

# Effect of Silane KH-550 to Polypropylene/Brucite Composite

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**ABSTRACT:** The effect of the KH-550 type silane coupling agent on the properties of polypropylene (PP)/brucite (BC) composite was studied. X-ray diffraction, scanning electron microscope, and polarization optical microscope indicated that morphology structure of PP changed with the addition of KH-550, which activated the heterogeneous nucleation centers on BC surface, which made the distribution of the spherulite diameter become narrow and uniform, and improved the dispersibility of BC in the matrix. Therefore, KH-550 enhanced the interfacial adhesion of matrix-filler and improved the compatibility of the

PP/BC composite, when KH-550's content was 2.9% the tensile strength and impact strength increased approximately by 90% and by 30%, respectively than PP/BC. Flow tests and IR proved that some reactions took place between BC and KH-550. All the investigations showed KH-550 was a proper coupling agent in the PP/BC composite. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 107: 1000–1005, 2008

**Key words:** silane coupling agent; polypropylene; brucite; compatibility

## INTRODUCTION

Polypropylene (PP) is a polymer of great importance in the industrial sector because of its low density, high water and chemical resistance, ease of processability, and being one of the most cost-effective polymers available today.<sup>1</sup> However, the susceptibility of PP to fire continues to be of great concern all over the world. As a fire-retardant, magnesium hydroxide (MH) is widely used in polymer composites by virtue of its high decomposition temperature, innocuity, restraining smoke, prevent dropping, and appending, etc. For PP/MH composites, much research has been focused on morphology, mechanical, and flame-retardant properties.<sup>1–4</sup> Because of the incompatibility of PP and MH, PP/MH composites exhibit poor mechanical properties; therefore, it is necessary to use coupling agents to improve the compatibility. Liauw et al.<sup>5</sup> have investigated that MH filler treated with vinytriethoxy-silane enhanced the rigid filler-matrix of PP/MH composites. Wang et al.<sup>6</sup> investigated that LLDPE grafted with dibutyl maleate (PE-g-DBM) compatilized LLDPE/MH composites; the results showed PE-g-DBM could improve the mechanical properties of the LLDPE/MH blends. Yang et al.<sup>7</sup> studied that coupling agent could not only help filler (MH) achieve better dispersion in polymer

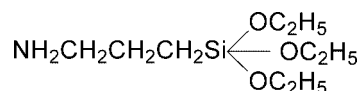
matrix, but also improve the roughness of the composites with good rigidity.

The main component of Brucite (BC) is magnesium hydroxide (MH). Although BC is inferior to MH in whiteness and purity, its abundant mineral resource and lower price make it more attractive for researchers.<sup>8–10</sup> The silane coupling agent was the most widely used coupling agent,<sup>11</sup> our previous work proved that KH-550 had a better effect among silane coupling agents<sup>12</sup> in PP/BC composite. So what is introduced in this article is to investigate the coupling action of KH-550 in the PP/BC composite.

## EXPERIMENTAL

### Materials and samples preparations

The PP [type T30S,  $d = 0.901 \text{ g/cm}^3$ ] was supplied by Tian-Jin United Chemical (Tianjin, China). BC:  $\text{Mg}(\text{OH})_2 \geq 93\%$ ,  $\text{SiO}_2 \leq 1.6\%$ ,  $\text{CaO} \leq 0.8$ ,  $\text{Fe}_2\text{O}_3 \leq 0.2$ ,  $d_{50}$ : 2.7–3.2  $\mu\text{m}$  were supplied by Haicheng Xin-da Mining Limited Company (Limited Ciaoning, China). The KH-550 silane coupling agent was supplied by Beijing Shenda Fine Chemical Company (Beijing, China), its structure is:



The BC powder and KH-550 were kneaded in a ZH-0.5 kneading at 130°C. The weight to weight

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ratios (w/w) of BC/KH-550 were 50 : 1, 50 : 2, 50 : 3 (mass ratio) and they were marked as BK. The composite samples were prepared by melt-blending on a two-roll mill at 180°C for 10 min. When PP melted, the BK was added into the mill, respectively. The ratios (w/w/w) of PP/BC/KH-550 were 50 : 50 : 1, 50 : 50 : 2, 50 : 3 (mass ratio). The composite samples were compressed in an electric-heat press for 3 min at 15 MPa and 180°C and cold-pressed at 5 MPa.

