Materials Research Express

PAPER

Effect of expandable Graphite/Hexaphenoxycyclotriphosphazene beads on the flame retardancy of silicone rubber foam

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Keywords: silicone rubber foam, expandable graphite, hexaphenoxycyclotriphosphazene, FLAME retardancy, thermal stability, mechanical properties

Abstract

Silicone rubber foam (SiF) with EG/HPCTP was prepared by high-temperature vulcanization. The flame retardancy of SiF was evaluated using the LOI (limiting oxygen index), UL-94, cone calorimetry test (CCT), thermogravimetric analysis (TGA), and mechanical properties. The results showed that EG/HPCTP could improve the LOI of SiF, and the SiF could pass the UL-94 V-0 rating. Compared with pristine SiF, EG/HPCTP could reduce the total heat release rate (THR), heat release rate (PHRR). Digital images of the char residues showed that the HPCTP was beneficial to promote the strength of SiF with EG. TGA showed that the branched decomposition temperature and main chain pyrolysis temperature of SiF were delayed. Mechanical properties analysis showed that EG and HPCTP could improve the mechanical properties of SiF. These indicated that the addition of EG/HPCTP was a good approach to prepare high effective flame-retarding SiF.

1. Introduction

Silicone rubber foam (SiF) is a new type of flexible, porous and functional polymeric elastomer made of silicone rubber after foaming [1, 2]. SiF not only retains the excellent performance of silicone rubber but also has the properties of light weight, sound absorption, heat insulation, high specific strength, sealing, shock absorption, etc. As a high-performance material used in applications such as damping, shock absorption, sound absorption, and heat insulation, it is widely used in defense, aerospace, transportation, electronics, agriculture, and other fields. Some functional, high-performance foams have played a significant role in cutting-edge fields, such as those of the military and aerospace, and are among the most interesting materials in the field of polymer foams [3–7]. However, the flame retardancy of SiF has limited its wide application in some fields. First, the side chain of silicone rubber is a hydrocarbon or substituted hydrocarbon organic group. These groups are first degraded during combustion to form flammable small molecules that promote the combustion of silicone rubber [8–10]. Second, silicone rubber has a silicon–oxygen bond as the main chain, and hydrocarbon organic groups cannot form an effective carbon layer for protection. Instead, silicone rubber relies on SiO₂ generated by the decomposition of the silicon–oxygen bond to form a protective layer, which further accelerates the destruction of the silicone rubber structure [11]. Third, SiF has a porous structure and a large specific surface area, which increases the contact between the silicone rubber and air and promotes combustion. To further improve the flame retardancy of SiF, some flame retardants, such as montmorillonite [12, 13], calcium carbonate [14], magnesium hydroxide [15], EG [16], polymer ammonium phosphate [17], graphite oxide [18], rare metals [19], etc; however, inorganic clays, metal hydroxides, and conventional phosphorus-based compounds are limited due to a large amount of required addition and low flame retardancy. EG and cyclophosphazene flame retardants are widely used due to environmental protection and high flame retardancy.

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EG is a typical halogen-free flame retardant and a graphite intercalation compound. Research on EG as a flame retardant is developing rapidly [20]. At a high temperature, when the material contains EG, the decomposition of the compounds between the EG layers generates gas to separate the graphite, which expands approximately one hundred times in volume, forming a ‘worm’-like structure, preventing the flame from damaging the substrate [21, 22]. Chen et al [23] used EG to synthesize functional expandable graphite (FEG) and then added it to silicone rubber to improve its compatibility with the silicone rubber matrix, which formed an organic–inorganic hybrid flame retardant system. When the amount of FEG was 15 wt%, the LOI of SR/FEG reached 53.8. The peak value of heat release rate (PHRR), total heat release rate (THR), smoke production rate (SPR), total smoke production (TSP), and influencing factors of smoke can effectively reduce the combustion characteristics. Although EG has excellent flame retardant effects, the worm-like structure layer that formed during the combustion process usually lacked sufficient adhesion and was therefore easily destroyed by thermal convection and flame pressure during the combustion process [24]. There are multiple hydroxyl functional groups on the surface of EG, so it can react with compounds such as PO to form phosphate esters [25].

