

An Efficient Way to Improve the Mechanical Properties of Polypropylene/Short Glass Fiber Composites

Hong-Quan Xie,¹ Shu Zhang,¹ Dong Xie²

¹Department of Chemistry, Huazhong University of Science and Technology, Wuhan 430074, China

²Department of Biomedical Engineering, Indiana University–Purdue University–Indianapolis, Indianapolis, Indiana 46202

Received 10 May 2004; accepted 16 November 2004

DOI 10.1002/app.21575

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

ABSTRACT: Enhancement of tensile strength, impact strength, and flexural strength of polypropylene/short glass fiber composites by treating the glass fibers with coupling agent, mixing with maleated polypropylene (MPP) for compatibilization and adhesion, and with nucleating agent for improvement of polypropylene crystallization was studied. The results showed that both the silane coupling agent and MPP enhance tensile strength, impact strength, and flexural strength. In the absence of MPP, the effect of silane coupling agent on the mechanical properties of the composites decreases in the following order: alkyl trimethoxy silane (WD-10) > γ -methacryloxypropyl trimethoxysilane (WD-70) > *N*-(β -aminoethyl)- γ -aminopropyl trimethoxysilane (WD-52), whereas in the presence of MPP, the order changes as follows: WD-70 > WD-10 > WD-52. When the glass fibers were treated with WD-52, 4,4-diamino-diphenylmethane bismaleimide (BMI) can further enhance the mechanical properties of the composite. The three kinds of strengths increase with

MPP amount to maximum values at 5% MPP. As a nucleating agent, adipic acid is better than disodium phthalate in improving the mechanical properties, except for the notched impact strength. Wide-angle X-ray diffraction showed that the adipic acid is an α -type nucleating agent, whereas disodium phthalate is a β -type nucleating agent. Blending with styrene–butadiene rubber can somewhat improve the notched impact strength of the composites, but severely lowers the tensile strength and bending strength. Scanning electron micrographs of the broken surface of the composite showed greater interfacial adhesion between the glass fibers and polypropylene in the modified composite than that without modification. © 2005 Wiley Periodicals, Inc. *J Appl Polym Sci* 96: 1414–1420, 2005

Key words: composites; polypropylene (PP); compatibilization, mechanical properties

INTRODUCTION

Polypropylene (PP) is widely used in many applications because of its low cost and versatile properties. However, PP obtained by bulk polymerization in small batch process does not have satisfactory properties, especially its poorer impact strength at low temperature and lower heat distortion temperature than that of commercial products obtained by other processes. Although polypropylene can be reinforced by glass fibers, properties of the composite are not uniformly satisfactory. Improvement of the interfacial adhesion between the glass fibers and PP is necessary for enhancement of mechanical properties.

Denault and Vu-Khauh¹ indicated that a kind of silane coupling agent induced nucleation at the glass filler–polyolefin matrix interface, attributed to the adsorbed layer of silane on the filler, and that at high concentration of glass filler, an improvement in the filler–matrix adhesion had a beneficial effect on the

fracture performance. Yue and Quek² showed that the maximum interfacial shear strength is obtained at a critical thickness of the silane layer on the treated glass fibers in PP composites. Severini et al.³ reported that PP modified with maleic anhydride served as a coupling substance in glass-reinforced polyolefin and showed that this product improved the adhesion between polyolefin matrix and glass fillers. Rijdsdijk et al.⁴ indicated that when maleated PP (MPP) was added to PP/long glass fiber composites, it can improve the adhesion between the PP matrix and the long glass fibers. Hausmann and Flaris⁵ showed that MPP can be used as a coupling material to enhance the impact strength of the glass fiber/PP composites. Nygård et al.⁶ used MPP to enhance the interfacial adhesion between amino-silane-treated glass fibers and PP matrix. Roux et al.⁷ indicated that highly grafted MPP promotes good adhesion in glass fiber-reinforced PP composites when used with γ -aminopropyl-triethoxysilane-treated glass fibers. Iroh and Berry⁸ achieved a significant improvement in crystallization of PP using sodium benzoate as nucleating agent for PP/glass fiber composites and indicated that a decreased interfacial shear strength and an increased inherent fiber

Correspondence to: H.-Q. Xie. (xiehongquan88@126.com)

orientation were obtained when the PP/glass fiber composite was premixed with sodium benzoate as a nucleating agent.⁹ Tjong et al.^{10–12} indicated that the incorporation of SEBS (styrene-*b*-ethylene-butylene-*b*-styrene) triblock copolymer or maleated SEBS (MSEBS) elastomers with MPP improved the impact strength of the short glass fiber (SGF)/SEBS/MPP and SGF/MSEBS/MPP hybrids. SEM observations revealed that the SGF surfaces of both SGF/SEBS/MPP and SGF/MSEBS/MPP hybrids are coated with a thin layer of matrix material, which implied that the MAH functional group of MPP improves the adhesion between SGF and PP, and between SGF and SEBS.

