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Thermal degradation mechanism and flame retardancy of MQ silicone/ epoxy resin composites

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Abstract A series of flame-retarded epoxy resins (EP) loaded with methyl MQ silicone resin and a novel hyperbranched polysiloxane(HPSi) acting as compatibilizer have been prepared. Compatibility of these EP composites was characterized by differential scanning calorimetry (DSC) and scanning electron microscopy (SEM). The results showed that HPSi significantly improved the compatibility of EP/MQ. The flame retardancy and thermal degradation behavior of these EP composites were investigated by limiting oxygen index (LOI), UL-94 vertical burning, themogravimetric analysis test (TG), FTIR and SEM. The results showed that the incorporation of MQ into EP can improve the thermal stability dramatically. It is observed that the LOI values of epoxy resins increased obviously (from 21 % to 31 %) with the MQ loading, which passed V-0 rating of UL-94. Specifically, its combustion residue at 700℃ was 14.5% weight , which exceed the value of neat EP resin(5.08% weight). Moreover, structural analysis of the remaining after vertical burning by FTIR spectra verified the formation of polyaromatic carbons. Additionally, morphology of the residue char showed the compact, smooth, and tight structure of EP composites systems. These outstanding integrated properties would make EP composites attractive for practical applications.

Keywords: Methyl MQ silicone resin; Flame retardancy; Thermal behavior.

Introduction

Epoxy resin (EP) has occupied a dominant position in the high performance materials owing to its outstanding mechanical stiffness, toughness, chemical resistance, and excellent adhesive properties and superior electrical properties [1-4]. To extend the applications of epoxy resins in the electrical industry, it is important to improve their flame resistance [5,6]. The non-halogen flame retardant epoxy resins have been widely researched and some have been applied in semi-conductor encapsulants, fiber reinforced composites and printed circuit boards for their excellent flame retardancy [7-11], among which silicon-containing compounds are considered as a promising choice due to their high flame-retardant efficiency [12,13].

MQ silicone resin, which can be prepared from the economic and easy-to-get soluble glass, is a kind of unique polysiloxane of double-layer compact globular structure. The extensive utilization of MQ silicone resin made it a novel promising organic silicon polymer. MQ resin is composed of the polycondensation chain unit of four functional siloxane (Q) and single functional siloxane chain (M). The proportion of M and Q in the molecule and the structure of M and Q determine the nature and scope of application of resin.

When the polysiloxane-modified epoxy resin is heated to high temperature, the conversion to the stable silicon dioxide in air can form an excellent protective char barrier to cut off the heat and oxygen transfer and improve the flame retardation of the polymers. However, due to large amounts of oxygen in this system, phase separation and substantial lower glass transition temperatures usually occur [14,15]. So hyperbranched polymer (HPSi) compatibilizer was utilized to improve the compatibility of MQ silicone resin/epoxy resin. The hyperbranched polymer possesses epoxy group, which can serve to increase the compatibility with polymers, improve the thermal stability and the flame retardancy of the EP composites. Integrated performances of HPSi/MQ silicone resin/epoxy resin systems including compatibilization, thermal degradation behavior, and the flame retardancy were intensively investigated.

