



Thermal conduction and fire property of glass fiber-reinforced high impact polystyrene/magnesium hydroxide/microencapsulated red phosphorus composite



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ABSTRACT

A series of flame-retarded high impact polystyrene/magnesium hydroxide/microencapsulated red phosphorus (HIPS/MH/MRP) composites reinforced with different amounts of glass fiber (GF) were prepared by melt compounding. The thermal conduction behavior of the GF-reinforced HIPS/MH/MRP composite before ignition was studied by embedding thermocouples inside the composite at different depths along the thickness direction. The fire property of the composite was investigated by limiting oxygen index (LOI), UL-94, cone calorimeter test (CCT), scanning electron microscopy (SEM) and thermogravimetric analysis (TGA). It has been shown that the presence of GF promotes thermal conduction from surface area to the interior in the composite and the thermal inertia of the composite increases with GF loading. Because of increase in thermal conductivity and decrease in specific heat capacity, the heat on the sample surface can be transferred to the interior of composite more quickly than neat polymer and the temperature near the surface area of the GF-containing composite is reduced appreciably, which leads to delayed thermal degradation of polymers and increased difficulty of ignition. The incorporation of GF can increase the thermal stability, flame retardancy and smoke suppression of this composite simultaneously. The introduction of 2 wt% GF can hinder melt dripping of the HIPS/MH/MRP (100/30/10) composite and upgrade the UL-94 rating of this composite from V-1 to V-0. Thermal degradation of the polymer is retarded and no wicking effect occurs in this composite. Furthermore, the composite containing GF produces more residue and less smoke upon combustion. On the whole, the addition of GF not only increases thermal conduction and mechanical property, it also further improves fire safety of the flame-retarded HIPS/MH/MRP composite. The optimal mass fraction of GF is around 30 wt%.

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1. Introduction

In recent years, there has been increasing concern worldwide over developing halogen-free flame-retardant (HFFR) polymer materials because the halogen-containing flame retardant materials produce heavy smoke and corrosive emission products during combustion. Extensive work has been performed to endow polymer materials with good flame retardancy [1–7]. In these studies, some cheap and cost-efficient flame retardants, such as magnesium hydroxide (MH), aluminum trihydrate (ATH), expandable graphite

(EG) and microencapsulated red phosphorus (MRP) were combined in a proper manner to impart flame retardancy to polymers. However, in many cases, the mass fraction of flame retardant (FR) incorporated in the polymer composite is up to 30 wt% or more in order to achieve satisfactory flame retardancy. Such high loading deteriorates the mechanical property of the composite because of the poor interfacial adhesion between polymer matrix and FRs. In some circumstances, the obtained flame-retarded polymer composites may even fail to function due to poor mechanical strength.

In order to overcome this deficiency, fiber reinforcement may be introduced to the flame-retarded composite. In this aspect, glass fiber (GF) is the most widely studied and used fiber reinforcement owing to its low cost, good thermal and chemical stability and excellent mechanical performances. Nonetheless, preparing an efficient HFFR GF-reinforced polymer composite is still a problem

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because of the wicking effect caused by GF. Although the GF (wick) filled in the polymer matrix doesn't burn in itself, it usually promotes burning of the polymer. As a result, the fire retardancy of the GF-filled composite is even worse than that of neat polymer [8–10]. So far, there have been some publications concerning GF-filled flame retardant polymer composites. The polymer matrices investigated include polypropylene [10], poly(1,4-butylene terephthalate) [11,12], poly(ethylene terephthalate) [13–15], polyamide [16–20], epoxy [21], etc. These studies indicate that the wicking effect caused by GF can be overcome through charring of the polymer composite upon burning. As the phosphorus-containing flame retardants (FRs) can promote charring of the polymer at high temperature, they are widely investigated in flame-retarded GF-reinforced polymer composites. As nanoclay is not only cheap and abundant in source, but also has good charring ability at high temperature, it is often combined with the phosphorus-containing FRs to prevent the occurrence of wicking effect.

