

Influence of Ammonium Polyphosphate Modified with 3-(Methylacryloxy) Propyltrimethoxy Silane on Mechanical and Thermal Properties of Wood Flour–Polypropylene Composites

Lin Zhou,¹ Chuigen Guo,² Liping Li¹

¹Heilongjiang Key Laboratory of Molecular Design and Preparation of Flame Retarded Materials, College of Science, Northeast Forestry University, Harbin 150040, China

²MOE Key Laboratory of Bio-Based Material Science and Technology, Northeast Forestry University, Harbin 150040, People's Republic of China

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ABSTRACT: In this article, the influence of ammonium polyphosphate (APP) and ammonium polyphosphate modified with 3-(Methylacryloxy) propyltrimethoxy silane (M-APP) on mechanical properties, flame retardancy, and thermal degradation of wood flour–polypropylene composites (WF/PP composites) have been investigated. Polypropylene grafted with *m*-isopropenyl- α,α -dimethylbenzyl-isocyanate (*m*-TMI-g-PP) was used to improve the adhesion of WF/PP composites. APP and M-APP were used as flame retardants. The experimental results demonstrated that addition of M-APP obviously enhanced mechanical properties of WF/PP composites. According to cone calorimetry results, M-APP is also an effective flame retardant for WF/PP composites, compared to that of APP. It was also found that M-APP decreased the 1% weight loss temperature and increased char residue. The

thermal degradation of wood flour based upon the first peak temperature of wood decreased from 329.3 to 322.9°C and the thermal degradation of PP based upon the second peak temperature of PP improve from 518.0 to 519.6°C, when M-APP was added to the WF/PP composites. From SEM results the char layer of the 25% M-APP systems is much more intumescent than that of the 25% APP systems, indicating that 3-(Methylacryloxy) propyltrimethoxy silane can improve the char-forming ability of WF/PP composites. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 122: 849–855, 2011

Key words: polypropylene; modified ammonium polyphosphate; mechanical properties; flame retardance; thermogravimetric analysis

INTRODUCTION

Wood–plastics composites (WPC) are known as a new generation materials for several fields such as housewares, automotive, and construction, etc. During recent years, they have been developed rapidly due to their favorable performance, low cost attributes of both wood (fiber) and plastics, environmentally friendly, and biodegradability.^{1–3} The interface compatibility between wood flour and polymers in WPC is a key problem.^{4–6} Hence, polymers grafted with *m*-isopropenyl- α,α -dimethylbenzyl-isocyanate (*m*-TMI)

were used as good compatibilizers to improve the mechanical properties of WPC.^{7,8} Polymers grafted with Maleic anhydride, silicones, fatty acids, and elastomers are also used as compatibilizers in WPC.^{9–12} However, flame retardancy of WPC is another important topic due to the easy flammability of both wood flour (fiber) and plastics, and demands of fire protection in their use. With increasing demand for WPC, the development of safe and environmentally flame-retarded WPC is of great importance.¹³

Recently, much research has been focused on intumescent flame-retardants (IFR), which is a growing group of halogen-free agent. On heating, fire-retardant intumescent material restricts the heat flux or flame transferred. The proposed mechanism is based on charred layer acting as physical barrier, which slows down the heat and mass transfer between the gas and the condensed phases.^{14,15} The conventional IFR additives (ammonium polyphosphate and ammonium polyphosphate with pentaerythritol-melamine, Char-forming agent, etc) are phosphorus-nitrogen containing materials, which are effective in the flame retardancy of WPC materials.^{16–20}

Correspondence to: L. Li (lilipingguo@nefu.edu.cn).

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TABLE I
Components of M-APP Samples

| Samples | APP/g | KH-570/g | Ethanol/mL |
|---------|-------|----------|------------|
| M-APP-1 | 100 | 0.5 | 150 |
| M-APP-2 | 100 | 1.0 | 150 |
| M-APP-3 | 100 | 1.5 | 150 |
| M-APP-4 | 100 | 2.0 | 150 |

However, those IFRs are highly polar systems and the compatibility between polar IFRs and nonpolar polyolefin is poor, so that the improvement of flame retardancy is obtained at the expense of the mechanical properties of flame-retarded materials. The high loading of IFR additives greatly damage the mechanical properties of materials, this result explains that IFR such as APP has a high molecular weight and are highly polar that was difficult to dispersed into the interface between wood-flour and PP, so high loading of IFR additives into WPC damage the mechanical properties of materials. Thus the flame retardancy of IFR needs to be further improved.^{21,22}