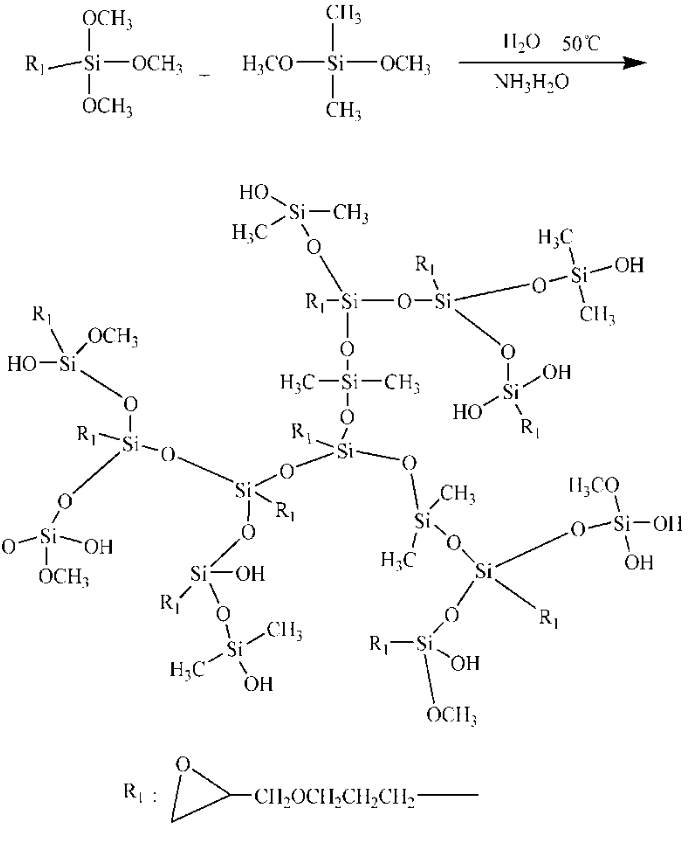
Experimental

Materials

Diglycidyl ether of bisphenol A (DGEBA, EP, epoxy value ¼ 0.44 mol/100 g) was purchased from Wuxi Bluestar Chemical Co.Ltd. China. Methyl MQ silicone resin was supplied by Chenguang. Chemical Co. Ltd. China. HPSi was synthesized by our laboratory [16]. Diethylto1uene diamine（DETDA）were obtained from Chengdu Kelong Chemical Reagent Factory. All materials were used without further purification.

Synthesis of polysiloxane (HPSi)

23.6g KH560, 18g DEMS and appropriate mass of anhydrous ethanol were added to a 250 mL three-necked round bottom flask equipped with a stirrer, a thermometer, and a condenser. Then 0.4g ammonia, 7.56g distilled water were both added into the flask. The temperature was raised to 50 °C and kept for 4.5 h. After that, a spot of 1-chlorotrimethylsilane was added as capping agent. Finally, the product was dried to eliminate methanol, ethanol, and water in a vacuum oven. Finally, a transparent and viscous liquid, hyperbranched polysiloxane (coded as HPSi), was obtained. The reaction mechanism of this system is shown in Scheme 1.



Scheme 1 The synthesis of organic silicon resin (HPSi) by hydrolysis and condensation.

Preparation of cured HPSi/EP/MQ blends

HPSi was blended with DGEBA with a ratio at 0.1:1(mass) by vigorous stirring at 130 °C for 20 min to obtain DGEBA-HPSi oligomer. Then an appropriate amount of MQ (0.3wt%, 0.5wt%, 1wt%, 3wt%, 5wt%,) was added into the above oligomer. Third, stoichiometric DETDA was mixed into the above-mentioned blends at 100 °C. Subsequently, the blends were cast into a preheated aluminium mold for curing and postcuring following the protocol of 130 °C/2.5 h+160 °C/2.5 h. Then epoxy resin specimens were obtained and subsequently machined to the desirable size for further testing.

Measurements

DSC measurements were conducted on a Netzsch Q-200 ranging from room temperature to 350℃ with a heating rate of 10℃ min^{-1} under nitrogen atmosphere. The morphologies of the fractured surfaces of samples and the surface morphology of the char obtained after the LOI tests were observed by Inspect-F SEM. TG analyses were conducted on Netzsch TG209 at a linear heating rate of 10 K min^{-1} under pure nitrogen within the temperature range from 30 to 800 °C. FTIR spectra were recorded between 400 and 4000 cm^{-1} on a Nicolet Magna-IR 560 spectrometer (Nicolet Instrument Co, USA). Thermogravimetric analysis/infrared spectrometry (TG-IR) of the samples was performed using the TGA Q5000 IR thermogravimetric analyzer that was interfaced to the Nicolet Magna-IR 560 spectrophotometer. About 5.0 mg of the sample was put in an alumina crucible and heated from 50 to 800℃ at a heating rate of 10℃/min (nitrogen atmosphere, flow rate of 45 ml/min). LOI data of all samples were obtained at room temperature on an oxygen index instrument (XYC-75) produced by Chende Jinjian Analysis Instrument Factory, according to GB/T2406-93 standard. The dimensions of all samples were 130 × 6.5 × 3 mm^3. Vertical burning tests were performed according to UL-94 standard with samples of dimensions 125 ×

12.5 × 3.2 mm. In this test, samples were classed as V-0, V-1, V-2, or unclassified according to their behaviour (dripping of burning material and burning time).

Compatibility of cured HPSi/EP/MQ blends

The influence of HPSi on the morphologies of HPSi/EP/MQ blend can be evaluated by measuring the glass transition temperature (T_g) of the blends. Figure 1 gives DSC curves of various mixtures. The curing systems all exhibit single T_g except EP/MQ. It is well known that the absolutely miscible blend presents homogeneous morphology exhibiting only one T_g but incompatible mixtures will exhibit more than one T_g. The homogeneous morphology of the samples provides the prerequisite for MQ resin to improve the flame retardancy property of epoxy resins. It can also be observed that with the increase of MQ, T_g of the whole system increases until reaches the maximum value of 142.8 ℃, and then decreases dramaticlly. MQ resin is a kind of compact ball with double-layer structure and usually has higher average free volume that plays a negative role in increasing the concentration of the chain segment of modified resins. At the same time, in the case of the EP/MQ/HPSi systems, there are strong interactions between MQ and HPSi, which can conspicuously increase the packing density of the polymer. The positive factor is dominant when the content of MQ is between 0.3 and 1 wt%, while when the content of MQ increases from 1 to 5 wt% the negative factor will offset this positive effect. From the data analysis of DSC, the addition of HPSi successfully improves the compatibility of EP/MQ, which provides the important prerequisite for MQ as a flame retardancy of EP.

