴

Figures 4 and 5 show the experimental tensile and flexural moduli of mineral-filler recycled-plastic

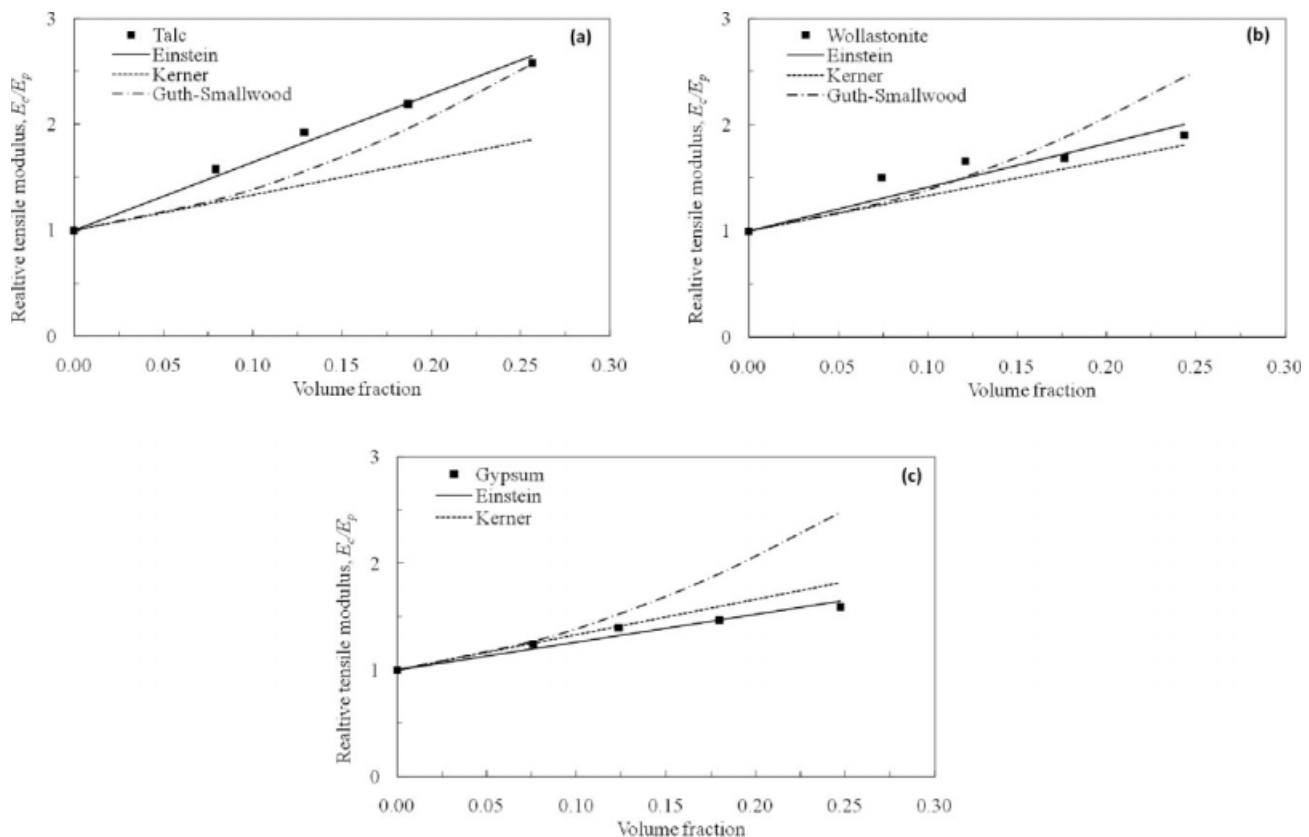


Figure 4 Relative tensile moduli of composites with (a) talc, (b) wollastonite, (c) gypsum fillers as functions of filler volume fraction and compared with the Einstein [eq. (3)], Kerner [eq. (4)], and Guth-Smallwood [eq. (5)] models.