The HPCTP molecular structure contains a phosphorus-nitrogen alternating structure and a large number of benzene ring structures, which give it good flame retardancy, and HPCTP has good compatibility with polymers, is environmentally friendly and thermal stable and has high performance and excellent flame retardancy among other characteristics [26, 27]. HPCTP has been applied to polyethylene, viscose fibers, ethylene-vinyl acetate copolymers, and so on. Shen et al [28] introduced HPCTP into UHMWPE/magnesium hydroxide to solve the problem that UHMWPE does not melt with magnesium hydroxide. The ignition time was delayed by 75 s, and the residual carbon and LOI were improved. Wang et al [29] studied the effect of HPCTP on viscose fibers. The addition of HPCTP can improve the LOI of viscose fibers and form a carbon layer on the surface of the fibers to improve the flame retardancy of viscose fibers. Shen et al [30] studied magnesium hydroxide and HPCTP as flame retardants and studied the synergistic flame retardant ethylene-vinyl acetate copolymer with magnesium hydroxide and HPCTP. The introduction of HPCTP could reduce the use of magnesium hydroxide from 50% to 20%, and the flame retardancy of the ethylene-vinyl acetate copolymer was improved. Therefore, the defects of the EG flame retardant could be solved by providing PO compounds by HPCTP decomposition.

As EG could enhance the oxygen barrier and thermal insulation performance of SiF, HPCTP could increase the viscosity of the matrix material at high temperature. In this study, EG and HPCTP were introduced to fabricate flame-retardant SiF. This article explored the effects of EG, HPCTP, and EG/HPCTP on the flame retardancy of SiF through LOI, UL-94, and CCT. The flame retardant mechanism was investigated by TGA, and the effects of EG and HPCTP on the mechanical properties of SiF were discussed. These results could provide theoretical foundation for EG/HPCTP flame retardant products and support their application in new energy vehicle sealing materials.

2. Materials and methods

2.1. Materials

Liquid silicone rubber, with a viscosity of approximately 95000–100000 mPas, was kindly donated by Salirone New Materials Co. (Huizhou, China); the platinum catalyst had a platinum content of 3000 ppm was purchased from Shenzhen Osbang New Materials Co. (Shenzhen, China); the methyl hydrogen silicone oil had a hydrogen content of 1.4 and a viscosity of 18–25 cps and was purchased from Xi’an Daosheng Chemical Technology Co., Ltd Expandable graphite, with an average particle size of approximately 80 mesh, was purchased from Qingdao Yanhai Carbon Material Co., Ltd Hexaphenoxy cyclotriphosphazene was purchased from Jinan Taixing Fine Chemical Co., Ltd 2-Methyl-3-butyne-2-ol was purchased from Maclean’s Chemical Reagent Company. The formulations are given in table 1.

2.2. Preparation of samples

A total of 150.00 g of liquid silicone rubber was accurately weighed into a plastic cup, flame retardant was added, and the mixture was stirred for approximately 3 min after transfer to a double-roll open mill (ZG-250, Dongguan Zhenggong Electromechanical Equipment Technology Co., Ltd); at a roll distance of 2 mm, the sample was mixed evenly, and 4.0 g of methyl hydrogen silicone oil and 0.01 g of 2-methyl-3-butyne-2-ol were added in sequence. After mixing for approximately 10 min, 0.5 g of platinum catalyst was added, and mixing was continued for 5 min. Then, the double roller opening was adjusted to a roller distance of more than 10 mm, and the mixed silica gel was quickly removed and transferred to the constant-capacity grinding tool (the mold volume was 160.0 mm × 160.0 mm × 12.0 mm) at 120 °C in a programmable high-temperature oven (model: DHG-9010, Suzhou Guangjun Electronic Technology Co., Ltd) for 10 min; the gas produced by the
The hydrosilylation reaction was SiF, and a flame-retardant SiF with a flat surface was obtained, the preparation process is shown in figure 1.

In this experiment, hydrogen-containing silicone oil was used as a cross-linking agent, 2-methyl-3-butyn-2-ol was used as an inhibitor, platinum was used as a catalyst, and EG and HPCTP were used as flame retardants.