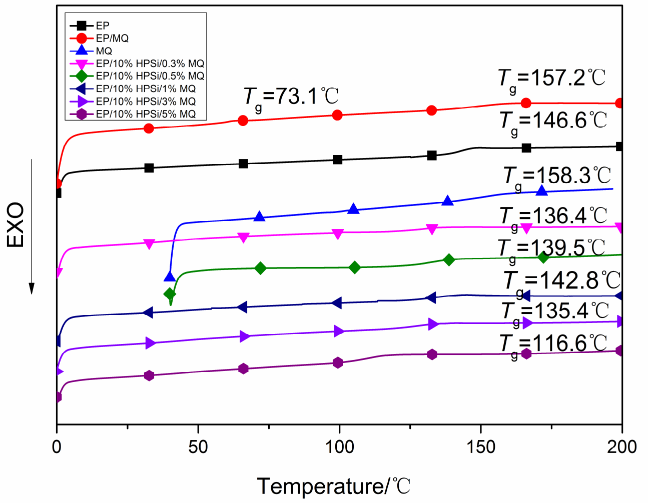


Figure 1 The DSC curves of the blending systems.

To further explore the influence of HPSi/MQ in the cured system, SEM morphologies of the fractured surfaces of samples are shown in Figure 2. Figure 2a is the SEM photograph of cured pure EP sample. Morphology of the EP/10%HPSi cured sample shows the compact, smooth, and tight surface in Figure 2b which demonstrates the outstanding compatibility of EP/HPSi. However, on macroscopical ，the sample（vertical burning tests samples of dimensions 125 × 12.5 × 3.2 mm）of EP/MQ resin is laminated with the translucent yellow bottom and the white opaque upper. In addition, reuniting in the process of blending was also responsible for the serious phase separation. Cured EP/10%HPSi/1%MQ is uniformly yellow. It can be observed in Figure 2c that small balls and circular holes appear on the fractured surface. These balls are evenly distributed in the matrix, which could be attributed to that the MQ is ideally dispersed in EP matrix in the presence of HPSi.

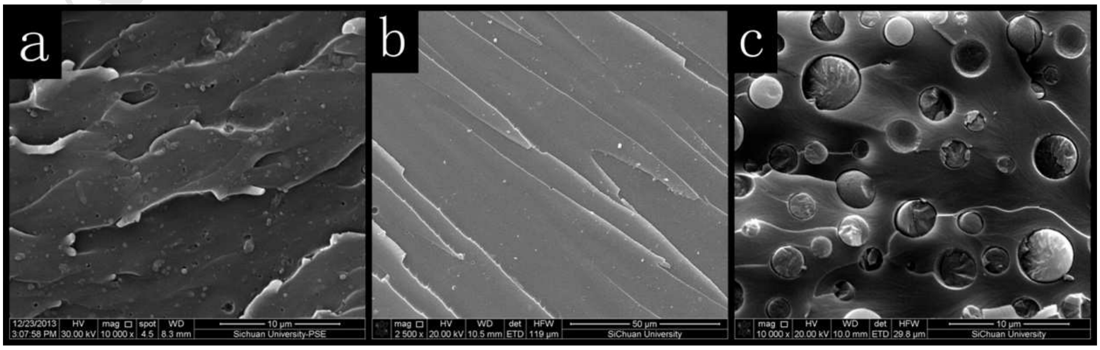


Figure 2 SEM of the section of the blending systems.（2a EP; 2b EP/10%HPSi; 2c EP/10%HPSi/1%MQ resin）

Flammability

The flame retardant properties of the cured epoxy resins were investigated by LOI test and UL94 test. Data are summarized in Table 1. The LOI value of pure EP is 21 % and the UL-94 rating is NOT V-2 with unsatisfying anti-drip property. Table 1 presents that a higher LOI value is obtained with the increase in MQ content. LOI increases drastically from 23 % to 31 % (meets the flame-retardant criterion [17]) when MQ content is increased from 0 to 5 wt %.When 10% compatibilizer and 1% MQ are added into the EP resin，LOI value comes to 31 % and at the same time UL94 V-0 ratings can be achieved.

