

# Performance Elevation of Linear Low-Density Polyethylene Highly Loaded with Aluminum Hydroxide by the Grafting of Methacrylic Acid to the Matrix

Zhan-Pai Su, Ping-Kai Jiang, Ping Wei, Gen-Ling Wang, Yong Zhang

School of Chemistry and Chemical Engineering, Shanghai Jiaotong University, Shanghai 200240, People's Republic of China

Received 4 December 2003; accepted 26 July 2004

DOI 10.1002/app.21350

Published online 27 January 2005 in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** The mechanical and electric performances of linear low-density polyethylene (LLDPE) highly loaded with aluminum hydroxide ( $\text{Al}(\text{OH})_3$ ) by the grafting of methacrylic acid (MAA) to the LLDPE matrix were studied. The results of Fourier transform infrared spectroscopy showed that the grafting reaction occurred by melt grafting. Mechanical testing of composites of LLDPE highly loaded with  $\text{Al}(\text{OH})_3$  showed that the strength and elongation at break were significantly improved after the grafting of MAA to the LLDPE matrix. The results of the electric tests showed

similar trends. The results of scanning electron microscopy showed better decentralization of  $\text{Al}(\text{OH})_3$  loaded in the LLDPE matrix in the tensile fractured surface and a transition layer between  $\text{Al}(\text{OH})_3$  and the LLDPE matrix in the fractured surface after the grafting of MAA to the LLDPE matrix. © 2005 Wiley Periodicals, Inc. *J Appl Polym Sci* 96: 162–168, 2005

**Key words:** flame retardance; graft copolymers; morphology; polyethylene (PE)

## INTRODUCTION

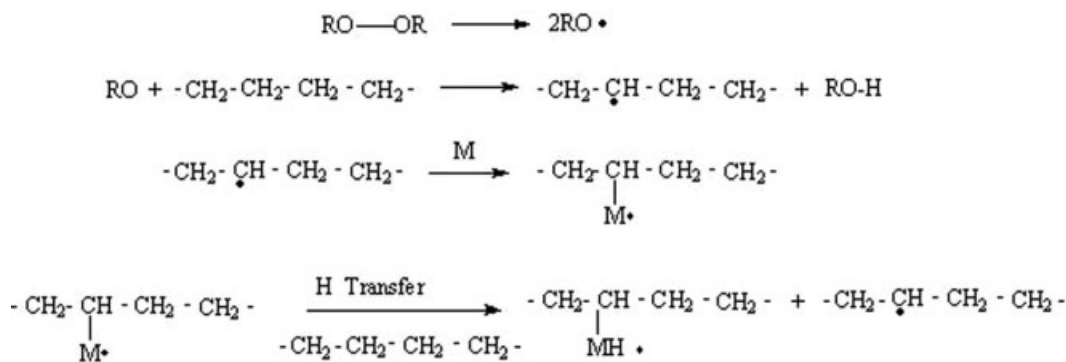
Polyethylene has long been known, because of its excellent dielectric and mechanical properties, for its use as insulation in power cable construction and other applications.<sup>1–3</sup> It has the advantages of excellent electric properties, excellent resistance to cold flow, ease of processing, adequate mechanical properties, and excellent value (cost and performance). The main drawback of PE is that it is a combustible material, and its limited oxygen index (LOI) is about 17.4; this limits its applications to some extent.  $\text{Al}(\text{OH})_3$  is an effective and environmentally friendly flame retardant that is acid- and halogen-free.<sup>4–6</sup>

Unfortunately, the high loading of  $\text{Al}(\text{OH})_3$  in PE leads to a reduction of the impact strength and elongation at break because of the poor interfacial adhesion between the two components.<sup>2,4,7–9</sup> In addition, aggregation induces the enhancement of permittivity ( $\epsilon$ ) when the particles are closely packed because of the intense polarity of  $\text{Al}(\text{OH})_3$  particles.<sup>10–12</sup> The interfacial behavior between the inorganic compounds and polymer matrices is recognized as a key factor influencing the mechanical and electric properties of composites. The mostly commonly used methods for

improving the interfacial adhesion of matrices and fillers are processing fillers with coupling agents or adding a compatibilizer. The traditional coupling agents used for PE/inorganic filler composites are silane, titanate, and functionalized polyolefins with unsaturated monomers, such as maleic anhydride, glycidyl methacrylate, acrylic acid, and its derivatives.<sup>3,13–15</sup> The drawbacks of these methods are insignificant effects at low concentrations or a deterioration of mechanical and electric properties, especially the strength at break and breakdown strength, at high concentrations. On the other hand, it is difficult to add a coupling agent to the composites. Therefore, choosing suitable matrix materials or changing the properties of the matrix is often done to improve the interfacial behavior between inorganic compounds and polymer materials.