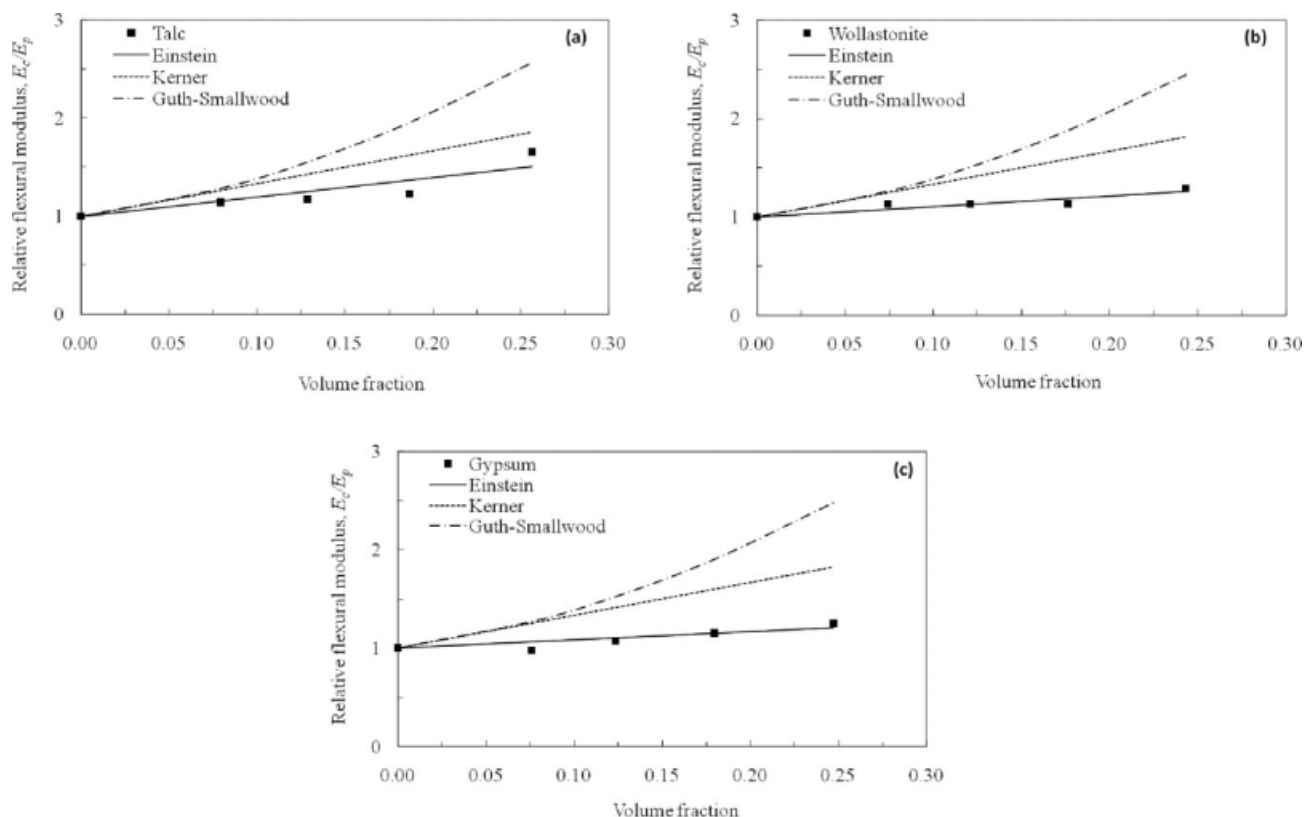


Figure 5 Relative flexural moduli of composites with (a) talc, (b) wollastonite, (c) gypsum fillers as functions of filler volume fraction and compared with the Einstein [eq. (3)], similar to Figure 4, Kerner [eq. (4)], and Guth-Smallwood [eq. (5)] models.

composites to those predicted by the Einstein [eq. (3)], Guth and Smallwood [eq. (5)], and Kerner [eq. (4)] models. The experimental data deviate greatly from the Guth-Smallwood and Kerner equations, whereas the Einstein equation gives the best fit for all fillers. Although the Einstein equation is considered to apply to composites having low concentrations of particulate fillers, the model gave good correlations over the entire range of volume fractions considered in this study.

Table III shows values for the adhesion constant a in the Einstein model applied to composites with mineral fillers. Again, talc exhibits the strongest interfacial adhesion compared to the other mineral fillers studied here. In addition, the values of a are higher for the tensile modulus than for the flexural modulus. In flexure testing, stress is sustained by the underside surface of the molded specimens, and surfaces normally contain less filler than the interior. This may cause the flexural modulus to be lower than the tensile modulus, and hence, values for adhesion constants are lower.

Morphology

Figure 6 contains SEM images of the base plastic obtained from secondary electron (SE) diffraction (a) and backscatter electron (BSE) diffraction (b). In Fig-

ure 6(b), the white specks appearing in the body of the matrix possibly indicate the presence of nonpolyethylene polymers in the mixed plastic (e.g., expandable polystyrene).

The inferior tensile and flexural properties of the gypsum-filled samples could be a result of severe agglomeration. This is shown in Figure 7 for the blends with 0.12 and 0.25 v/v gypsum. Gypsum agglomerates formed at all weight fractions and their extent increased with increasing filler additions, as observed in Figure 7(a,b). These large agglomerates are believed to disrupt interphase bonding and, therefore, create large voids upon loading; subsequent debonding further weakens the structure and may initiate crack propagation.²⁷

TABLE III
Values of the Constant a in the Einstein [eq. (3)] Model for Mineral Fillers

Filler	a (Tensile modulus)	ΔR^a (%)	a (Flexural modulus)	ΔR^a (%)
Gypsum	2.61	1.03	0.84	2.79
Wollastonite	4.12	3.68	1.07	3.93
Talc	6.43	0.96	1.97	5.34

^a Mean value of relative differences between experimental and computed (using the Einstein [eq. (3)] model) tensile/flexural strengths of composites.

