

# Halogen Free Flame-Retardant Glass Fiber Reinforced PA 66 / PPO Alloys

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## Research Article

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# Abstract

Halogen-free flame retardants are ideal plastic additives that meet carbon neutral requirements. In this work, halogen free flame retardant with glass fiber reinforced PA66/PPO composites were prepared by using coated red phosphorus (FRM-150B) and phosphorus-silicon flame retardant (WR6002). The mechanical properties, heat resistance, friction and wear properties and flame retardancy were carried out to evaluate the performances of composites using as structural parts that require heat resistance, dimensional stability and accuracy. It was found that the friction coefficient and wear volume of the composites were improved with the contents of glass fiber increased, as a result, PA66/PPO composites was obtained with excellent comprehensive performance when the content of compatilizer is 7%, the glass fiber was 30%, the content of FRM is 8% and the phosphorous-silicon flame retardant is 16%. The flame retardant effect of FRM-150B and WR6002 in PA66/PPO was presented in the condensed phase, the results showed that the composite material with 16% WR6002 forms a carbon layer with excellent thermal stability. On the other hand, the mechanical properties of composites were hardly affected, has important prospects in automotive components and household appliances

## 1. Introduction

Polyphenylene Oxide (PPO) also known as poly (2, 6-dimethyl-1, 4-phenylene oxide), is one of the five major engineering plastics with good thermal stability and mechanical strength. Polyphenylene ether has flame-retardant properties due to its molecular structure, and the oxygen index is 28<sup>[1-2]</sup>. The glass transition temperature (T<sub>g</sub>) of polyphenylene ether is 210°C, which can maintain its good mechanical properties, flame retardant properties and electrical properties in a large temperature range. The dielectric properties of PPO are the best among the five engineering plastics<sup>[3-5]</sup>. In addition, the dimensional stability of polyphenylene oxide is good at plastic cage, but the disadvantage of polyphenylene oxide is that it has a large melt viscosity and it is difficult to process. And the products are prone to stress cracking due to residual stress.

Nylon 66 is a thermoplastic, one of the five major engineering plastics, too, and it mostly used in electronic devices and automotive parts due to its excellent mechanical properties, outstanding molding characteristics<sup>[6]</sup>, good chemical resistance and electrical insulating features<sup>[7-12]</sup>. However, the oxygen index of nylon 66 is only 24, which belongs to the flammable class. The flame retardancy of nylon 66 is very poor, and it will burn with the release of smoke and severe melting and dripping during the combustion process, which is the reason that greatly limits its application range<sup>[13-14]</sup>. However, nylon 66 has excellent processing fluidity due to it contains a large number of amide bonds in its main chain<sup>[15]</sup>. Moreover, with low melt viscosity. But the high-water absorption is the major disadvantage of nylon 66.

PPO and PA66 has a similar melting point. The high melt viscosity of PPO could be deduced by incorporating PA66. However, the mechanical properties of the simple blend need to be improved due to the poor interface compatibility between PA66 and PPO. Therefore, some compatibilizers, such as high

impact polystyrene and styrene-butene copolymer, were introduced to enhance the compatibility of the PPO/PA66 [2-3, 5].

Although pure polyphenylene oxide will extinguish by itself within a period of time after ignition. However, the flame-retardant performance of the materials would be greatly reduced. after blended with PA66. It is necessary to perform flame retardant modification treatment on composites [7]. In order to meet the market's requirements for flame retardant performance in electronic devices and other aspects. Though halogen-based flame retardants still occupy the main market, its threat on environment and human health has attracted more and more people's attention.

As the country pays more and more attention to environmental protection, halogen-free flame retardants such as phosphorous flame retardants, nitrogen flame retardants etc. rise in response to the proper time and conditions [10-14, 16].

In this study, the coated red phosphorus and phosphorus-silicon flame retardants were blended into PA66/PPO, respectively. The glass fibers were added during the extrusion of the composites to strengthened the mechanical properties. Then, a halogen-free flame-retardant glass fiber reinforced PA66/PPO with excellent comprehensive performance was prepared [18-29]. The oxygen index test, vertical combustion test, friction and wear performance, mechanical properties and thermal properties were carried out to appraise effect of blending.

## 2. Experimental

### 2.1 Materials and chemicals

PPO (polyphenylene oxide) was provided by Nantong Xingchen Synthetic Materials Co., Ltd. China. Polyamide66 (PA66) was purchased by Ping-dingshan Shenma Engineering Plastics Co., Ltd. China. High impact polystyrene (HIPS) was supplied by Shanghai SECCO Petrochemical Co., Ltd. Maleic anhydride was bought by Tianjin Komio Co., Ltd. Non-alkali non-twisted glass fiber provided by Jushi Group Co., Ltd. The coated red phosphorus (FRM-150B) and Phosphorus-silicon flame retardant (WR6002) was provided by Zibo Wanrong Chemical Co., Ltd. HIPS-g-MAH was made by the laboratory.

### 2.2 Synthesis of HIPS-g-MAH

The HIPS resin was dried in a vacuum drying oven at 95°C for 12 hours, and then the HIPS resin and maleic anhydride were melt-blended in an internal mixer at 180°C for 8 minutes at a speed of 20 r/min. Finally, the melt-blended polymer was irradiated with a dose of 10 kGy in the gamma rays emitted by the <sup>60</sup>Co source.

### 2.3 Preparation of PA66/PPO composite

PA66 and PPO were dried under a vacuum oven at 110°C for 4 hours prior to extrusion. The required additives and flame retardant materials were uniformly mixed in different proportions. And the additives,

PA66 and PPO were extruded through a Giant SHJ-36 twin-screw extruder and pelletized. The pellets were dried in a vacuum oven at 120°C for 4 hours and then injection molded into samples with an injection molding machine (PL860/260, Wuxi Haitian Machinery Co., Ltd.). The detailed processing conditions for the extrusion and injection molding are listed in Table 1.