On the other hand, as the combustion of polymer materials always starts from thermal degradation and ignition of polymers on surface area and the spread of fire is essentially a process of repeated ignition of the material, the ignitability of the material plays a crucial role in the flame retardation of polymer materials. Thus, the ignitability of polymers has acquired increasing attention in the study of flame-retardant polymer materials in recent years [22–26]. Raising the difficulty of ignition is undoubtedly propitious to fire safety of the materials. As the ignition of polymer materials results from reaching a critical mass loss rate or reaching a characteristic ignition temperature, the surface temperature of the polymer sample is vital to its ignition. If the heat coming from external heat source can be conducted to the interior or other parts of the material effectively, the temperature near the surface area will be reduced. This will hinder thermal degradation of the polymer on the surface, diminish production of combustible gases and retard ignition of the polymer material. Namely, the time to reach thermal degradation temperature on the sample surface can be postponed by increasing thermal conduction of the material. Therefore, the thermal conduction behavior of the polymer composite has great influence on its ignition and flame retardancy. There have been several papers discussing the influence of thermal conduction on the time to ignition (TTI) of polymer composites [27–30]. It has been shown that thermal conductivity and heat absorption of the material yield opposite effects on ignition times. An increase in thermal conductivity increases thermal conduction from high temperature sample surface to the interior of the sample and thereby delays the onset of sample mass loss and prolongs the ignition time. The ignition time of polymer materials usually increases with the values of thermal conductivity and thermal inertia. As reported in the literature [31,32], in order to study temperature change inside polymer materials, thermocouples were embedded inside the composites to measure temperature inside burning specimen, which was radiated either by the cone heater in a cone calorimeter or by a heated electrical panel.

As far as the GF-filled flame retardant polymer composites are concerned, as GF itself has different properties from the other ingredients in the composite, the polymer composite containing GF will display a series of different properties, such as heat capacity, thermal conductivity and viscosity, from the virgin polymer. Thus, the presence of GF inevitably has some influence on the thermal conduction and fire behaviors of the flame-retarded polymer composites. Nevertheless, to the best of our knowledge, there has been no report on thermal conduction of GF-reinforced flame-retarded polymer composite prior to ignition in the literature. In this paper, high impact polystyrene (HIPS) was used as polymer matrix and MH and MRP were chosen as FR. Different amounts of GF were introduced to the flame-retarded HIPS/MH/MRP

composite with a mass ratio of 100/30/10. A series of thermocouples were embedded at different depths of the polymer sample along its thickness direction to detect the internal temperatures of the polymer composite upon heated by the external heat source via different modes (direct contact mode or radiation mode). By measuring temperature change at different depths along the thickness direction prior to ignition, the influence of GF on the thermal conduction and ignition of the various composites can be obtained. In comparison with what reported in the literature [31,32], the emphasis of this work is to investigate the thermal conduction before ignition. This experimental method to characterize thermal conduction not only applies to GF-reinforced flame-retarded polymer composite, it can also be used to other polymer composites. The result of this work has clarified the relationship between thermal conduction and ignition in fiber-reinforced polymer composites.

2. Experimental

2.1. Materials

High impact polystyrene (HIPS), with a brand of PH-88 and melt flow rate of 11.9 g/10 min (255 °C, 2.16 kg), was purchased from Zhenjiang Qimei Chemical Corporation (Jiangsu, China). Magnesium hydroxide (MH), with an average particle size of 3 μm and modified with silane coupling agent (dodecyl trimethoxysilane), was supplied by Jinan Chenxu Chemical Corporation (Shandong, China). Microencapsulated red phosphorus (MRP), with an average particle size of 19 μm, was obtained from Lianyungang Pengrui Chemical Company (Jiangsu, China). Chopped glass fiber (GF), with a brand of 534A and an average diameter of 13 μm and length of 4.5 mm, was provided by Jushi Group Corporation, China.

2.2. Sample preparation

HIPS/MH/MRP/GF composites with different compositions were prepared by melt-mixing HIPS resin, the MH and MRP flame retardants and GF on a two-roll mill at a temperature of 180 °C for 15 min, followed by hot-pressing the as-prepared HIPS composites under 10 MPa for 10 min at 180 °C into sheets of 3 mm thick, which were subsequently cut into suitable sizes for various analyses. The composition of the various samples is listed in Table 1, where it can be seen that the mass ratio of HIPS/MH/MRP in all the samples is fixed at 100/30/10 and the mass fraction of GF ranges from 0 wt% to 40 wt%.

The HIPS/MH/MRP/GF composites embedded with thermocouples at different depths along the thickness direction were prepared according to the following procedure. First of all, the above-mentioned HIPS-based composites were hot-pressed in two different circular molds under 10 MPa for 5 min at 180 °C into sheets with a diameter of 80 mm and a thickness of 3 mm and 1 mm, respectively. The thickness of the sheet was controlled by the

Table 1
The composition of different composites.

Sample code	Mass ratio of HIPS/MH/MRP/GF	Mass fraction of GF (wt%)
0%	100/30/10/0	0
1%	100/30/10/1.4	1
2%	100/30/10/2.9	2
5%	100/30/10/7.4	5
7%	100/30/10/10.5	7
10%	100/30/10/15.6	10
30%	100/30/10/60	30
40%	100/30/10/93.3	40

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