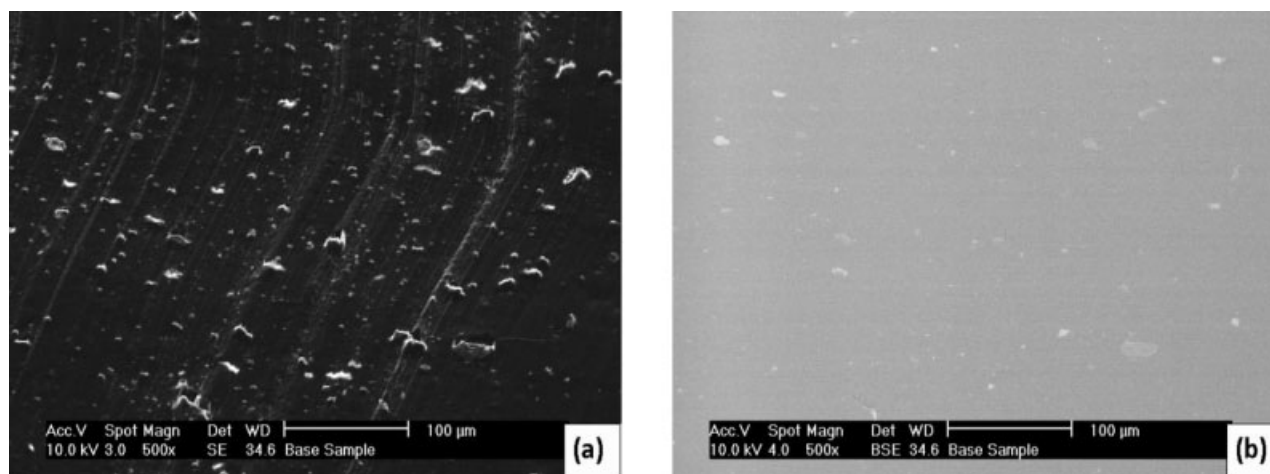


Figure 6 SEM images of the base plastic: (a) SE and (b) BSE diffraction.

The SEM micrographs of the composites filled with 0.12 and 0.24 v/v wollastonite are shown in Figure 8. Microvoids are observed on the vicinity of wollastonite particles. The voids are larger in size and more common in the blend with 0.24 v/v wollastonite. In comparison to gypsum and wollastonite, talc tends to disperse better in the matrix and to create less voids in the matrix as depicted in Figure 9. This might be the reason for talc to have the highest strength and modulus.

Extensive agglomeration in gypsum could result because the gypsum particles have an affinity to agglomerate more than talc and wollastonite. The micrographs of filler particles show that primary particles of gypsum naturally clump together [Fig. 10(a)], whereas particles of talc [Fig. 10(b)] and wollastonite [Fig. 10(c)] barely come into contact with each other. Agglomeration leads to nonuniform filler dispersion and inhomogeneous mechanical proper-

ties; moreover, it inhibits the activity of filler particles on the polymer matrix.¹⁸

Agglomeration of filler particles is complex and is influenced by many variables, including particle size, chemical groups on the particle surface, filler production method, moisture level, and surface treatment.³ Some fillers possess a natural tendency to agglomerate, especially when strong van der Waals bonds form among particles.³ In addition, small particles have a stronger tendency to agglomerate^{1,3,19}; as a result, the smaller particle size of gypsum might have intensified the formation of agglomerates.

In the case of glass fiber, Figure 11 shows the orientation of glass fiber in a cross-section of the matrix. At 0.13 v/v, the fibers seem to be aligned in parallel along the longitudinal axis. At 0.26 v/v glass fiber [Fig. 11(b)], the spacing between the fibers becomes smaller; in fact, the fibers come into contact and become less oriented.