This article reports the study of the enhancement of the tensile strength, impact strength, and bending strength of the PP/short glass fiber composites by increasing the interfacial adhesion between the glass fibers and PP, using silane coupling agents for glass fibers and addition of maleated polypropylene (MPP), and also improving the crystalline properties of PP by addition of nucleating agents for PP. The morphology of the composites was studied by SEM, WAXD, and polarizing light microscopy (PLM).

EXPERIMENTAL

Materials

Polypropylene (PP; melt index: 1.9 g/10 min) was produced by Wuhan Petroleum Chemical Factory (China), using batch bulk polymerization in small scale. Alkali-free short glass fibers (6 mm length; 9 μ m diameter) were supplied by Chongqing Glass Fibers Co. (China). Antioxidant 1010 [sorbitol tetra[β -(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionate], antioxidant dilauryl thiodipropionate (DLPT), and 4,4-diaminodiphenylmethane bismaleimide (BMI) were of industrial grade. The silane coupling agents, WD-10 (alkyl trimethoxy silane), WD-52 [*N*-(β -aminoethyl)- γ -aminopropyl trimethoxy silane], and WD-70 (γ -methacryloxypropyl trimethoxy silane) were supplied by the Chemical Factory of Wuhan University. Xylene, benzoyl peroxide (BPO), maleic anhydride (MAH), and acetic anhydride were chemically pure. Ethanol, isopropanol, Na₂CO₃, KOH, and concentrated hydrochloric acid were analytical reagents. Disodium phthalate was prepared by reaction of phthalic acid with equal moles of Na₂CO₃ in an ethanol/water solution.

Maleation of PP

A mixture of 87 g PP, 10.5 g MAH, and 680 mL xylene, after being purged with nitrogen, was heated to 130 \pm 5°C with stirring until PP and MAH were dissolved. A xylene solution of 0.53 g BPO was added dropwise within a period of 90 min. Then a xylene solution of 2.6 g MAH was added, followed by dropping a xylene

solution of 0.26 g BPO within 40 min. The reaction was carried out for an additional 30 min. After cooling, the precipitate was filtered and washed with acetone three times. The product was dried under an infrared lamp and in a vacuum oven, successively. The maleated PP (MPP) was dissolved in hot xylene containing 10% ethanol.

Determination of MAH content in MPP

A sample of MPP (0.1–0.2 g) was weighed accurately into 15 mL ethanol solution of 0.1 mol/L KOH and 150 mL water-saturated xylene. The mixture was refluxed for 5 h. Then the hot mixture was titrated with isopropanol solution of 0.1 mol/L HCl. The combined MAH can be calculated as follows:

$$\text{MAH combined (\%)} = (M_{\text{KOH}} \times \text{mL}_{\text{KOH}} - M_{\text{HCl}} \times \text{mL}_{\text{HCl}}) \times \text{MW}_{\text{MAH}} / (20 \times W_s)$$

where M is the molar concentration, MW_{MAH} is the molecular weight of maleic anhydride, and W_s is the weight of sample.

Surface treatment of glass fibers with coupling agent

The glass fibers were first baked at 250–300°C for 2 h to remove the lubricant oil on the surface. Then the temperature was raised to 400–500°C for 2 h. After cooling, the glass fibers were immersed immediately in a 1/1 ethanol/water solution of 1% silane coupling agent, the pH of which was adjusted to 5, for several min. Then the glass fibers were dried under an infrared lamp and at 80°C in an oven for 30 min, followed by heating in the oven at 150°C for 20 min.

Blending of PP, MPP, and glass fibers

A mixture of PP and MPP in various proportions was blended with 0.3% antioxidant 1010, 0.2% antioxidant DLPT, and 1% calcium stearate on a mill at 160–170°C for 2 min. Then the glass fibers, treated with coupling agent, were added and the mixture was blended together for about 10 min, until the visible disappearance of the glass fibers. The blend was melted in a mold on a hot press, pressed at 180°C for 3–4 min, and then cooled to room temperature.