### Measurements of mechanical properties of composites

The testing specimens were shaped with a universal clipper. The tensile strengths were measured following the GB/T-1040-1992 standard by an LJ-5000N test apparatus, and the impact strengths were measured following the GB/T-1843-1993 standard by an XCJ-40 test apparatus. The testing was conducted at room temperature. Values were an average of five testing.

### Polarization microscope

Polarization optical microscope (POM) micrographs were obtained with a XSZ-H (Chongqing, China) polarized optical microscope equipped with a DSC-V1 (SONY, Japan) camera. Compression-molded film was sandwiched between two cover glasses. The samples were heated from room temperature to 200°C, held at this temperature for 5 min to allow complete melting on a hot-stage, then transferred to a 200°C oven, and cooled hermetically and naturally to 145°C for isothermal crystallization for 3 h. The samples were observed in the condition of being magnified 100 times.

### Scanning electron microscope

After being coated with gold, the tensile fracture surfaces morphology of the composites was investigated with a scanning electron microscope (SEM) (KYKY-2800B, USA). The electron micrographs were taken, using an acceleration voltage of 25.0 kV.

### Rheological measurements

The melt viscosity of samples under processing conditions was measured using a high-pressure capillary rheometer (mode XLY-1, China), with a capillary diameter of 1 mm, and  $L/D$  ratio of 40. The experiment temperature was fixed at 180°C, with experimental loads of 100, 120, 140, 160, and 180 kg/cm<sup>2</sup>.

### X-ray diffraction

The aforementioned composite samples were pressed 4-mm thick sheets for X-ray diffraction (XRD) (model Y-4Q, China) analysis.

### Infrared spectroscopy

The sample of BC/KH-550 (BC modified by KH-550) was prepared according to the ratio (w/w) 1 : 1. The prepared sample was packed with filter paper and extracted with acetone in a Soxhlet apparatus for 72 h in order to wash off unreacted KH-550, and then dried for IR analysis by IR Spectroscopy Instrument (vector-22 Germany Bruker).

## RESULTS AND DISCUSSION

### Mechanical properties

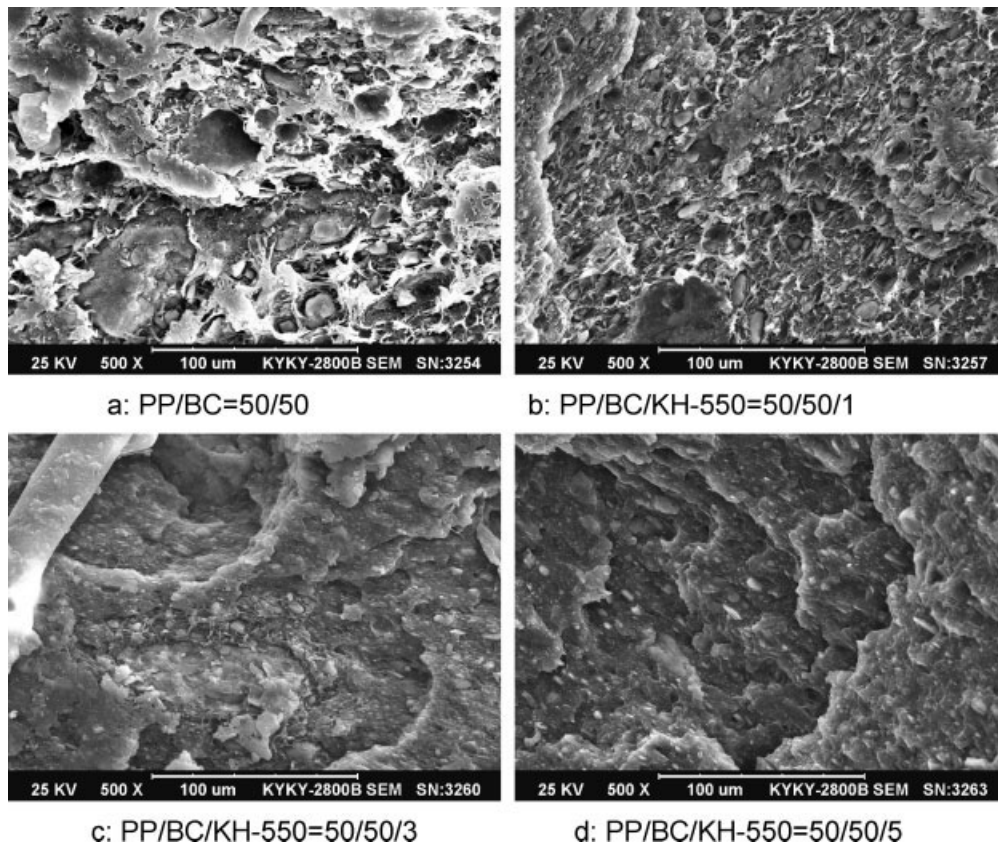
Generally speaking, the PP/MH has obvious effect of flame retardancy when MH content exceeds 50%,<sup>13</sup> so PP/BC = 50/50 was chosen. The mechanical properties of PP/BC/KH-550 composites were shown in Table I. Because of the poor compatibility between PP and BC, the mechanical properties of PP/BC decreased obviously than pure PP. High loading of BC particles embedded in the PP matrix results in reduced ability to absorb impact and tensile energy. This is due to the filler effect which damage PP matrix continuity. However, the mechanical properties were improved obviously when BC was modified by silane coupling agent KH-550. BC is sized with silane coupling agent, which can adhere to the PP matrix. Especially, the tensile strength and impact strength of composite E (PP/BC/KH-550 = 50/50/3) were enhanced by 34.9% and by 91.5%, respectively than PP/BC.