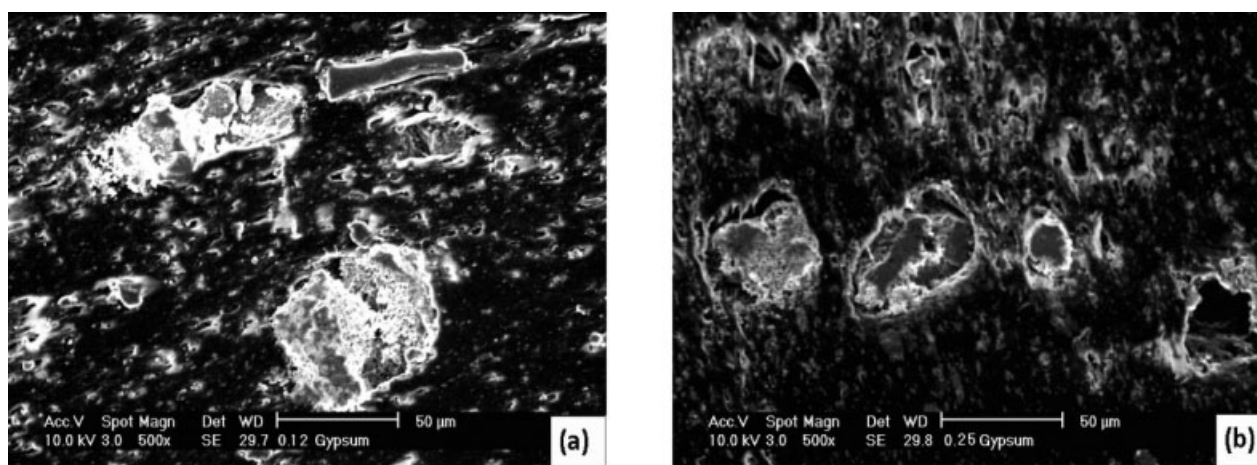


Figure 7 SEM images of 0.12 (a) and 0.25 (b) v/v gypsum blends.

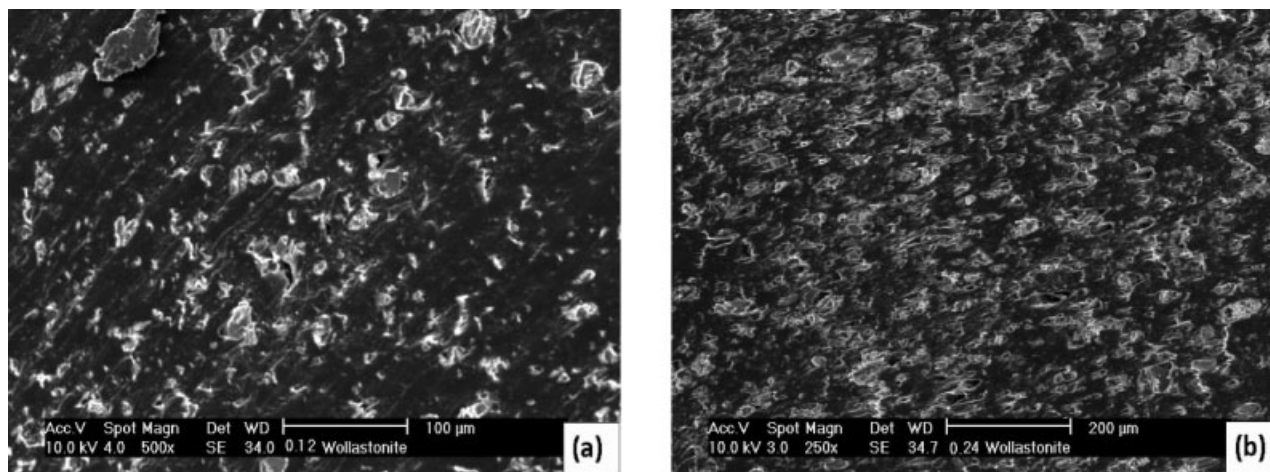


Figure 8 SEM images of 0.12 (a) and 0.24 (b) v/v wollastonite blend.

Effect of silane treatments on behavior of mineral fillers

The results in Morphology section show that using talc or wollastonite filler led to considerable improvements in tensile and flexural properties. Therefore, the study of coupling agents then focused on talc and wollastonite-recycled-plastic composites. Small quantities of the silanes (0.01, 0.05, and 0.03 of filler volume) were added to 0.13 v/v talc and 0.12 v/v wollastonite composites.

Mechanical properties

The effect of silane coupling agents on tensile and flexure properties of the blends of 0.13 v/v talc is given in Table IV. Different types and contents of silane coupling agents slightly improved (less than 10%) the tensile properties of the untreated blend. Furthermore, both silane coupling agents appear to have no effect on the flexural properties of the talc

blends (Table IV). Therefore, the optimum additions considering the largest increase in tensile properties for γ -MPS and γ -APS were found at 0.01 and 0.03 v/v, respectively.

Tensile and flexural properties of silane-treated wollastonite samples are given in Table V. The tensile behavior of the wollastonite blends improves with the addition of both silanes. The additions of γ -MPS and γ -APS at 0.03 v/v show the optimum tensile properties. In general, γ -APS gives slightly better tensile strength and modulus than γ -MPS, although the differences are only around 5%. As found in the case of silane-treated talc, the flexural properties of the wollastonite blends were not affected by the silane coupling agents.

Morphology

To improve the mechanical properties of a polymer, a type of filler needs to be “active” or be able to

