Effects of the Content of Silane Coupling Agent KH-560 on the Properties of LLDPE/Magnesium Hydroxide Composites

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ABSTRACT: Based on traditional idea of molecular coupling effect, surface modifiers were helpful to improve the performances of inorganic/organic composites. However, it was also widely accepted that the content of surface modifier should be controlled in a suitable range, and more or less modifiers could not reach to optimal properties. The intrinsic reason for this phenomenon was not clear until now. In this article, the influences of the content of surface modifier: silane coupling agent KH-560, on the final performances of linear low-density polyethylene (LLDPE)/magnesium hydroxide (MH) composites, have been studied. The performance tests of LLDPE/MH composites, including mechanical properties measurements, thermal oxidative stability analysis, and surface morphology observation, all confirmed that there was an optimal content range of surface modifier. However, further morphology investigation of the modified MH by particle size and particle size distribution analyzer showed that, the particle size and particle size distribution of MH both increased as the content of KH-560 increased. ATR-FTIR analysis proved that silane grafting was achieved on the MH particle surface, which not only improved the compatibility between MH and LLDPE for molecular coupling effect, but also caused bigger particle size and wider particle size distribution, which were disadvantageous to improve the performances of the composites. The two opposite effects of the surface modifiers mentioned above affected the final performances of the composites. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 118: 2634–2641, 2010

Key words: silane coupling agent; LLDPE/MH composites; flame retardancy; compatibility; morphology

INTRODUCTION

Flame retardant polymer materials were of great importance to save time for survival in case of fire accident, because these materials had characteristics of difficult ignition, slow combustion flame propagation, low-quality of losses, and heat-release rate.[1–5] Compared with halogen-based flame retardants, halogen-free flame retardants (HFFR), such as metal hydroxides, have been paid much more attention recently, as they were environment-friendly and unharmful materials.[1–7] Magnesium hydroxide (MH) and aluminum trihydroxide (ATH) as HFFR were kept growing demands for their effective balance in cost versus performance. Further, MH had a higher endothermic decomposition temperature around 340°C, whereas that of ATH was quite low and usually in the range of polymer processing temperature, so MH was more promising as flame retardant than ATH.[8–12]

However, in the actual application of MH as flame retardants, there were still some problems. The biggest one was probable great reduction of mechanical properties for the poor compatibility between inorganic and organic materials after loading high-content of MH in the polymer matrix,[1–7] as more than 60 wt % MH filled in the composites was generally required for guaranteeing acceptable combustion resistance.[13] It was well-known that good dispersibility of inorganic particles in polymer matrix was determinant for satisfactory mechanical properties of the composites. Furthermore, the inorganic particles with smaller size and more uniform size distribution were helpful to improve the performances of composites.[6,14,15] But self-aggregation would always counteract particles dispersing into the polymer matrix homogenously.

To improve the compatibility between inorganic fillers and polymer materials, surface modifiers have been applied including macromolecular surface modifiers[16–18] and small-molecular-weight ones, such as surface active agents, silane coupling agents, and titanate coupling agents.[13,19–28] Compared with macromolecular surface modifiers, small-molecular-weight ones had characteristics of low-price and facile processing, so they have been applied more popularly. As for the polyolefin (PO)/MH systems, researchers have tried many different kinds of small-molecular-weight surface modifiers to the composites

including stearate,[19,20] oleate,[21,22] and silane coupling agents,[13,23–27] and found that silane coupling agents were quite efficient to improve the compatibility of polyolefin/MH composites for the active hydroxyl groups of MH.[13,23–27] Based on various functional groups on the hydrophobic chain, different kinds of silane coupling agents had been tried.[13,23–27] Ma et al.[23] did a systematic study on the effects of KH-550 (γ-aminopropyl triethoxysilane) on PP/brucite composites, and found that some chemical reactions took place between KH-550 and brucite, according to the experimental results of flow test and IR, which enhanced the interfacial adhesion of matrix-filler and improved the compatibility of PP/brucite. They pointed out that, the optimal content of KH-550 in this system was about 2.9%. Chen et al.[13] tried silane coupling agent KH-560 (γ-(2,3-epoxypropoxy)propyl trimethoxysilane) and dimethyl silicone oil to PP/MH composites, respectively, and also investigated the effects of the content of surface modifiers on the final properties of the composites. Yang et al.[24] studied that silane coupling agent KH-570 (γ-methacryloxypropyl trimethoxysilane) could not only make fillers achieve better dispersion in polymer matrix but also improve the roughness of the composites with good rigidity at the same time.