It implies that the compatibility between the BC dispersed phase and the PP matrix phase can be improved by silane coupling agent KH-550.

TABLE I  
Effect of Silane Coupling Agent Contents  
on Properties of PP/BC

Sample	PP/BC/KH-550 (w/w/w)	Impact strength (KJ/m <sup>2</sup> )		Tensile strength (MPa)	
		Data	S*	Data	S*
A	100/0/0	4.97	0.19	38.07	0.45
B	50/50/0	2.96	0.13	25.76	0.56
C	50/50/1	3.66	0.18	30.33	0.60
D	50/50/2	4.06	0.24	26.82	0.53
E	50/50/3	5.67	0.27	34.75	0.37
F	50/50/4	4.01	0.15	27.77	0.80
G	50/50/5	3.21	0.25	26.71	0.75

S\*: standard deviation.



**Figure 1** Scanning electron micrographs of tensile fracture surface of composites.

### Morphology analysis

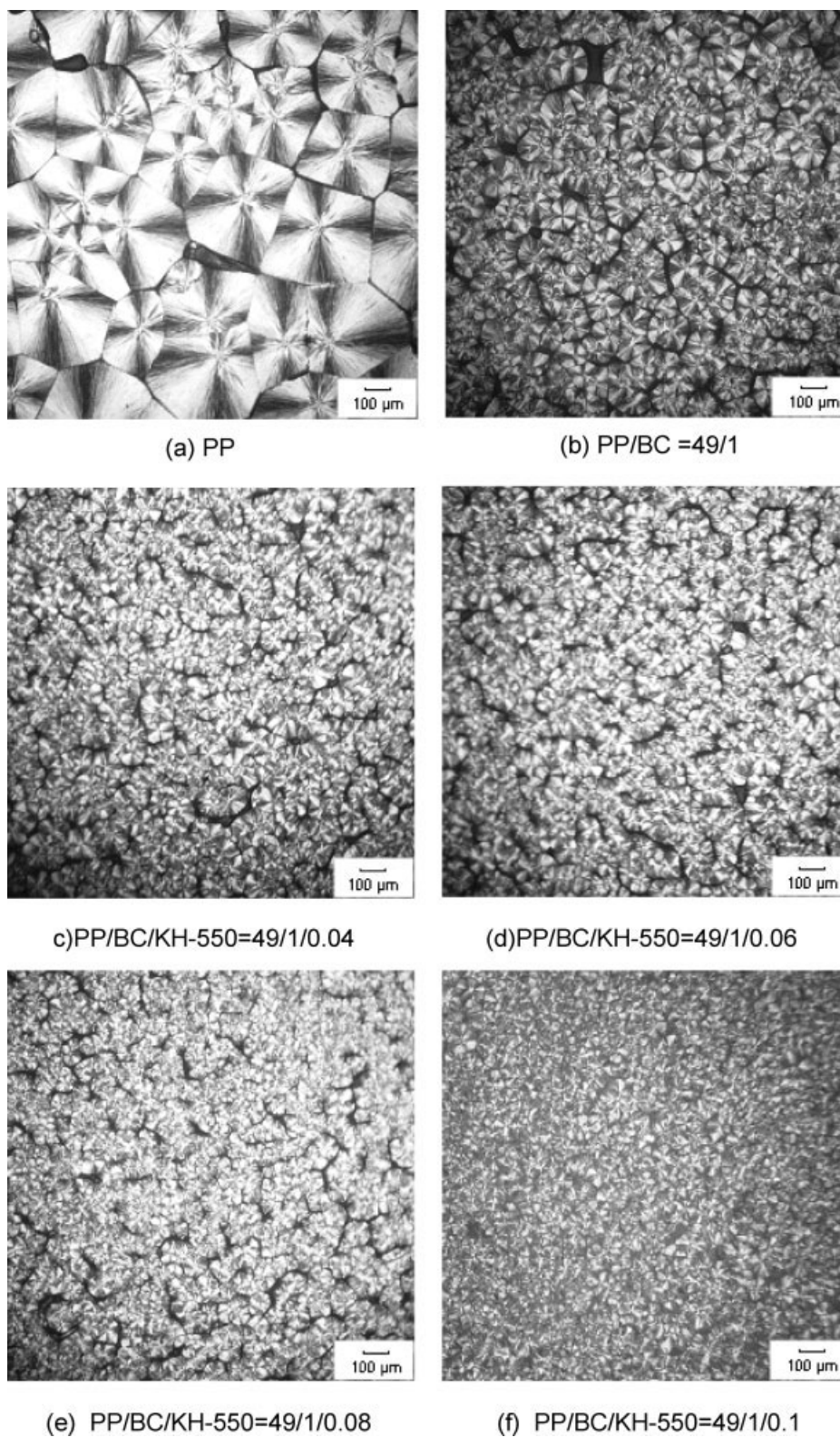
The morphology is an important way to characterize the compatibility of the composites directly. Here it is investigated by SEM, POM, and XRD.

Figure 1 shows the SEM of the tensile fracture surfaces of the composite samples. It can be seen from Figure 1(a), the picture of PP/BC composite, the interface between BC and PP can be clearly observed, and their surfaces with the remaining holes appeared to be very smooth. Because BC had a high surface energy and PP had a low surface energy, the adhesion was poor between them. As was shown in Figure 1(b–d), with the silane coupling agent KH-550 content increased, the fracture surfaces of composites became rough, there were drapes appeared and the interfaces became blurred which illustrated that the adhesion was improved.