It is clear that the epoxy resins with both HPSi and MQ gain enhanced flame retardancy. After adding MQ resin, the amount of smoke significantly reduces when under combustion. What’s more, the residue with MQ resin can keep the original shape and form hollow shell after burning. The dripping are no longer observed for all the samples and the self-extinguishing is observed with 0.5 wt % - 3 wt % MQ loading. The best UL-94 rating (t_1=2 s, t_2= 2 s), which is a V-0 rating, can be got when the content of MQ is 1 wt %.

Table 1 The values of LOI and UL-94 for finally cured systems

Sample	LOI（%）	UL-94	t_1/s	t_2/s
EP	21	NR	_	_
EP/10%HPSi	23	NR	_	_
EP/10%HPSi/0.3%MQ	26	V-1	11.49	3.47
EP/10%HPSi/0.5%MQ	30.5	V-0	4.38	3.49
EP/10%HPSi /1%MQ	31	V-0	6.38	3.22
EP/10%HPSi /3%MQ	30	V-1	8.36	5.01
EP/10%HPSi /5%MQ	28	V-1	7.62	5.34

Thermal analysis

The thermal stability and decomposition behavior of cured epoxy resins comprising the MQ siloxane and HPSi were investigated by TG. Figure 3 shows plots of weight versus temperature from 0 to 800 °C for the EP/HPSi/MQ systems in nitrogen atmosphere and the corresponding $T_{5\%}$, the T_{max}, and char yield (Y_c) at 800 °C are summarized in Table 2.

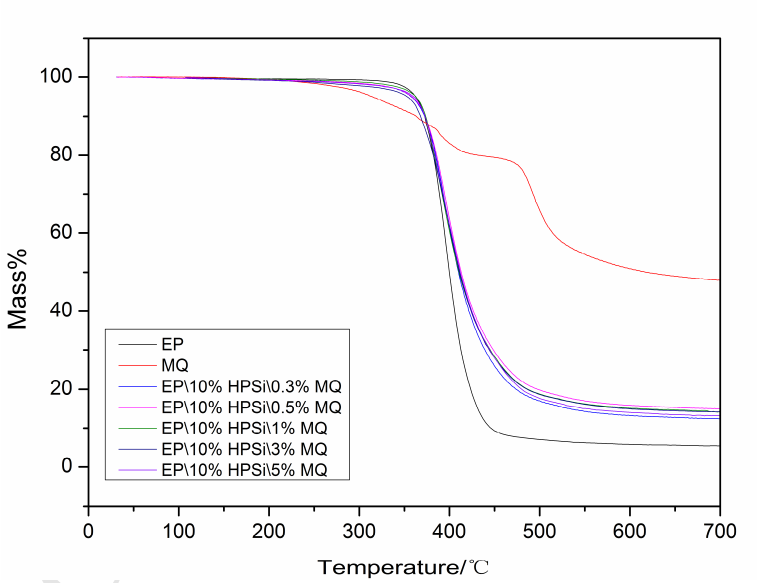
In a nitrogen atmosphere, MQ begins to slowly degrade at 250 ℃, then it undergoes a rapid weight loss of 35% in the range 400-500℃, and the residue at 800℃ is 45.4% showing that MQ has a high charring yield. The T_{max1} of MQ is 392℃(close to $T_{5\%}$ of EP), which indicates that MQ can exert its flame-retardant action before the fast degradation of EP.

It can be observed from Figure 3 that all cured resins have one-stage decomposition process, demonstrating that the original and modified EP resins have similar degradation processes. It was found that almost no weight loss was detected below 350 °C. The initial decomposition temperatures at 5% weight loss were observed in the range of 350–358 °C in nitrogen.

As for pure EP, the thermal oxidative degradation process has one stage. The stage is in the temperature range of 350-450 ℃ corresponding to a strong DTG peak at 396 ℃ (T_{max1}) and the weight loss is 91.4%. These indicate that some complex chemical reactions take place at the stage. Bits of decompositions appear before 300 °C, which can be attributed to the decomposition of unstable alkyl groups in the hybrids. The free radical chain scission of the isopropylidene linkage and the branching and crosslinking reactions of molecular chains take place, leading to the release of EP [18].

TG shows that with the increase of MQ resin, the initial thermal decomposition of

EP/HPSi/MQ systems, occurring between 350 and 500 °C, are wider than that of EP resin. Char residue at 800℃ remarkably increases from 5.08 % to 14.54 % with MQ content increases from 0% to 5 %. $T_{5\%}$ and T_{max1} of MQ are lower than that of EP. As being discussed above, this phenomenon originates from the decomposition of MQ at inferior temperature that leads to the formation of the silicone-containing group, which will participate in the cross-linked carbonization. Silicone resins have the ability to convert the usual organic decomposition to partially inorganic decomposition by forming the carbon-silicon residue acting as thermal insulation [19,20] to prevent gas evolution, and achieve ultimate improvement on flame retardation of this silicone-containing epoxy system. Moreover, the Si–O group of MQ is able to absorb more thermal energy and its vibration could dissipate the thermal decomposition energy.