In this study, methacrylic acid (MAA) was grafted to a linear low-density polyethylene (LLDPE) matrix by melt grafting to change its compatibility with  $\text{Al}(\text{OH})_3$  particles. The results of Fourier transform infrared (FTIR) showed that the grafting reacting was carried out. The mechanical and electric properties and the microstructures of the composites were studied. The relationship between the macroproperties and microstructures of the composites was also analyzed. Moreover, because LLDPE is an important insulator material, the relationship between the electric properties and morphology of LLDPE-filled with  $\text{Al}(\text{OH})_3$  particles was also studied.

Correspondence to: Z.-P. Su (pingwei@sjtu.edu.cn).



Notes: RO-OR:DCP; —CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>- :LLDPE; M: MAA

Figure 1 Reaction mechanism of MAA grafting onto LLDPE.

EXPERIMENTAL

Materials

The polyethylene used here was LLDPE supplied by Saudi Arabian Basic Industries (Riyadh, Saudi Arabia) with a melt index (GB 3682-83) of 2 g/10 min and a density of 0.918 g/cm<sup>3</sup>. Al(OH)<sub>3</sub> powder with a mean particle size of 1.2 μm and a nitrogen surface area of 6 m<sup>2</sup>/g (Brunauer-Emmett-Teller method) was provided by Sou Le Enterprise Co., Ltd. (Hong Kong). MAA was supplied by Shanghai Chemical Reagent Co. (Shanghai, China). Dicumyl peroxide (DCP) with a half-life of 1 min at 170°C was provided by the Institute of Chemistry and Engineering (Harbin, China).

Sample preparation

First, DCP, MAA, and the third additive were dissolved in acetone. Then, LLDPE was blended with the

solution. The third additive was dimethylformamide, which was used to suppress the crosslinking of LLDPE. A certain amount of LLDPE, MAA, and DCP in a mixture (LLDPE/MAA/DCP = 100:1.95:0.02 w/w/w) was fed into a Haake Rheocord 90 internal mixer (Karlsruhe, Germany) for melt grafting at 170°C and 80 rpm for 5 min. Some of the sample was taken out for the analysis of the grafting degree (GD) after melt grafting, and then Al(OH)<sub>3</sub> was added. The samples were obtained when the torque remained constant.

Characterization

FTIR

First, the obtained samples were pressed into films 100 μm thick. Next, the films were dipped into distilled water for 24 h to remove the nonreactive MAA, and this was followed by Soxhlet extraction with ethanol for 24 h to remove any adhering homopolymer. Fi-

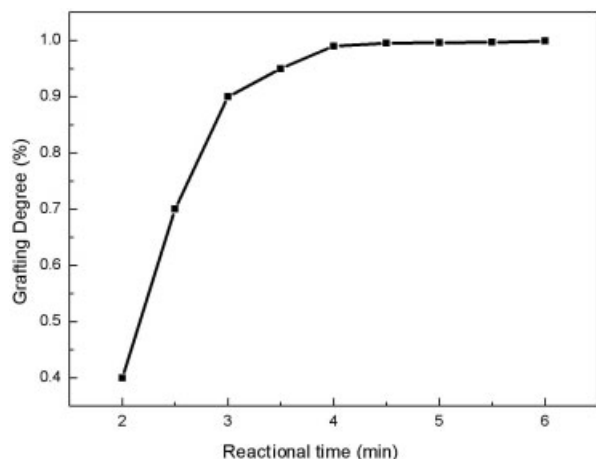


Figure 2 GD of MAA-g-LLDPE versus the reaction time in a Haake mixer at 170°C and 80 rpm with an LLDPE/MAA/DCP weight ratio of 100:1.95:0.02.

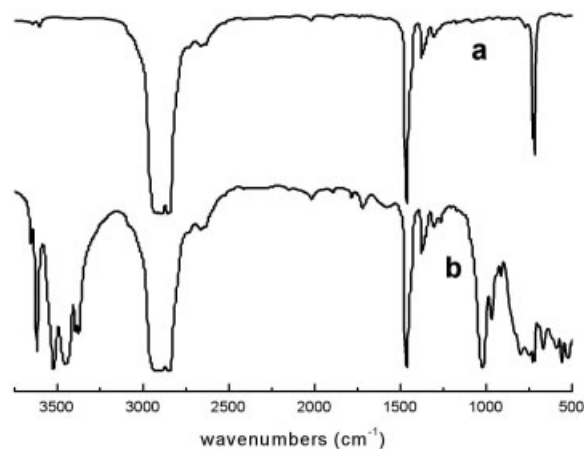
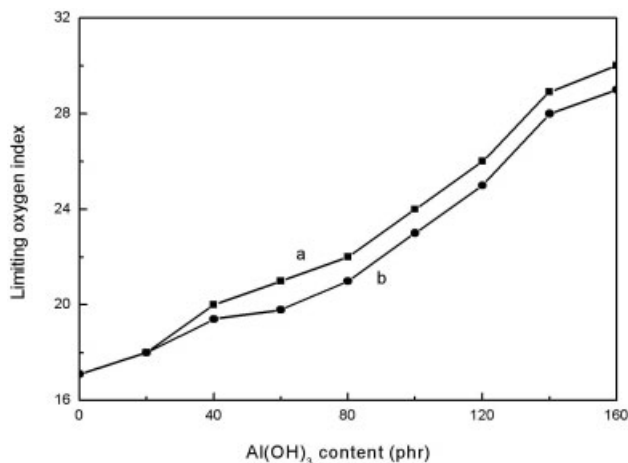


Figure 3 FTIR spectra of (a) neat LLDPE and (b) MAA-g-LLDPE/Al(OH)<sub>3</sub> (100:20) composites.