As mentioned above, surface modifiers could increase the compatibility between inorganic fillers and polymer matrix, and improve the final performances of the composites efficiently. Furthermore, only a proper content of surface modifier applied could reach to the optimal properties of composites, whereas more or less surface modifiers was detrimental to final properties. However, the intrinsic reasons for this effect have not been made clear yet. In this article, silane coupling agent KH-560 was chosen as surface modifier to MH filled in linear low-density polyethylene (LLDPE), which could be applied widely in housing, transportation, electrical engineering, and many other fields.[1–7] The effects of the content of KH-560 on the final mechanical properties and thermal oxidative stability of LLDPE/MH composites were studied, respectively. Furthermore, the morphologies of modified MH, with different content of KH-560, were also investigated because the initial morphologies of inorganic fillers, before mixing into polymer matrix, could influence the structures and morphologies of composites.[6,14,15] The intrinsic reasons for the effects of the content of surface modifier on the performances of composites have been discussed in detail.

EXPERIMENTS

Materials

LLDPE (type DFDA-7042, $d = 0.92$ g/cm^3) was supplied by Sinopec Yangzi Petrochemical (Nanjing,

China). MH (BET surface area is 6.79 m^2/g) was supplied by Chuangye (Yixing, China). Silane coupling agent KH-560 was supplied by Xiangzhifei Fine Chemical (Nanjing, China).

Stearic acid (A.R.) was supplied by Huakang Technology (Nanjing, China). Zinc borate was supplied by Wenhua Flame retardant materials (Wenzhou, China) as synergists, and the chemical formula for this grade is $2ZnO \cdot 3B_2O_3 \cdot 3.5H_2O$.

Preparation of LLDPE/MH samples

First, the MH powder, KH-560, and other synergic agents, including stearic acid (1 wt % in MH) for better processing ability[1,19,29] and zinc borate (5 wt % in MH) as synergist,[1,8,30] were mixed in a high-speed mixer (Type FW100; Taisite Instrument, Tianjin, China) for 20 min. The weight percentage of KH-560 in MH were 0, 1, 3, 5, 7, and 10%, respectively for different samples. The modified MH samples were marked as MK. Then, the LLDPE/MH composites were prepared by melt-blending on a twin-roll open mill (Type XK-160; New Jinling Rubber Machinery; Nanjing, China) at ~ 125°C for 5 min. After LLDPE melted, the MK was added into the mill. The ratio (w/w) of LLDPE/MK was 35 : 65, which was commonly used in industrial production. At last, the composites with standard size for further test were prepared in a lamination of PVC plastic press tester (Type GT-7014-P; GOTECH Testing Machine; Dongguan, China), and the mold of 100 mm × 100 mm × 2 mm was used for each sample. The detailed preparing process was given below: the composites were compressed in heat press for 5 min at 500 psi and 150°C at the first step; then in heat press for another 5 min at 1100 psi and 150°C; finally in cold press at 300 psi until the samples reached room temperature.

Mechanical properties measurements

The testing specimens were stamped with an ASTM D1078-06 standard die. The mechanical properties (tensile strength and elongation at break) were measured at a tensile speed of 50 mm/min at 12°C under 42% humidity by a universal material tester (Type Instron-4466; Instron Test Machine Trading, Shanghai, China). Six specimens were tested for each sample.

Thermal analysis

Thermogravimetric analysis of composites (about 10 mg) were carried out under air atmosphere purge (30 mL/min) from 20 to 600°C at a scanning rate of 10°C/min by thermal analyze instrument (Type STA-449C; NETZSCH Instrument, Selb, Germany).

The temperature error and mass error of TGA is less than 1°C and 0.1 mg, respectively.

Scanning electron microscope (SEM)

The composites were broken after cooling below their glass transition temperature by liquid nitrogen for keeping the original morphologies of composites. After being sputter coated with gold, the morphologies of the fracture surfaces was observed directly with a SEM (Type SSX-550; Shimadzu; Kyoto, Japan). The electron micrographs were taken with an acceleration voltage of 25.0 kV.

Attenuated total reflectance/fourier transform infrared spectroscopy (ATR/FTIR)

The modified MH for ATR/FTIR test was prepared according to the MH/KH-560 mass ratio (w/w) 1 : 1. The prepared sample was packed with filter paper and extracted by acetone in Soxhlet apparatus for 60 h to wash off unreacted KH-560. After the sample was dried, ATR-FTIR spectra was recorded using FTIR spectrometer (Type vector-22; Bruker; Karlsruhe, Germany).

