

# Stearic Acid Surface Modifying Mg(OH)<sub>2</sub>: Mechanism and Its Effect on Properties of Ethylene Vinyl Acetate/Mg(OH)<sub>2</sub> Composites

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**ABSTRACT:** In this article, FTIR spectra and ESEM images were employed to evaluate the effect of stearic acid surface modification of Mg(OH)<sub>2</sub>. As a result, the absorbing peak intensity of organic group on Mg(OH)<sub>2</sub> increased with the coating amount of stearic acid increasing and there was no so-called surface saturation as expected. The results indicated stearic acid surface treatment of Mg(OH)<sub>2</sub> belonged to the acid–base reaction between stearic acid and Mg(OH)<sub>2</sub>, and it would not stop until Mg(OH)<sub>2</sub> was reacted completely. In addition, stearic acid surface treatment of Mg(OH)<sub>2</sub> had remarkable influence on the properties of ethylene vinyl acetate/Mg(OH)<sub>2</sub> composites. With the increasing coating

amount of stearic acid, the composites had decreased tensile strength, increased elongation at break, and deteriorated flame retardancy, compared with the composites filled with the uncoated Mg(OH)<sub>2</sub>. However, stearic acid surface treatment of Mg(OH)<sub>2</sub> benefited processing ability of composites, and the composites had better processing ability as the coating amount of stearic acid increased. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 107: 3325–3331, 2008

**Key words:** ethylene-vinyl acetate copolymer; magnesium hydroxide; stearic acid; surface modification; flame retardancy

## INTRODUCTION

Ethylene polymer and copolymer are widely used in many fields, such as, electrical products, decorating products, even packing products, etc. Owing to their chemical compositions, these polymers are easily combustible and their flammability has restricted their application. Therefore, the flame-retarding science and technology of polymers are becoming more and more important and necessary. Halogen-type flame retardants were popular for the excellent flame-retarding efficiency before, and yet they are banned in some situation like RoHS dictate because of their toxicity and the release of toxic gas and smoke of flame-retarded composites once they are heated and fired.<sup>1</sup> Consequently, it is a trend to apply and develop halogen-free flame retardants instead of halogen-containing flame retardants.

Nowadays, the most popular method of preparing halogen-free flame-retarded polymeric materials is to

fill inorganic flame retardant into polymers, including magnesium hydroxide, aluminum hydroxide, etc. Compared with Al(OH)<sub>3</sub>, Mg(OH)<sub>2</sub> is widely used and exhibits some merits,<sup>2</sup> such as, higher decomposed temperature, better smoking suppressing capability, etc. To endow polymer/Mg(OH)<sub>2</sub> composites with excellent flame retardancy, for instance, to get to UL-94 for V-0 rating, the filling level of Mg(OH)<sub>2</sub> is usually required to reach over 50 wt % in total, which easily results in processing difficulties and marked deterioration in mechanical performances due to the poor interfacial adhesion between Mg(OH)<sub>2</sub> and polymer. Therefore, the best way to compromise among the mechanical properties of composites, the modified combustion characteristics, and processing properties when preparing halogen-free flame retarded composites is the surface modification of Mg(OH)<sub>2</sub>.<sup>3–7</sup> Stearic acid is a kind of anion surfactant and widely used as surface modifier without coupling structure. Many investigations focusing on CaCO<sub>3</sub> about the application of stearic acid as surface modifier show that CaCO<sub>3</sub> can be effectively treated with stearic acid, and the related mechanism is that a basic salt of calcium stearate is formed on the filler surface. Stearic acid surface treatment of CaCO<sub>3</sub> can better mechanical performance of composites by diminishing particle–particle interaction and enhancing the dispersion of filler and compatibility between filler and matrix.<sup>8–12</sup> Therefore, stearic acid