The 3-(methylacryloyl) propyltrimethoxy silane is a kind of macromolecule, which contains Si—O and Si—C bonds as its trunk chain and the organic groups such as methyl, ethylene as the side chain. It provides lots of good properties such as water resistance, chemical stability, and thermal stability. Siloxane itself could be also used as flame-retardants; however, the flame retardancy of WPC was not obviously increased when it was used as a flame-retardant alone. Hence, the IFR treated with the organic-silicon shows great potential in flame-retardant areas and may be an effective method to improve the disadvantage of traditional IFR.²³

In this study, silane was chosen as a compatibilizer to modify the ammonium polyphosphate which could improve the highly polar properties and flame-retardant properties. The purpose of this article is to prepare a new intumescent flame-retardant (ammonium polyphosphate modified with 3-(Methylacryloyl) propyltrimethoxy silane) and then incorporate this IFR into WF/PP composites. The mechanical

properties, flame retardancy, and thermal decomposition were studied based on mechanical instrument, cone calorimetry, and thermogravimetric analysis (TGA). The surface morphology of char residues of the flame-retardant WF/PP composites after cone calorimetry test was examined by SEM.

EXPERIMENT

Materials

Wood flour (WF) was supplied by Harbin Yongxu (Harbin, China). Polypropylene (PP) was supplied by Daqing PetroChemical Company (Daqing, China). Ammonium polyphosphate (APP) and ethanol were supplied by Hengyu (Tianjin, China). The 3-(methylacryloyl) propyltrimethoxysilane (KH-570) was supplied by Changzhou Yabang Company (Jiangsu, China). Antioxidant-1010 was supplied by Jiangsu Hanguang (Jiangsu, China). Polypropylene grafted with *m*-isopropenyl- α,α -dimethylbenzyl-isocyanate (*m*-TMI-*g*-PP) and ammonium polyphosphate modified with 3-(methylacryloyl) propyltrimethoxysilane (KH-570 (M-APP) were prepared in our laboratory.

Preparation of M-APP

The recipe of synthesis of M-APP was shown in Table I. The APP, KH-570, and ethanol were placed in a 250-mL three-necked round-bottomed flask fitted with a stirrer and reflux condenser. The reaction was conducted for 1 h at 60°C. Then the products were dried at 105°C for 3 h in a vacuum oven.

The preparation of samples

The wood flour and APP was dried at 105°C for 8 h in an oven before the experiment. A fixed ratio of amount of WF/PP/*m*-TMI-*g*-PP and WF/PP/*m*-TMI-*g*-PP (60 : 40 : 6) was used through the treatment. The components of samples were shown in Table II. All samples in Table II were prepared by mixing PP, WF, *m*-TMI-*g*-PP, APP, M-APP and

TABLE II
Components of Samples

| Samples | PP/g | WF/g | <i>m</i> -TMI- <i>g</i> -PP/g | 1010/g | APP/g | M-APP-1/g | M-APP-2/g | M-APP-3/g | M-APP-4/g |
|---------|------|------|-------------------------------|--------|-------|-----------|-----------|-----------|-----------|
| 1 | 40 | 60 | 6 | 1 | 0 | 0 | 0 | 0 | 0 |
| 2 | 40 | 60 | 6 | 1 | 25 | 0 | 0 | 0 | 0 |
| 3 | 40 | 60 | 6 | 1 | 0 | 25 | 0 | 0 | 0 |
| 4 | 40 | 60 | 6 | 1 | 0 | 0 | 25 | 0 | 0 |
| 5 | 40 | 60 | 6 | 1 | 0 | 0 | 0 | 25 | 0 |
| 6 | 40 | 60 | 6 | 1 | 0 | 0 | 0 | 0 | 25 |
| 7 | 40 | 60 | 6 | 1 | 0 | 0 | 0 | 5 | 0 |
| 8 | 40 | 60 | 6 | 1 | 0 | 0 | 0 | 10 | 0 |
| 9 | 40 | 60 | 6 | 1 | 0 | 0 | 0 | 15 | 0 |
| 10 | 40 | 60 | 6 | 1 | 0 | 0 | 0 | 20 | 0 |
| 11 | 40 | 60 | 6 | 1 | 0 | 0 | 0 | 30 | 0 |