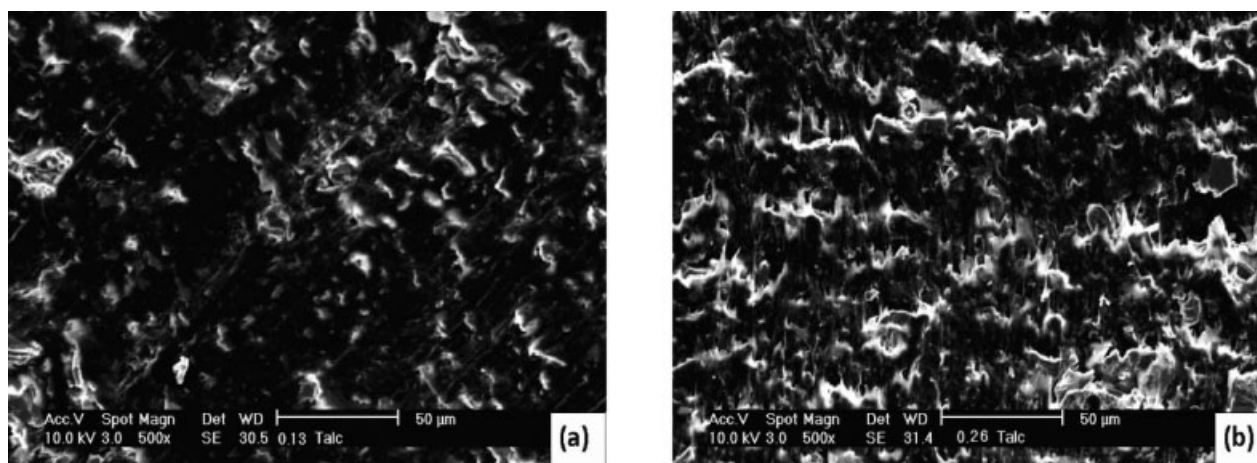


Figure 9 SEM images of 0.13 (a) and 0.26 (b) v/v talc blends.

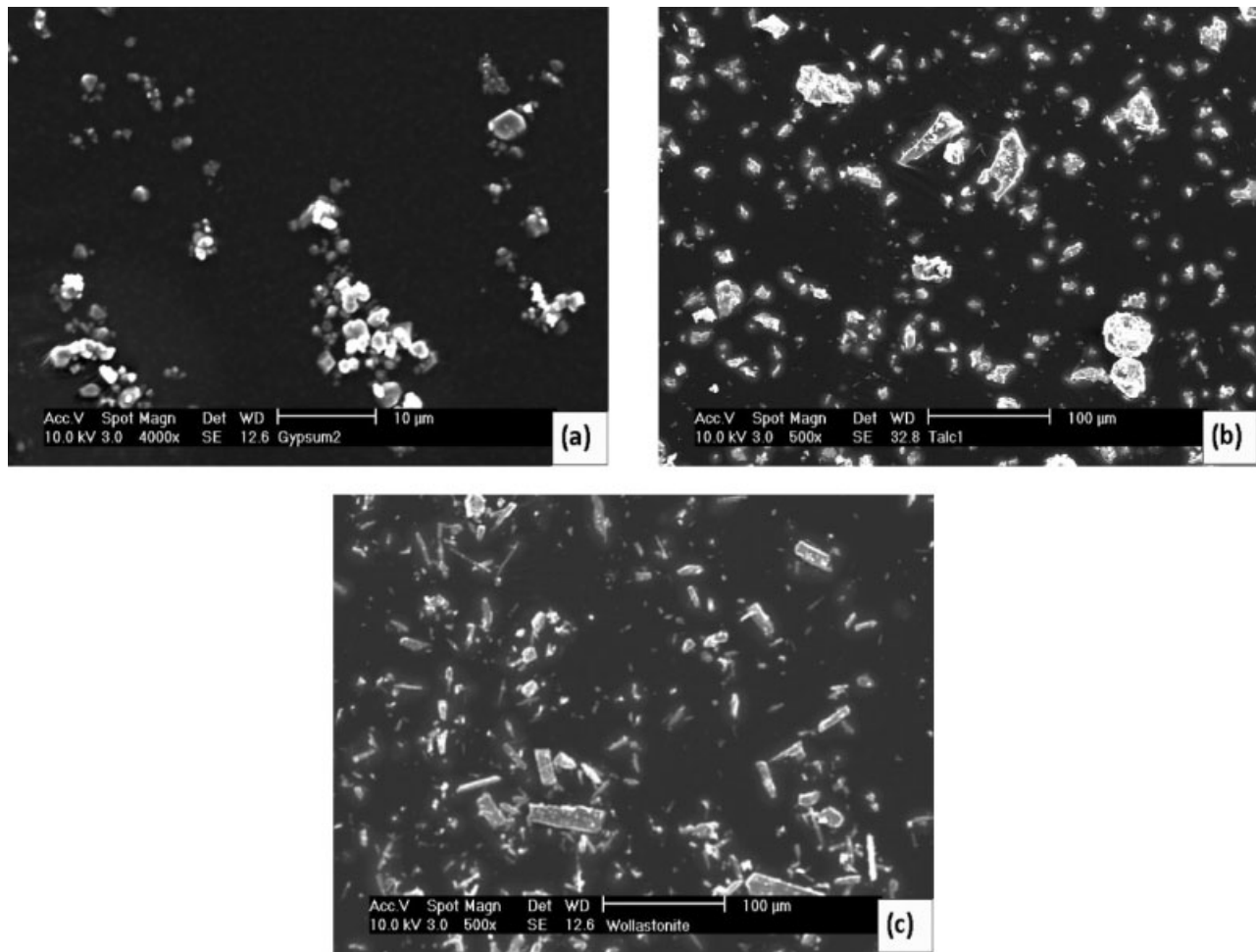


Figure 10 SEM micrographs of (a) anhydride gypsum, (b) talc, and (c) wollastonite.