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surface treatment of  $\text{CaCO}_3$  is by means of calcium stearate staying on the surface of inorganic filler. The surface modification method usually is aqueous coating; however, dry coating techniques have also received little scientific study.<sup>12</sup> Stearic acid surface treatment of  $\text{Mg}(\text{OH})_2$  is also reported and the surface saturation occurs due to the low shear forces of 7000 rpm by dry coating technique,<sup>13</sup> but the effect of treatment on the surface properties of  $\text{Mg}(\text{OH})_2$  and the effect of stearic acid surface treatment on the mechanical properties and flame retardancy of composites have been rarely mentioned before.<sup>13–15</sup>

In this work, different amounts of stearic acid were applied to treat  $\text{Mg}(\text{OH})_2$  by dry coating techniques, and FTIR and ESEM results had been used to study the surface treatment effect of  $\text{Mg}(\text{OH})_2$ . In addition, the effect of surface treatment on the mechanical properties, flame retardancy, and processing ability of ethylene vinyl acetate (EVA)/ $\text{Mg}(\text{OH})_2$  composites were also discussed. It was found that the surface treatment had remarkable influence on the properties of composites. This investigation is expected to be useful for the preparation and processing of halogen-free flame-retarded polymer materials.

## EXPERIMENTAL

### Materials

Ethylene vinyl acetate (EVA) copolymer (Elvax 460<sup>(R)</sup>, with vinyl acetate of 18% and melt flow index of 2.5 dg/min) was bought from Dupont Company, USA.  $\text{Mg}(\text{OH})_2$  with a particle size of 2.03  $\mu\text{m}$  and specific surface area of 3.65  $\text{m}^2/\text{g}$  was provided by Beijing Fine Chemical Plant of BUCT (Beijing City, China).

Basic formula (phr): EVA, 100; Antioxidant 1010 (Tetrakis[ $\beta$ -(3,5-ditertiary-butyl-4'-hydroxyphenyl)propionate]pentaerythritol ester), 1.6;  $\text{Mg}(\text{OH})_2$  with surface modification by different amounts of stearic acid, 150.

### $\text{Mg}(\text{OH})_2$ surface modification

Magnesium hydroxide was modified by dry coating technique. Magnesium hydroxide was coated by stearic acid using a rotor speed of 2900 rpm in a high-speed Mixer (GH-10, 10 L, Plastic Machine Factory of Beijing, China). Firstly, magnesium hydroxide was heated to 80°C in the mixer gradually, and then different amounts of stearic acid dissolved in the solvent were injected and coated on the surface of  $\text{Mg}(\text{OH})_2$ . The fillers coated by stearic acid were transferred and roasted in the oven at 80°C for 4 h. The coated  $\text{Mg}(\text{OH})_2$  would be used to prepare EVA/ $\text{Mg}(\text{OH})_2$  composites.

### Preparation of composites

EVA and coated  $\text{Mg}(\text{OH})_2$  were blended together in the SK-160B two-roll mixer with the dimension of  $\Phi 160 \times 320 \text{ mm}^2$  and minimum clearance of 0.1 mm (Shanghai Rubber Machine Factory, China) at 130°C for 20 min. The resulting compound was compressed for 10 min at 15 MPa at 160°C, and then it was transferred to another pressing machine and pressed for 10 min at 15 MPa at an ambient temperature. For different testing, the molds with different dimension were applied. The relative mechanical properties and flame retardance test were conducted.

### Measurements

#### Mechanical properties measurement

The mechanical properties measurement was performed by using CMT4101 testing machine (SANS Company, China) equipped with a 1000 N load cell at 23°C. Five dumb-bell shaped specimens with the dimension of 115 mm  $\times$  6 mm  $\times$  2 mm were tested at a tensile speed of 250 mm/min according to the ASTM D-638, and the median was determined as the final mechanical properties data.

