

Effect of Silane-Grafted Polypropylene on the Mechanical Properties and Crystallization Behavior of Talc/Polypropylene Composites

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ABSTRACT: Talc-filled polypropylene (PP) composites coupled with silane-grafted polypropylene (PP-*g*-Si) were prepared. Effect of PP-*g*-Si on the mechanical properties, crystallization, and melting behavior of PP composites was investigated. Compared with the uncoupled composites, the mechanical properties of Talc/PP composites coupled with a small amount of PP-*g*-Si were increased to some extent. Meanwhile, PP-*g*-Si can promote crystallization rate and increase crystallization temperature of PP in the composites. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 77: 2974–2977, 2000

Key words: silane-grafted polypropylene; polypropylene; talc; composite; mechanical properties; crystallization behavior

INTRODUCTION

Talc-filled polypropylene composites have been studied.^{1–7} The properties of the composites strongly depend on the interfacial adhesion between filler and matrix. The interfacial adhesion and properties of the interphase play an crucial role in the determination of composite properties. A good interfacial adhesion may improve the mechanical properties of composites, while a poor adhesion may decrease the mechanical strength. To enhance the mechanical properties of the composite materials, coupling agents were used to modify the surface of the filler. The ordinary coupling agents are silane and titanate coupling agent, and other small molecular coupling agents. Another method often used is the modification of

the matrix. Grafting of different chemical moieties to a polyolefin chain is a frequently applied method to improve the compatibility of the immiscible polymer blends and the interfacial adhesion in polymeric composites. Polypropylene (PP), an apolar polymer, is usually grafted with polar groups like, acrylic acid, maleic anhydride, acrylic esters, etc. Functionalized polyolefins were used as a coupling agent in filler/polyolefin composites.⁸ The interaction between matrix and filler was improved and the mechanical properties of composites were enhanced. The shortcoming of the small molecular coupling agents was overcome. Low molecular coupling agents cannot form a strong adhesion bond between polyolefin and filler. Short molecules at these substances, although they can create a relatively strong bond with the filler surface, do not form a chemical bond with either the polyolefin molecules and physical entanglements causing an improvement in polyolefin cohesive strength.⁹ Functionalized polyolefins can create strong bonds with the filler surface and form physical entanglements with the matrix in the bulk. The effect of functional-

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ized polyolefins on PP composites filled with mica,^{10–13} CaCO₃,^{9,14–18} Mg(OH)₂,^{9,14,17,19} and reinforced with glass fiber^{20–23} have been investigated.

Silane grafted polypropylene (PP-g-Si) as a coupling agent on glass fiber/PP composites had been studied by other authors.²⁴ Results showed that the mechanical properties of composites were obviously increased, which indicate that the PP-g-Si is an efficient macromolecular coupling agent of PP and glass fiber. In the present investigation, vinyltrimethoxy silane-grafted polypropylene was used as a coupling agent on talc/PP composites, and their mechanical properties and crystallization and melting behavior were measured.

EXPERIMENTAL

Materials

Polypropylene was commercial product F401, obtained from Guangzhou petrochemical Complex. Talc powder (1250 mesh), antioxidant 1010 (AO), and calcium stearate (Cast) were all commercial products. Silane-grafted polypropylene (PP-g-AA) were prepared by others.²⁴

Preparation of Talc/PP Composites

The composites of talc-filled polypropylene pellets and additives were conducted in a SHJ-53 twin screw compounding extruder (L/D = 28/1, D = 18.9 mm). The feed compositions are given in Table I. The feed ratio of polypropylene (including PP-g-Si) to talc was 70 : 30 by weight. The temperature range in extruder was 200 to 220°C, and the final composite extruded was cooled in-line in a water-bath, dried, and granulated less than 3 mm.

Table I Feed Composition of Talc/PP Composite

Sample	PP, wt %	PP-g-Si, wt %	AO, phr	Cast, phr
M0	100	0	0.2	0.2
M1	95	5	0.2	0.2
M2	90	10	0.2	0.2
M3	80	20	0.2	0.2
M4	70	30	0.2	0.2

^a PP + PP-g-Si : Talc = 70 : 30 by weight.