Testing for the mechanical properties of the composite

Tensile strength and bending strength of the composite were measured on an XL-2500 tensile tester (Changsha Materials Tester Co., China) with an extension rate of 200 mm/min. Notched impact strength of

the composite was measured on a Charpy XCL-500 impact strength tester (Changsha Materials Tester Co.). All the tests were carried out at 15–20°C.

Determination of crystallinity by WAXD

Crystallinity of PP and PP in the composite was measured on a Rigaku 3015 X-ray diffractometer (Rigaku, Tokyo, Japan). The characteristic peaks for the α -type crystal are at $2\theta = 14.1, 17.0,$ and 18.6° , whereas the characteristic peak for the β -type crystal is at $2\theta = 16.1^\circ$. A third possible crystal structure, γ , has peaks at $2\theta = 21.3$ and 21.9° . The crystallinity (X_c) and contents of the different types of crystals were calculated as follows¹³:

$$X_c(\%) = I_c \times 100 / (I_c + 1.25I_a) \quad (1)$$

$$K_\alpha(\%) = I_\alpha / (I_\alpha + I_\beta + I_\gamma) \quad (2)$$

$$K_\beta(\%) = I_\beta / (I_\alpha + I_\beta + I_\gamma) \quad (3)$$

$$K_\gamma(\%) = 100 - K_\alpha - K_\beta \quad (4)$$

where I_a represents the diffraction intensity of the amorphous part and I_c that of the crystalline part; K_α , K_β , and K_γ are the % content of α -, β -, and γ -type crystals, respectively.

Morphology characterization

The surface of a fractured section of the PP composite sample tested for tensile strength was coated with a thin layer of gold with a thickness of about 100 Å. The morphology was observed under a JSM-35C scanning electron microscope (JEOL, Tokyo, Japan). The spherulitic morphology of PP was observed under a XPT-7 type polarizing microscope (Jiangnan Optics & Electronics Co., China), after melting a 0.3 mg sample at 200°C that was pressed for 1 min to a thickness of about 0.2 mm, maintained at 130°C for 3 h, and cooled to room temperature.

RESULTS AND DISCUSSION

Effects of silane coupling agents for glass fibers and MPP on the mechanical properties of the polypropylene/glass composite

PP, made by the bulk polymerization in a small batch process, exhibited a very low notched impact strength of 1.9 kJ/m², even lower than that of the general commercial product. Addition of 30% short glass fibers to the PP enhanced the impact strength to 3.1 kJ/m². Improvement of the tensile strength, impact strength, and flexural strength of the glass fiber-reinforced PP composites, by coupling the glass fiber with WD-10 and addition of

TABLE I
Effects of WD-10 and MPP on Mechanical Properties of Glass Fibers (GF)-Reinforced PP Composites^{a,b}

WD-10/GF (wt %)	MPP/composite (wt %)	s^b (MPa)	e (%)	i (kJ/m ²)	t (MPa)
0	0	33.1	160	3.1	41.8
0	10	48.6	10	5.1	60.0
0.5	5	48.8	12	5.0	63.4
0.75	5	50.0	8	7.5	68.5
1.0	5	56.2	9	9.3	68.6
1.5	5	53.9	6	9.3	68.8
1.0	0	45.3	14	5.2	53.4
1.0	3	53.0	7	8.7	61.6
1.0	5	56.2	8	10.4	70.0
1.0	10	51.7	8	9.3	72.2
1.0	15	49.0	10	5.2	57.4
1.0	20	30.7	25	3.5	41.6

^a Glass fiber (GF)/composite = 30 wt %, MAH content of MPP = 11.3%.

^b s , e , i , and t represent tensile strength, ultimate elongation, notch impact strength, and flexural strength, respectively.