Particle size and particle size distribution analysis

Particle size and particle size distributions of modified MH samples marked as MK were analyzed by particle size and particle size distribution analyzer (Type MasterSizer 2000; Malvern Instruments, Worcestershire, UK). The statistical analysis for the particle size and particle size distribution of MH particles was assumed a spherical particle structure. The volume weighted mean diameter (D_v) and surface area weighted mean diameter (D_a) were provided directly by data processing software from manufacturer, and they were calculated, respectively according to the eqs. (1) and (2) as shown below:

$$D_v = \frac{\sum D^4}{\sum D^3} \qquad (1)$$

$$D_a = \frac{\sum D^3}{\sum D^2} \qquad (2)$$

Here D in the equations was the diameter of equivalent sphere. The index of particle size distribution (d) was defined and calculated according to the eq. (3) as shown below:

$$d = \frac{D_v}{D_a} \qquad (3)$$

RESULTS AND DISCUSSION

Mechanical properties

The mechanical properties of the varied composites with different contents of KH-560, including tensile

TABLE I
Mechanical Properties of LLDPE/MH Composites with Different Content of KH-560[a]

Content of KH-560 (%)	Tensile strength (MPa)	Elongation at break (%)
0	8.85 ± 0.65	3.32 ± 0.74
1	7.22 ± 0.31	5.01 ± 0.54
3	8.71 ± 0.22	4.52 ± 0.66
5	11.63 ± 0.64	21.61 ± 0.80
7	11.89 ± 0.53	20.15 ± 0.89
10	10.21 ± 0.80	16.17 ± 0.81

[a] The tensile strength and elongation at break of neat LLDPE are 25.63 MPa and 1297.22%, respectively.

strength and elongation at break, were investigated, respectively. The final results were listed in Table I. From Table I, it was found that, after MH was filled into LLDPE, the tensile strength and the elongation at break of composites both decreased greatly, compared with neat LLDPE resin. This was mainly because of the high-content loading of MH particles that greatly deteriorated the mechanical properties of the composites for poor compatibility between LLDPE and MH and resulted in the decrease of tenacity.

However, after KH-560 was applied, the elongation at break of composites improved. Simultaneously, the tensile strength of composites was also enhanced. The improved mechanical properties implied that surface modifier enhanced the compatibility between the MH and LLDPE efficiently.

Furthermore, ATR-FTIR measurement was applied to investigate the molecular coupling effect[7,19,23,24,26,27,31] of KH-560 for the enhancing mechanism. Figure 1 gave the ATR-FTIR spectra of unmodified MH, KH-560, and modified MH samples, respectively. It was found in Figure 1(a) that there was a sharp and intense peak at 3689 cm^{-1}, which was attributed to the O—H vibration of MH. The ATR-FTIR spectrum of KH-560, as shown in Figure 1(b), represented the characteristic peaks of methylene (>CH$_2$, 2941 cm^{-1} and 2840 cm^{-1}) and Si—O (1190 cm^{-1}), respectively. The spectrum of modified MH showed intense characteristic peaks of KH-560 in Figure 1(c) proved that KH-560 grafted on MH surface successfully. Therefore, the organic segment of the silane coupling agent on MH particle surface, entwisted with polymer segments, which would increase the adhesion between inorganic fillers and polymers.

In addition, based on Table I, it was found that the LLDPE/MH composites modified by 5–7% KH-560, showed better mechanical properties among all measured composites, which was because of good compatibility and dispersibility of MH in polymer matrix. However, more or less KH-560 applied was
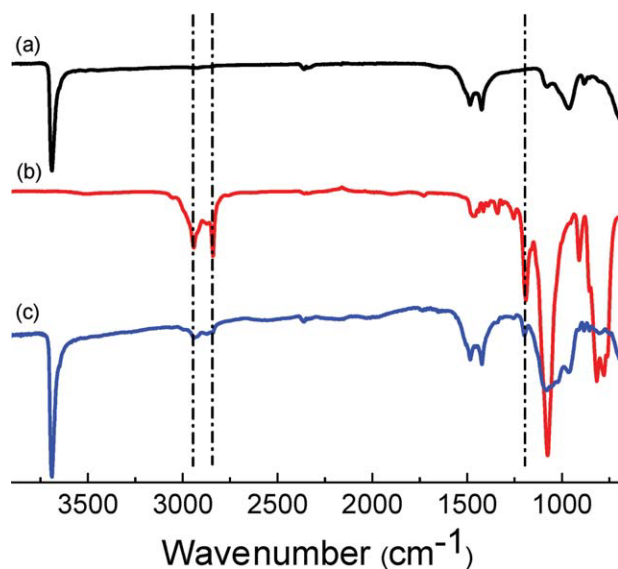
Figure 1 ATR-FTIR spectra of (a) MH, (b) KH-560, and (c) MH modified by KH-560. [Color figure can be viewed in the online issue, which is available at www.interscience. wiley.com.]

Figure 2 TG curves of LLDPE and LLDPE/MH composites with different content of KH-560 under air flow [the contents of KH-560 in MH are (a) 0%, (b) 1%, (c) 3%, (d) 5%, (e) 7%, and (f) 10%, respectively.] (Insert Figure: TG curves of neat LLDPE: ……, LLDPE/MH composites without KH560: ——). [Color figure can be viewed in the online issue, which is available at www. interscience. wiley.com.]

useless to improve the mechanical properties of composites.