**Figure 4** LOI of composites of polyethylene and polyethylene filled with various amounts of  $\text{Al}(\text{OH})_3$  samples: (a) LLDPE/ $\text{Al}(\text{OH})_3$  composites and (b) MAA-g-LLDPE/ $\text{Al}(\text{OH})_3$  composites.

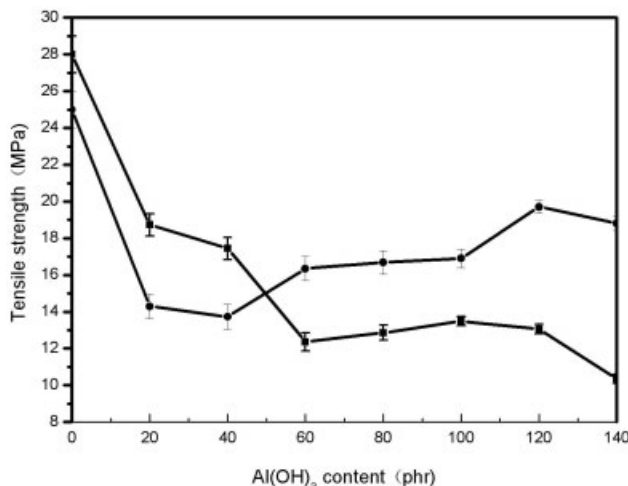
nally, samples were dried in an oven *in vacuo* at  $80^\circ\text{C}$  for 24 h before the testing. Infrared transmission spectra were obtained with a Paragon 1000 FTIR spectrometer from PerkinElmer Corp. (Massachusetts, USA) with a resolution of  $4\text{ cm}^{-1}$ .

#### GD

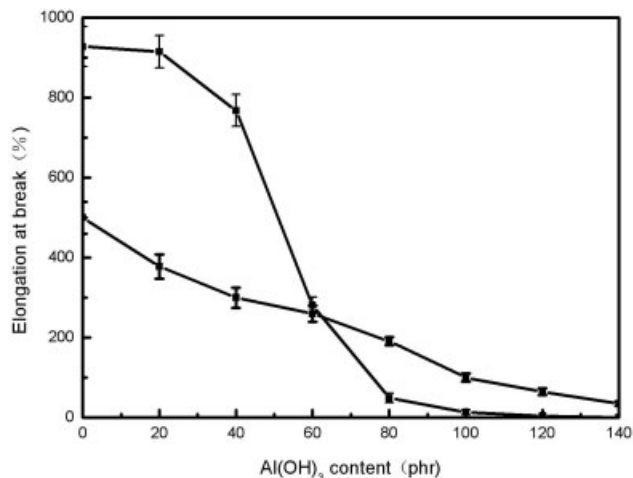
GD was determined by a titration procedure. About 1 g of the purified product was dissolved in 100 mL of hot toluene, and then this was titrated with 0.1 mol/L NaOH in methanol. Phenolphthalein was used as an indicator.

#### Mechanical properties

The mechanical properties were measured on an Instron 4302 tensile tester (Massachusetts, USA) according to Chinese Standard GB/T 1042-92.



**Figure 5** Tensile strength of LLDPE and MAA-g-LLDPE filled with various amounts of  $\text{Al}(\text{OH})_3$ : (a) LLDPE/ $\text{Al}(\text{OH})_3$  composites and (b) MAA-g-LLDPE/ $\text{Al}(\text{OH})_3$  composites.



**Figure 6** Elongation at break of LLDPE and MAA-g-LLDPE filled with various amounts of  $\text{Al}(\text{OH})_3$ : (a) LLDPE/ $\text{Al}(\text{OH})_3$  composites and (b) MAA-g-LLDPE/ $\text{Al}(\text{OH})_3$  composites.

#### Electric properties

The samples were made according to GB/T1042-92. The volume resistivity was measured on a ZC-36 megohmmeter (Shanghai Precision and Scientific Instrument Corp., Shanghai, China).  $\epsilon$  and the dissipation factor were measured on QS30 high-voltage bridge (Shanghai huguang Corp., Shanghai, China). The breakdown strength was measured on a AHDZ-10/100 alternating-current dielectric strength tester (Shanghai Ianpotronics Corp., Shanghai, China).

#### Morphology observations

The phase morphology of the composites was examined with scanning electron microscopy (SEM; model S520, Hitachi, Tokyo, Japan). Specimens were prepared by the immersion of test pieces in liquid nitrogen before they were broken. The fractured surfaces of the test pieces were coated with gold.