MPP to the composite, is shown in Table I. When WD-10 and MPP are not used, the three kinds of strengths for the PP/glass fiber (30%) composites are relatively low. However, after the glass fibers were treated with WD-10 or MPP was added, not only the tensile strength but also the impact strength and flexural strength increased significantly. The ultimate elongation of the modified composite was <12%, unless a large excess MPP was used, whereas that of the unmodified composite was >150%. When the WD-10-treated glass fibers and MPP were used simultaneously in the composite, the three kinds of strengths increased with increasing amount of WD-10 used in treatment of the glass fibers, up to the optimum amount of 1.0 wt % based on the glass fibers. Excess WD-10 (>1.5%) somewhat lowered the strengths. When the glass fibers was treated with 1% WD-10, the three kinds of strengths also increased with increasing amount of MPP amount, up to 5 wt % based on the composite. An excess amount of MPP (>10%) also lowered the strength of the composites. When the amount of MPP reached 20 wt %, the strengths of the composites were decreased to almost the same levels as those in the the composite containing glass fibers without silane treatment and no MPP.

The above phenomena can be explained as follows: either silane coupling agent-treated glass fibers or MPP can improve the adhesion at the surfaces between the glass fibers and PP because the alkyl group of WD-10 and the main chain of MPP have an affinity for PP molecules, whereas the Si—O part of WD-10 and the MAH groups or COOH group from its hydrolysis product have a hydrogen bond interaction and esterification with the OH groups of the glass fibers, respectively. The interfacial interaction and reaction can promote stress transfer from the matrix to the

TABLE II
Effects of WD-52 and MPP on the Mechanical Properties of Glass Fiber (GF)-Reinforced PP Composites^{a,b}

WD-52/GF (wt %)	MPP/composite (wt %)	BMI/composite (wt %)	s^b (MPa)	i (kJ/m ²)	t (MPa)
0	0	0	33.1	3.1	41.8
1	0	0	36.3	5.8	46.6
1	10	0	52.4	6.0	50.0
1	3	1	48.1	6.7	55.7
1	5	1	52.0	9.4	60.8
1	10	1	64.1	10.4	72.7
1	15	1	58.3	11.5	77.9
1	20	1	55.6	17.8	86.7

^a Glass fiber (GF)/composite = 30 wt %; MAH content of MPP = 11.3%.

^b s , i , and t represent tensile strength, notch impact strength, and flexural strength, respectively.

glass fibers during mechanical properties testing. Excess coupling agent overlaps the first layer of coupling agent on the surface of glass fibers, whereas excess MPP destroys the crystallinity of PP, both resulting in a lowering of strengths. If BMI is added to the composite during blending, no favorable effect on the strength of composite can be seen.

Table II shows the effects of amounts of WD-52 and MPP on the tensile strength, impact strength, and bending strength of the glass fiber-reinforced PP composite. The results indicate that when PP was blended with only WD-52-treated glass fibers without using MPP, the strengths of the composite increased somewhat, but were lower than those of composites containing WD-10-treated glass fibers without using MPP, probably because the alkyl groups of WD-10 are more compatible with the methyl groups of PP. However, when MPP was added simultaneously, the tensile strength increased substantially.

When BMI was blended together with PP, WD-52-treated glass fibers, and MPP, then the three kinds of strengths substantially increased. This is contrary to the case of WD-10-treated glass fibers, as stated above. This fact may be attributed to the existence of nitrogen-containing groups in both BMI and WD-52 and of maleic groups in MPP and BMI. BMI assumes the role of a bridge between MPP and WD-52-treated glass fibers. However, WD-10 contains no amino groups and has no interaction with BMI. In the presence of BMI, both impact strength and flexural strength increase with increasing MPP, whereas a maximum tensile strength occurs at 10% MPP. Excess MPP diminishes the strengths of the composite. The latter phenomenon is probably attributable to the destruction of crystallinity arising from excess MPP.

Table III lists the effects of amounts of WD-70 and MPP on the tensile strength, impact strength, and bending strength of the glass fiber-reinforced PP com-

TABLE III
Effects of WD-70 and MPP on the Mechanical Properties of Glass Fibers-Reinforced PP Composites^{a,b}

WD-70/GF (wt %)	MPP/composite (wt %)	BMI/composite (wt %)	s^b (MPa)	i (kJ/m ²)	t (MPa)
0	0	0	33.1	3.10	41.8
1	0	0	41.0	9.36	45.5
1	10	0	67.9	13.9	69.1
1	3	1	45.3	13.1	69.6
1	5	1	58.9	15.4	78.4
1	10	1	66.9	15.2	74.6
1	15	1	60.1	12.3	67.4
1	20	1	59.6	9.4	49.9

^a Glass fiber (GF)/composite = 30 wt %; MAH content of MPP = 11.3%.