Moreover, considering a single modifier molecular layer covering for the effective treatment of filler at least, the theoretic optimal content of KH-560 could be estimated according to eq. (4) primarily as shown below:

$$C_{\text{theory}} = \frac{A \cdot M_{\text{KH-560}}}{\pi R_O^2 \cdot N_A} \times 100\% \qquad (4)$$

Here A was the BET surface area of MH particles; $M_{\text{KH-560}}$ was the molecular weight of KH-560; R_O was the van der Waals radius of oxygen atom around 140 pm; and N_A was Avogadro constant. The calculated result was about 4.3 wt %, which was quite near to the experimental results as mentioned above.

Thermogravimetric analysis

Then, the further thermo-analysis for these LLDPE/ MH composites was carried out. TGA and DTG curves of LLDPE and LLDPE/MH composites with different contents of KH-560 measured in air atmosphere were shown in Figure 2 and 3, respectively.

The peak temperatures in DTG curves of different composites shown in Figure 3 were summarized in Figure 4. It was found that the neat LLDPE started to decompose at about 240°C, and two degradation peaks appeared in its DTG curve were corresponding to the LLDPE samples with low-molecular weight and high-molecular weight, respectively, which underwent completely thermal oxidative degradation at temperature up to about 530°C without any residue. However, compared with neat LLDPE,
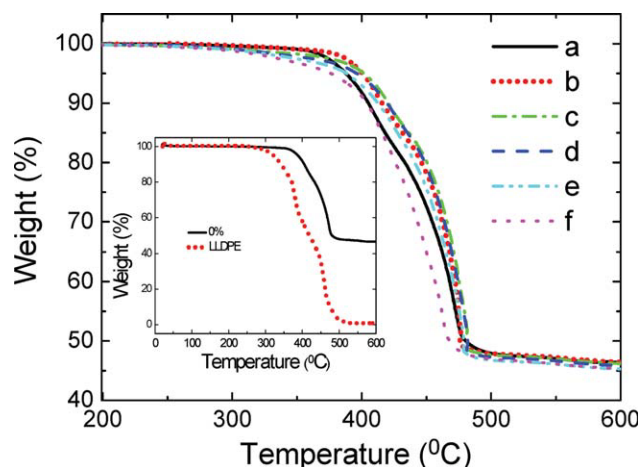
all the LLDPE/MH composites almost showed no mass loss below 350°C. Furthermore, only one degradation peak appeared in their DTG curves, which indicated that MH could effectively improve thermal oxidative stability of composites.

In addition, it could be found from Figure 4 that, except the composites modified by 10% of KH-560, the peak temperatures of thermal oxidative degradation of those composites were higher than that of composite without KH-560, which was ascribed to improvement of the compatibility between MH and
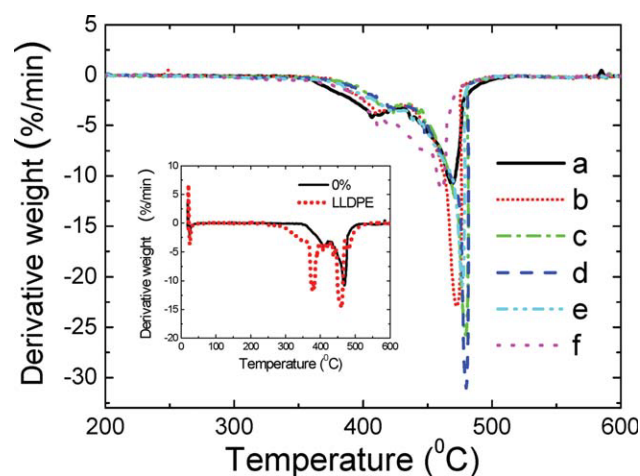

Figure 3 DTG curves of LLDPE and LLDPE/MH composites with different content of KH-560 under air flow. [the contents of KH-560 in MH are (a) 0%, (b) 1%, (c) 3%, (d) 5%, (e) 7%, and (f) 10%, respectively.] (Insert Figure: TG curves of neat LLDPE: ……, LLDPE/MH composites without KH560: ——). [Color figure can be viewed in the online issue, which is available at www.interscience. wiley.com.]
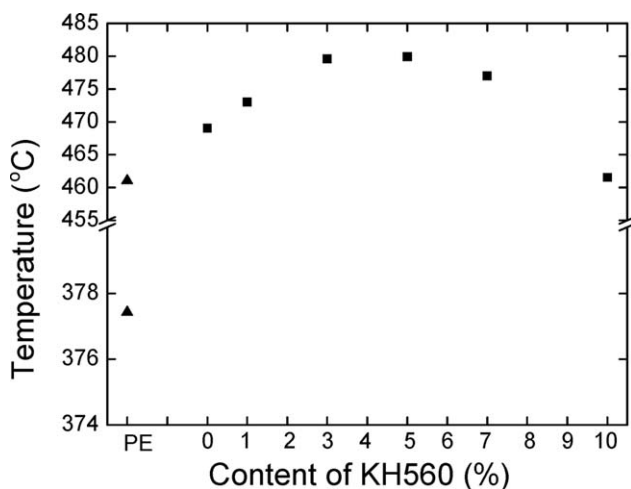
Figure 4 Peak temperatures in DTG curves of LLDPE and LLDPE/MH composites with different content of KH-560.