Table 1 A  
Processing conditions

Composition		Barrel temperature (°C)								
PA66/PPO	GF	1	2	3	4	5	6	7	8	Export
75/20	0	245	255	260	260	265	265	270	270	265
	30%	275	275	270	270	265	265	265	265	265

Table 1 B  
Injection conditions

Composition		Barrel temperature (°C)				
PA66/PPO	GF	1	2	3	4	5
75/20	0	257	262	265	260	255
	30%	267	272	275	270	265

## 2.4 Characterization

The limit oxygen index (LOI) values were measured by a JF-3 digital oxygen index meter (Nanjing Jionglei Instrument Equipment Co., Ltd.) with sheet dimensions of 80 mm×10 mm×4 mm according to GBT 2406.1–2008. The UL-94 vertical burning level was tested on a 5402 horizontal vertical burning analyzer (Suzhou, China) according to GB/T 2408 – 2008 and the dimensions of all samples were 130 mm×13 mm×3 mm. The tensile strength and bending strength were conducted on an electronic universal test system according to GB/T 1040-92 using the (RGM-3010, Shenzhen Rigel Instrument Co., Ltd.) with the spline size was 80 mm×10 mm×4 mm. The tensile strength test rate was 20 mm/min, and the bending strength test rate was 2 mm/min with the test span was 64 mm. The notched impact strength is tested by the izod impact tester according to the standard of GB/T 1843–2008, the spline size was 80 mm×10 mm×4 mm, the notch shape is a V-shaped mouth of 45°, There are five patterns in each group, and the average value is calculated. The morphology of the fracture surface obtained after broken by izod impact tester were observed by scanning electron microscopy (SEM, S-4800 Japan) at an accelerating voltage of 15 kV. The surface of samples was sputter-coated with gold before examination. The friction coefficient and wear rate were conducted on a High-speed reciprocating friction and wear testing machine according

to GB/T3960-2016 using the (MGW02, Lanzhou Zhongke Kaihua Technology Development Co., Ltd.) with the spline size was 80 mm×10 mm×4 mm. And the load of the sample is 10 N, the running speed was 500 rpm, the reciprocating friction time was 1h, the wear radius was 3 mm. The thermogravimetric analysis (TGA) was carried out on the STA-409 PC synchronous thermal analyzer (NETZSCA, Germany) using a heating rate of 10°C/min in nitrogen atmosphere. The heat distortion temperature (HDT) was tested according to GB/T 1642–2004 by Thermal Deformation Vicat Softening Temperature Tester (HDT/V1002, Chengde), the load was 1.80 MPa, the heating rate was 2°C / min.

## 3. Results And Discussion

### 3.1 Flame retardancy of PA66/PPO/FRM blends

Although numerous tests have been developed to examine the flame retardant properties of blends, the vertical burning test (UL-94) and LOI test are the most commonly used methods for testing the flame retardant properties of polymer materials. The limit oxygen index (LOI) and vertical burning testing (UL-94) were carried out to investigate the flame retardancy of the PA66/PPO/FRM blends. The corresponding results were listed in Table 2 and Table 3, respectively. Obviously, the PA66/PPO composites was highly flammable with a low LOI value (24 vol.%) and no rating at the UL-94 testing accompanied by severe melt dripping. It can be seen from Table 2 that the oxygen index of the material reached 39.5% when 10 parts of coated red phosphorus added, and the flame retardant passed V-0 rating. Because the appropriate red phosphorus was added to the composites, a membrane will be formed in the carbon layer to prevent external oxygen and some combustibles when the sample was burned, thereby an effective preventing combustion can be achieved. However, the limit oxygen index of the composites was decreased with more FRM added. This phenomenon maybe caused by the fact that red phosphorus itself was a kind of flammable material. it would release a lot of heat during combustion when too much red phosphorus added, which will make the material easy to burn, and decrease the flame retardancy of the composites.

WR6002 is a commercialized flame retardant that contains both phosphorus and silicon. Phosphorus and silicon can form a denser carbon layer during combustion, thereby achieving better effects in flame retardant. Phosphorus-based flame retardants and silicon-based flame retardants are non-toxic green flame retardants with good smoke suppression effects. They can form an expanded carbon layer during combustion to achieve heat insulation and oxygen isolation to prevent the burning of materials. Table 2, 3 and the Fig. 1 were the results of the flame retardant properties of PA66/PPO/30% GF with different content of phosphor-silicon flame retardants. The analysis of the table showed that as the number of phosphor-silicon flame retardants increases, the limiting oxygen index of the composite material gradually increase. When the glass fiber content was 30% and the flame retardant content is 20%, the limiting oxygen index of the material reaches the maximum 35.7%, and the flame retardant grade was V-0, which meets the flame retardant standard. However, in conjunction with the analysis of the mechanical properties of the material in Fig. 5, the overall performance of the materials is optimal when 30% glass fiber is added and the content of the phosphor-silicon flame retardant is 16%.