engage autonomously with the polymer host.²⁵ Where the filler is not readily active in a matrix of polymer or weak interphase adhesion is present,

chemical bonding needs to be established between those two phases.²⁵ This can be achieved through the addition of coupling agents.²⁸

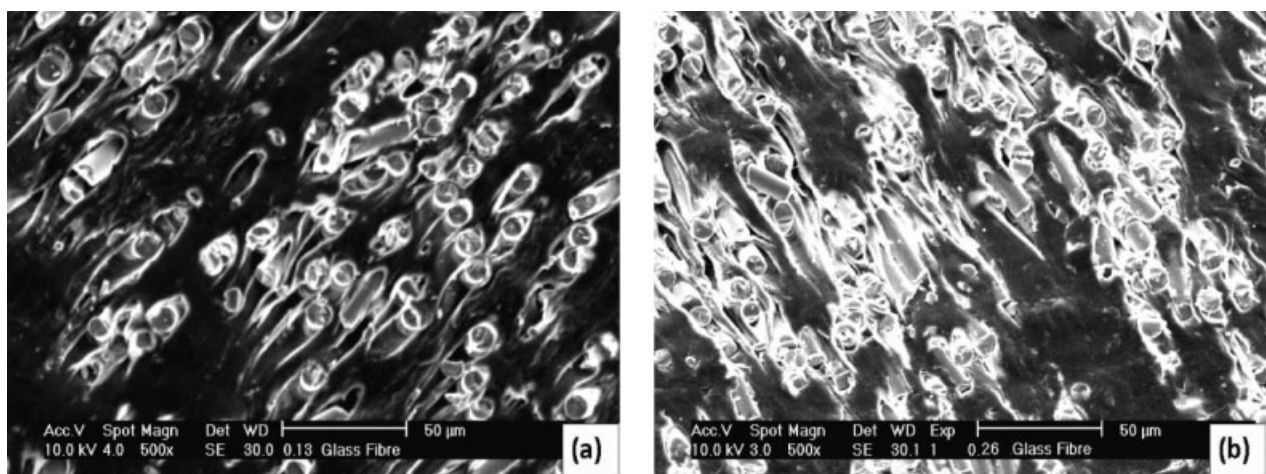


Figure 11 SEM images of 0.13 (a) and 0.26 (b) v/v glass fiber blends.

TABLE IV
Effect of Silane Coupling Agent on Tensile and Flexure Properties of 0.13 v/v Talc Blends

Silane type and content	Tensile strength (MPa) ^a	Tensile modulus (MPa)	Flexure strength (MPa)	Flexure modulus (MPa)
No silane	15.9	925.9	18.9	533.4
.01-M	17.4	964.4	19.9	562.9
.03-M	16.4	959.3	18.7	552.3
.05-M	16.8	1009.3	19.3	561.7
.01-A	16.8	890.1	18.3	541.4
.03-A	17.8	1030.4	18.2	542.0
.05-A	17.9	1049.1	17.6	530.3

^a The figures 0.01, 0.03, and 0.05 indicate the additions of 0.01, 0.03, and 0.05 v/v of silane, respectively; whereas the alphabets M and A denote γ -MPS and γ -APS, respectively.

TABLE V
Effect of Silane Coupling Agent on Tensile and Flexure Properties of 0.12 v/v Wollastonite Blends

Silane type and content	Tensile strength (MPa) ^a	Tensile modulus (MPa)	Flexure strength (MPa)	Flexure modulus (MPa)
No silane	14.8	794.9	17.2	514.6
.01-M	16.4	884.0	14.5	487.6
.03-M	16.7	935.5	15.5	520.5
.05-M	15.8	837.1	17.5	550.6
.01-A	17.0	917.4	17.0	524.4
.03-A	17.4	972.4	16.3	496.0
.05-A	17.5	949.8	17.2	518.9

^a The figures 0.01, 0.03, and 0.05 indicate the additions of 0.01, 0.03, and 0.05 v/v of silane, respectively; whereas the alphabets M and A denote γ -MPS and γ -APS, respectively.