Figure 5 SEM of fracture surface of neat LLDPE under lower magnification (×500).

LLDPE, since better compatibility would result in higher thermal oxidative stability. Furthermore, as well as the mechanical properties, the thermal oxidative stabilities of composites also showed optimal values only occurred in a proper content range of KH-560 around 3–7% according to Figure 4.

Morphology analysis: SEM measurement

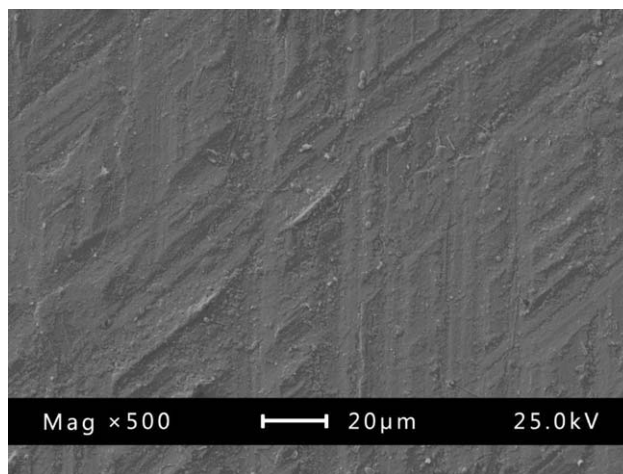It was well-known that, the most important factor affecting the final performances of the composites was the morphology of composites. Therefore, the effects of the content of KH-560 on the morphologies of composites were also investigated by direct observation using SEM. Figure 5 showed the SEM image of neat LLDPE, and a very flat and homogenous fracture surface could be observed.

Figure 6 showed the lower magnification (×500) SEM images of varied LLDPE/MH composites with different contents of KH-560. It was found from Figure 6(a) that, the MH particles without KH-560 were not able to disperse into LLDPE matrix well for the poor compatibility. Compared with the surface of
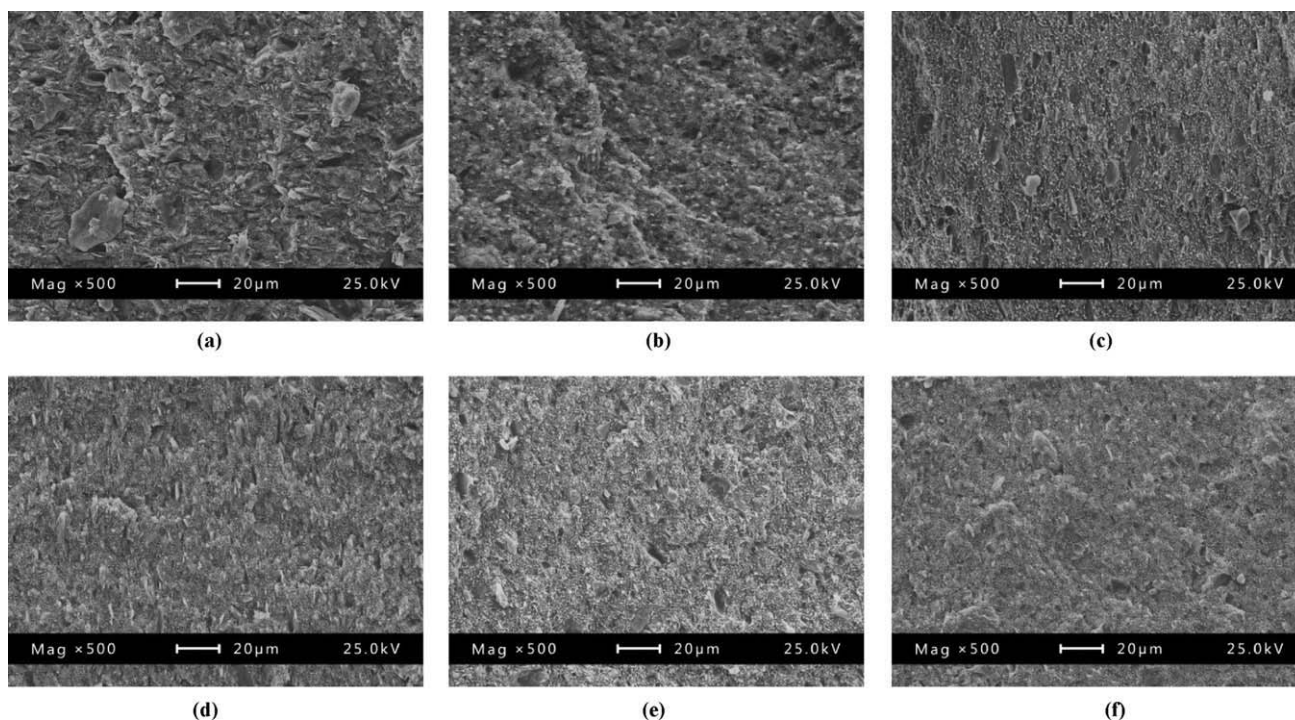