### 2.3. Characterization

The flame retardancy, thermal stability, morphology and mechanical properties of EG- and HPCTP-amended SiF were tested by a pull-up test, LOI, a UL-94 test, CCT, TGA, and mechanical property evaluation.

The LOI was determined by a JF-3 oxygen index meter (Nanjing Jiangning Analytical Instrument Co., Ltd) following ISO4589. The sample size was 150.0 mm × 10.0 mm × 10.0 mm. Each sample was tested 5 times, and the average was taken.

UL-94 was measured by a CZF-3 horizontal and vertical tester (Nanjing Jiangning Analytical Instrument Co., Ltd) according to the UL-94 standard. According to the ASTM D3801-2010 test standard, the sample size was 127.0 mm × 12.7 mm × 3.0 mm. Each sample was tested 3 times, and the average was taken.

The experiments were conducted using a cone calorimeter (Motis Fire Technology Co., Ltd, Kunshan, China) at an external heat flux of 35.0 kW m$^{-2}$ by the ISO 5660 standard. All sample sizes were 100.0 mm × 100.0 mm × 5.0 mm.

TGA was performed using a synchronous thermal analyzer (TGA/DSC) (METTLER TOLEDO Instruments, Switzerland). At a heating rate of 20.0 °C min$^{-1}$, 8.0 mg–10.0 mg of sample was heated from room temperature to 1000.0 °C in an air atmosphere.

The mechanical properties (the tensile strength and elongation at break) of the samples were determined on an electronic tensile testing machine (XZW5000N, Jiangsu Xinzenwei Testing Machinery Co., Ltd, China) with a constant speed of 100 mm min$^{-1}$ at room temperature. Each sample was tested 5 times, and the average was taken. All specimens were tested according to GB/T 528-1998 (China).

### 3. Results and discussion

#### 3.1. Analysis of char expansion

Figure 2 shows the residue of a series of SiF after combustion in the cone calorimeter test. The SiF-EG0%-HPCTP0% sample was completely brittle and severely cracked. After adding EG, a dense spherical silica protective layer was formed on the surface of the sample after burning, which effectively prevented the sample from burning, such as SiF–EG15%–HPCTP0%. The backs of the SiF–EG15%–HPCTP0% sample was intact and had a certain strength, indicating that the addition of EG could effectively inhibit the combustion of SiF. From the SiF–EG0%–HPCTP15% sample, after adding HPCTP, a dense silica protective layer was formed in the
middle of the surface of the SiF, and a little spherical protective layer was formed around the sample. However, from the back of the SiF-EG0%-HPCTP15% sample, it was completely broken. When EG and HPCTP were combined, the compactness of the silica protective layer on the surface of the sample was reduced and had a certain strength after combustion, as shown by SiF-EG7.5%-HPCTP7.5%. Compared with SiF-EG15%-HPCTP0%, the spherical coating on the surface of SiF-EG7.5%-HPCTP7.5% was discontinuous, because the pyrolysis products of HPCTP could increase the adhesion of between EG and SiF, suppress the pyrolysis of SiF. In addition, we found water stamps on the back of SiF-EG7.5%-HPCTP7.5% samples, which further showed that HPCTP improved the adhesion of EG, and EG/HPCTP possessed good synergistic flame retardant effect.

3.2. Flame retardancy
To evaluate the flame retardancy of SiF, the LOI and UL-94 tests were used. The LOI is the lowest oxygen content that can sustain the combustion of a material. The higher the LOI, the better the flame retardancy [31]. Table 2 shows the LOI and UL-94 test results of EG, HPCTP, and EG/HPCTP. A proportion of 5 wt% EG was able to make the LOI of SiF 38.4%, and the UL-94 test passed the V-0 rating. Compared with pristine SiF, the LOI increased by 47.7%. As the amount of EG continued to increase, the maximum LOI reached 40.2% when the amount of EG was 25 wt%, which was an increase of 54.6% compared with that of pristine SiF. When HPCTP was added, the LOI gradually increased with increasing HPCTP proportion. When HPCTP was added at 25 wt%, the maximum LOI was 30.0%; compared with pristine SiF, the
LOI increased by 15.4%, and the UL-94 test passed the V-0 rating. By comparing the addition of EG and HPCTP, EG was conducive to improving the LOI and UL-94 levels. This was similar to the research results by Shen, Chen, et al\cite{28, 30, 32}, and phosphazene flame retardants have limited contributions to LOI and UL-94. To further verify whether EG and HPCTP have a synergistic effect, the flame retardancy of different ratios of EG/HPCTP (15 wt%) on SiF was studied.