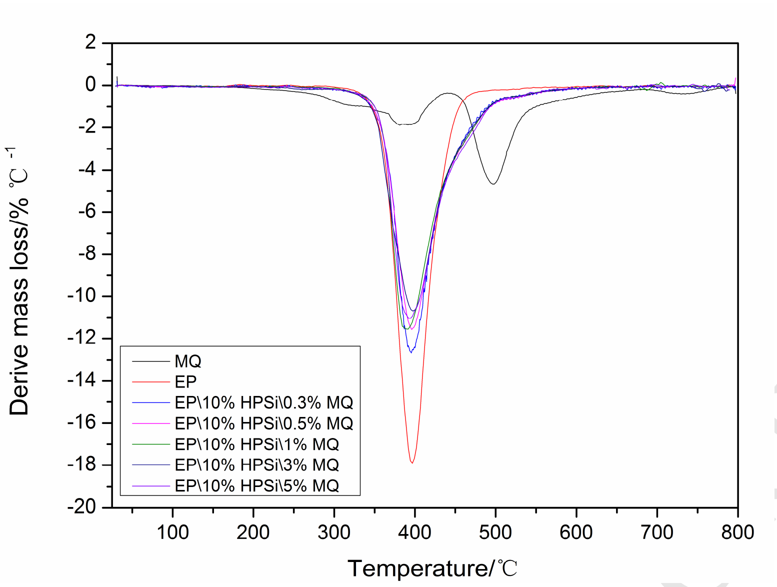


Figure 3 The TG and DTG data of different systems under N_2 atmosphere.

Table 2 TG and DTG data of thermal degradation of systems and char residue

Sample	$T_{5\%}$	R_{1peak}	T_{max1}	Y_c at 700℃	
	/ ℃	/ (%℃$^{-1}$)	/ ℃	Cal.	Exp.
EP	362.4	-17.90	396	——	5.08
MQ	314.3	-5.28	493.8	——	48.49
EP/10%HPSi/0.3%MQ	357.3	-12.43	359.4	12.1	12.47
EP/10%HPSi/0.5%MQ	358.2	-11.57	396.1	13.54	14.04
EP/10%HPSi/1%MQ	350.2	-4.91	388	15.62	14.5
EP/10%HPSi/3%MQ	357.9	-4.81	399	16.22	14.17
EP/10%HPSi/5%MQ	348.8	-5.05	376.5	17.53	13.27

$T_{5\%}$ represents the temperature at which mass loss is 5 % for the system (the initial decomposition temperature), T_{max1} and T_{max2} represents the temperature of the first maximum mass loss rate for the system and the second maximum mass loss rate for the system, respectively. Y_c represents the char yields at 700 °C.

Figure 4 shows the experimental and theoretical TG curves of EP/10%HPSi/1%MQ system in N_2. The theoretical curve was calculated based upon the mass percentage of the ingredient in the EP system. The formula is as follows:

$$Mcal=wt_{HPSi}\%*M_{HPSiexp} + w_{EP}\%*M_{EPexp}+ w_{MQ\%}*M_{MQexp}$$

M_{cal} is the theoretical amount of carbon residue, M_{Exp} refers to the actual amount of carbon residue, wt% refers to the corresponding proportion of ingredients.