^b s , i , and t represent tensile strength, notch impact strength, and flexural strength, respectively.

posite. When the composite contains 30% of glass fibers treated with WD-70, all three strengths increase, especially the impact strength. Addition of 10% MPP enhances the three strengths even more substantially. The addition of BMI seems to have less effect on the strengths than in the case of the composite containing glass fibers treated with WD-52. This can be explained by the fact that there are fewer similar groups between WD-70 and BMI than between WD-52 and BMI. The three kinds of strengths increase with increasing MPP and a maximum value of tensile strength occurs at 10% MPP, whereas maximum values of impact strength and bending strength occur at 5% MPP. In comparison among Tables I, II, and III, in the absence of MPP, the best coupling agent seems to be WD-10, whereas in the presence of MPP, especially in the range of 5–10% MPP, the best coupling agent is WD-70. This fact can be explained by greater compatibility among the groups of WD-70, MPP, and PP than that between WD-70 and PP.

TABLE IV
Effect of Amount of Glass Fibers, Treated with WD-70, on the Mechanical Strength of PP Composites

Glass fiber ^a /composite (wt %)	MPP ^a /composite (wt %)	s^b (MPa)	i (kJ/cm ²)	t (MPa)
0	0	28.5	1.9	32.4
5	10	34.1	2.1	40.6
10	10	40.5	2.8	45.9
20	10	50.9	7.8	51.8
30	10	67.9	13.9	69.1
35	10	67.9	14.2	69.5
50	10	70.2	14.2	69.0

^a Glass fibers treated with 1% WD-70; MAH content of MPP = 11.3%.

^b s , i , and t represent tensile strength, notch impact strength, and flexural strength, respectively.

TABLE V
Effect of Nucleating Agents (NA) on the Mechanical Properties of PP and the Glass Fiber-Reinforced PP Composites

NA	NA/PP or composite (wt %)	Glass fiber ^a / composite (wt %)	s^b (MPa)	i (kJ/cm ²)	t (MPa)
Adipic acid	0	0	28.5	1.9	32.4
	0.1	0	40.6	6.1	82.7
	0.2	0	41.2	8.7	90.6
	0.5	0	42.1	10.8	92.1
	1.0	0	41.3	8.0	84.3
	1.2	0	35.4	5.4	78.5
	0	30	56.2	9.3	68.6
	0.1	30	60.9	9.4	85.3
	0.5	30	68.3	9.4	92.5
	0.67	30	70.7	11.4	97.3
Disodium phthalate	0.83	30	71.2	12.1	110.5
	1.0	30	72.4	13.2	123.4
	0.17	0	40.8	12.4	74.6
	0.5	0	43.5	16.3	80.4
	0.8	0	41.3	—	86.5
	1.1	0	40.6	13.6	90.3
	1.5	0	39.2	3.0	81.9
	1.67	0	33.7	2.5	76.5
	0.1	30	55.4	10.7	86.4
	0.5	30	50.9	11.9	98.7
0.83	30	49.6	12.3	106.5	
1.0	30	48.7	14.6	116.8	

^a The glass fibers were treated with 1% WD-10 and mixed with 5 wt % MPP in the composite.

^b s , i , and t represent tensile strength, notch impact strength, and flexural strength, respectively.

Effect of amount of glass fibers on the mechanical properties of the composites

The effect of the amount of the WD-70-treated glass fibers on the tensile strength, impact strength, and bending strength is shown in Table IV. All three kinds

TABLE VI
Effect of Different Nucleating Agents on the Crystalline Properties of PP Using WAXD Analysis

Nucleating agent	Amount/PP (%)	Crystallinity (%)	K_α (%)	K_β (%)
Adipic acid	0.2	90.1	69.1	0
	0.5	94.8	67.6	0
	1.0	89.7	59.8	0
Disodium phthalate	0.2	83.6	50.2	17.5
	0.5	92.4	55.9	23.4
	1.0	90.1	56.1	16.7

of strengths obviously increase with the amount of glass fibers < 30% but very slowly when the amount is >30% glass fibers. The optimum amount of glass fibers is 30–35 wt % based on the composite.