Table 2 shows that when the amount of added HPCTP was dominant, the LOI decreased rapidly. When the amount of EG/HPCTP was 1.5 wt%/13.5 wt%, the LOI was only 31.0. When a small amount of HPCTP was added, that is, when EG/HPCTP was 13.5 wt%/1.5 wt%, the LOI was the largest, reaching 44.0. Compared with the pristine SiF, the LOI increased by 69.2%, and compared with the EG addition amount of 15 wt%, the LOI increased by 10.3%. In addition, when the EG/HPCTP addition amount was 7.5 wt%/7.5 wt% or more, the LOI performance was better than that when only 15 wt% EG was added. The above results were due to the different flame retardant mechanism of EG and HPCTP. In SiF-EG15%-HPCTP0%, SiF-EG13.5%-HPCTP1.5% and SiF-EG7.5%-HPCTP7.5%, the condensed phase flame retardant mechanism of the EG played main role, and it expanded rapidly when be heated, which worked with the effects of heat insulation and oxygen barrier in short time. The LOI increased correspondingly. Moreover, the strength of the expanded coating layer increased

![Digital images of flame-retardant SiF samples.](image-url)
further after HPCTP added, but expansion rate decreased. Hence, LOI decreased with HPCTP increased, and LOI maintained optimal state at SiF-EG13.5%-HPCTP1.5%. The above analysis showed that adding a small amount of HPCTP can help improve the LOI of SiF, and EG/HPCTP exhibited a synergistic flame retardant effect.

3.3. Fire hazard analysis
CCT is an important tool for studying the flame retardancy of materials, and it can highlight key parameters in the combustion process. Compared with the LOI and UL-94, CCT can better reflect the true combustion state [33–35]. Therefore, the CCT was used to obtain the HRR, THR, fire performance index (FPI) and fire growth index (FGI) diagrams of the SiF, as shown in figures 3–4. Table 3 shows the ignition time (TTI), PHRR, heat release rate peak time (PHRRT), and THR.

Table 2. The mechanical properties and flame retardancy of silicone rubber foam.

| Sample                  | LOI/ (%) | UL-94 | Tensile strength/(KPa) | Elongation at break/(%) |
|-------------------------|----------|-------|------------------------|------------------------|
| SiF-EG0%-HPCTP0%        | 26.0 ± 0.3 | V-2   | 192.3 ± 16.7 | 399.9 ± 18.6 |
| SiF-EG5%-HPCTP0%        | 38.4 ± 0.2 | V-0   | 554.5 ± 17.3 | 681.4 ± 16.4 |
| SiF-EG10%-HPCTP0%       | 39.4 ± 0.2 | V-0   | 597.8 ± 17.2 | 649.1 ± 15.7 |
| SiF-EG15%-HPCTP0%       | 39.9 ± 0.2 | V-0   | 481.6 ± 19.8 | 621.7 ± 17.3 |
| SiF-EG20%-HPCTP0%       | 40.0 ± 0.3 | V-0   | 552.2 ± 17.3 | 757.5 ± 17.2 |
| SiF-EG25%-HPCTP0%       | 40.2 ± 0.3 | V-0   | 534.4 ± 16.9 | 767.4 ± 18.1 |
| SiF-EG0%-HPCTP5%        | 26.2 ± 0.2 | V-1   | 479.5 ± 16.4 | 777.6 ± 15.4 |
| SiF-EG0%-HPCTP10%       | 26.6 ± 0.3 | V-1   | 414.6 ± 17.2 | 455.3 ± 17.8 |
| SiF-EG0%-HPCTP15%       | 27.8 ± 0.4 | V-0   | 281.7 ± 16.6 | 469.0 ± 16.1 |
| SiF-EG0%-HPCTP20%       | 29.0 ± 0.3 | V-0   | 268.5 ± 17.8 | 453.0 ± 16.9 |
| SiF-EG0%-HPCTP25%       | 30.0 ± 0.2 | V-0   | 292.3 ± 17.4 | 330.3 ± 17.2 |
| SiF-EG13.5%-HPCTP1.5%   | 44.0 ± 0.4 | V-0   | 441.5 ± 16.4 | 427.3 ± 17.6 |
| SiF-EG10.5%-HPCTP4.5%   | 42.5 ± 0.2 | V-0   | 420.2 ± 19.3 | 535.69 ± 18.1 |
| SiF-EG7.5%-HPCTP7.5%    | 42.0 ± 0.3 | V-0   | 377.1 ± 18.6 | 477.7 ± 16.7 |
| SiF-EG4.5%-HPCTP10.5%   | 36.0 ± 0.2 | V-0   | 303.8 ± 16.7 | 528.6 ± 17.4 |
| SiF-EG1.5%-HPCTP13.5%   | 31.0 ± 0.2 | V-0   | 274.3 ± 17.3 | 700.4 ± 16.9 |