Table 2  
LOI and UL-94 test results of the PA66/PPO/FRM-150B blends

Sample	LOI (%)	UL-94
PA66/PPO/30%GF	25	No
PA66/PPO/30%GF/8%FRM	40.29	V-1
PA66/PPO/30%GF/10%FRM	39.488	V-0
PA66/PPO/30%GF/12%FRM	38.272	V-0
PA66/PPO/30%GF/14%FRM	36.65	V-0

Table 3  
LOI and UL-94 test results of the PA66/PPO/WR6002 blends

Sample	LOI (%)	UL-94
PA66/PPO/30%GF	25	No
PA66/PPO/30%GF/4%P-Si	31	V-2
PA66/PPO/30%GF/8% P-Si	33	V-1
PA66/PPO/30%GF/12% P-Si	33.6	V-1
PA66/PPO/30%GF/16% P-Si	34.6	V-0
PA66/PPO/30%GF/20% P-Si	35.7	V-0

## 3.2 The influence of compatibilizer on the properties of PA66/PPO composite

Compared with the query data, it was found that MAH has the characteristic peak of C = O of cyclic anhydride at about  $1850\text{ cm}^{-1}$  and  $1780\text{ cm}^{-1}$ , and the characteristic peak of carboxycarbonyl (C = O) after hydrolysis of cyclic anhydride at about  $1700\text{ cm}^{-1}$ . The graft compatibilizer HIPS-g-MAH has corresponding characteristic peaks of maleic anhydride in corresponding places, indicated that the graft compatibilizer has the characteristic peak of maleic anhydride, indicating that HIPS and MAH have successfully undergone the grafting reaction. Compared with pure HIPS, the purified reaction blend has three more characteristic peaks, and these peaks were just the characteristic peaks of maleic anhydride, indicated that the high impact polystyrene was grafted with maleic anhydride. The above infrared spectra prove that the grafting reaction between high impact polystyrene and maleic anhydride has formed the graft HIPS-g-MAH.

The previous work verified the influence of different contents of compatibilizer on the tensile strength and impact strength of the matrix material. The experiment shows that with the increase of the compatibilizer content, the tensile strength of the PA66/PPO composite material first increases and then decreases. When the content of the compatibilizer is 7%, the tensile strength of the material reaches 71.95 MPa. This is because HIPS-g-MAH is used as a compatibilizer. Maleic anhydride is a functional monomer. It will react with the terminal amine groups of PA66 to form HIPS-g-PA66 at the two-phase interface, which makes the interface layer thicker, reduces the interfacial tension and improves the adhesion. However, because the added amount of the compatibilizer is too high, unreacted compatibilizer may accumulate in the interface layer, which will affect the tensile properties of the material. The notched impact strength of the composite material increases with the increase of the compatibilizer content.

The measurement of thermal deformation temperature (HDT) and the measurement of tensile strength and impact strength were carried out to study the thermal and mechanical properties of PA66 and FRPA66. It can be seen from Fig. 2 that the tensile strength and impact strength of composites have been greatly improved with the increase of glass fiber content. When the glass fiber content is 30%, the impact strength of the composites reaches the maximum and the performance is optimal. It was obvious that the blending of glass fiber in the PA66/PPO matrix not only improved the bonding force of the interface, but the physical properties of the composites were improved. The matrix material is firmly bonded to the matrix after being blended with the glass fiber, and the material will preferentially transfer the load to the glass fiber when it was loaded. The interface plays the role of transferring stress and gradually spreading the load to the entire composites, greatly improved the load bearing capacity, and the size has become relatively stable.

It can be found from Fig. 3 that the thermal deformation temperature of the composites was 70°C when the glass fiber content was 0. With the addition of glass fiber, the heat distortion temperature of composites has been greatly improved, and with the increase of glass fiber content, the heat distortion temperature of composites has been continuously improved. The reason was that the glass fiber itself has high heat resistance, and after blending with the matrix material, it increases the cohesive force of the components of the composites. Moreover, the glass fiber plays a role in the heterogeneous nucleation of the composites, which allows the molecular chains of the matrix to be arranged in an orderly manner within a short distance, and the movement was hindered, so more energy was required to make the polymer chains slip and movement. Therefore, the overall heat resistance of the material was improved, and the heat distortion temperature is also greatly increased.

### **3.3 Friction and Wear Performance**

Polymers are more prone to adhesive wear when subjected to friction. Figure 4 shows the friction coefficient graph and the analysis graph of the wear rate of composites with different glass fiber content. It can be seen from Fig. 3 that with the glass fiber content increased from 0 to 10%, the friction coefficient of the composites decreased from 0.47 to 0.34, and the friction coefficient has been improved with the glass fiber content increased. The friction coefficient is 0.3, and the wear rate also drops from 14.4–1.1% when the glass fiber content is 30%. This is because glass fiber is a kind of wear-resistant material.

## 3.4 Mechanical properties of PA66/PPO/GF/FRM

The mechanical properties of microcapsule-coated red phosphorus applied to PA66/PPO composites were shown in Fig. 5. It can be seen that after only adding coated red phosphorus, the tensile strength of the composites did not change significantly with the increase of the coated red phosphorus content. However, the impact strength of the material began to decrease as the content of coated red phosphorus increased. Because after coated red phosphorus added to the composites, these red phosphorus particles were likely to become stress concentration points, which weaken the resistance of the material when subjected to external forces. With the content of coated red phosphorus increased, the stress concentration point also increases, resulting in a gradual decrease in impact performance. After 30% glass fiber added, the mechanical properties of the flame-retardant composites have been significantly improved. When the content of coated red phosphorus is 8%, the impact strength of the composites without glass fiber was  $5.8 \text{ kJ/m}^2$ , and the impact strength of glass fiber reinforced red phosphorus flame retardant material reached  $7.08 \text{ kJ/m}^2$ , increased 22%, a composite with excellent comprehensive mechanical properties was obtained.

Figure 6. Tensile properties (left picture) and impact properties (right picture) of composites with different phosphorus-silicon (WR6002) flame retardant content.