Effect of nucleating agent on the strengths of PP and of glass-reinforced PP composites

The effects of two kinds of nucleating agents on the tensile strength, impact strength, and flexural strength of PP and glass fiber-reinforced PP composites are shown in Table V. In the absence of glass fibers, with increasing amount of adipic acid used as nucleating agent, the three kinds of strengths all exhibit a maximum value at 0.5% adipic acid, whereas with increasing disodium phthalate as nucleating agent, maximum values occur at 0.5% for both tensile strength and impact strength, but at 1.1% for flexural strength. Addition of nucleating agent causes the size of spherulites to diminish, resulting in enhancement of strengths. However, excess nucleating agent may cause the spherulites to become too fine and have many defects, resulting in lowering the strengths of PP. In the presence of glass fibers, the tensile strength and the flexural strength of the composite increase

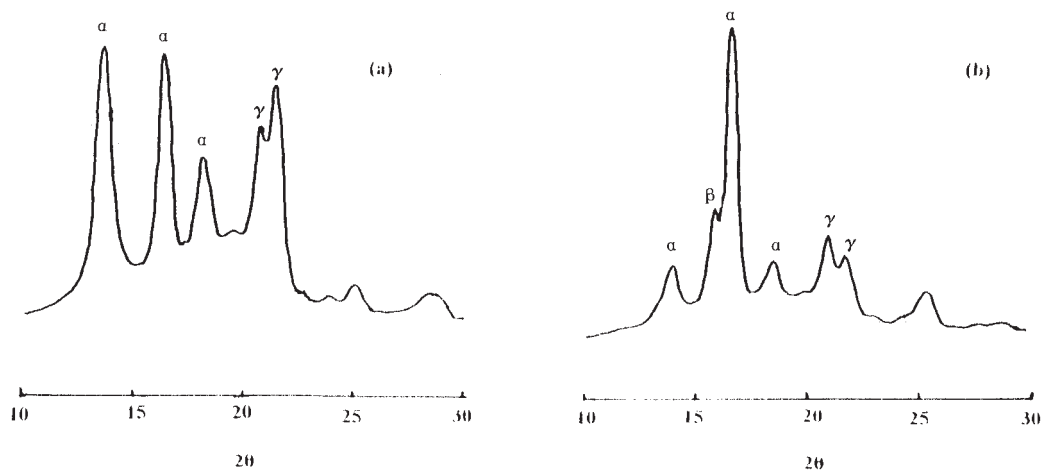


Figure 1 WAXD spectra of PP with addition of (a) 0.5% adipic acid and (b) 0.5% disodium phthalate.

TABLE VII
Effect of Different Elastomers on the Mechanical Properties of Glass Fiber-Reinforced PP Composites^{a,b}

Elastomer	Elastomer/composite (wt %)	<i>s</i> (MPa)	<i>i</i> (kJ/m ²)	<i>t</i> (MPa)
—	0	72.4	13.2	123.4
SBR	10	54.1	16.5	95.7
	15	39.3	10.8	78.9
<i>cis</i> -1,4-PB	10	44.5	13.7	84.8
	15	37.6	7.5	65.6

^a The composite contains 30% glass fibers treated with WD-70, 10% MPP with 11.3% MAH content, and 1% adipic acid as nucleating agent.

^b *s*, *i*, and *t* represent tensile, notch impact strength, and flexural strength, respectively.

with increasing adipic acid. However, the tensile strength decreases and the flexural strength increases with increasing disodium phthalate. The notched impact strength of the composite increases more obviously with increasing disodium phthalate than that with increasing adipic acid. The difference arises from the fact that they yield different types of crystals; adipic acid is a nucleating agent for α -type crystallization, but disodium phthalate yields β -type crystals, which are beneficial to the impact strength. The optimum tensile strength, impact strength, and flexural strength can reach 72.4 MPa, 13.2 kJ/m², and 123.4 MPa, respectively, using 1% adipic acid as nucleating agent, 30% WD-70-treated glass fibers, and 10% MPP. The optimum amount of disodium phthalate is 1.0% with respect to the notched impact strength and flexural strength, although the tensile strength decreases.

Effect of different nucleating agents on the crystallinity and type of PP crystals

Crystallinity and type of PP crystals can be determined by using WAXD analysis. The WAXD spectra of PP, with addition of 0.5% adipic acid and 0.5% disodium phthalate, are shown in Figure 1(a) and (b), respectively. Table VI details the effects of amount of adipic acid and disodium phthalate on crystallinity and crystal type of PP. With increasing nucleating agent, the crystallinity of PP increases up to a maximum value about 93% at 0.5% nucleating agent. Excess nucleating agent causes the crystallinity to decrease because of the formation of too small PP spherulites with defects. As shown in Table VI, adipic acid favors the formation of α -type crystals without formation of β -type crystals, but an excess amount favors the formation of γ -crystals. On the contrary, disodium phthalate favors the formation of β -type crystals (as shown in Table VI), which exhibits a sheaflike structure.