#### FTIR analysis

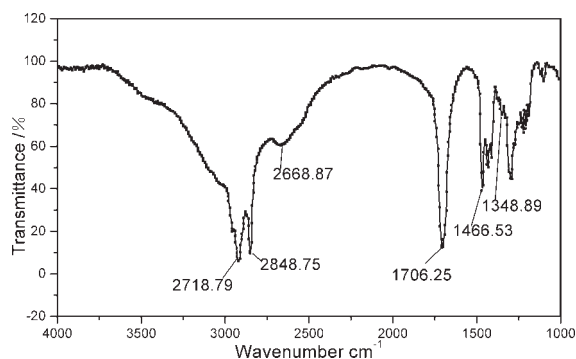
The surface treatment of  $\text{Mg}(\text{OH})_2$  by stearic acid was analyzed using a Nicolet 210 Fourier transform infra red spectrometer. The coated  $\text{Mg}(\text{OH})_2$  samples was mixed and mullied with KBr to obtain the FTIR spectra. Spectra were analyzed to give quantitative data concerning coating amount.

#### Flame retardance testing

Three standard test methods were utilized to evaluate the flame retardance of composites.

Limiting oxygen index (LOI) is an indicator of the minimum oxygen concentration that is needed to cause the material combusting in an oxygen–nitrogen atmosphere through downward burning of a vertically mounted specimen. Test specimen required is 70–150 mm in length, 6.5 mm in width, and 3 mm in thickness, according to Chinese state standard GB/T 2406-93, using a Jiangning Analyzer Plant instrument JF-3, China.

Horizontal fire test and vertical fire test of the specimen (125 mm  $\times$  13 mm  $\times$  3 mm) is conducted in ambient atmosphere according to the Chinese state standard GB/T 2408-1996, using a Jiangning Analyzer Plant instrument CZF-3, China. For the horizontal fire test, there are two marks on the specimen: one lies in 25 mm to the left end, and the other lies in 100 mm to the left end. During testing, the specimen is kept in a horizontal position, and the left end



**Figure 1** FTIR spectrum of pure stearic acid.

of the specimen is burned for 30 s. Then fire may spread from the left to the right, passing the two marks. Four levels (FH-1, FH-2, FH-3, and FH-4) are defined by whether fire spreads across the two marks or not and the spread speed. If the fire does not pass the first mark, it is identified as FH-1, standing for the best. If the fire passes the first sign, but does not pass the second sign, it is identified as FH-2. If the fire passes the second mark, there must be a spread speed through the two marks. If the speed is less than 40 mm/min, it is identified as FH-3, but when the fire spreading speed is more than 40 mm/min, the FH-4 rating is reached, which is the worst rating. Three specimens are needed at least in each experiment.

For the vertical fire test, the specimen is kept in a vertical position, and the bottom is exposed to fire. According to the time in which burning stops after two applications of 10 s each of a flame to a test bar and whether flaming drips are allowed or not, four ratings (FV-0, FV-1, FV-2, and -) are identified according to Chinese state standard GB2408-1996. If the burning stops within 10 s after two applications of 10 s each of a flame to a test bar and no flaming drips are allowed, it is identified as FV-0 rating. If burning stops within 30 s after two applications of 10 s each of a flame to a test bar and no flaming drips are allowed, it is identified as FV-1 rating; however, if flaming drips are allowed, the composites can only reach FV-2 rating. Finally, if burning stops over 30 s after two applications of 10 s each of a flame to a test bar, it is identified as “-” rating. FV-0 stands for the best rating, while “-” stands for the worst. The test result is equal to the UL-94 standard. For example, the FV-0 rating corresponds to UL 94 standard V-0 rating. At least five specimens are needed in each experiment.

#### Rheological property measurement

Rheological properties of the composites were measured at 200°C by an Instron Capillary Viscometer

INSTRON-3211 (Instron, UK). The dimension of the capillary is 1.196 mm in diameter and 51.11 mm in length.