#### Flammability

The flammability behavior was characterized by LOI with a Stanton-Redcroft LOI FTA II instrument (Rheometric Scientific, Ltd., West Sussex, UK) according to ASTM Standard D 2863. LOI was defined as the minimum molar percentage of oxygen in a mixture of oxygen and nitrogen necessary to barely support flaming combustion of a material initially at room temperature under the conditions of the test method.

## RESULTS AND DISCUSSION

### Preparation and FTIR study of MAA-g-LLDPE

It is generally accepted that MAA is grafted onto polyolefins via a free-radical mechanism.<sup>16-18</sup> The

**TABLE I**  
**Values of Stress Concentration Parameters in LLDPE/Al(OH)<sub>3</sub> and MAA-g-LLDPE/Al(OH)<sub>3</sub> Composites**

Composite or polymer	Theoretical stress at break with <i>S</i> (MPa)	Volume fraction (%)	Stress at break (MPa)	<i>S</i>
LLDPE	28	0	28	1
LLDPE-60	20.16	28	12.36	0.61
MAA-g-LLDPE-60			16.35	0.81
LLDPE-80	18.48	34	12.86	0.69
MAA-g-LLDPE-80			16.68	0.90
LLDPE-100	16.80	40	13.49	0.80
MAA-g-LLDPE-100			16.9	1.01
LLDPE-120	15.68	44	13.06	0.83
MAA-g-LLDPE-120			19.71	1.26
LLDPE-140	14.56	48	10.35	0.71
MAA-g-LLDPE-140			18.82	1.29

mechanism of MAA grafting onto the LLDPE backbone in the presence of DCP is shown in Figure 1. Figure 2 shows the GD of MAA-g-LLDPE with the progress of the reaction in a Haake mixer at 170°C and 80 rpm with an LLDPE/MAA/DCP weight ratio of 100:1.95:0.02. The grafting reaction could exceed 99%, and the GD was kept at 1% within an error of 0.1%.

Figure 3 shows FTIR spectra of neat LLDPE and MAA-grafted LLDPE (MAA-g-LLDPE; the GD of MAA-g-LLDPE in this study was kept at 1 wt %) loaded with 20 phr Al(OH)<sub>3</sub> composites. The MAA grafting reactions were shown to occur because the peak at 1719.35 cm<sup>-1</sup> was assigned to the functional carbonyl group. Because nonreactive MAA and its adhering homopolymer were removed previously, the peak at 1719.35 cm<sup>-1</sup> showed that MAA grafting of LLDPE in the Haake instrument occurred.

### Flammability of the composites

The dependence of LOI of Al(OH)<sub>3</sub>-filled LLDPE samples on the Al(OH)<sub>3</sub> concentration is shown in Figure 4. Similarly to results reported previously,<sup>4-6</sup> LOI of LLDPE filled with Al(OH)<sub>3</sub> improved with an increase in the filler concentration, and LOI of neat LLDPE and LLDPE filled with 140 phr Al(OH)<sub>3</sub> were 17.1 and 28.9, respectively [the LOI of MAA-g-LLDPE filled with 140 phr Al(OH)<sub>3</sub> was 28.2]. This demonstrated that only

when highly filled with Al(OH)<sub>3</sub> could LLDPE confer an adequate level of flame retardancy.

The composites of MAA-g-LLDPE filled with Al(OH)<sub>3</sub> had lower LOIs than the composites of neat LLDPE filled with corresponding Al(OH)<sub>3</sub>. In the LLDPE/Al(OH)<sub>3</sub> composites, the flame was easily cut by the isolated Al(OH)<sub>3</sub> agglomerate because of the poor adhesion. By the grafting of MAA to the matrix, the formation of a transition layer sustained the continuity of the flame.

### Mechanical properties

The mechanical properties of LLDPE and MAA-g-LLDPE loaded with various amounts of Al(OH)<sub>3</sub> were measured, and the results are shown in Figures 5 and 6.

By a comparison of Figures 5 and 6, we can see that the strength at break of neat LLDPE filled with the Al(OH)<sub>3</sub> composites was less than 13.5 MPa and that the elongation at break was less than 10% when the concentration of aluminum hydroxide exceeded 60 phr; moreover, after the grafting of MAA to the LLDPE matrix, the strength at break of the composites was larger than 16 MPa, and the elongation at break was larger than 20%, even when the concentration of aluminum hydroxide in the composites was larger than 60 phr and up to 140 phr. Figure 5 also shows that the tensile strength of MAA-g-LLDPE/Al(OH)<sub>3</sub> was minimal when the Al(OH)<sub>3</sub> concentration was 40 phr