TABLE III
Mechanical Properties of WF/PP Composites

| Samples | Tensile strength (MPa) | Flexural strength (MPa) | Notched impact strength (kJ m^{-2}) |
|---------|------------------------|-------------------------|--|
| 1 | 23.27 \pm 0.15 | 60.29 \pm 0.12 | 3.94 \pm 0.02 |
| 2 | 21.88 \pm 0.12 | 59.87 \pm 0.15 | 3.39 \pm 0.07 |
| 3 | 24.93 \pm 0.06 | 61.98 \pm 0.08 | 4.33 \pm 0.05 |
| 4 | 25.35 \pm 0.20 | 63.03 \pm 0.22 | 4.45 \pm 0.05 |
| 5 | 28.04 \pm 0.15 | 65.53 \pm 0.12 | 5.75 \pm 0.03 |
| 6 | 25.20 \pm 0.25 | 60.75 \pm 0.18 | 4.11 \pm 0.08 |
| 7 | 23.98 \pm 0.08 | 61.30 \pm 0.05 | 4.46 \pm 0.06 |
| 8 | 25.56 \pm 0.16 | 63.20 \pm 0.15 | 4.65 \pm 0.03 |
| 9 | 26.62 \pm 0.15 | 64.25 \pm 0.22 | 4.90 \pm 0.06 |
| 10 | 27.10 \pm 0.24 | 65.68 \pm 0.18 | 5.50 \pm 0.12 |
| 11 | 26.31 \pm 0.22 | 62.46 \pm 0.15 | 4.88 \pm 0.07 |

antioxidant-1010 on a SK-100 two-roll mill (produced by Harbin Special Plastic, China) at 175°C for 10 min, then pressing on a hot press machine at 175°C for 1 min to form sheets with the dimension of 200 \times 200 mm².

Mechanical properties test

The tensile strength, flexural strength was performed using a TA-20 computer controlled universal testing machine (produced by Shenzhen Reger Instrument, China). The Izod impact strength was carried on a XJC-25D impact testing machine (produced by Chengde Precision Testing Machine, China). Five specimens were tested to obtain the average value for each treatment. Tensile strength tests were examined according to ASTM D638 with a crosshead speed of 5 mm min⁻¹. Flexural strength tests were performed according to ASTM D790 with a crosshead speed of 2 mm min⁻¹ and a support span length of 64 mm. Izod impact strength tests were conducted according to ASTM D256.

Thermal degradation behavior

Thermal degradation behavior test were carried out by a Perkin-Elmer Pyris 1 Thermal Analyzer at a linear heating rate of 10°C min⁻¹ under nitrogen. The range of temperature was from 30 to 800°C. The weight of samples was kept within 3–5 mg.

Cone calorimeter

The cone calorimeter (CONE) tests were performed according to the ASTM 1356-90 standard. Each specimen with the dimensions of 100 \times 100 \times 3 mm³ was wrapped in aluminum foil and exposed horizontally to an external heat flux of 50 kW m⁻².

Scanning electron microscopy

The surface morphology of char residues of the flame-retardant WF/PP composites after cone calo-

rimeter were observed with a Quanta 200 scanning electron microscopy (SEM) using 15 kV and 60 mA electric current. The char was coated with gold/palladium alloy ready for imaging.

RESULTS AND DISCUSSION

Mechanical properties

The influences of APP and M-APP on the mechanical properties of WF/PP composites are shown in Table III, whose components were based on Table II. Comparison with the results of Samples 1 and 2, the tensile strength, flexural strength, and Izod impact strength of WF/PP composites decreased when APP was added. Because of the highly polar system of APP, poor compatibility and dispersion with WF/PP composites, the mechanical properties of WF/PP composites was dramatically destroyed with addition of APP.

Comparison with the results of Samples 1 and 3–6, addition of M-APP could properly enhanced tensile strength, flexural strength, and Izod impact strength of WF/PP composites. M-APP-3 effectively increased mechanical properties compared with the untreated WF/PP composites, tensile strength represented an increase of 20.5%, flexural strength exhibited an increase of 8.7%, and Izod impact showed an improvement of 45.9%. This result demonstrated that KH-570 was an effective compatibilizer and dispersant for improving the highly polar properties and dispersion of APP, and more interfacial function occurred between KH-570 in M-APP and PP, WF and APP. Thus the addition of M-APP effectively increased mechanical properties of WF/PP composites, especially for the M-APP-3 (APP modified with a loading of 1.5% KH-570). Hence, M-APP-3 was used as a flame-retardant in WF/PP composites.