Figure 2 shows the POM of the composite samples. It is unfavorable to observe the crystal morphology of PP/BC with high content BC, so the low content BC composites were prepared to process POM analysis. From Figure 2(a), we can see the pure PP revealed well-defined and large spherulitic morphology. The perfect sphere crystal of pure PP and spherulites which impinging each other were observed. Figure 2(b) showed the crystal morphol-

ogy of the PP was greatly affected by the addition of BC, the spherulites size decreased significantly and dispersed nonuniform, and the spherulites were small in the place where more BC dispersed, but large where there were less or no BC. Comparing with Figure 2(b), Figure 2(c–f) showed that the size distribution of the spherulites became narrow and uniform, which may be due to the KH-550 activating the heterogeneous nucleation centers on the filler surface.

Figure 3 shows the XRD patterns for PP, BC, and PP/BC/KH-550. The strong diffraction peaks were at about  $2\theta$  of  $14.00^\circ$ ,  $16.79^\circ$ ,  $18.48^\circ$ , and  $21.80^\circ$ , of which the former three peaks corresponded to (1 1 0), (0 4 0), and (1 3 0) planes, respectively and were characteristic of the typical  $\alpha$ -form monoclinic structure of PP.<sup>14</sup> Figure 3(c) showed that when BC was added the diffraction intensity of PP at about  $2\theta = 18.48^\circ$  increased and partial reason is the overlapping of (0 0 1) plane ( $2\theta = 18.53^\circ$ ) in BC and (1 3 0) plane ( $2\theta = 18.48^\circ$ ) in PP; in Figure 3(d–h), the intensity of (0 4 0) and (1 3 0) planes increased and then decreased with the increase of KH-550 content, which indicated the planes of (0 4 0) and (1 3 0) in PP were preferential crystallographic orientation. All the above showed KH-550 improved the interaction between BC and PP.