**TABLE II**  
**Electric Properties of LLDPE and LLDPE Filled with Various Amounts of Al(OH)<sub>3</sub>**

[Al(OH) <sub>3</sub> ] (phr)	0	20	40	60	80	100	120	140
Volume resistance (Ω cm)	8 (±1) × 10 <sup>18</sup>	6 (±0.8) × 10 <sup>16</sup>	4 (±0.5) × 10 <sup>16</sup>	1.6 (±0.1) × 10 <sup>15</sup>	4 (±0.2) × 10 <sup>14</sup>	1 (±0.1) × 10 <sup>13</sup>	9 (±1) × 10 <sup>12</sup>	2.5 (±0.5) × 10 <sup>11</sup>
Dissipation factor	0.0001 (±0.0001)	0.016 (±0.001)	0.041 (±0.001)	0.043 (±0.001)	0.076 (±0.001)	>0.11	>0.11	>0.11
ε	2.26 (±0.01)	2.38 (±0.01)	2.64 (±0.01)	2.74 (±0.01)	3.31 (±0.01)	3.87 (±0.01)	4.14 (±0.01)	5.78 (±0.01)
Breakdown strength (kV/mm)	42 (±2)	41 (±3)	39 (±2)	38 (±1)	37 (±1)	37 (±1)	37 (±1)	35 (±1)

**TABLE III**  
**Electric Properties of LLDPE and MAA-g-LLDPE Filled with Various Amounts of Al(OH)<sub>3</sub>**

[Al(OH) <sub>3</sub> ] (phr)	0	20	40	60	80	100	120	140
Volume resistance (Ω cm)	5 (±1) × 10 <sup>18</sup>	1.6 (±0.1) × 10 <sup>16</sup>	1.4 (±0.1) × 10 <sup>16</sup>	4 (±0.4) × 10 <sup>15</sup>	8 (±1.5) × 10 <sup>14</sup>	4 (±0.35) × 10 <sup>14</sup>	2 (±0.2) × 10 <sup>14</sup>	1.4 (±0.15) × 10 <sup>13</sup>
Dissipation factor	0.001 (±0.0001)	0.0019 (±0.0001)	0.008 (±0.001)	0.008 (±0.001)	0.009 (±0.001)	0.025 (±0.001)	0.026 (±0.001)	0.028 (±0.001)
ε	2.28 (±0.01)	2.28 (±0.01)	2.28 (±0.01)	2.3 (±0.01)	2.4 (±0.01)	3.15 (±0.01)	3.20 (±0.01)	3.29 (±0.01)
Breakdown strength (kV/mm)	42 (±2)	40 (±1)	40 (±1)	40 (±1)	39 (±1)	39 (±1)	38 (±1)	38 (±1)

because the grafting reaction destroyed the crystallization of LLDPE and caused LLDPE,<sup>17,19</sup> with an increase in the Al(OH)<sub>3</sub> concentration beyond 40 phr, the Al(OH)<sub>3</sub> concentration significantly affected the rigidity of the composites.

Because of the percolation and aggregation of aluminum hydroxide particles,<sup>20</sup> the matrix was not continuous, and impure particles were formed. The mechanical properties of the LLDPE/Al(OH)<sub>3</sub> composites were determined by the interfacial adhesion between the particles and LLDPE matrix. In general, the stress concentration caused the practical strength to much less than the theoretical strength, and the stress was caused by weakness. When there was stress concentration in the composites, these parts endured much more stress than the even stress. Thus, before the theoretical strength was reached, the practical broken strength was reached, and the material was destroyed at those points and macroscopic breakdown was caused. Predictive models were used to analyze the tensile strength of the polymer composites to assess the level of interfacial interaction.<sup>21,22</sup>

Neilsen's first law model is<sup>21</sup>

$$\frac{\sigma_b}{\sigma_p} = (1 - \phi_1)S \quad (1)$$

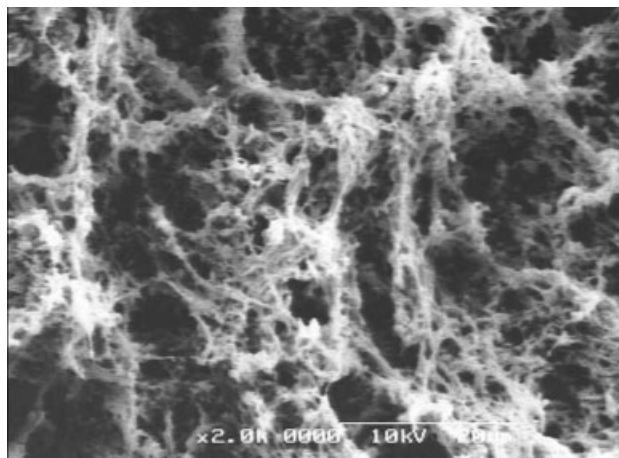
where  $\sigma_b$  and  $\sigma_p$  are the strength at break of the composites and the neat LLDPE, respectively;  $\phi_1$  is the volume fraction of Al(OH)<sub>3</sub> in the composites; and  $S$  is Neilsen's parameter in the first power-law model. The maximum value of  $S$  in eq. (1) is unity for no stress concentration effect. The lower  $S$  is, the greater the stress concentration effect is or the poorer the adhesion is; this is valid for filled polymer composites or blends. The values of  $S$  are listed in Table I, which compares the experimental data and theoretical models.