Figure 3. HRR curves of SiF composites at a flux of 35 kW m⁻².
3.3.1. Heat release test (HRR)

HRR, especially the PHRR, is an important parameter for evaluating fire safety, which can reflect fire intensity and determine other key parameters [36, 37].

Figure 3 shows the HRR of SiF-EG0%-HPCTP0%, SiF-EG15%-HPCTP0%, SiF-EG0%-HPCTP15%, SiF-EG13.5%-HPCTP1.5%, SiF-EG10.5%-HPCTP4.5%, and SiF-EG7.5%-HPCTP7.5%. In combination with the characteristic data in Table 3, EG, HPCTP and EG/HPCTP could effectively inhibit the heat release rate of the sample.

When no flame retardant was added, the TTI of the sample was 54 s, and the PHRRT and PHRR were 83 s and 93.42 kW m$^{-2}$, respectively. At the same time, from the curve point of view, the two peaks occurred because a large amount of combustible gas accumulated inside the SiF during the combustion process and destroyed the surface-protected silica protective film, causing the second peak (this can be seen in the photograph in figure 2(a)).

When EG was added, the TTI and PHRRT were delayed to 130 s and 167 s, respectively, and the PHRR dropped to 64.77 kW m$^{-2}$, which was 27.8% lower than that of pristine SiF. When HPCTP was added, the TTI prolonged significantly, reaching 270 s, and the time of PHRR was delayed to 317 s, but the PHRR exceeded the 93.42 kW m$^{-2}$ for the pristine SiF and reached 99.72 kW m$^{-2}$. When EG and HPCTP were combined, it was found that when the EG/HPCTP mass fraction was 1:1, the PHRR was at least 49.92 kW m$^{-2}$, and the PHRR decreased by 46.56%. Additionally, compared with SiF-EG0%-HPCTP0%, SiF-EG15%-HPCTP0%,

![Figure 4. THR curves of SiF composites at a flux of 35 kW m$^{-2}$. Fire performance index (FPI) and fire growth index (FGI).](image-url)

Table 3. Flame retardant characteristic parameters of silicone foam.

| Sample                  | TTI (s) | $T_{PHRR}$ (s) | PHRR (KW m$^{-2}$) | THR (MJ m$^{-2}$) | FGI (KW m$^{-2} \cdot s^{-1}$) | FPI (m$^2$ · s K$^{-1}$ W$^{-1}$) |
|-------------------------|--------|----------------|--------------------|-------------------|-------------------------------|----------------------------------|
| SiF-EG0%-HPCTP0%        | 54     | 83             | 93.42              | 25.80             | 1.13                          | 0.58                             |
| SiF-EG15%-HPCTP0%       | 130    | 167            | 64.77              | 11.36             | 0.39                          | 2.01                             |
| SiF-EG0%-HPCTP15%       | 270    | 317            | 99.72              | 13.77             | 0.31                          | 2.71                             |
| SiF-EG13.5%-HPCTP1.5%   | 80     | 115            | 65.83              | 16.84             | 0.57                          | 1.22                             |
| SiF-EG10.5%-HPCTP4.5%   | 87     | 122            | 72.06              | 21.84             | 0.59                          | 1.21                             |
| SiF-EG7.5%-HPCTP7.5%    | 77     | 114            | 49.92              | 12.94             | 0.44                          | 1.54                             |