**Figure 2** Polarization optical micrographs for the crystal of the composites.

From above analysis, it can be concluded that KH-550 enhanced the matrix-filler interfacial adhesion, improved the dispersibility of BC in the matrix,

which moreover influenced the crystallization behavior of the PP matrix, and increased mechanical properties as shown in Table I.

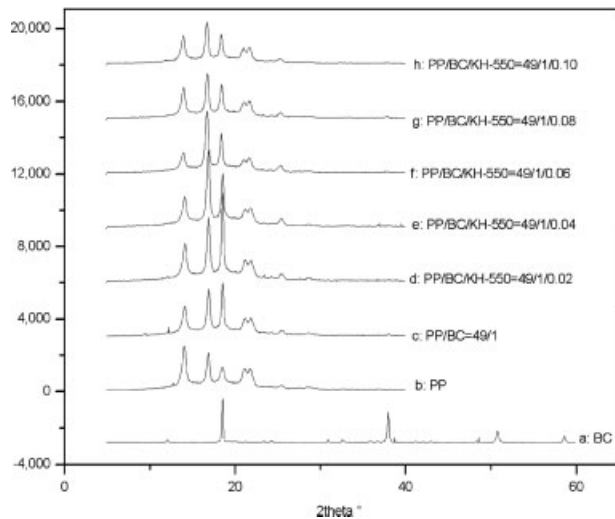


Figure 3 XRD patterns of PP/BC/KH-550 composites.

### Rheological behavior of composites

A plot of apparent viscosity  $\ln \eta_a$  versus shear rate  $\ln \dot{\gamma}_w$  was given in Figure 4. The  $\eta_a$  of PP/BC composite increased comparing with pure PP, because BC restricted the motion of PP molecular. With the addition of KH-550, the apparent viscosity of composites decreased first; When 3% KH-550 was added, the apparent viscosity of composite E was the smallest which indicated KH-550 played the role of lubricant; but when more KH-550 was added, the apparent viscosity was increased, which implied some actions took place.

In addition, the apparent viscosity of the composites decreased with the shear rate ( $\ln \dot{\gamma}_w$ ) increased and the rule demonstrated was linearity, which implied that the melt is a pseudoplastic liquid and has good processibility.

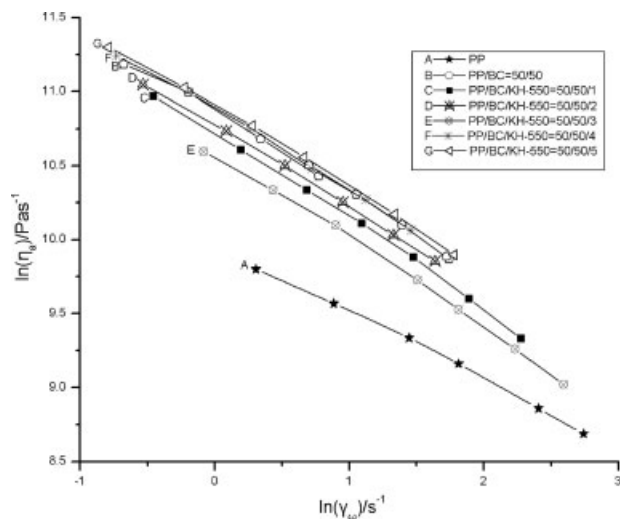


Figure 4 Rheological behavior of composites.