^b The PP-g-Si was obtained by the reactive grafting process in the extruder with the PP, A-171, DCP, and Cast in the mass ratio of 100 : 6 : 1 : 0.2 by weight.

Table II Processing Parameters

Injection time (s)	50 (26)
Injection pressure (MPa)	115
Screw speed (rpm)	60
Cooling time(s)	30 (20)
Mold temperature (°C)	60
Melt temperature (°C)	
Zone I	230
Zone II	230
Zone III	220

Preparation of the Specimens

The prepared talc/PP composite granular were dried at 80°C for 8 h before injection molding. The granular were injection molded by using a CJ-150M-NC screw injection-molding machine. The processing conditions are specified in Table II. The size and shape of test specimens are described elsewhere.²⁴

Test Procedures

The methods of tensile, flexural, and impact tests were described elsewhere.²⁴

Differential Scanning Calorimetry

DSC measurements were performed with a Perkin-Elmer DSC-7 instrument in a nitrogen atmosphere. Samples were held at 230°C for 3 min to ensure complete melting of crystals before cooling to 50°C at 20°C/min. The cooling scanning and the second heating scanning were record.

RESULTS AND DISCUSSION

Mechanical Properties

Tensile, flexural, and impact results of talc/PP composites are presented in Table III. Compared with neat PP, the mechanical properties of the composites are enhanced when PP was filled with talc, in particular, tensile modulus and flexural modulus due to the stiffening effect of the filler. The marked fall in elongation at break is due to the presence of the filler acting as stress precursors.¹⁹ Coupled with small amount of PP-g-Si (less than 10 wt % in the PP matrix), the mechanical properties of the composites were further improved. Although the tensile modulus of PP composite decreases with increasing the PP-g-Si content, yet still higher than that of PP parents. Tensile strength and flexural properties show a

Table III Mechanical Properties of Talc/PP Composites

	Neat PP	M0	M1	M2	M3	M4
Tensile strength (MPa)	33.0	34.9	38.5	38.7	38.2	36.5
Tensile modulus (GPa)	1.84	3.89	3.16	3.12	3.21	3.19
Elongation at break (%)	>250	69	29	23	18	11
Flexural strength (MPa)	53.5	59.4	63.6	68.4	65.6	64.4
Flexural modulus (GPa)	1.60	3.28	3.67	4.07	4.07	3.46
Izod impact strength (J/m ²)	4.9	5.9	6.2	5.0	4.3	3.8

maximum at about 10 wt % PP-g-Si content. When the PP-g-Si content in the PP matrix is over a certain degree, the mechanical properties of the composite were all decreased. With increasing the PP-g-Si content, the thickness of the interphase increases; it becomes more diffuse and stress transfer improves significantly. This results in a slight decrease of the modulus.¹⁰ In the talc/PP composite, due to the lack of polarity and reactivity in the PP chain, there is poor compatibility between PP matrix and talc, which results in weak interaction at the interface, also the low mechanical properties of the composite. It is known that there a strong interaction exists between silane and talc; meanwhile, there is good compatibility between the PP and PP segment of PP-g-Si. A diffuse interlayer with excellent adhesion can be formed between the components. In this interlayer, polarity gradually decreases from the surface of the filler to the matrix polymer. Therefore, addition of PP-g-Si to the talc/PP composite may improve the interaction of the talc/PP interphase, and thus improve the mechanical properties of the composites. There exists a critical amount of PP-g-Si in the composite at which the talc surface was covered with PP-g-Si, followed by the PP matrix because the order of the surface free energy of them is talc > PP-g-Si > PP, and PP-g-Si acts as the coupling agent or compatibilizer. Over the critical amount, there is a negative effect of mechanical properties of the composite due to the degradation of PP during the grafting process and incompatibility between PP and trimethoxy silane groups in PP-g-Si. The previous results indicated that after grafting, the melt flow index of the PP increased from 3.2 to 8.7 g/min. The unnecessary PP-g-Si acts no longer as the coupling agent, even as the foreign matter, and results in the decreasing of the mechanical properties.