## RESULTS AND DISCUSSION

### Surface modification of Mg(OH)<sub>2</sub> with stearic acid

#### FTIR spectrum of stearic acid

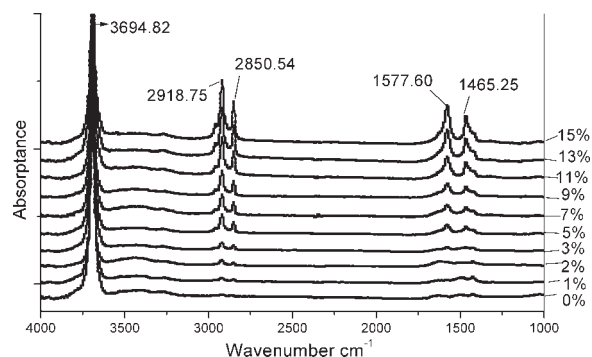
Figure 1 shows the FTIR spectrum of stearic acid. It can be seen that the absorptions at 1706 cm<sup>-1</sup> is C=O stretching vibration of carboxyl and the broad peak around 2900 cm<sup>-1</sup> is C-H stretching vibration.

#### FTIR spectra of the modified Mg(OH)<sub>2</sub>

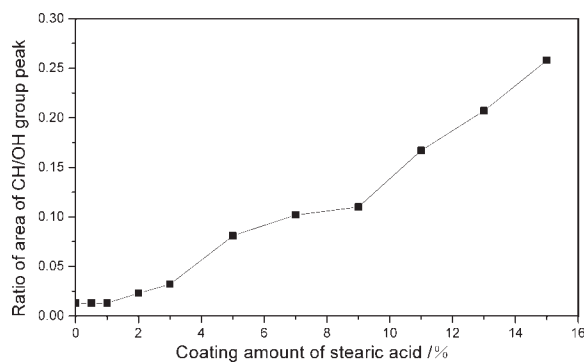
The surface treatment is essentially the reaction between the acidic group (-COOH) of stearic acid and the -OH of Mg(OH)<sub>2</sub>, according to the following equation. The surface properties of Mg(OH)<sub>2</sub> is changed by the surface modification.



Therefore, there must be the absorption of C=O stretching vibration of carboxylate and C-H stretching vibration on FTIR spectra of Mg(OH)<sub>2</sub> treated by stearic acid. Figure 2 shows the FTIR spectra of uncoated Mg(OH)<sub>2</sub> and surface-treated Mg(OH)<sub>2</sub> with different amounts of stearic acid. The sharp band at 3694 cm<sup>-1</sup> is due to the -OH group of magnesium hydroxide. There are various features observed on the spectra of Mg(OH)<sub>2</sub>. The C-H stretching vibration at around 2900 cm<sup>-1</sup> occurs and increases with increasing coating amount of stearic acid. Moreover, the absorption of C=O stretching vibration of carboxylate is also found at around 1577 and 1466 cm<sup>-1</sup>, and its intensity increases as the coating amount of stearic acid increases. However,



**Figure 2** FTIR spectra of Mg(OH)<sub>2</sub> and coated Mg(OH)<sub>2</sub> with different amounts of stearic acid.



**Figure 3** Curve of ratio of CH/OH to coating amount of stearic acid.

the absorption of C=O stretching band of carbonyl at about  $1706\text{ cm}^{-1}$  disappears in the treated sample, which means that no unreacted stearic acid exists.

The peaks area of the —OH group is divided by the peaks area of C—H group from the substrate and the results are presented in Figure 3. It indicates that the ratio of C—H to —OH group absorption peak area is constantly increasing with the increasing coating amount of stearic acid, reflecting a gradual increase in aggradation of magnesium stearate on the surface of the  $\text{Mg}(\text{OH})_2$  samples, where the so-called “plateau” or “surface saturation” does not exist.