## 3.5 TGA and DTG of blends

In an inert atmosphere (nitrogen), the thermal decomposition processes on the TGA curves displayed for PA66/PPO, PA66/PPO/GF, PA66/PPO/GF/P-Si, and PA66/PPO/GF/FR composites in Fig. 7. It can be seen from the figure that although the mechanical properties of the material enhanced after the glass fiber added. However, due to the "candle heart effect" of glass fiber, the weight loss of the material was increased compared to PA66/PPO. The introduction of WR6002 and FRM into the PA66/PPO/GF was found to reduce the temperature of the onset decomposition temperature of PA66/PPO/GF. The weight loss of the material increased from 28.8–52% after adding phosphorous and silicon flame retardant. The  $T_{\max}$  of the PA66/PPO/GF was  $472^\circ\text{C}$ , but after WR6002 added, the  $T_{\max}$  was improved to  $504^\circ\text{C}$ . With the addition of flame retardants, the more stable and higher quality carbon layer was formed. It can be seen from the curve that the carbon residue rate of the material also improved with the increase of the flame retardant addition. This is because with the addition of the coated red phosphorus flame retardant and phosphorus silicon flame retardant, the material can form polyphosphate during combustion, which plays a role in blocking oxygen and heat transfer. WR6002 is a phosphorous silicon flame retardant. Due to the joint action of Si-O-Si and phosphorous elements, the isolation layer formed was more stable than red phosphorous, which achieved a better flame retardant effect.

## 3.6 Fracture appearance of blends

SEM images of notch impact surface of different samples after test were presented in Fig. 8. It can be seen from A and B that the particle size distribution between the two dispersed phases before adding the compatibilizer was small, and the distribution was relatively clear, the parabolic dimple phenomenon was

more obvious, and the two phases penetrate the uniform sea-island structure. With the addition of the compatibilizer, we can clearly see that the phase morphology of the blends is improved. The phase interface of the blends becomes blurred, which reduced the interfacial tension and refined the dispersed phase, resulting in enhanced material compatibility. The broken surface presents a co-continuous phase structure, and the thicker the two-phase interface layer, the more obvious the compatibilization effect, which is consistent with the trend of increasing mechanical properties. The C is the composites with glass fiber. From the D and E, it can be seen that a clear cavity structure after the glass fiber was broken. After the glass fiber was pulled out, it can be clearly seen that the adhesive materials were taken out, indicating that the glass fiber has improved the phase of the composites. Capacitance improves the mechanical properties of composites. After the flame retardant added, the morphology of the fracture site has changed significantly. It can be seen from Fig. 8C that the fracture surface of the composites after adding the flame retardant was bark-like, which will damage the mechanical properties of the blends.

Based on the above results, a flame-retardant mode of action was proposed (Fig. 9). The Si-O-Si will promote the degree of carbonization of flame-retardant materials in providing good flame retardancy of PA66/PPO blends. The cooperative of Si and P enhanced the stability of char layer, contributed to the phosphorus and silicon-rich cross-linked char, which play the flame retardant effect in condensed phase. In addition, released incombustible gases during the combustion, including ammonia and steam, could dilute the oxygen concentration and took the heat away, contributing to the gas-phase mode of action. Therefore, FRM exert the flame retardant effect both in gas phase and condensed phase.

## 4. Conclusions

In summary, by adding irradiated HIPS-g-MAH and glass fiber to PA66/PPO, the composite material was compatibilized and modified by glass fiber, and PA66/PPO was reinforced and modified. The composite material was modified by adding flame retardant (IFR) FRM-15B and WR6002 respectively. The flame retardant efficiency and mechanical properties of PA66/PPO have been significantly improved. Different contents of glass fiber and compatibilizer have a significant impact on the mechanical properties of PA66/PPO composites. According to the experimental data, when the compatibilizer content is 7% and the glass fiber content is 30%, the composite material can obtain the greatest interfacial adhesion and the best mechanical properties. As the content of red phosphorus coated in the composite material increases, the oxygen index of the composite material first increases and then decreases. When FRM-15B is added to the composite material, a glass-like isolation layer of metaphosphoric acid is formed on the surface of the material during combustion to block the combustion. When the content of FRM-15B is 10%, the flame retardant performance reaches the V0 level, the flame retardant performance is the best, and the impact on the mechanical properties of the composite material is minimal. After research, it is found that the oxygen index of the spline after adding glass fiber is higher than that of the spline without glass fiber, which proves that glass fiber can increase the flame retardant performance of the material while enhancing the mechanical properties of the material. Experiments show that the flame retardant ability of

phosphorous silicon flame retardant is weaker than that of coated red phosphorus. However, when the phosphorus-silicon flame retardant content is 16%, the composite material can also be extinguished in a short time. The flame retardant performance of the composite material after adding 16% phosphorous silicon flame retardant reaches the V0 level, and the carbonization after combustion and the isolation layer on the surface of the material are better than the blended material with red phosphorus. In addition, the overall performance of composite materials is better than that of adding coated red phosphorous materials.

## Declarations

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## Conflicts of Interest:

The authors declare no conflict of interest.