Crystallization and Melting Behavior

Figure 1 is the crystallization and melting curves of DSC for talc/PP composites. The measured re-

sults of DSC analyses for PP matrix and talc/PP composites are shown in Table IV. In a heating run of the sample, the onset temperature (T_{om}) for neat PP is 156.0°C, and the T_{om} of uncoupled talc/PP composite (M0) is 158.1°C. As shown in Table IV with addition of a small amount of PP-g-Si to the composite, the T_{om} of the composite decreased, which is attributed to the change of the shape of melting peak due to the nucleation of the filler. Furthermore, the T_{om} increased with increasing of the PP-g-Si content in the compos-

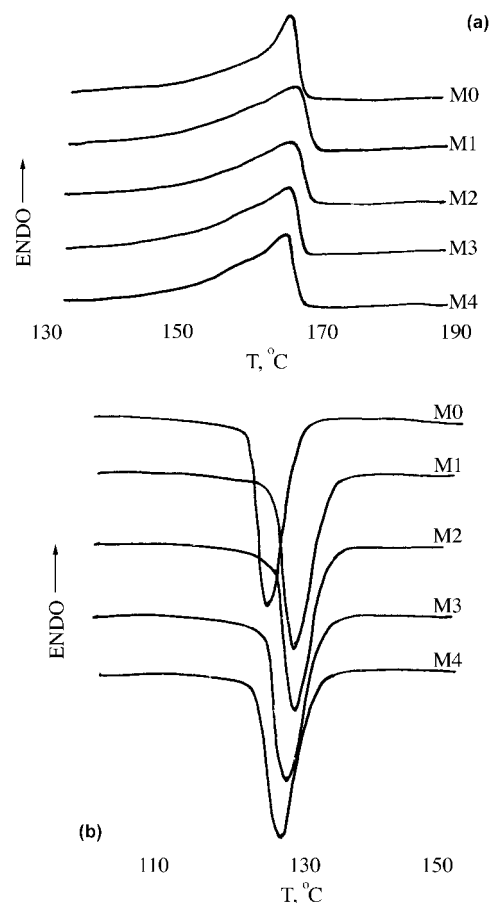


Figure 1 The melting (a) and crystallization (b) curves of talc/PP composites.

Table IV DSC Results of Talc/PP Composites

Samples	Heating Run			Cooling Run			$T_m - T_c$ (°C)
	T_{om} (°C)	T_m (°C)	ΔH_m (J/g)	T_{oc} (°C)	T_c (°C)	ΔH_c (J/g)	
Neat PP	155.3	162.1	83.0	111.8	106.5	-92.3	55.6
M0	158.8	164.8	95.4	130.1	121.6	-102.4	43.2
M1	151.9	165.2	86.6	134.6	125.1	-88.6	40.1
M2	151.4	165.0	95.0	134.3	125.5	-96.7	39.5
M3	154.3	165.0	85.2	133.4	123.2	-90.5	41.8
M4	156.3	164.0	92.3	132.6	122.8	-98.5	41.2

ites. Although the temperature of melting peak (T_m) of the talc/PP increased slightly compared with the neat PP due to the nucleation of the filler, the PP-*g*-Si has almost no influence on the temperature of the melting peak.

In a cooling run of the neat PP, the onset temperature (T_{oc}) of the crystallization is 111.8°C and the crystallization peak temperature (T_c) is 106.5°C. When filled with talc, the T_{oc} and T_c of the PP in talc/PP composites increased remarkably. Addition of PP-*g*-Si in the composite further increases the crystallization peak temperature and the onset temperature of the crystallization of PP in the composites. This may be due to an increase in the interactions between the talc particles and the PP matrix, improving the nucleating activity of the filler and promoting faster crystallization during the cooling process.²⁵ The almost constant T_m values and the increasing of the T_c values in the composites result in a reduced supercooling ($T_m - T_c$), which shows an increase in the rate of crystallization. This behavior is the result of an improved nucleating activity of the talc as a nucleating agent for the PP by addition of PP-*g*-Si. Because of improvement the talc nucleating activity, the formation of a finer and more resistant crystalline texture of PP in the composites results in an increase the mechanical properties of talc/PP composites.

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