Figure 6 Scanning electron micrographs of fracture surface of LLDPE/MH composites under lower magnification (×500) [the contents of KH-560 in MH are (a) 0%, (b) 1%, (c) 3%, (d) 5%, (e) 7%, and (f) 10%, respectively].
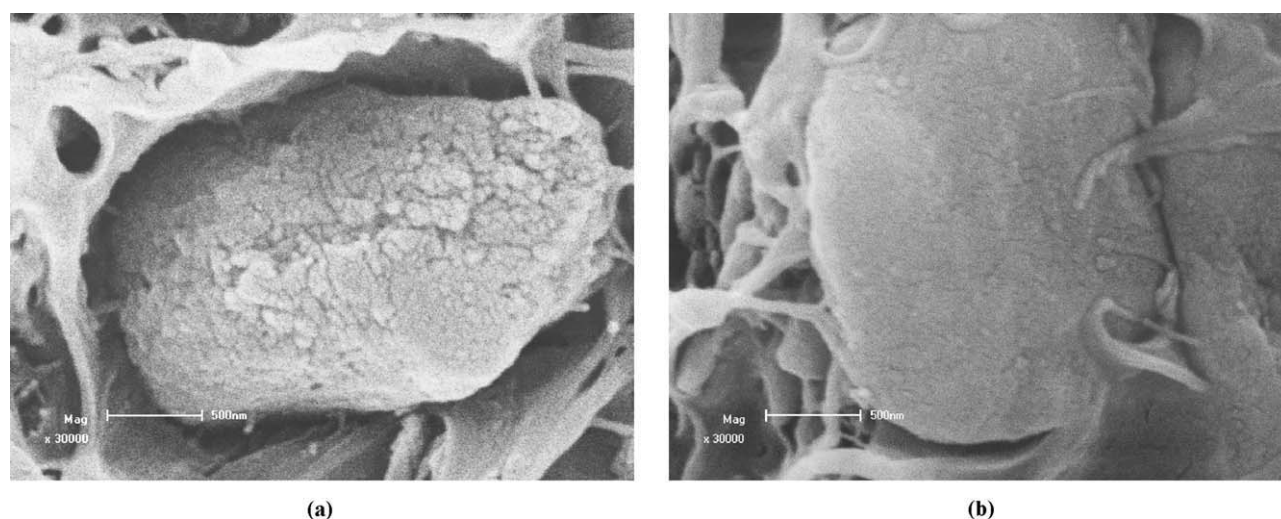
Figure 7 Scanning electron micrographs of fracture surface of LLDPE/MH composites modified, (a) without KH-560 and (b) with 5% of KH-560, under higher magnification (×30000).

neat LLDPE as shown in Figure 5, the interface between MH and LLDPE could be observed clearly. Furthermore, many debonded MH particles and the accidented surface containing some holes have been observed in Figure 6(a), which all indicated bad adhesion and poor compatibility between MH fillers and polymers. The inhomogenous morphology of composites was the intrinsic reason for lower performances of materials. However, compared with the surface of the composite without KH-560, those of the composites, modified by KH-560 as shown in Figure 6(b–f), had fewer holes and looked more homogenous wholly, and no clear interface between MH and polymer could be found also.

Furthermore, the surface morphologies indicated that composites modified by 3–5% KH-560 obviously had better dispersibility of MH in polymer matrix, as shown in Figure 6(c,d). It was coincident with the results from the mechanical properties measurement and thermal oxidative stability analysis, as mentioned above. The suitable content of surface modifier would result in the homogenous morphology of composites and the final performances.

For further investigation of 5% KH-560 additive on the morphology of composite, higher magnification (×30000) SEM images of fracture surface of LLDPE/MH composites treated without and with 5% KH-560

were both shown in Figure 7. Evidently it was found that, the surface of MH particle without KH-560 was very rough, and there was large gap between LLDPE and MH because of poor compatibility. However, for the composite modified by KH-560, as shown in Figure 7(b), the surfaces of MH particles became smooth and had some branch-like binding LLDPE closely around for good adhesion and compatibility between MH and LLDPE, which was because of molecular coupling effect of KH-560.

Summary of various performance tests data

Above all, the optimal content ranges of KH-560 based on various performance tests were summarized in Table II.