#### Analysis of surface modification of $\text{Mg}(\text{OH})_2$

Generally, it is considered that the surface saturation exists when  $\text{Mg}(\text{OH})_2$  is treated by surface modifiers like silicon coupling agent and titanate coupling agent, and the optimal coating amount is corresponded to surface saturation. However, analyzing the results gained from Figures 2 and 3, it is very obvious that the reaction between  $\text{Mg}(\text{OH})_2$  and stearic acid continues with the increasing coating level of stearic acid and the reaction will not stop at any coating amount of stearic acid until  $\text{Mg}(\text{OH})_2$  is consumed completely.

Figure 4 shows the FTIR spectrum of the blends in which  $\text{Mg}(\text{OH})_2$  and stearic acid are mixed simply with equal quantity without any energy at room temperature. It is seen that the absorption of C=O stretching vibration of carboxyl at around  $1705\text{ cm}^{-1}$  still exists; however, the absorption of C=O stretching vibration of carboxylate at  $1582\text{ cm}^{-1}$  also appears, which reflects that the reaction between  $\text{Mg}(\text{OH})_2$  and stearic acid has occurred. Therefore, the experiment strongly confirms that the reaction between  $\text{Mg}(\text{OH})_2$  and stearic acid should belong to the acid–base reaction. Moreover, analyzing the above results, it is concluded that the chemical reac-

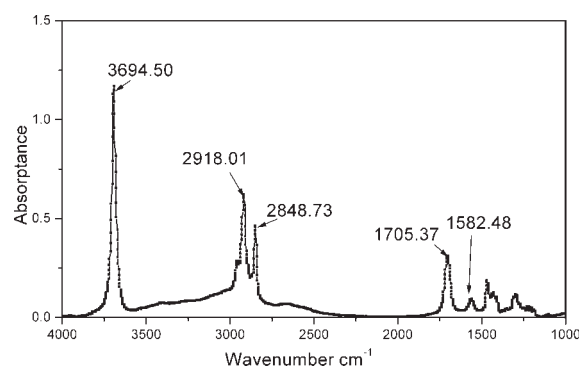
tion may finish as neutralization reaction between stearic acid and  $\text{Mg}(\text{OH})_2$ .

During the surface modification of  $\text{Mg}(\text{OH})_2$ , it is considered that the surface modifier can change the surface properties of  $\text{Mg}(\text{OH})_2$ . But when  $\text{Mg}(\text{OH})_2$  is surface treated by excessive stearic acid, it is observed that the appearance of  $\text{Mg}(\text{OH})_2$  is also changed and the surface of  $\text{Mg}(\text{OH})_2$  is obviously eroded, as shown in Figure 5. It is seen that the surface of uncoated  $\text{Mg}(\text{OH})_2$  is smooth and has obvious interphase; however, the surface of coated  $\text{Mg}(\text{OH})_2$  with 15 wt % stearic acid is very coarse and has blurry interphase. In addition, according to the reaction mechanism of stearic acid surface treatment of  $\text{Mg}(\text{OH})_2$ , magnesium stearate is produced and stearic acid binding  $\text{Mg}(\text{OH})_2$  is not by covalent bonds, but by the ionic bond. The ionic bond is so weak that the magnesium stearate on the surface of  $\text{Mg}(\text{OH})_2$  can be easily washed by hot alcohol of  $75^\circ\text{C}$ , as shown in Figure 6. Sharp absorption peaks of C=O stretching vibration of carboxylate is detected at around  $1577$  and  $1466\text{ cm}^{-1}$  and C—H stretching vibration at around  $2900\text{ cm}^{-1}$  in Fig. 6 (curve a); however, these peaks disappear in Fig. 6 (curve b) after stearic acid surface treatment of  $\text{Mg}(\text{OH})_2$  was washed by hot alcohol. This result shows that the magnesium stearate staying on the surface of the  $\text{Mg}(\text{OH})_2$  samples belongs to absolute physical absorption, and magnesium stearate may come off from  $\text{Mg}(\text{OH})_2$  surface during surface modification and preparation of composites at high temperature.