The analysis of  $S$  showed that all the values of the LLDPE/Al(OH)<sub>3</sub> composites were less than those of the MAA-g-LLDPE/Al(OH)<sub>3</sub> composites. This showed that all the MAA-g-LLDPE/Al(OH)<sub>3</sub> blends could take excessive stress because the interfacial adhesion was improved in comparison with that of the MAA-g-LLDPE/Al(OH)<sub>3</sub> composites.

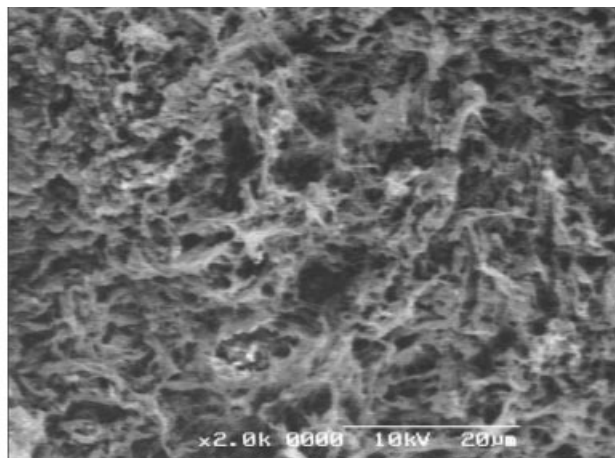
### Electric properties

The electric properties of LLDPE and MAA-g-LLDPE loaded with various amounts of Al(OH)<sub>3</sub> were measured, and the results are shown in Tables II and III.

Table II shows that the concentration of aluminum hydroxide had a significant effect on the electric properties of LLDPE/Al(OH)<sub>3</sub>. With an increase in the

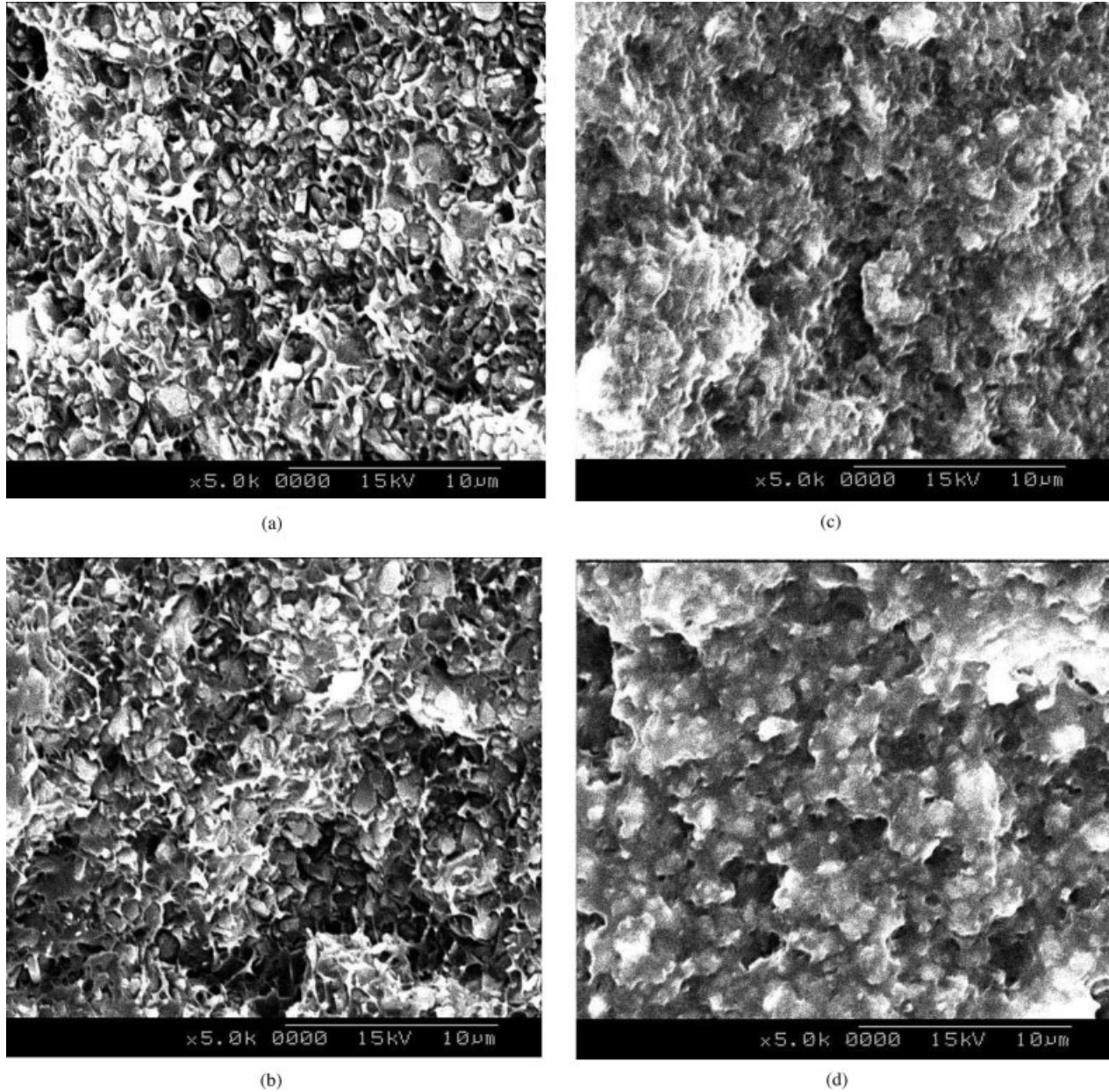


(a)