TABLE II
Summary of the Modifier's Optimal Content Ranges Based on Various Performance Tests

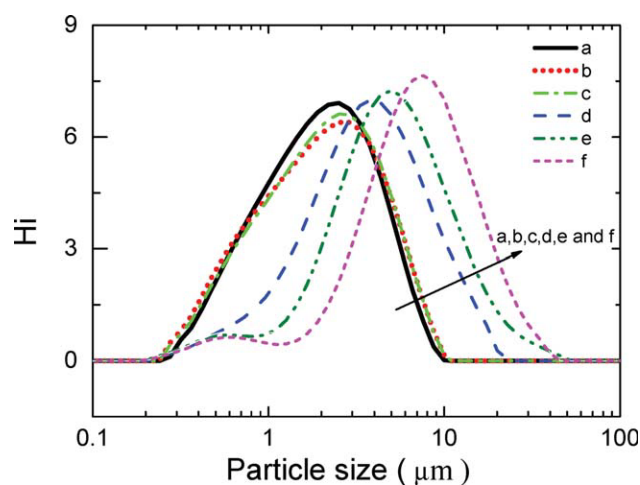| Performance tests of the composites	Mechanical properties	Thermal stability	Morphology of fracture surfaces
Optimal content ranges of modifier (wt % in MH)	5–7	3–7	3–5

Figure 8 Particle size distribution curves of varied MH samples [the contents of KH-560 in MH are (a) 0%, (b) 1%, (c) 3%, (d) 5%, (e) 7%, and (f) 10%, respectively]. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

TABLE III
Statistical Analysis Results of Particle Size and Particle Size Distribution of Different MH Samples

MH/KH-560 (w/w)	100/0	100/1	100/3	100/5	100/7	100/10
Diameter at 10% (μm)	0.71	0.57	0.60	1.07	1.53	2.25
Diameter at 50% (μm)	2.45	1.86	1.91	3.39	4.58	6.63
Diameter at 90% (μm)	6.92	4.76	4.77	8.91	12.28	16.05
D_v (μm)	2.21	2.32	2.35	4.32	6.10	8.13
D_a (μm)	1.31	1.26	1.30	2.18	2.72	3.52
d	1.68	1.84	1.81	1.98	2.22	2.31

It could be inferred from Table II that the suitable range of KH-560 for LLDPE/MH composites should be around 3–7 wt % in MH. And higher content of modifiers applied could not reach to better performances of composites. Therefore, it was believed that, besides molecular coupling effect, there must be another effect of surface modifiers on the composites.

Particle size and particle size distribution analysis

Based on the experimental results above, it could be concluded that, only a proper amount of surface modifier, neither more nor less, would give the composites with satisfactory properties. To find the intrinsic reasons for this phenomenon, the effects of the content of KH-560 on MH were investigated further.

The morphologies of the varied MH samples with different contents of KH-560 were measured by particle size and particle size distribution analyzer. The curves of particle size distribution for various modified MH were shown in Figure 8. Interestingly, it could be found that the peaks of the curves moved to larger scale with the increase of the content of KH-560. Further statistical analysis results of the particle size and particle size distribution were summarized in Table III. Here, value d based on eq. (3) was applied to express the particle size distribution, and bigger value d referred to wider distribution of particles. Based on Table III, it was found that the more KH-560 was used, the bigger particle size and the wider particle size distribution of MH were. On the one hand, the grafting of silane coupling agent on MH might result in the increase of particle size and particle size distribution; on the other hand, at higher content of KH-560, self-polymerization among surface modifiers would occur easily,[32] which induced MH particles congregating together to form complexes with bigger size facilely.

It was believed that, bigger particle size and wider particle size distribution of the inorganic fillers were deleterious to improve the dispersibility of inorganic particles into polymer matrix,[6,14,15,18,25] and deteriorated the properties of composites.

However, the final performances of composites depend on not only the initial morphologies of inorganic fillers but also the molecular coupling effect of modifiers as mentioned above. At lower content of KH-560, the molecular coupling effect was dominated, and the final performance of composites improved with the increase of KH-560; Whereas at higher content of KH-560, the initial morphologies of inorganic fillers was deteriorated further, and the bigger particle size and wider particle size distribution resulted in the reduction of both the mechanical properties and thermal oxidative stabilities of composites.

CONCLUSIONS

Based on the experimental facts as mentioned above, it was believed that, to prepare high-performance inorganic/organic composites, it was very important to control the content of surface modifier in a suitable range (in this article, 3–7 wt % of KH-560 in MH for LLDPE/MH composites). The intrinsic reasons for this phenomenon were ascribed to two opposite effects of surface modifiers: On the one hand, the surface modifiers would improve the adhesion and dispersion of inorganic fillers in polymer matrix and counteract self-aggregation of MH efficiently for molecular coupling effect; on the other hand, with the content of KH-560 increase, the particle size of MH turned bigger and its distribution became wider, which were both disadvantageous to improve the performances of the composites. Therefore, a proper content range of surface modifier was necessary to get satisfactory performances of the composites.