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## Figures



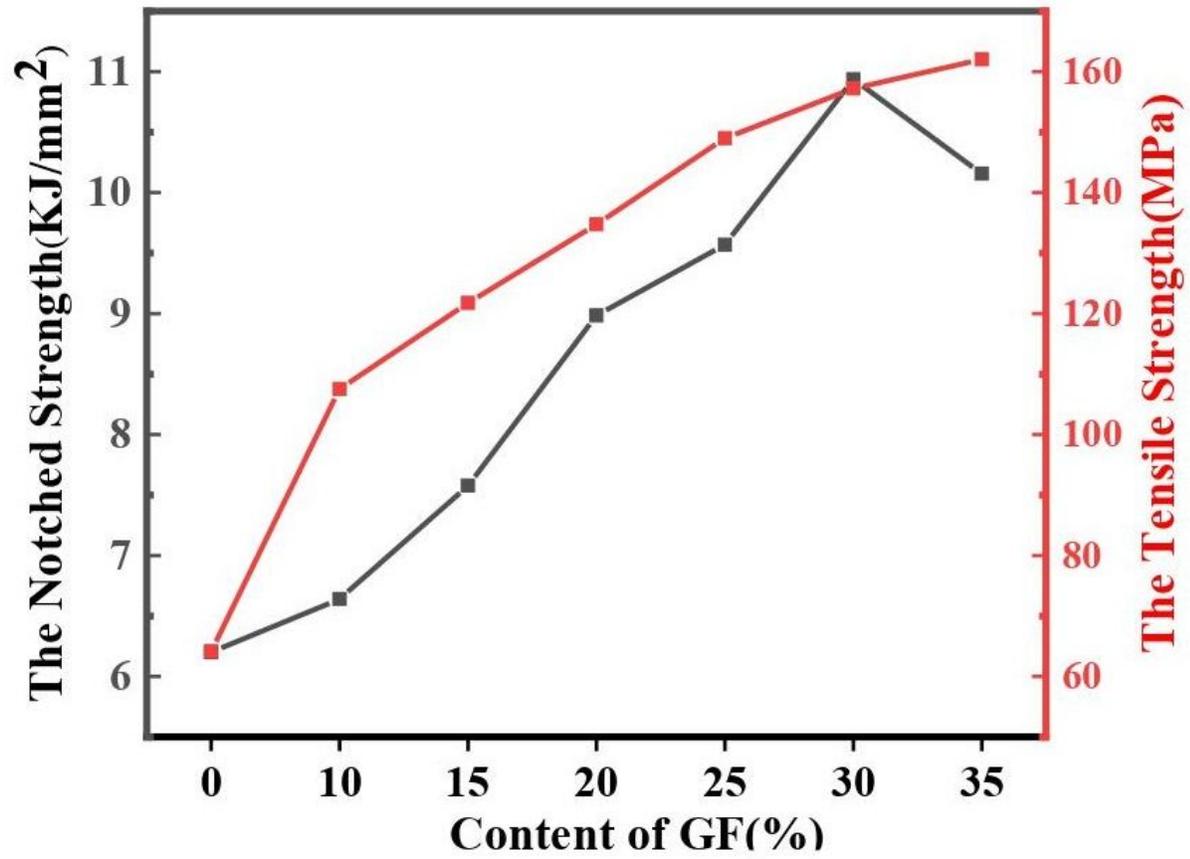


Figure 2

The influence of glass fiber content on the mechanical properties of PA66/PPO

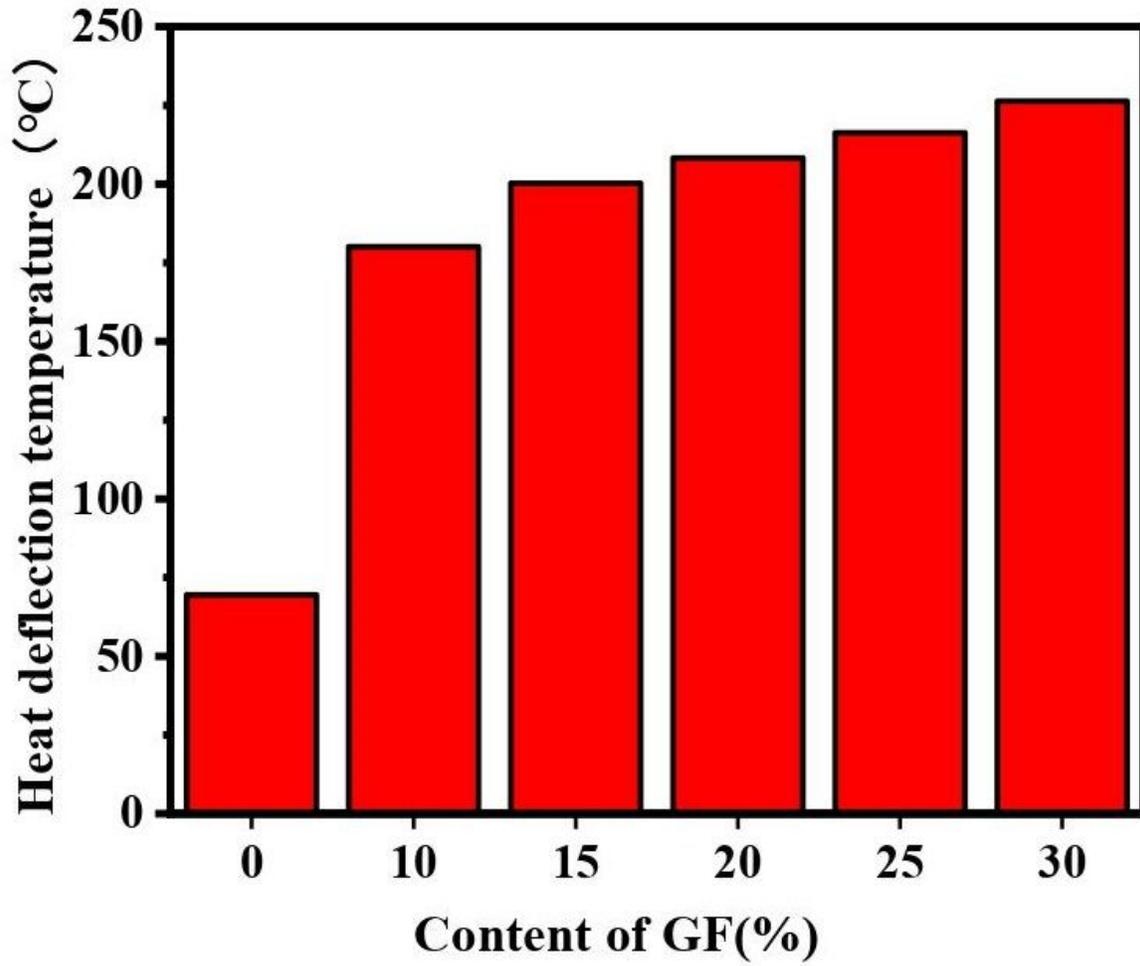


Figure 3

The influence of glass fiber content on the thermal deformation temperature of PA66/PPO