Comparison of Samples 1, 5, and 7–11, all the composites showed better mechanical properties than the untreated composites. As the content of M-APP-3 increased gradually, the mechanical properties of WF/PP composites increased first then reduced, but it was still higher than that of Sample 1, showing better interfacial interaction among WF, PP, and M-APP-3. The tensile strength of the composites reached its maximum with 25% M-APP-3 loading (based on WF+PP mass). The flexural strength of the composites reached its maximum with 20% M-APP-3 loading and the notched impact strength of the composites reached its maximum with 25% M-APP-3 loading. The loading of 25% M-APP-3 can reach UL-94(Underwriters Laboratories) flammability standard, V-0 rating. Thus the content of M-APP-3 or APP in the composites was fixed as 25% (based on WF+PP mass) for TGA and cone analysis.

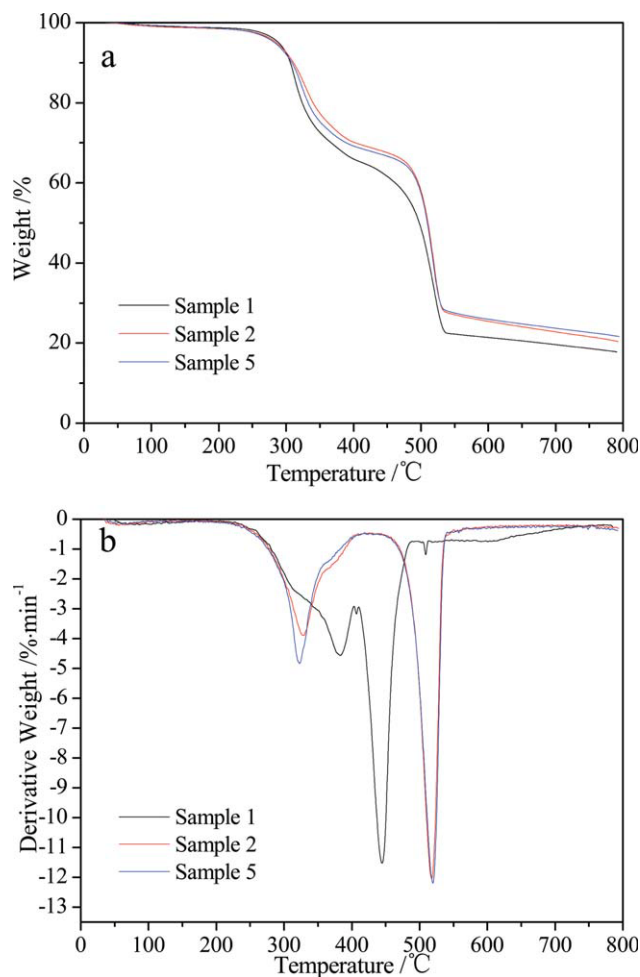


Figure 1 TGA (a) and DTG (b) curves of composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Thermal degradation behavior

The TGA and DTG curves of Sample 1, 2, and 5 were shown in Figure 1. The 1% weight loss temperature ($T_{1wt\%}$), the first peak temperature of wood (T_{max1}), and the second peak temperature of PP (T_{max2}), and the char residue at 750°C were listed in Table IV. The TGA curves showed that they underwent two-step thermal degradation processes. In the first step, thermal degradation of wood flour took place, and the thermal degradation of PP in WF/PP composites took place mainly at the second step. From their DTG curves, when the M-APP or APP was added, the decomposition temperature of wood flour and PP shifted to lower and higher temperature, respectively. According to the data of Table IV, addition of APP or M-APP-3 made the thermal degradation of WF/PP composites system take place earlier and promoted char formation. The 25% M-APP decreased $T_{1wt\%}$ from 110.7 to 102.3°C and also promoted the T_{max1} from 329.3 to 322.9°C, the T_{max2} from 518.0 to 519.6°C in comparison with 25%

APP addition. The residue char weight of WF/PP composites at 750°C increased from 21.5 to 22.7%. Because addition of APP or M-APP-3 made PP in WF/PP composites more stable, and APP or M-APP-3 decomposed to produce phosphoric acid, HPO, and PO radicals, which are considered to be free radical scavengers. Thus, HPO and PO can trap the free radicals produced from wood flour, and APP eliminated the influence of wood flour on stability of PP.¹⁶ However, M-APP-3 with Si—O and Si—C bond and organic groups such as methyl, ethylene could also produce free radicals which could eliminate the influence of wood flour on stability of PP, compared with that of APP. It is possible that the presence of M-APP can promote char formation in the WF/PP composites and form a char layer of better quality which can endure the higher temperatures and protect PP from decomposing. Thus the thermal degradation property was improved with the addition of M-APP.