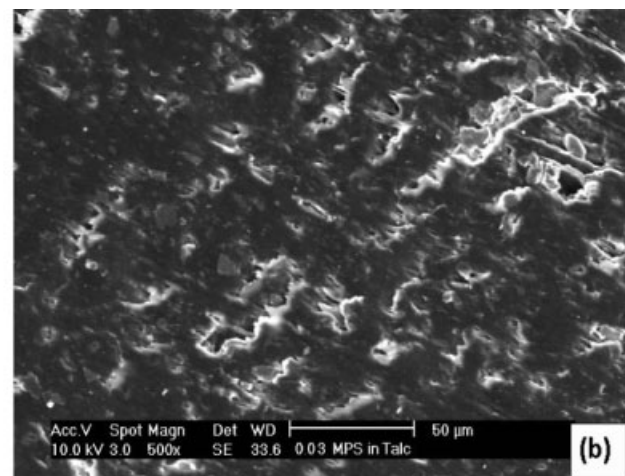
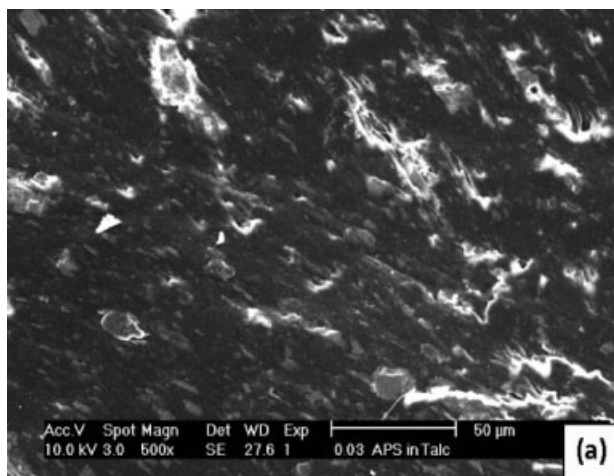


Figure 12 SEM images of talc composites blended with: 0.03 v/v γ -APS (a) and 0.03 v/v γ -MPS (b).

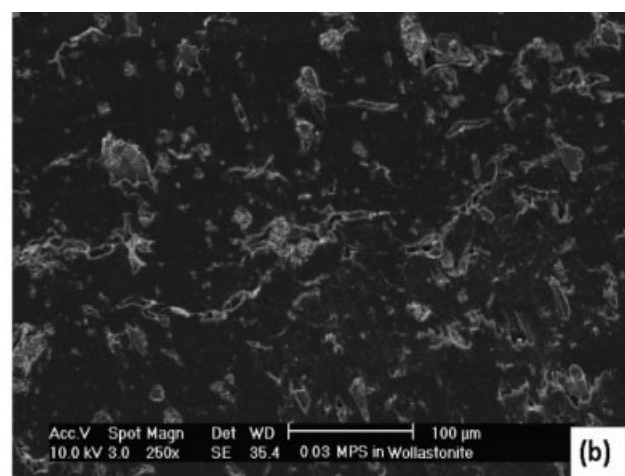
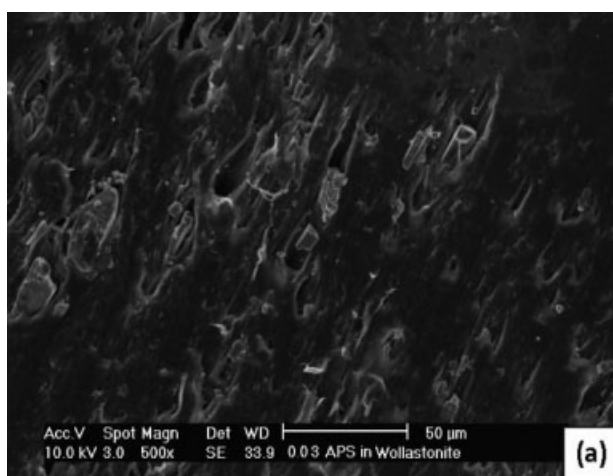


Figure 13 SEM images of wollastonite composites blended with: 0.03 v/v γ -APS (a) and 0.03 v/v γ -MPS (b).

Some degree of improvement in the tensile properties found in the γ -APS-treated talc composites might be explained by SEM observations on the sections near the fracture surface of the tensile-tested specimens (Fig. 12). An observation of the 0.03 v/v γ -APS article [Fig. 12(a)] under SEM shows that the talc particles seem to be covered by the matrix, indicating that interphase bonding between the two phases is sufficient. This might result in the large improvement of the 0.03 v/v γ -APS talc blend.

The blends prepared with γ -MPS did not result in as high an increase in tensile behavior as those treated with γ -APS. This might be explained by a SEM analysis of the talc blend prepared with 0.03 v/v γ -MPS as shown in Figure 12(b). It is apparent that the voids in the vicinity of the talc particles are generally larger in scale and more widespread in the matrix than that found in the γ -APS blends.

In the case of the blends containing silane-treated wollastonite, which can be seen from Figure 13, the wollastonite particles seem to bond better with the matrix, when compared with the unmodified wollastonite mix. Less microvoids surrounding the filler particles were observed in the matrix. As found in the modified talc blends, the γ -APS blends [Fig. 13(a)] have lesser microvoids, which interrupt the adhesion between the matrix and filler, in comparison to the blends containing γ -MPS [Fig. 13(b)]. This may have caused the γ -APS blends to have slightly higher tensile properties than the γ -MPS blends.