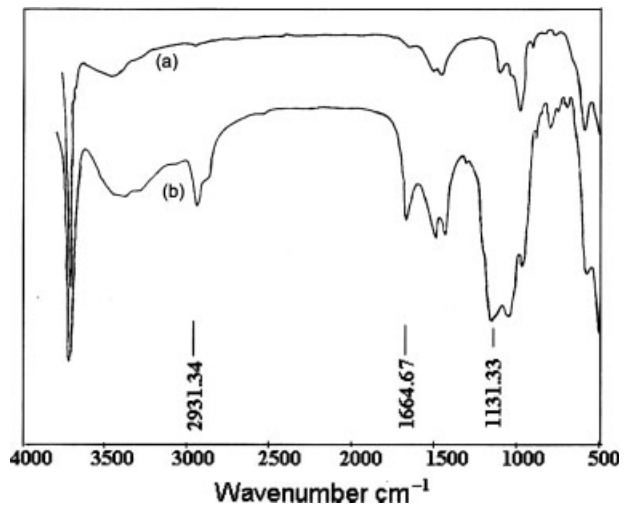


Figure 5 FTIR Spectra of BC. (a): BC (b): BC modified by KH-550.

### Coupling mechanism

To explain clearly the coupling mechanism, the sample of BC/KH-550 according to the ratio (w/w) 1 : 1 was prepared for FTIR spectrum. As can be seen from Figure 5, the spectrum of (a) and (b) have many similarities. The appearances of peaks at  $2931.34 \text{ cm}^{-1}$ ,  $1664.67 \text{ cm}^{-1}$ , and  $1131.33 \text{ cm}^{-1}$  are due to C—H stretching vibration, bending vibration of N—H and stretching vibration of Si—O, respectively, which proved the KH-550 grafted on BC. The reactions might be the hydroxy of BC through dehydration, can react with the  $[-\text{Si}(\text{OC}_2\text{H}_5)_n(\text{OH})_{3-n}]$  that can be obtained from the  $[-\text{Si}(\text{OC}_2\text{H}_5)_3]$  hydrolysis of KH-550 silane coupling agent. In addition, the organic segment of KH-550 entwists with PP segments. So the compatibility of BC and PP was improved, and the mechanical properties of composites were increased.

### CONCLUSIONS

From the above results the following conclusions were made.

1. KH-550 coupling agent can be used as a compatibilizer in the PP/BC composites, which increases not only the tensile strength, but also the impact strength of the composites.
2. The SEM micrographs give the evidence that KH-550 can improve the adhesion between BC and PP, and make BC particles well dispersed into the polymer matrix.
3. POM and XRD illustrated that KH-550 activated the heterogeneous nucleation center on the BC surface, made BC dispersion become uniform and affect the crystal morphology of PP.

4. IR proved that some reactions took place between BC and KH-550 and rheological behavior showed the adhesion of PP/BC composite improved with the addition of KH-550.

In a word, KH-550 was a good coupling agent for PP/BC composites, which improved the interactions and the compatibility of PP and BC.

#### References

1. Shehata, A. B. *J Appl Polym Sci* 2004, 85, 577.
2. Dvir, H.; Gottlieb, M.; Daren, S.; Tartakovsky, E. *Comp Sci Technol J* 2003, 63, 1865.
3. Tai, C. M.; Li, Robert K. Y. *Mater Des* 2001, 22, 15.
4. Qiu, Y.; Lin, Z.; Mai, K. *J Appl Polym Sci* 2003, 88, 2148.
5. Liauw, C. M.; Lees, G. C.; Hurst, S. J.; Rotheron, R. N.; Ali, S. *Composites* 1998, 29A, 1313.
6. Wang, Z. Z.; Qu, B. J.; Fan, W. C. *Polym Degrad Stab J* 2002, 76, 123.
7. Yang, R.; Liu, Y.; Wang, K.; Yu, J. *J Anal Appl Pyrolysis* 2003, 70, 413.
8. Heng, L. *J Liaoning Normal Univ* 2002, 25, 223.
9. Xiang, S.; Zang, K.; An, Y.; et.al. *Plast Addit J* 2004, 4, 21.
10. Gaoxiang, D.; Shuilin, Z.; Yang, L. *China Plast J* 2004, 18, 69.
11. Gaoxiang, D.; Shuilin, Z.; Yang, L. *J Chin Ceram Soc* 2005, 33, 659.
12. Ma, Z. L.; Wang, J. H.; Ma, C. Y.; Shi, H. *J Heibei Univ (National Science Edition)*, to appear.
13. Wang, Z. Z.; Qu, B. J.; Fan, W. C.; Li, Z. *J Funct Polym* 2001, 14, 45.
14. Wang, D.; Gao, J. *J Appl Polym Sci* 2006, 99, 670.