Effect of elastomer on mechanical properties of the composite

The effect of elastomer, that is, styrene-butadiene rubber (SBR) and *cis*-1,4 polybutadiene (PB), on the strengths of the composite containing WD-70-treated glass fibers, PP, MPP, and adipic acid is shown in Table VII. In the presence of elastomer both tensile strength and bending strength severely decrease; only the impact strength increases at 10% elastomer. SBR seems better than *cis*-1,4-PB. The excess amount of elastomer causes a deterioration in the impact strength, a phenomenon that implies that the elastomer can increase the toughness of the composite, but

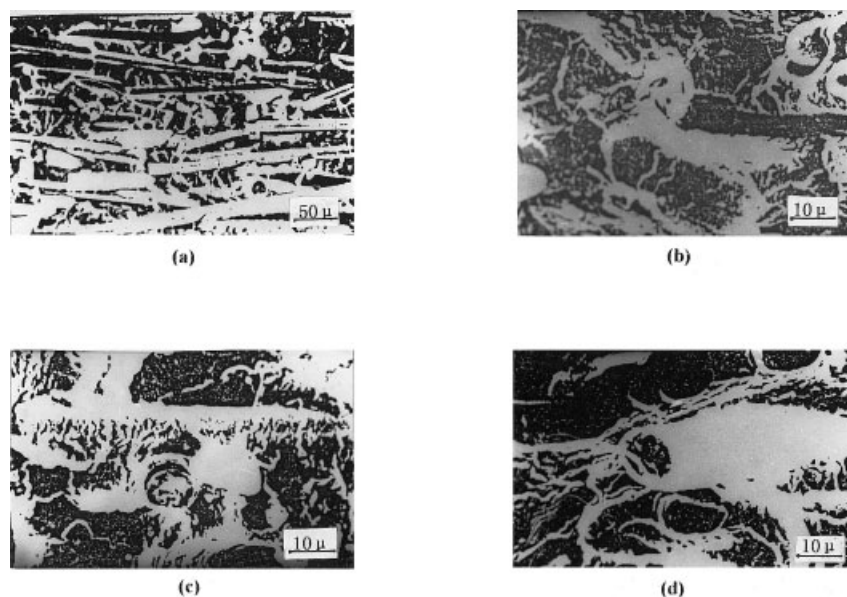


Figure 2 SEM micrographs of PP composites, containing glass fibers (a) without treatment, (b) treated with WD-70, (c) treated with WD-52 in the presence of BMI, and (d) containing MPP and glass fibers without silane treatment.

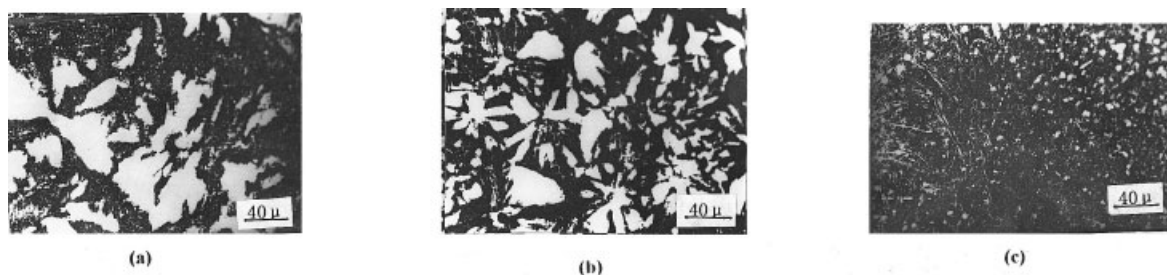


Figure 3 Polarized light micrographs of (a) pure PP, (b) PP containing 0.1% adipic acid as nucleating agent, and (c) composite containing 30% WD-70-treated glass fibers and 5% MPP.

an excess amount of elastomer hinders the interfacial adhesion among PP, MPP, and glass fibers.