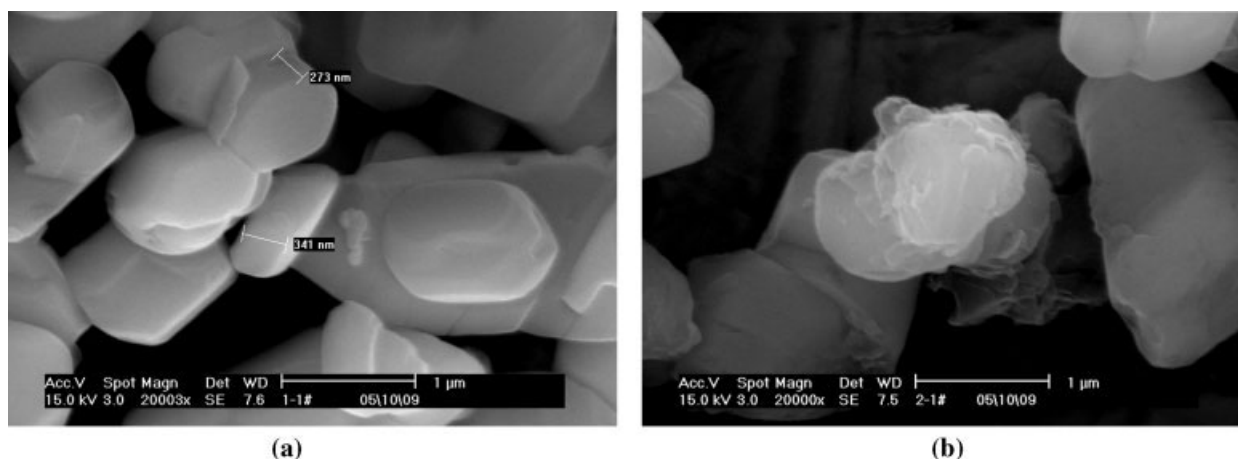
#### Effect of stearic acid surface treatment of $\text{Mg}(\text{OH})_2$ on properties of EVA/ $\text{Mg}(\text{OH})_2$ composites

Effect of stearic acid surface treatment of  $\text{Mg}(\text{OH})_2$  on mechanical properties of composites

Figure 7 exhibits the effect of coating amount of stearic acid on mechanical properties of EVA/ $\text{Mg}(\text{OH})_2$  composites. It is seen that the tensile strength of composites decreases with the increasing coating



**Figure 4** FTIR spectrum of blend of stearic acid and  $\text{Mg}(\text{OH})_2$  by simply mixing.



**Figure 5** ESEM images of (a) uncoated Mg(OH)<sub>2</sub> and (b) 15 wt % stearic acid-coated Mg(OH)<sub>2</sub>.

amount of stearic acid. In the elongation of composites, when Mg(OH)<sub>2</sub> is treated with small quantities of stearic acid, the elongation at break of composites has little change. However, as the coating amount of stearic acid increases, the elongation at break increases sharply to a maximum value of 600%, corresponding to 7 wt % stearic acid. When the coating amount of stearic acid varies from 7 to 15 wt %, the elongation at break has little decrease and then arrives at a stable value of about 500%.

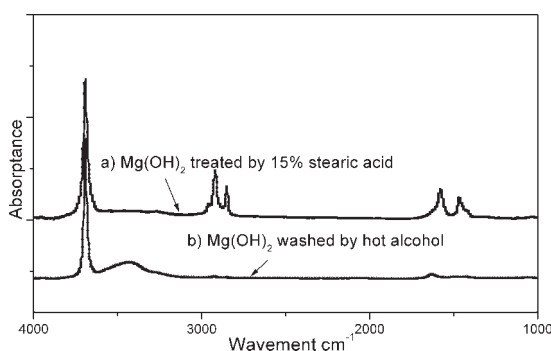
On the one hand, as stated above, the interaction between Mg(OH)<sub>2</sub> and stearic acid belongs to weak physical absorption; on the other hand, the interaction between filler and matrix is by stearic acid chains entangling with macromolecular chains, which also have no means of chemically reacting. Surface modification can enhance the dispersion of Mg(OH)<sub>2</sub> in matrix, but the interaction between Mg(OH)<sub>2</sub> and EVA is weak and produce low bond strengths between filler and matrix. This results in filler debonding at relatively low stress levels, although there can sometimes be significant improvement in properties such as elongation.<sup>12</sup> Moreover, the excessive magnesium stearate come off from the surface