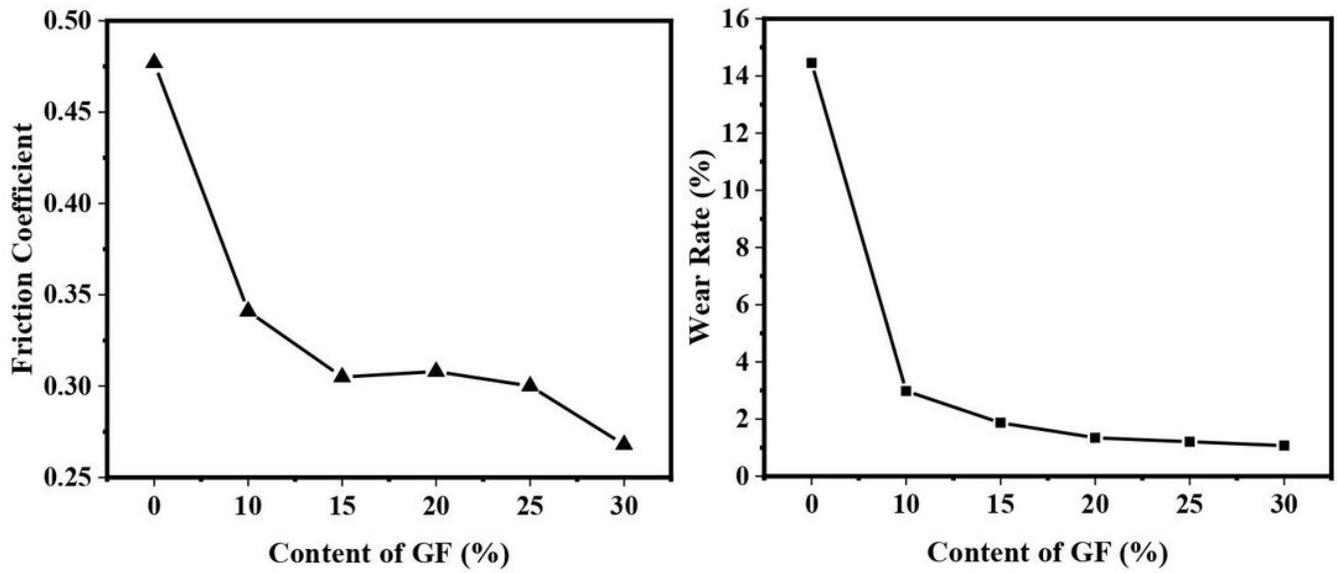


Figure 4

The effect of glass fiber content on the friction and wear properties of PA66/PPO

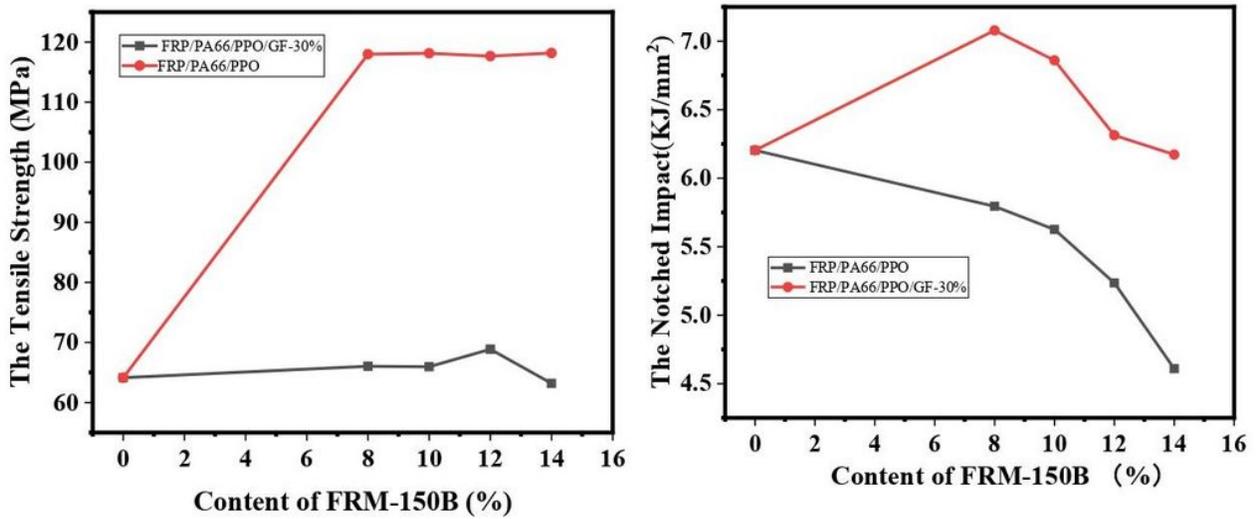


Figure 5

Tensile properties (left picture) and impact properties (right picture) of composites with different red phosphorus (FRM-150B) content