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Figure 3 shows the HRR of SiF-EG0%-HPCTP0%, SiF-EG15%-HPCTP0%, SiF-EG0%-HPCTP15%, SiF-EG13.5%-HPCTP1.5%, SiF-EG10.5%-HPCTP4.5%, and SiF-EG7.5%-HPCTP7.5%. In combination with the characteristic data in Table 3, EG, HPCTP and EG/HPCTP could effectively inhibit the heat release rate of the sample.

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and SiF-EG0%-HPCTP15%, when EG/HPCTP was added, the PHRR was quickly reduced and steadily and slowly continued to decrease. The reason was that the condensed phase flame retardant effect played a leading role with the increase of HPCTP, but the gas phase flame retardant effect also rose and repressed gas phase combustion. The decomposition products of HPCTP improved strength of EG, and presented the optimal PHRR.

3.3.2. Total heat release (THR)

3.4. Thermal stability analysis

TGA is an important method for evaluating the thermal stability of polymer materials. The key parameters, such as initial decomposition temperature, maximum thermal weight loss temperature, maximum thermal weight loss rate, and residual carbon rate, can reflect the thermal stability of the material [38, 42]. Figures 5(a) and (b) show the TG curve and DTG curve of the SiF material, and table 4 shows the main characteristic parameters of TGA.

From table 3, when EG, HPCTP, and EG/HPCTP were added, FGI decreased and FPI increased, indicating that EG, HPCTP, and EG/HPCTP all exhibited flame retardant effects. When HPCTP was added, the effect was the best, and when EG and HPCTP were combined, as the amount of EG increased, FGI gradually decreased, while FPI gradually increased, indicating that HPCTP and EG could effectively improve the safety of the sample. Because HPCTP could generate large amount of free radicals, which quickly repressed the gas phase combustion on the sample surface and increased the ignition time, thereby performed excellently in FGI and FPI.
maximum weight loss rate of porous SiF was 367 °C, which was 21 °C, 11 °C, 11 °C, 21 °C, and 27 °C lower than that of SiF-EG15%-HPCTP0%, SiF-EG13.5%-HPCTP1.5%, SiF-EG10.5%-HPCTP4.5%, and SiF-EG7.5%-HPCTP7.5%, respectively. The addition of HPCTP effectively delayed the decomposition of alkyl branches in SiF, but the rates corresponding to the maximum weight loss temperature were all larger than those of SiF-EG0%-HPCTP0%, further promoting branched carbonization and forming an effective protective layer. The third stage of weight loss of 450 °C–600 °C was mainly the decomposition of silicon-oxygen bonds. The maximum weight loss temperature of SiF-EG0%-HPCTP0% was 476 °C, which was 44 °C, 34 °C, 30 °C, 51 °C

Figure 5. TGA and DTG curves of flame-retardant SiF composites at a constant heating rate of 20 °C min⁻¹.
| Sample                  | $T_1$/$^\circ\text{C}$ | $T_{1\text{ma}}$/$^\circ\text{C}$ | $T_{2\text{max}}$/$^\circ\text{C}$ | $T_{3\text{max}}$/$^\circ\text{C}$ | $T_{4\text{max}}$/$^\circ\text{C}$ | PMLR$_{1\text{min}}$/min | PMLR$_{2\text{min}}$/min | PMLR$_{3\text{min}}$/min | PMLR$_{4\text{min}}$/min | Residues at 900.0/$^\circ\text{C}$/% |
|-------------------------|-------------------------|-----------------------------------|-----------------------------------|-----------------------------------|-----------------------------------|---------------------------|---------------------------|---------------------------|---------------------------|-------------------------------|
| SiF-EG0%-HPCTP0%        | 356                     | —                                 | 367                               | 476                               | —                                | 0.228                     | 0.164                     | —                         | —                         | 69.7                          |
| SiF-EG15%-HPCTP0%       | 353                     | 271                               | 388                               | 520                               | 778                               | 0.048                     | 0.295                     | 0.113                     | 0.110                     | 57.0                          |
| SiF-EG0%-HPCTP15%       | 345                     | —                                 | 378                               | 510                               | —                                | 0.386                     | 0.108                     | —                         | —                         | 59.4                          |
| SiF-EG13.5%-HPCTP1.5%   | 291                     | 261                               | 378                               | 506                               | 795                               | 0.111                     | 0.312                     | 0.047                     | 0.120                     | 54.7                          |
| SiF-EG10.5%-HPCTP4.5%   | 326                     | 275                               | 387                               | 527                               | 840                               | 0.071                     | 0.372                     | 0.040                     | 0.095                     | 55.4                          |
| SiF-EG7.5%-HPCTP7.5%    | 305                     | 263                               | 393                               | 536                               | 864                               | 0.079                     | 0.397                     | 0.043                     | 0.079                     | 54.7                          |