of Mg(OH)<sub>2</sub> as lubricant, which also brings about the lower strength and longer elongation at break. Therefore, the composites have increasing elongation and decreasing tensile strength with the increasing coating amount of stearic acid.

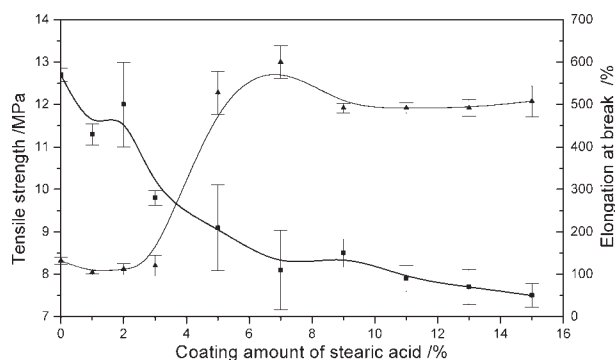
Effect of coating amount of stearic acid on flame retardancy of composites

Flame retardancy of composites indicates the capacity to resist fire. Generally speaking, the stronger the interaction among macromolecular chains of composites is, the higher heat resistance ability the composites have. Higher heat-resistance of composites is believed to benefit the flame retardancy through the resistance to heat-distortion and heat-flow.<sup>16</sup>

Table I shows the effect of stearic acid with different coating amount surface treatment of Mg(OH)<sub>2</sub> on flame retardancy of composites. As the stearic acid amount increases, the flame retardancy of composites deteriorates gradually, such as, decreasing LOI values and lowering horizontal fire rating and vertical fire rating. When the coating amount of stearic acid is less than 7 wt %, stearic acid surface treat-



**Figure 6** FTIR spectra of coated Mg(OH)<sub>2</sub> (a) before washing; (b) after washing.



**Figure 7** Effect of stearic acid on mechanical properties of EVA/Mg(OH)<sub>2</sub> composites.

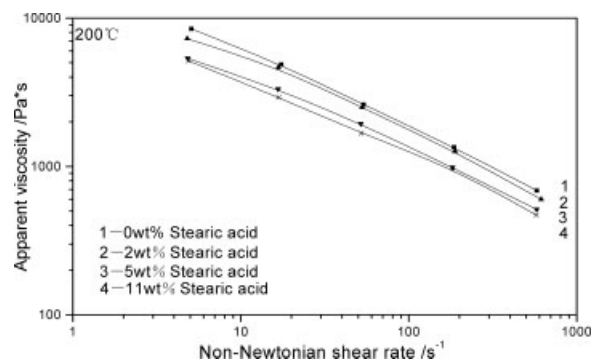
ment of  $\text{Mg}(\text{OH})_2$  has little influence on the LOI values of the composites; however, when the coating amount of stearic acid changes from 7 to 15 wt %, the LOI of composites decreases by 4. When the stearic acid amount is too high, the horizontal fire rating of composites cannot reach FH-1 rating. In vertical fire test, the rating of the composites cannot reach FV-0 at 2 wt % stearic acid and FV-1 rating at 3 wt % stearic acid.