Cone calorimeter study

The results from cone were shown in Figure 2, and the important data were summarized in Table V. From Figure 2(a), it can be seen that WF/PP composites burn very fast after ignition at heat flux of 50 kW m⁻². The heat release rate (HRR) peak appears at a rate of heat release of 357.5 kW m⁻² and the total heat release (THR) reaches 102.1 MJ m⁻² within 450 s. For flame retarded WF/PP composites, the peak HRR values decreased remarkably, especially for the 25% M-APP additives. Moreover, the ignition time (IT) of WF/PP-M-APP-3 25% (34 s) was longer than that of WF/PP-APP 25% (28 s). It can be found that process was prolonged in comparison with Samples 1, 2. It is very clear that a significant flame retardant effect was obtained with flame retardant treatment on WF/PP composites. WF/PP-M-APP-3 25% showed a HRR peak at about 154.9 kW m⁻² while the HRR peaks of WF/PP-APP 25% were 196.2 kW m⁻². It is well known that the samples containing phosphorus can release phosphoric acid which catalyzes the dehydration and carbonization of wood, resulting in formation of less flammable products and correspondingly more char. Meanwhile, Si—O, Si—C bond and organic groups such as methyl, ethylene also produce free radicals that

TABLE IV
TGA Data Obtained for WF/PP Composites

| Samples | $T_{1wt\%}$ (°C) | T_{max1} (°C) | T_{max2} (°C) | Char residue at 750°C (wt %) |
|---------|------------------|-----------------|-----------------|------------------------------|
| 1 | 118.4 | 383.2 | 444.8 | 12.6 |
| 2 | 110.7 | 329.3 | 518.0 | 21.5 |
| 5 | 102.3 | 322.9 | 519.6 | 22.7 |