(b)

**Figure 7** SEM macrographs of tensile fractured surfaces of LLDPE and MAA-g-LLDPE filled with 140 phr Al(OH)<sub>3</sub>: (a) LLDPE/Al(OH)<sub>3</sub> composites and (b) MAA-g-LLDPE/Al(OH)<sub>3</sub> composites.



**Figure 8** SEM macrographs of fractured surfaces of LLDPE and MAA-g-LLDPE filled with various amounts of Al(OH)<sub>3</sub>: (a) 100:120 LLDPE/Al(OH)<sub>3</sub>, (b) 100:120 MAA-g-LLDPE/Al(OH)<sub>3</sub>, (c) 100:140 LLDPE/Al(OH)<sub>3</sub>, and (d) 100:140 MAA-g-LLDPE/Al(OH)<sub>3</sub>.

concentration of aluminum hydroxide, all the electric properties deteriorated, especially the volume resistivity, dissipation factor, and  $\epsilon$ . When the concentration of aluminum hydroxide was greater than 100 phr, the volume resistivity was less than  $10^{13} \Omega \text{ cm}$ , the dissipation factor was larger than 0.11, and  $\epsilon$  was larger than 3.8. These electric properties were so bad that the composites almost could not meet the demand of a cable insulator being flame retardant with a volume resistivity of  $10^{12} \Omega \text{ cm}$ , a dissipation factor of 0.1, and an  $\epsilon$  value of 3.5 (GB8815-1984 for a soft PVC cable insulator was referenced).

In general, the basic electric properties of a polymer include the volume resistance, dissipation factor,  $\epsilon$ ,

and breakdown strength. Among these parameters,  $\epsilon$  is essential and predominates over the other parameters.  $\epsilon$  of an isotropic medium is given by the well-known Clausius–Mossotti approximation when only dipole interactions are present:<sup>12,23</sup>

$$\frac{\epsilon - 1}{\epsilon + 2} = \frac{N_A \rho_m \alpha}{3M} \quad (2)$$

where  $N_A$  is Avogadro’s number,  $\rho_m$  is the mass density, and  $\alpha$  is the molecular polarizability.

Equation (2) showed that  $\alpha$  was the crucial factor for determining  $\epsilon$  of the composites and that it was pos-

sible to improve the electric properties through changes in the polarizability of the  $\text{Al}(\text{OH})_3$  filler. For the composites of MAA-grafted LLDPE filled with  $\text{Al}(\text{OH})_3$ , after the grafting of MAA to the matrix, because MAA-grafted LLDPE acted as a compatibilizer that reduced the surface energy of  $\text{Al}(\text{OH})_3$  particles, a transition layer formed between the LLDPE matrix and  $\text{Al}(\text{OH})_3$  particles. The transition layer acted as an insulator layer and reduced the polarizability of the polar  $\text{Al}(\text{OH})_3$  particles. On macroscopes, the dissipation factor and dielectric constant decreased, and the volume resistivity and breakdown strength were significantly improved, after the LLDPE matrix was grafted with MAA.

Almost all the electric properties were improved after the grafting of MAA to the LLDPE matrix according to a comparison of Tables II and III. For the MAA-g-LLDPE/ $\text{Al}(\text{OH})_3$  composites, the volume resistivity was not greater than  $1.4 \times 10^{13} \Omega \text{ cm}$ , the dissipation factor was less than 0.028,  $\epsilon$  was not more than 3.3, and the breakdown strength was greater than 37 kV/mm, with an aluminum hydroxide concentration as high as 140 phr.

### Morphologies of the composites

From this analysis, the conclusion could be drawn that the mechanical and electric properties of LLDPE composites highly loaded with  $\text{Al}(\text{OH})_3$  could be improved by the grafting of MAA to the LLDPE matrix. It could be also deduced that the interfacial adhesion was improved by the grafting of MAA to the LLDPE matrix. To clarify the microscopic mechanism, we performed a morphological analysis by SEM, and the results are shown in Figure 7(a,b) and Figure 8(a–d).