References

1. Weil, E. D.; Levchik, S. V. J Fire Sci 2008, 26, 5.
2. Hornsby, P. R. Int Mater Rev 2001, 46, 199.
3. Horrocks, A. R.; Price, D., Eds. Fire retardant materials, Woodhead Publishing Ltd: Cambridge, 2001, 264.
4. Cai, J.; Yang, Z.; Lv, D.; Zhou, D.; Chen, B. F.; Yang, H.; Guo, X. F.; Ding, W. P.; Cheng, R. S. Polym Bull 2009, 6, 22.
5. Ou, Y. X.; Han, T. J.; Li, J. J. Handbook of flame retardant plastic; National Defence Industry Press: Beijing, 2008, 16, 77.
6. Chen, T.; Lynch, T. J. Proc Conf Recent Adv Flame Retardancy Polym Mater 2004, 15, 228.
7. Kim, S. J Polym Sci Part B: Polym Phys 2003, 41, 936.
8. Zhang, Y. X., Ed. Manual of optimal design of flame retardants production formula and application of new materials, Yinsheng Audio-video Publishing House: Changchun, 2005, 287, 311.

9. Wang, Z. Z.; Hu, K. L.; Hu, Y.; Gui, Z. Polym Int 2003, 52, 1016.
10. Hornsby, P. R.; Watson, C. L. Plast Rubber Process Appl 1989, 11, 45.
11. Rothon, R. N.; Hornsby, P. R. Polym Degrad Stab 1996, 54, 383.
12. Wang, Z. Z.; Qu, B. J.; Fan, W. C.; Huang, P. J Appl Polym Sci 2001, 81, 206.
13. Chen, X. L.; Yu, J.; Guo, S. Y. J Appl Polym Sci 2006, 102, 4943.
14. Jia, X. W., Ed. Nano-sized flame retardant materials, Chemical Industry Press: Beijing, 2005, 46, 184.
15. Huang, H. H.; Tian, M.; Liu, L.; Liang, W. L.; Zhang, L. Q. J Appl Polym Sci 2006, 100, 4461.
16. Wang, Z. Z.; Chen, Z.; Fan, W. C.; Nie, W. Y. Polym Plast Technol Eng 2006, 45, 191.
17. Wang, Z. Z.; Qu, B. J.; Fan, W. C.; Hu, Y.; Shen, X. F. Polym Degrad Stab 2002, 76, 123.
18. Nachtigall, S. M. B.; Miotto, M.; Schneider, E. E., Mauler, R. S.; Forte, M. M. C. Eur Polym J 2006, 42, 990.
19. Huang, H. H.; Tian, M.; Yang, J.; Li, H. X.; Liang, W. L.; Zhang, L. Q.; Li, X. L. J Appl Polym Sci 2008, 107, 3325.
20. Liu, L. H.; Song, Y. H.; Chen, J. M.; Guo, F. J Beijing Univ Chem Technol 2004, 31, 31.
21. Liauw, C. M.; Dumitru, P.; Lees, G. C.; Clemens, M. L.; Rothon, R. N. Macromol Mater Eng 2003, 288, 412.
22. Zhao, L. M.; Zhao, J. H.; Zhang, Q.; Chen, S. Y. Fire Sci Technol 2007, 26, 80.
23. Ma, Z. L.; Wang, J. H.; Zhang, X. Y. J Appl Polym Sci 2008, 107, 1000.
24. Yang, R.; Liu, Y.; Wang, K. H.; Yu, J. J Anal Appl Pyrolysis 2003, 70, 413.
25. Zhang, L.; Li, C. Z.; Zhou, Q. L.; Shao, W. J Mater Sci 2007, 42, 4227.
26. Wu, W.; Chen, J. F.; Qu, Y. X. J Chin Ceram Soc 2004, 32, 570.
27. Schofield, W. C. E.; Hurst, S. J.; Lees, G. C.; Liauw, C. M.; Rothon, R. N. Compos Interfaces 1998, 5, 515.
28. Lu, K. L.; Qiu, G. X.; Liu, X. M.; Pan, T. X. Mod Plast Process Appl 2005, 17, 20.
29. Zweifel, H. Plastic additives handbook; 5th ed.; Carl Hanser Verlag: Munich, 2001, 405.
30. Genovese, A.; Shanks, R. A. Polym Degrad Stab 2007, 92, 2.
31. Uotila, R.; Hippi, U.; Paavola, S.; Seppala, J. Polymer 2005, 46, 7923.
32. Plueddemann, E. P., Ed; Liang F. S.; Xie, S. J., Translators. Silane and titanate coupling agents, Shanghai Science and Technology Literature Press: Shanghai, 1987, 39.