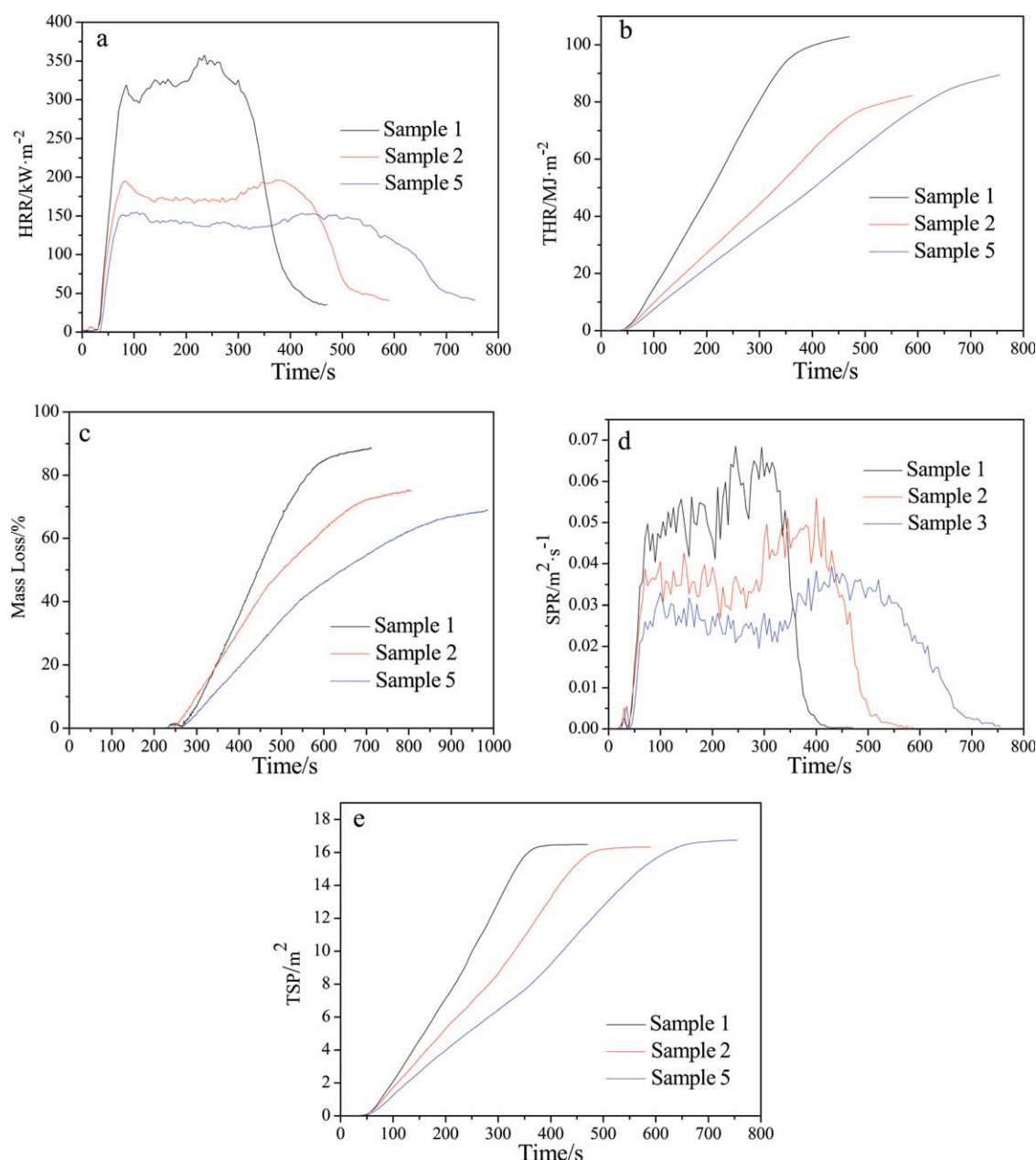


Figure 2 HRR (a), THR (b), Mass loss (c), SPR (d), and TSP (e) curves of WF/PP composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

they relatively increase thermal stabilization of PP, which suppresses the mass loss. M-APP includes simultaneously phosphoric acid and KH-570. So their synthetic actions resulted in the synergistic effect on flame retardance.

Figure 2(b) showed the total heat release (THR) for Samples 1, 2, and 5. The slope of THR curves can be assumed as representative of fire spread. The lower the THR value is, the safer the material is. It is very clear that the flame spread of Samples 2 and 5 have decreased, and the flame spread of Sample 5 was comparatively the lowest. The values were listed in Table V. The THR within 450 s, decreased

by 44.2% with M-APP-3 and by 29.5% with APP compared to untreated WF/PP composites, suggesting a significant effect of M-APP-3 on flame retardance in WF/PP composites. The incorporation of KH-570 to APP enhanced the flame retardancy by

TABLE V
Cone Calorimeter Test Data of WF/PP Composites

| Sample | Pk-HRR (kW m^{-2}) | THR (MJ m^{-2}) | Mass loss (%) |
|--------|-------------------------------|----------------------------|---------------|
| 1 | 357.5 | 102.1 | 89 |
| 2 | 196.2 | 72.0 | 75 |
| 5 | 154.9 | 57.2 | 68 |