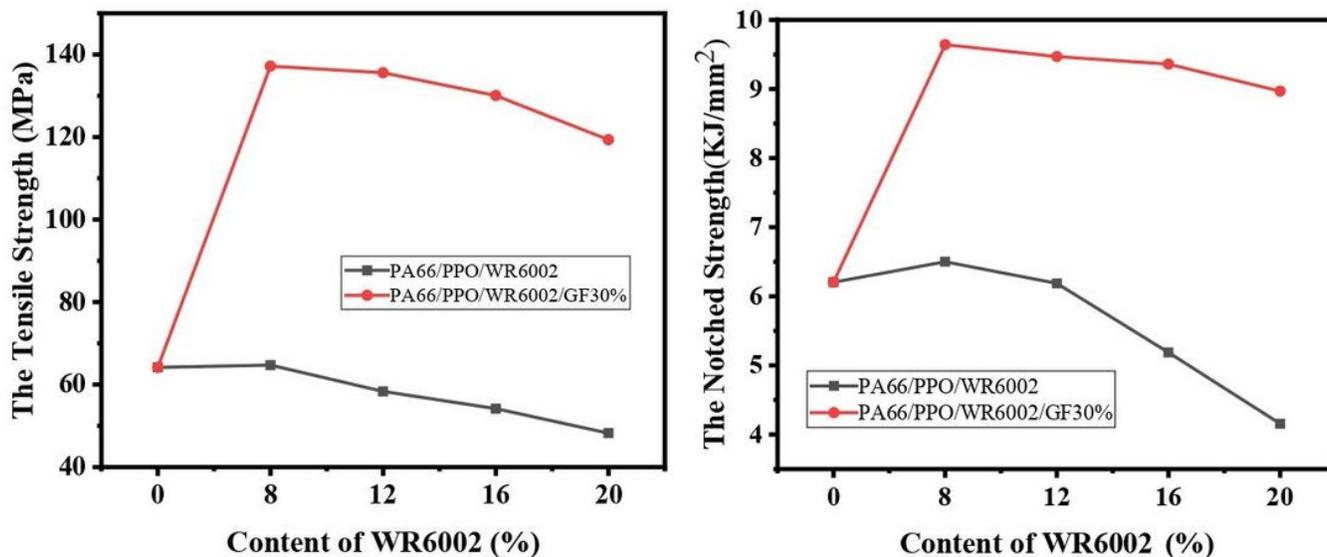


Figure 6

Tensile properties (left picture) and impact properties (right picture) of composites with different phosphorus-silicon (WR6002) flame retardant content.

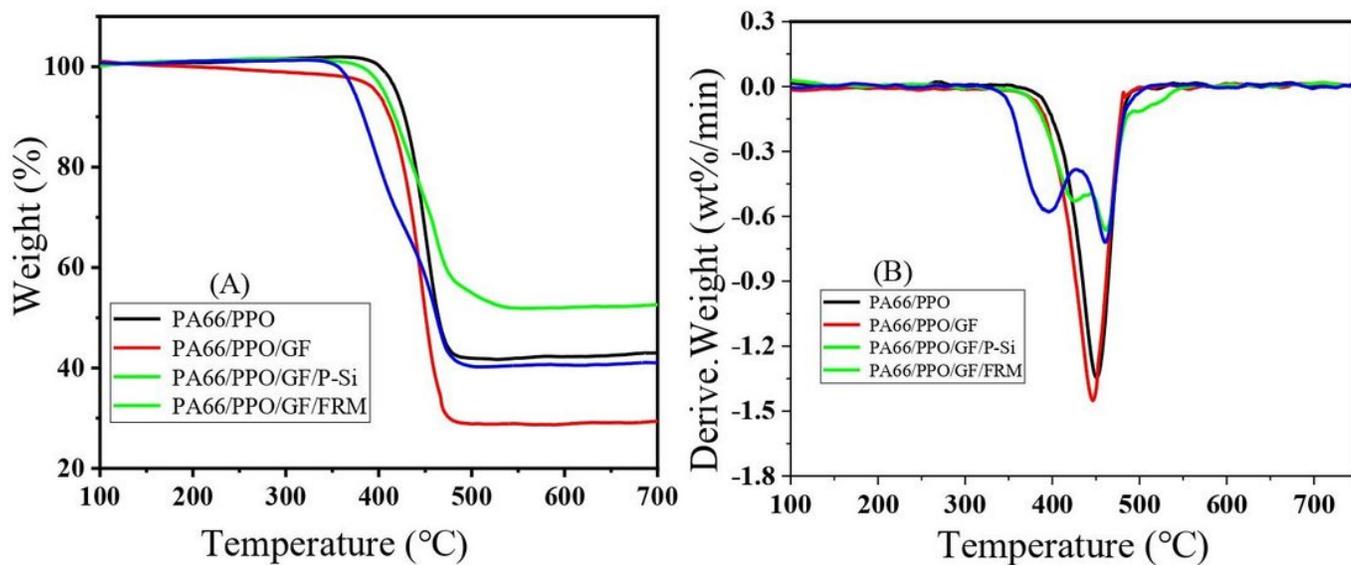


Figure 7

A, TGA and B, DTG curves of different samples.