CONCLUSIONS

The results obtained for the fillers show that talc gives the best improvement to the mechanical properties of recycled plastic when compared with wollastonite and gypsum. It was believed that filler dispersion and adhesion were the main factors contributing to the observed trends. Agglomeration of filler found in the gypsum filled blends interrupted the adhesion of the matrix and hence reduced the tensile strength. Furthermore, the interphase adhesion for talc particles was better than wollastonite, as indicated by the lower formation of voids.

Established mathematical models including those of Turcsanyi and Einstein can satisfactorily predict the mechanical strength and stiffness of the commingled postconsumer plastic composites, respectively.

Silane coupling agents have little effect in improving the mechanical properties of commingled-recycled plastic composites. The addition of γ -APS coupling agent gives greater improvement in the mechanical properties than that from γ -MPS. This was confirmed by SEM observation showing fillers

treated with γ -APS are better bonded and do not develop as many voids compared to γ -MPS.

The authors acknowledge Advanced Plastic Recycling of South Australia (APR) of South Australia for the material made to this project.

References

1. Plastics and Chemicals Industries Association (PACIA) 2007. National Plastics Recycling Survey (2006 Calendar year), Hyder Consulting Pty Ltd, Melbourne, http://www.pacia.org.au/_uploaditems/docs/15.plasticsrecycling2006 (accessed December 2007).
2. Camacho, W.; Karlsson, S. *Polym Eng Sci* 2001, 41, 1626.
3. Pawlak, A.; Morawiec, J.; Pazzagli, F.; Pracella, M.; Galeski, A. *J Appl Polym Sci* 2002, 86, 1473.
4. Breslin, V. T.; Senturk, U.; Berndt, C. C. *Resour Conserv Recycl* 1998, 23, 243.
5. Perugini, F.; Mastellone, M. L.; Arena, U. *Environ Prog* 2005, 24, 137.
6. Fortelny, I.; Michalkova, D.; Krulis, Z. *Polym Degrad Stab* 2004, 85, 975.
7. Leong, Y. W.; Abu Bakar, M. B.; Mohd Ishak, Z. A.; Pukanszky, B. *J Appl Polym Sci* 2004, 91, 3315.
8. Turcsanyi, B.; Pukanszky, B.; Tűds, F. *J Mater Sci Lett* 1988, 7, 160.
9. Wypych, G. *Handbook of Fillers*; Chemtec Publishing: Toronto, 1999.
10. Ismail, H.; Rozman, H. D.; Jafri, R. M.; Ishak, A. M. *Eur Polym J* 1997, 33, 627.
11. Khalil, H. P.; Sharifah, S. A.; Shahnaz, S. B.; Ratnam, M. M.; Ahmad, F.; Fuaad, N. A. *J Reinforced Plast Comps* 2006, 25, 1291.
12. Tselios, C. H.; Bikiaris, D.; Savidis, P.; Panyiotou, C.; Larena, A. *J Mater Sci* 1999, 34, 385.
13. Bose, S.; Mahanwar, P. A. *J Mater Sci* 2005, 40, 6423.
14. Teixeira, S. C.; Moreira, M. M.; Lima, A. P.; Santos, L. S.; da Rocha, B. M.; de Lima, E. S.; da Costa, R. A. A. F.; da Silva, A. L. N.; Rocha, M. C. G.; Coutinho, F. M. B. *J Appl Polym Sci* 2006, 101, 2559.
15. Hiljanen-Vainio, M.; Heino, M.; Seppala, J. V. *Polymer* 1998, 39, 865.
16. Jesionowski, T.; Bula, K.; Janiszewski, I. J.; Jurga, J. *Compos Interfaces* 2003, 10, 225.
17. Nicolais, L.; Narkis, M. *Polym Eng Sci* 1971, 11, 94.
18. Akin-oktem, G.; Tinçer, T. *J Appl Polym Sci* 1994, 54, 1103.
19. Bikiaris, D.; Matzinos, P.; Prinos, J.; Flaris, V.; Larena, A.; Panayiotou, C. *J Appl Polym Sci* 2000, 80, 2877.
20. Demir, T.; Tincer, T. *J Appl Polym Sci* 2000, 79, 827.
21. Bertin, S.; Robin, J.-J. *Eur Polym J* 2002, 38, 2255.
22. Adewole, A.; Dackson, K.; Wolkowicz, M. *Adv Polym Tech* 1994, 13, 219.
23. Ulutan, S.; Gilbert, M. *J Mater Sci* 2000, 35, 2115.
24. Bliznakov, E. D.; White, C. C.; Shaw, M. T. *J Appl Polym Sci* 2000, 77, 3220.
25. Lipatov, Y. S. *Polymer Reinforcement*; Chemtec Publishing: Toronto, 1995.
26. Wong, T. L.; Barry, C. M. F.; Orroth, S. A. *J Vinyl Add Technol* 1999, 5, 235.
27. Zuideveld, W. C. J.; Westzaan, C.; Huetnik, J.; Gaymans, R. *J Polym* 2003, 44, 261.
28. Plueddemann, E. P. *Silane Coupling Agents*; Plenum Press: New York, 1991.