The surface modification of  $\text{Mg}(\text{OH})_2$  by stearic acid and magnesium stearate can weaken the interaction among macromolecular chains and the interaction between  $\text{Mg}(\text{OH})_2$  and EVA, which results in the lower heat-resistance and poor flame retardancy of composites. Another reason is that the surface treatment of  $\text{Mg}(\text{OH})_2$  by excessive stearic acid can bring forth the decrease of actual content of  $\text{Mg}(\text{OH})_2$  in composites, which can also deteriorate the flame retardancy of composites.

#### Effect of coating amount of stearic acid on rheological property of composites

As shown in Figure 8, the viscosity of the composites decreases with the increasing coating amount of stearic acid. That is to say, as the coating amount of stearic acid increases, the processing ability of EVA/ $\text{Mg}(\text{OH})_2$  composites improves. Compared with the composites filled with  $\text{Mg}(\text{OH})_2$  coated by 2 wt % stearic acid, the apparent viscosity of composites filled with 5 wt % stearic acid-coated  $\text{Mg}(\text{OH})_2$  decreased sharply; however, stearic acid changing from 5 to 11 wt % has little influence on the viscosity of composites. Therefore, it confirms that the  $\text{Mg}(\text{OH})_2$  coated by stearic acid can work as lubricant and the lubricating effect improves with the increasing coating amount of stearic acid.

Considering the above results, addition of stearic acid did cause the deterioration of the flame retardancy. But, more importantly, the surface modification of  $\text{Mg}(\text{OH})_2$  by stearic acid can enhance the



**Figure 8** Effect of coating amount of stearic acid on viscosity of composites.

processing ability of composites, which is critical when the filling level of the inorganic flame retardant is very high, e.g., 50 wt % or more. Therefore, the coating amount of stearic acid should be about 5 wt % to get correspondingly good mechanical properties and flame retardancy at the same time. And the flame retardancy of composites can be further improved by filling other flame retardant synergist.

## CONCLUSIONS

FTIR spectra were employed to evaluate the surface modification of  $\text{Mg}(\text{OH})_2$  by stearic acid with different coating amounts. When coating level of stearic acid varied from 1 to 15 wt %, the absorption of C—H stretching vibration at around  $2900\text{ cm}^{-1}$  and C=O stretching vibration of carboxylate at  $1577\text{ cm}^{-1}$  appeared and intensity increased with the increasing amount of stearic acid. No unreacted stearic acid was detected even when very high coating concentrations were used, and the so-called “saturation coverage” did not exist. When  $\text{Mg}(\text{OH})_2$  is surface-modified by excessive stearic acid,  $\text{Mg}(\text{OH})_2$  was eroded clearly. All the experiments revealed that the related reacting mechanism between stearic acid and  $\text{Mg}(\text{OH})_2$  belonged to the acid–base reaction between acidic group of stearic acid and alkalescent group of  $\text{Mg}(\text{OH})_2$ .

The surface modification of  $\text{Mg}(\text{OH})_2$  by stearic acid brought about great changes in properties of composites, including mechanical properties, flame retardancy, and rheological property. As the coating amount of stearic acid increased, the tensile strength of composites decreased, and the elongation at break of composites increased sharply to a maximum value and then decreased to a stable value. With flame retardancy of composites, it deteriorated with the stearic acid amount increasing. However, stearic acid surface treatment of  $\text{Mg}(\text{OH})_2$  benefited the processing ability of EVA/ $\text{Mg}(\text{OH})_2$  composites.

**TABLE I**

**Effect of Coating Amount of Stearic Acid on Flame Retardancy of Composites**

Amount of stearic acid (%)	LOI	Horizontal fire test	Vertical fire test
0	34.6	FH-1	FV-0
1	34.4	FH-1	FV-0
2	34.5	FH-1	FV-1
3	34.4	FH-1	—
5	34.3	FH-1	—
7	34.0	FH-1	—
9	32.3	FH-1	—
11	31.4	FH-1	—
13	30.3	FH-1	—
15	30.5	FH-3	—

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