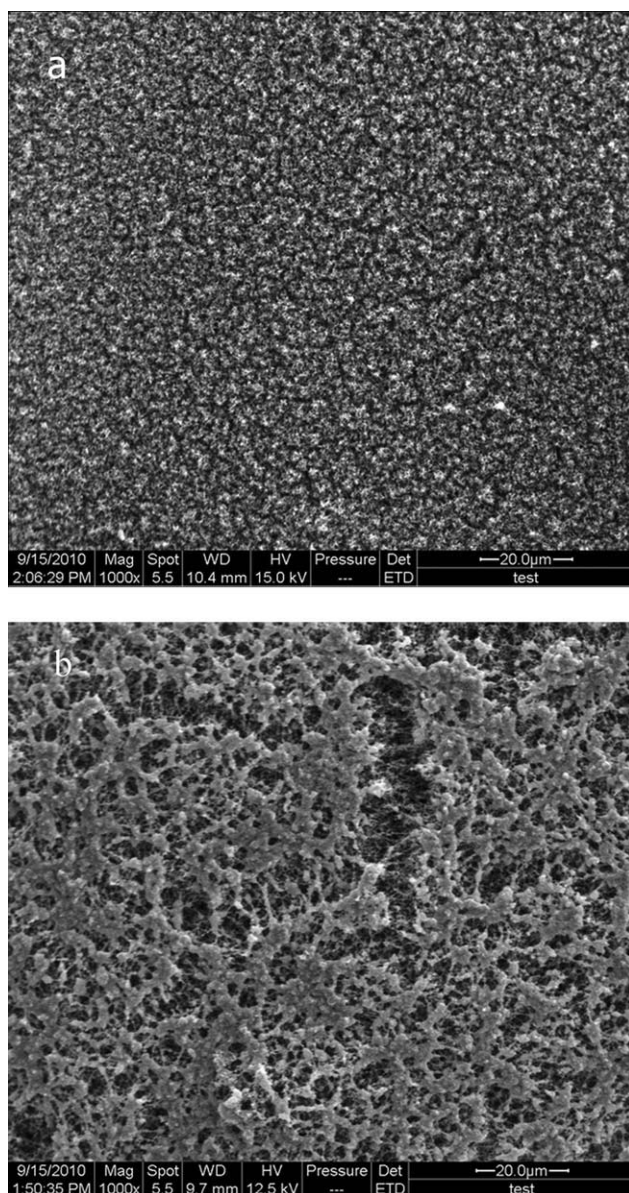


Figure 3 SEM micrographs of the outer surface of intumescent carbon layer from WF/PP composites after cone calorimeter test. (a) WF/PP-APP 25%; (b) WF/PP-M-APP-3 25%.

promoting char formation and reduced the THR during burning process.

Figure 2(c) shows the mass loss (%) of composites. It can be seen that the mass loss (%) value of 25% M-APP-3 were smaller than addition of 25% APP, decreasing by 9.3% from 75 to 68%. It is well known that the smaller the mass loss, the better is the flame retardants, indicating that addition of M-APP-3 effectively decreased the thermal degradation of WF/PP composites. It was implied that during the heating process, the intumescent flame-retardant generated a cellular charred layer on the surface of the composite, which protected the underlying material from the action of the heat flux or flame and of

oxygen toward the composites, which lead to an increase of char residues and a decrease in HRR and THR.

The smoke production rate (SPR) curves in Figure 2(d) showed that the maximum value of the smoke production rate for WF/PP composites was larger than that of flame-retarded WF/PP composites, and WF/PP-M-APP-3 25% were obviously smaller than WF/PP-APP-25%. It shows an improved effect on reduction in the amount of smoke generated. It showed that the fire-retardant effectiveness of phosphorus-containing compounds goes through a maximum with increasing phosphorus concentration. It was attributed to the competition between the char-forming fire-retardant action and evolution of combustible gases. When the composites was heated, M-APP decomposed and then produced incombustible material such as SiO_2 and NH_3 to barrier or dilute oxygen, retarding combustion, exhibiting more effective inhibition of oxidation of combustible gases. Hence, it took a longer time for WF/PP-M-APP-3 25% to reach the flame out and release less smoke. Similar result can be obtained from the TSP in Figure 2(e). The results of HRR, THR, mass loss (%), SPR, and TSP showed that M-APP 25% was the better flame retardant in WF/PP composites compared with the APP.

Morphological characterization

To further investigate effect of the APP and M-APP on the char formation of WF/PP composites during combustion, the morphologies of chars obtained from the sample extinguished during cone calorimeter test were examined by SEM. Figure 3 showed the morphologies of the chars of WF/PP-APP 25% (a) and WF/PP-M-APP-3-25% (b). From SEM pictures, it can be seen that for WF/PP-M-APP 25%, the surface was covered by carbonaceous char. The carbon layer is much more intumescent than that of WF/PP-APP 25% and the formation of this carbonaceous layer could create a higher physical barrier toward process of fire along the sample, indicating that 3-(methylacryloxy)propyltrimethoxy silane can improve the compatibility and the flame-retardant properties of the WF/PP composites. So that the dispersion of APP was increased, leading to the formation of more intumescent char layers. With the presence of efficient protective char, the amount of heat transferred to PP matrix and combustible gas escaping from the matrix decrease greatly, so the further combustion of inner PP is prevented. Thus, the flame retardant property of APP was improved with the addition of 3-(methylacryloxy) propyltrimethoxy silane. The network structure of carbon layer is the dominant reason for its advantageous influence on the flame retardancy. This result is

TABLE VI
Residue Components of WF/PP-APP 25% (a) and
WF/PP-M-APP-3 25% (b)

| Elements | a-Wt% | a-At% | b-Wt% | b-At% |
|----------|-------|-------|-------|-------|
| C | 86.97 | 91.68 | 43.04 | 54.91 |
| P | 5.21 | 2.13 | 18.68 | 9.24 |
| O | 7.82 | 6.19 | 36.68 | 35.14 |
| Si | 0 | 0 | 0.61 | 0.34 |

consistent with cone analysis. This result suggested that the interactions of WF/PP-M-APP may play an important role in improving not only mechanical properties, but also flame retardant.