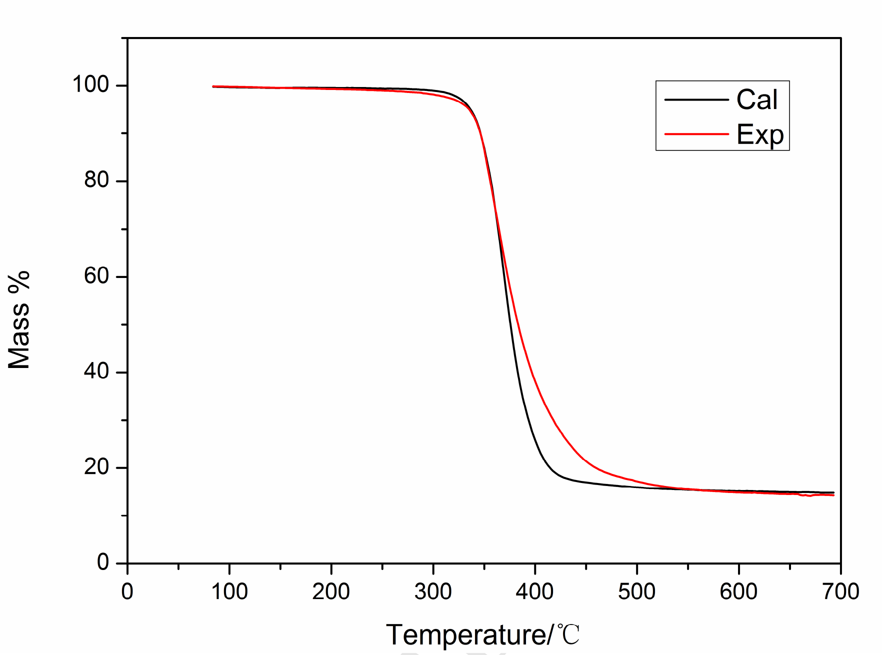


Figure 4 the experimental and theoretical TG curves of EP/10%HPSi/1%MQ system in N_2.

As is shown in N_2, when the temperature is below 400 ℃, the experimental and theoretical curves are similar. When the temperature is higher than 400 ℃The experimental and theoretical TG curves of EP/HPSi/MQ system are slightly different: the experimental mass of residual char surpasses the theoretical one. This is because that organic silicon generated more stable substance at high temperatures. This kind of material acting as isolation belt can traps the flammable small molecule diffusion and heat transfer prevent the underlying substance further decomposed and increase the amount of carbon residue. This effectively improves the thermal stability of the matrix.

Higher char yield of MQ resin indicates that the carbonization mechanism indeed plays an important role in flame retardation. It is well known that the general flame retarding mechanism of organic silicone resin-based materials is thought to be the formation of protective barrier during combustion. The improved flame retardancy can be explained by a flame retarding mechanism: providing a barrier for heat and mass transfer in the condensed phase at the same time preventing melted EP from burning [21,22].

Structural analysis and pyrolysis gas products of EP/HPSi/MQ

From The TGA-FTIR spectra we can figure out the main compositions of gas products in the thermal degradation process and further disclose the pyrolysis information [23].

As is shown in Fig. 5, the evolved gas analysis for EP/10%HPSi/1%MQ at maximum decomposition rates is similar to that of EP. Specifically, characteristic bands of H_2O and/or phenol, CO_2, hydrocarbons ($-CH_3$ and $-CH_2$ groups: 2950-2850 and 1100-1300 cm^{-1}) and compounds containing aromatic rings (1624 cm^{-1}) [24-25]. When the temperature reaches 400 °C, the MQ resin begin to decompose, so the absorption peaks for carbonyl compound (1747 cm^{-1}) and alkane (2882-2970 cm^{-1}) are quite strong. Fig. 6 shows the 3D TG-FTIR spectrum of gas phase in the thermal degradation of EP/10%HPSi/1%MQ. Obviously, the main gas products including H_2O (3649 cm^{-1}) and CO_2 (3100-2600 cm^{-1}) can be detected over the whole degradation process of EP/10%HPSi/1%MQ. It is well-known that depolymerization is the main process in the thermal degradation of polymers. In the process of depolymerization, the main decomposition products are CO_2, phenol, and hydrocarbons, etc.