Table 4. TG parameters of SiF composites.
and 60 °C lower than that of SiF-EG15%-HPCTP0%, SiF-EG13.5%-HPCTP1.5%, SiF-EG10.5%-HPCTP4.5%, and SiF-EG7.5%-HPCTP7.5%, respectively, showing that EG formed an effective protective layer on the surface of SiF. Comparing SiF-EG0%-HPCTP0% with SiF-EG15%-HPCTP0%, SiF-EG13.5%-HPCTP1.5%, SiF-EG10.5%-HPCTP4.5%, and SiF-EG7.5%-HPCTP7.5%, the decomposition rate of flame-retardant samples at this stage decreased significantly. When EG and HPCTP were mixed, the decomposition temperature was higher and the rate was lower than when they were added alone, indicating that EG and HPCTP had a synergistic effect. The weight loss of 600 °C–850 °C in the fourth stage was mainly the oxidative decomposition of EG. The addition of HPCTP could delay the decomposition of EG and reduce the decomposition rate.

From the perspective of the carbon residue rate, the carbon residue rate of SiF-EG0%-HPCTP0% was significantly higher than that of SiF with added flame retardants. The main reasons were as follows: first, the sample was only approximately 10 mg during the TGA test, and the protective layer formed by EG and HPCTP was not enough to protect the SiF sample for a long time [42, 43]; second, EG began to oxidize at 600 °C and was lost after ashing at 800 °C.

3.5. Mechanical properties
Mechanical properties directly affect the use of materials. To study the effects of EG and HPCTP on SiF, tensile strength and elongation at break were determined for the SiF samples. The test results are shown in table 2.

It can be seen from table 2 that the tensile strength and elongation at break of the samples increased after the addition of EG. However, as the amount of EG added increased, the tensile strength and elongation at break of the samples stabilized within a range. Mainly, there were a large number of hydroxyl groups on the surface of EG, which could form chemical bonds with SiF, thereby improving the mechanical properties of SiF. As the amount of added HPCTP increased, both the elongation at break and tensile strength decreased, but they were still better than those of SiF-EG0%-HPCTP0%. This showed that adding a small amount of HPCTP helped improve the mechanical properties of SiF, but with excessive addition, P, N, and other elements poisoned the platinum catalyst and reduced the crosslinking of SiF, so the mechanical properties of SiF decreased. When EG and HPCTP were combined, as HPCTP increased, the elongation at break of the sample increased, but the tensile strength decreased. This showed that the combination of HPCTP and EG could help improve the toughness of SiF samples.

4. Conclusions
In this study, heat-cured SiF with EG/HPCTP was obtained, and the SiF material was analyzed in detail through LOI, UL-94, CCT, TGA, and mechanical property evaluation. The detailed conclusions were as follows: First, EG, HPCTP, and EG/HPCTP could improve the flame retardancy of SiF, and HPCTP could enhance the intensity of EG. Second, the thermal stability of the SiF could be improved by adding EG/HPCTP. Furthermore, EG and HPCTP helped to improve the mechanical properties of SiF. In summary, the EG/HPCTP synergistic flame-retardant system for SiF composites was effective.

Acknowledgments
The work was financially supported by the National Natural Science Foundations of China (5177–4232).

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