Morphology of the glass fiber-reinforced composites and PP

The morphology of the PP composites, containing glass fibers without treatment, treated with WD-70, treated with WD-52 in the presence of BMI, and containing MPP and glass fibers without silane treatment are shown, respectively, in Figure 2(a)–(d). In Figure 2(a) the clean and smooth glass fibers and very poor interfacial bonding between the glass fibers and matrix can clearly be identified. In Figure 2(b), (c), and (d) no smooth glass fibers can be observed. Rather, the glass fibers are surrounded by fibrils of matrix; that is, small pieces of matrix are bonded firmly to the glass-fiber surfaces, indicating that a strong interfacial adhesion developed between the silane-treated glass fibers and PP or MPP. The MAH functional groups of MPP can react with OH groups on the glass-fiber surfaces during compounding. There are different types of interfacial interactions that develop between the glass fibers and MPP, coupling agent and glass fibers, coupling agent and PP, and coupling agent and MPP. Most of the MPP is miscible with PP. SEM micrographs of the fractured surface of the composite showed greater interfacial adhesion between the modified glass fiber and modified polypropylene than that without modification.

Polarized light micrographs in Figure 3(a), (b), and (c) show, respectively, the spherulites of pure PP, PP containing 0.1% adipic acid as nucleating agent, and a composite containing WD-70-treated glass fibers and 5% MPP. The pure PP shows large spherulites, whereas PP containing a small amount of nucleating agent exhibits smaller spherulites. Large spherulites of PP are responsible for poor impact strength. The composite containing both WD-70-treated glass fibers and MPP shows very small spherulites, probably because the short and fine glass fibers with large surfaces in the presence of MPP favor heterogeneous nucleation by acting as nucleation sites for crystallization. The polarized micrographs correlate well with the impact strength of the materials.

CONCLUSIONS

Both the silane coupling agent and MPP enhance the tensile strength, notched impact strength, and flexural strength of PP/glass fiber composites. In the absence of MPP, the effect of silane coupling agent on the mechanical properties of the composites decreases in the following order: WD-10 > WD-70 > WD-52, whereas in the presence of MPP, the order changes as follows: WD-70 > WD-10 > WD-52. When the glass fibers were treated with WD-52, BMI can increase the mechanical properties of the composite. The three kinds of strengths increase with increasing amount of MPP, to maximum values at 5% MPP. Addition of adipic acid as an α -nucleating agent to the composite can further enhance the mechanical properties, whereas addition of disodium phthalate as a β -nucleating agent enhances only the notched impact strength. Blending with SBR elastomer can somewhat improve the notch impact strength of the composites, but severely lowers the tensile strength and flexural strength. SEM micrographs of the broken surface of the composite showed greater interfacial adhesion between the glass fibers and polypropylene in the modified composite than that without modification.

References

- Denault, J.; Vu-Khau, B. *Polym Compos* 1988, 9, 360.
- Yue, C. Y.; Quek, M. Y. *J Mater Sci* 1994, 29, 2487.
- Severini, F.; Pegoraro, M.; Ricca, G. S.; Di, L. L. *Makromol Chem Macromol Symp* 1989, 28, 217.
- Rijsdijk, H. A.; Contant, M.; Peijs, A. *Compos Sci Technol* 1993, 48, 161.
- Hausmann, K.; Flaris, V. *Polym Compos* 1997, 5, 113.
- Nygård, P.; Redford, K.; Gustafson, C.-G. *Compos Interfaces* 2002, 9, 365.
- Roux, C.; Denault, J.; Champagne, M. F. *Annu Tech Conf Soc Plast Eng* 1999, 57, 2656.
- Iroh, J. O.; Berry, J. P. *Polymer* 1993, 34, 4747.
- Iroh, J. O.; Berry, J. P. *Polymer* 1993, 34, 2303.
- Tjong, S. C.; Xu, S. A.; Li, R. K. Y.; Mai, Y. W. *J Appl Polym Sci* 2002, 86, 1303.
- Tjong, S. C.; Xu, S. A.; Li, R. K. Y.; Mai, Y. W. *Compos Sci Technol* 2002, 62, 831.
- Tjong, S. C.; Xu, S. A.; Mai, Y. W. *J Polym Sci Part B: Polym Phys* 2002, 40, 1881.
- Zhu, S.-N. *Analysis of Polymeric Materials*; Science Publishers: Beijing, 1988 (in Chinese).