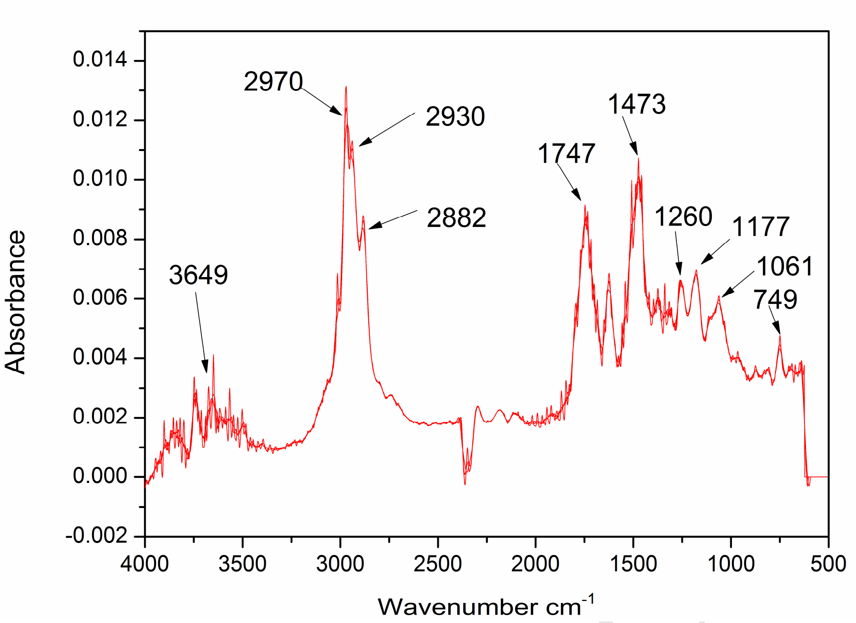


Figure 5 gas analysis for EP/10%HPSi/1%MQ at maximum decomposition rates

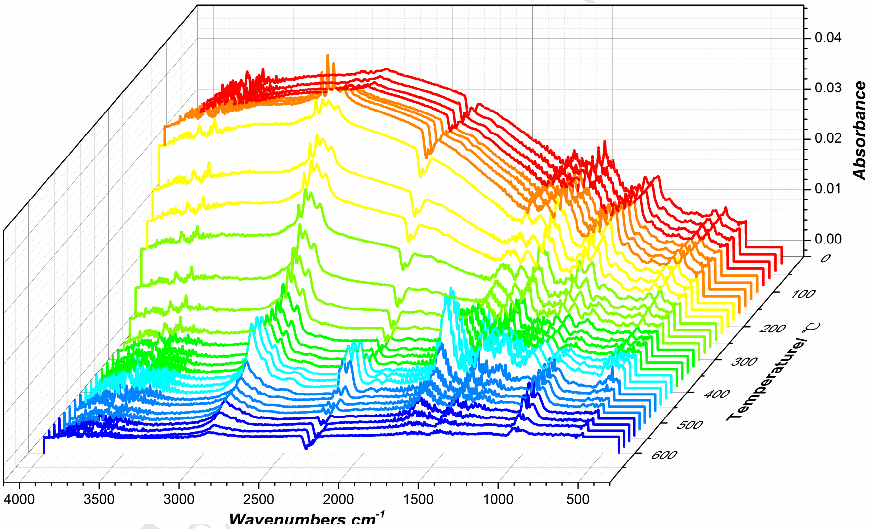


Figure 6 3D TG-FTIR spectrum of gas phase in the thermal degradation of EP/10%HPSi/1%MQ

In order to further study the chemical alteration in the condensed phase of EP/10%HPSi/1%MQ system, the combustion residue after thermogravimetry under nitrogen atmosphere were investigated by FTIR. The FTIR spectra at different degradation temperatures (400 and 550 °C) are shown in Figure 7. At 400 °C, the main products of the thermal decomposition of EP/10%HPSi/1%MQ are compounds containing -OH (such as H_2O, phenol; 3500-3600 cm^{-1}), hydrocarbons (C-H stretching at 2929 cm^{-1}), compounds containing aromatic rings (1612, 1436, cm $^{-1}$), C–O stretching vibration (1035,1155 cm^{-1}) etc ,which are the characteristic absorptions of pure epoxy resin [26]. At 550 °C, it is found that the absorption peaks at 2929, 1436, 1155 and 1035 cm^{-1} disappear, indicating that the main decomposition happened in this stage, which is consistent with the TG results. According to the shown information, the absorption of aliphatic components and the C–C stretching vibration of aromatic ring at 1510 cm^{-1} totally disappear at 800 °C. However, bending vibration peak belonging to Si-O-Si groups centered at 1100 cm^{-1} become stronger and wider and characteristic peaks belonging to polyaromatic centered at 800 cm^{-1} and 1630 cm^{-1} become broader, indicating the formation of polyaromatic carbons.

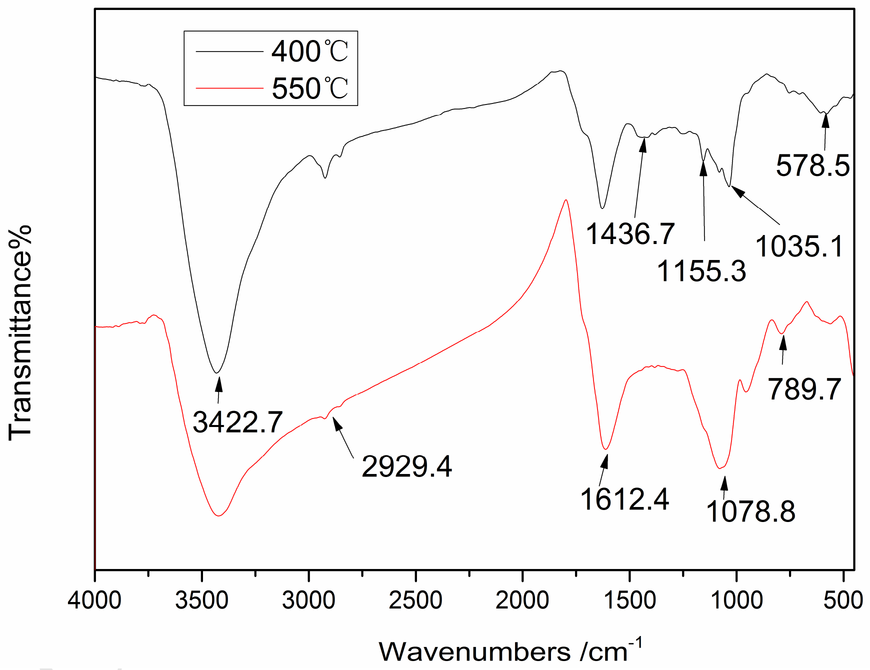


Figure 7 The FTIR spectra of condensed phase of EP/10%HPSi/1%MQ system at 400 and 550 °C