The analysis of components in char layer with energy dispersive spectrometer (EDS) was shown in Table VI. It can be seen that for WF/PP-M-APP-3 25% system, silicon existed in carbon layer which was propitious to slow down the heat and mass transfer between the gas and the condensed phases, and the amount of phosphoric and oxygen species were larger, probably due to the formation of Si—O—P, which can produce incombustible material to prohibit the oxygen into carbon layer, and improve the flame-retardant properties of WF/PP composites.

CONCLUSIONS

The purpose of this article was to investigate the effect of APP and M-APP on the fire retardancy of WF/PP composites. The modified effect of APP with KH-570 in the flame retardancy, mechanical properties, and thermal degradation of WF/PP composites has been studied. The optimal content of KH-570 in this study is 1.5% with the increase of the mechanical properties. And M-APP as a flame retardant enhanced the mechanical properties of WF/PP composites, such as tensile strength, flexural strength, and Izod impact strength; while addition of APP in WF/PP composites reduced mechanical properties of WF/PP composites. The TGA and DTA showed that the 25% M-APP systems can make more char formation and have a higher thermal stability compared with 25% APP. The results of the cone calorimeter show that the Pk-HRR, THR, mass loss,

SPR, and TSP decreased significantly with the addition of 25% M-APP. The Pk-HRR decreased 21% from 196.2 to 154.9 kW m⁻² compared to that of adding APP. The SEM photographs demonstrate that in the presence of M-APP, the residual char is more intumescent. All the results showed that M-APP has good compatibility and good flame retardancy ability in WF/PP composites. The addition of M-APP may be one of the most promising ways for designing new efficient flame retardancy in WF/PP composites.

References

- Clemons, C. *Forest Prod J* 2002, 25, 10.
- Jacob, A. *Reinf Plast* 2006, 50, 32.
- Ashori, A. *Bioresource Technol* 2008, 99, 4661.
- Lei, Y.; Wu, Q. L. *Bioresource Technol* 2010, 101, 3665.
- Bengtsson, M.; Oksman, K. *Compos A Appl S* 2006, 37, 752.
- Harper, D.; Wolcott, M. *Compos A Appl S* 2004, 35, 385.
- Guo, C. G.; Wang, Q. W. *J Appl Polym Sci* 2008, 109, 3080.
- Karmarkar, A.; Chauhan, S. S.; Modak, J. M.; Chanda, M. *Compos A Appl S* 2007, 38, 227.
- Nachtigall, S. M. B.; Cerveira, G. S.; Rosa, S. M. L. *Polym Test* 2007, 26, 619.
- Kim, H. S.; Lee, B. H.; Choi, S. W.; Kim, S.; Kim, H. J. *Compos A Appl S* 2007, 38, 1473.
- Lai, S. M.; Yeh, F. C.; Wang, Y.; Chan, H. C.; Shen, H. F. *J Appl Polym Sci* 2003, 87, 487.
- Bengtsson, M.; Gatenholm, P.; Oksman, K. *Compos Sci Technol* 2005, 65, 1468.
- Seymour, R. B. *Polym Degrad Stabil* 2010, 95, 1903.
- Bourbigot, S.; Bras, M. L.; Duquesne, S.; Rochery, M. *Mater Eng* 2004, 289, 499.
- Chou, C. S.; Lin, S. H.; Wang, C. I. *Adv Powder Technol* 2009, 20, 169.
- Li, B.; He, J. M. *Polym Degrad Stabil* 2004, 83, 241.
- Liu, Y.; Wang, D. Y.; Wang, J. S.; Song, Y. P.; Wang, Y. Z. *Polym Adv Technol* 2008, 19, 1566.
- Ma, Z. L.; Zhao, M.; Hu, H. F.; Ding, H. T.; Zhang, J. *J Appl Polym Sci* 2002, 83, 3128.
- Wang, M.; Hu, Y. C. *Plastic Technol* 2010, 38, 104 (In Chinese).
- Lv, P.; Wang, Z. Z.; Hu, K. L.; Fan, W. H. *Polym Degrad Stabil* 2005, 90, 523.
- Zhang, Y.; Zhang, D. P. *China Annual Report for Flame Retardant*, 2006 (In Chinese).
- Liu, Y.; Wang, Q. *Polym Degrad Stabil* 2006, 91, 2513.
- Wang, X. G.; Li, Y.; Liao, W. W.; Gu, J.; Li, D. *Polym Adv Technol* 2008, 19, 1055.