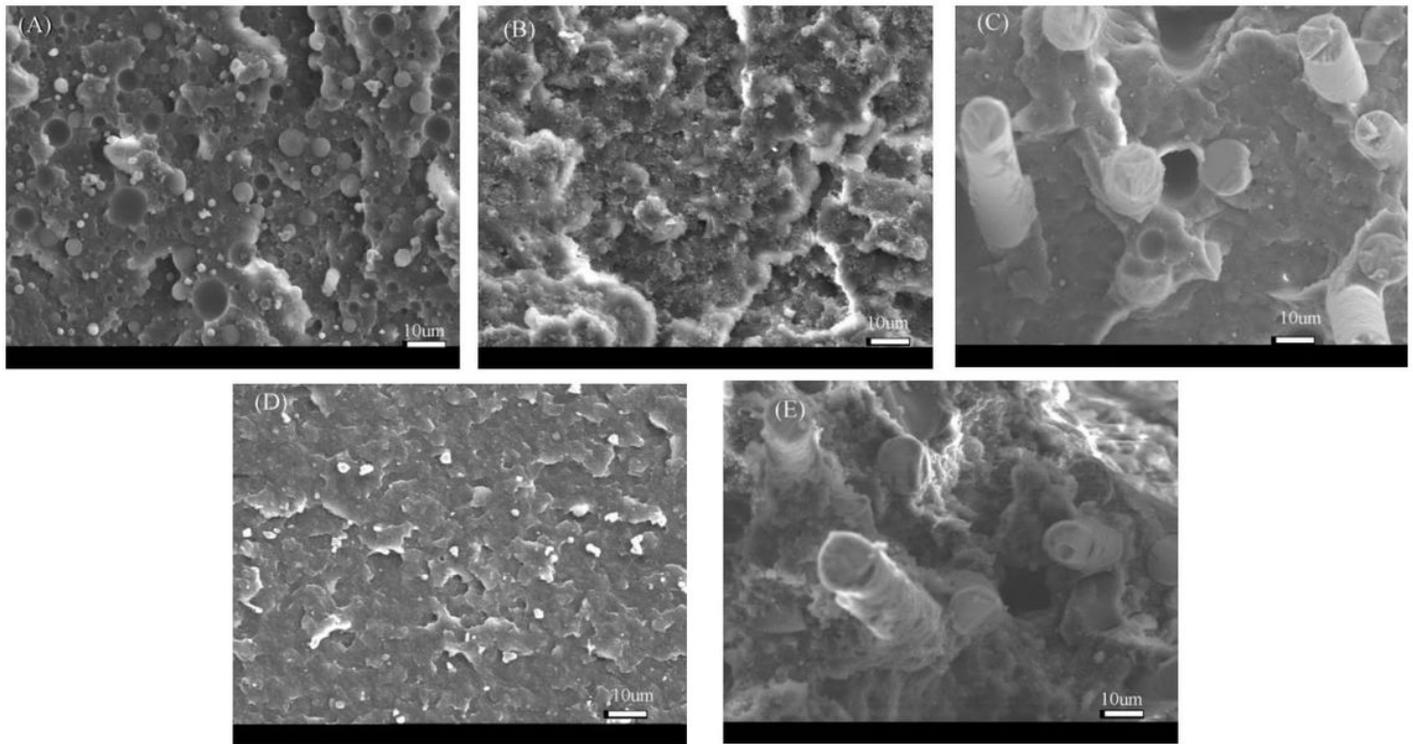
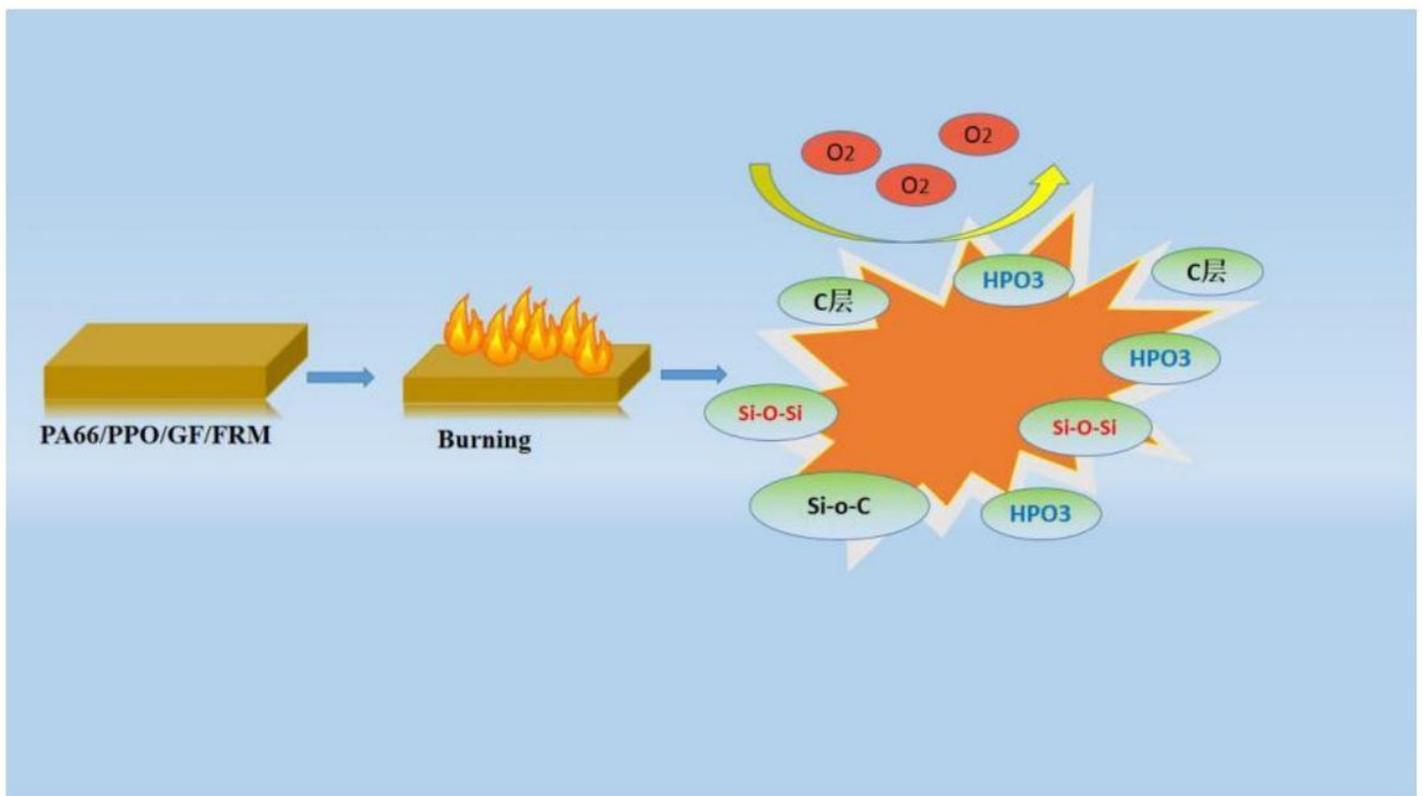


Figure 8

SEM images of Notch impact surface  
 A PA66/PPO  
 B PA66/PPO/HIPS-g-MAH  
 C PA66/PPO/HIPS-g-MAH/GF  
 D PA66/PPO/HIPS-g-MAH/WR6002  
 E PA66/PPO/HIPS-g-MAH/WR6002/GF



## Figure 9

The flame-retardant mechanism of PA66/PPO/FR composites

## Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

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