Figure 7(a,b) shows the tensile fractured surfaces of LLDPE and MAA-g-LLDPE filled with 140 phr  $\text{Al}(\text{OH})_3$  particles. Figure 7(a) clearly shows that  $\text{Al}(\text{OH})_3$  was badly dispersed, and much particle aggregation could be found because of the incompatibility with the LLDPE matrix. Because of this aggregation, it was very easy for composites to generate weakness and to be destroyed by this weakness; it was then inevitable for the toughness to be lost. Figure 7(b) shows a much more evenly dispersed morphology of  $\text{Al}(\text{OH})_3$  particles well dispersed in the polymer matrix because of the better surface adhesion and enhanced compatibility of the filler particles and grafted MAA.

Figure 8(a–d) shows the fractured surfaces of LLDPE and MAA-g-LLDPE filled with various amounts of  $\text{Al}(\text{OH})_3$  particles. Figure 8(a,c) shows the clear two-phase morphology of the matrix and filler. Figure 8(b,d) shows that the color contrast between the LLDPE matrix and  $\text{Al}(\text{OH})_3$  particles became much more

blurry than that of Figure 8(a,b) because the MAA-grafted LLDPE acted as a compatibilizer, reducing the surface energy of  $\text{Al}(\text{OH})_3$  particles and the transition layer between the LLDPE matrix and  $\text{Al}(\text{OH})_3$  particles.

### CONCLUSIONS

The results of infrared spectroscopy showed that a graft reaction occurred. After the grafting of MAA to the matrix, the LOI of LLDPE filled with  $\text{Al}(\text{OH})_3$  slightly decreased. Mechanical tests showed that for the composites of LLDPE highly loaded with  $\text{Al}(\text{OH})_3$ , the strength and elongation at break were significantly improved after the grafting of MAA to the matrix. The results of the electric tests showed a similar trend. The results of SEM showed better decentralization of  $\text{Al}(\text{OH})_3$  in the LLDPE matrix in the tensile fractured surface and a transition layer between  $\text{Al}(\text{OH})_3$  and the LLDPE matrix in the fractured surface after the grafting of MAA to the LLDPE matrix.

### References

- Mansour, S. H. *J Elast Plast* 2000, 32, 248.
- Tabtiang, A.; Venables, R. *Eur Polym J* 2000, 36, 137.
- Wang, G. L.; Jiang, P. K.; Zhu, Z. K.; Yin, J. *J Appl Polym Sci* 2002, 85, 2485.
- Field, A.; Friday, A. *Plast Rubber Compos Process Appl* 1998, 27, 46.
- Yeh, J.; Yang, M. J.; Hsieh, S. H. *Polym Degrad Stab* 1998, 61, 465.
- Wang, G. L.; Jiang, P. K.; Zhu, Z. K.; Yin, J. *J Appl Polym Sci* 2002, 86, 2544.
- Rong, M. Z.; Zhang, M. Q.; Zheng, Y. X.; Zeng, H. M.; Friedrich, K. *Polymer* 2001, 42, 3301.
- Liauw, C. M.; Lees, G. C.; Hurst, S. J.; Rotheron, R. N.; Dobson, D. C. *Plast Rubber Compos Process Appl* 1995, 24, 249.
- Rychly, J.; Dalinkevich, A. A.; Janigova, I.; Rychla, L. *J Therm Anal* 1994, 41, 115.
- Yeh, J. T.; Yang, M. J.; Hsieh, S. H. *Polym Degrad Stab* 1998, 61, 465.
- Xue, Q. Z.; Lu, G. W. *Acta Mater Compos Sinica* 2001, 18, 55.
- Grannan, D. M.; Garland, J. C.; Tanner, D. B. *Phys Rev Lett* 1981, 46, 375.
- Ma, Y. Q.; Li, Z. Y. *Acta Phys Sinica* 1990, 39, 457.
- Huang, H. L.; Yang, Z. H.; Wang, Y.; Yin, D. A. *J Appl Sci* 2001, 80, 2538.
- Huang, H.; Zhu, C. Y.; Zhou, Z. F.; Liu, N. C. *React Funct Polym* 2001, 50, 49.
- Heinen, W.; Rosenmoler, C. H.; Wenzel, C. B.; Groot, H. J. M.; Lugtenburg, J. *Macromolecules* 1996, 29, 1151.
- Wang, Z. Z.; Qu, B. J.; Fan, W. C.; Hu, Y.; Shen, X. F. *Polym Degrad Stab* 2002, 76, 123.
- Zhu, S.; Zhang, Y.; Zhang, Y. X. *Polym Polym Compos* 2002, 10, 447.
- Seo, Y.; Kim, J.; Kim, K. U.; Kim, Y. C. *Polymer* 2000, 41, 2639.
- Ma, D. Z.; He, P. S.; Xu, Z. D.; Zhou, Y. Q. *The Structure and Properties of Polymer*; Science: Beijing, 2000; Chapter 5.
- Neilsen, L. E. *J Appl Polym Sci* 1966, 10, 97.
- Bhattacharyya, A. R.; Ghosh, A. K.; Misra, A. *Polymer* 2003, 44, 1725.
- Doyle, W. T. *Phys Rev B* 1990, 42, 9319.