Morphology of the residue char

In order to explore how the structure of char determines the flame retardancy of EP, we investigated the residues of char after LOI testing by SEM. Figure 6 presents SEM micrographs of char residue of EP/HPSi/MQ and EP. According to Figure 8a, for neat

EP, there are many big holes due to insufficient char formation or less condensed char during the burning process. This poor char quality could not effectively protect the underlying EP from degradation during combustion; therefore, EP can’t pass UL 94-V0 rating. However, the char surface of EP with EP/10%HPSi/1%MQ, illustrated in Figure 8b, is compact, smooth, and tight. This structure of the char for EP composites could prevent heat transfer between the flame MQ and the substrate and thus protect the underlying materials from further burning and pyrolysis, because of which the material have much higher LOI values. In addition, this char structure can offer a good shield to prevent melted EP from burning, which was proved in vertical flammability tests (Table 1).

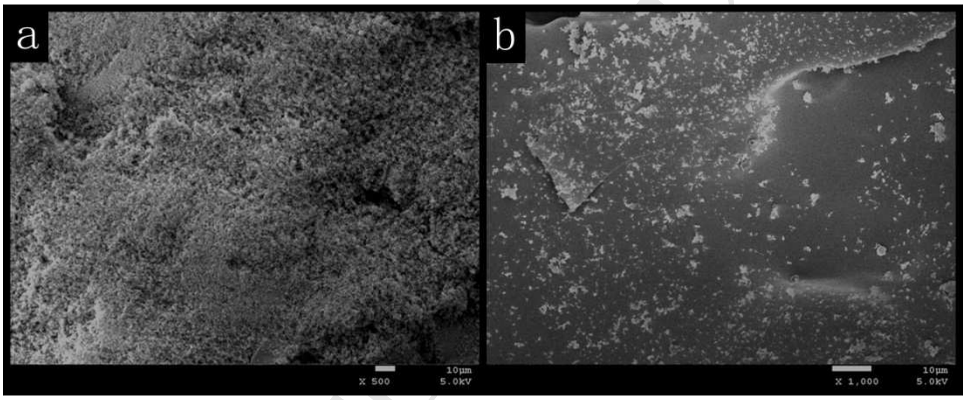


Figure 8 SEM of the char layer of EP and EP/10%HPSi/1%MQ system.

Conclusions

Flame-retarded epoxy resin (EP) composites were obtained by incorporation of laboratory synthesized compatibilizer (HPSi) and MQ resin into EP. By the incorporation of MQ and HPSi into EP, the LOI value of EP/10%HPSi/1%MQ increased to 31 %, which was about 1.4 times of the corresponding value of neat EP, and a V-0 grade for the UL-94 can be achieved.

The addition of both HPSi and MQ to EP leads to remarkable enhancement of the quantity of the char residues in TG. The high silicon content and double-layer compact globular structures of MQ contributed an excellent flame retardancy to epoxy resins. Specifically, structural analysis of the combustion residue by FTIR proved the formation of polyaromatic carbons which slowed heat and mass transfer between the gas and condensed phases. Based on these, the speculated degradation process of silicon resin was obtained. These properties would make this epoxy attractive for practical applications such as flame-retardant laminates.

Acknowledgments

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Scheme 1 The synthesis of organic silicon resin (HPSi) by hydrolysis and condensation.

Figure 1 The DSC curves of the blending systems.

Figure 2 SEM of the section of the blending systems.

Figure 3 The TG and DTG curves of the cured systems under N_2 atmosphere.

Figure 4 Experimental and theoretical TG curves of EP/10% HPSi/1%MQ under N_2 atmosphere.

Figure 5 The gas analysis for EP/10%HPSi/1%MQ at maximum decomposition rates

Figure 6 3D TG-FTIR spectrum of gas phase in the thermal degradation of EP/10%HPSi/1%MQ.

Figure 7 Figure 6 The FTIR spectra of condensed phase of EP/10%HPSi/30%MQ system at 400 and 550 °C

Figure 8 SEM of the char layer of EP and EP/10%HPSi/1%MQ system.