



Effect of glass frit with low softening temperature on the properties, microstructure and formation mechanism of polysiloxane elastomer-based ceramizable composites



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ARTICLE INFO

Article history:

Received 25 September 2016

Received in revised form

10 December 2016

Accepted 24 December 2016

Available online 25 December 2016

Keywords:

Ceramics

Glass frit

Polysiloxane elastomers

Sintering

Scanning electron microscopy

X-ray diffraction

ABSTRACT

The glass frit with low softening temperature, acting as one of the fluxing agents, is a key material to determine the microstructure of polysiloxane elastomer-based ceramizable composites. The effect of the glass frit with the softening temperature of 480 °C on the properties of the ceramic polysiloxane elastomer was investigated. The appearance of the ceramic residues was observed by a digital camera. The morphology of the impact fracture surface was observed by scanning electron microscopy (SEM). The ceramic mechanism of the ceramic residue was revealed by X-ray diffraction (XRD) analysis. The results showed that the linear shrinkage, as well as flexural strength and impact strength of the ceramic residue increased as the content of the glass frit increased from 0 to 30phr. SEM analysis showed that the structure of the ceramic residue was converted from sea-island phase to a uniform matrix with the increased content of the glass frit. XRD analysis showed that quartz and MgSiO₃ crystals were generated in the ceramic residues via the eutectic reaction between the mica and the glass frit.

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1. Introduction

Recently, ceramizable polymers were regarded as some novel fire-protection materials with wide application in electric wires and cables [1]. Ceramizable polymers ideally have the dual performance of polymers at room temperature and ceramics at elevated temperature. They can resist high temperature of more than 1000 °C due to the formation of ceramic layers to protect the inner copper of the wires. The ceramizable polymers are comprised by polymer matrix, ceramic fillers, fluxing agents and other additives [2–4]. Ceramic polysiloxane elastomer, as one of the ceramizable polymers, shows the same flexibility and extensibility as the common silicone rubber before ceramification. However, it may be transformed to some self-supporting ceramic residues to prevent the flame propagation at elevated temperature [5].

The formation of ceramic polysiloxane elastomers greatly depends on the ceramic conditions, such as sintering temperature or sintering rate [6]. In our former work [7], it was found that the weight loss and linear shrinkage, as well as the flexural strength

and impact strength of the ceramic residue, increased as the sintering temperature increased from 700 to 1100 °C.

In addition, the most effective method to improve the strength of ceramic residue is introducing some fluxing agents into the silicone rubber matrix [8–12]. The fluxing agents, including glass frit, zinc or boron oxide, zinc borate etc., with a low softening temperature melt at elevated temperature to form liquid phases, and stick refractory mineral fillers together, creating physical or chemical links between them [13]. Rafal Anyszka et al. [14] investigated silicone rubber-based composites with fluxing agent (B₂O₃) and it was found that the thermal stability of silicone rubber decreased significantly with incorporation of the fluxing agent due to the lower melting temperature (450 °C) of B₂O₃. However, the acidic and hydrophilic character of B₂O₃ can cause some problems during compounding and processing of the composites [15].

Furthermore, many research works had been focused on the addition of low melting temperature glass frit in the ceramizable polysiloxane elastomers. e.g., Imiela et al. [16] used glassy oxide frit containing large amount of Na₂O, which can significantly decrease the softening point, as the fluxing agents of the polysiloxane composites. Mansouri et al. [17] added glass frit with different soften points into silicone-based composites to improve low

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temperature ceramification at elevated temperatures. The ceramic residues showed a decline in volume resistivity with the addition of the glass frit.

Although many research works had been focused on the effect of fluxing agents and ceramic fillers on the properties of ceramizable polysiloxane elastomers, the effect of the glass frit content on the microstructure and properties of the ceramic residues has not been reported in detail, and the ceramic mechanism of the ceramizable composites with the addition of the glass frit was not revealed clearly.

In the present work, the effect of the glass frit content on the microstructure of polysiloxane elastomer-based composites is reported using various technologies including FT-IR, SEM, EDS and XRD analysis. The effects of the glass frit content on the weight loss, linear shrinkage, as well as flexural strength and impact strength of polysiloxane elastomer-based composites were investigated.

2. Experimental

2.1. Materials

Polysiloxane elastomer (model 110-2, methyl vinyl silicone rubber with the molecular weight of 600,000) was purchased from Shenzhen Square Silicone Co. Ltd., China. Fumed silica (CAB-O-SIL M-5) was purchased from Cabot Co. Ltd., USA. Fluorophlogopite mica ($[\text{KMg}_3(\text{AlSi}_3\text{O}_{10})\text{F}_2]$) was purchased from Sanbaoguangjing Mica Technology Co. Ltd., Shantou, China. Glass frit with the softening temperature of 480 °C, was prepared in the lab. 2,5-dimethyl-2,5-bis(tert butyl peroxy) hexane (abbreviated as DBPMH, model C-15) was supplied by Caiyuan Silicone Materials Co. Ltd., Donguang, China. The silicone gum compounds are summarized in Table 1. The formulation symbols are labeled as G0, G7.5, G15, G22.5 and G30.

2.2. Preparation of polysiloxane elastomer based ceramizable composites

All of the compounds were prepared on a two-roll mill (model XK-160, Zhanjiang Machinery Factory, China) with a gear ratio of 1:1.4 at room temperature. The polysiloxane elastomer was first softened, and then the fumed silica, mica and glass frit were added until a homogenous batch was obtained. The curing agent DBPMH was then added and processed until a visually good dispersion was achieved. The total mixing time is about 20 min. The polysiloxane elastomer compounds were cured with flat sheets by compression molding in a hydraulic press (Model KSHR100T, Kesheng Industry Co. Ltd., China) at 170 °C for 15 min under 15 MPa pressure. The post curing of samples was carried out in an oven at 180 °C for 2 h.

The ceramifying of flat sheet samples (50 mm long, 4 mm thick and 6 mm wide) was performed using a muffle furnace (Shenjia KL-12B, China). The samples were heated from room temperature to 1000 °C at a heating rate of 10 °C·min⁻¹, held for 30 min, and then cooled with the muffle furnace.

Table 1
Basic formulation of polysiloxane elastomer compounds.

Ingredients (phr)	Symbols				
	G0	G7.5	G15	G22.5	G30
Polysiloxane elastomer	100	100	100	100	100
Fumed silica	50	50	50	50	50
Fluorophlogopite mica	30	30	30	30	30
Glass frit	0	7.5	15	22.5	30
Hydroxyl silicone oil	2.5	2.5	2.5	2.5	2.5
DBPMH	2	2	2	2	2

*Parts per hundreds of rubber.

2.3. Characterization of the composites and ceramic residue

The flexural strength of pyrolysis samples was determined by a flexural method using a universal testing machine (Zwick/Roell Z010, Germany) with a cross-head speed of 0.5 mm min⁻¹ according to ASTM D790-10. The impact strength was evaluated on an electronic charpy impact tester (Suns, China) according to ASTM D256-10e1. The weight loss of the ceramic residue was calculated according to Eq. (1) where W is the weight loss, and m_1 and m_2 are the mass of the sample before and after sintering, respectively.

$$W = (m_1 - m_2)/m_1 \times 100\% \quad (1)$$

Shrinkage of the ceramic residue was measured by heating flat sheet samples to 1000 °C for 30 min in a muffle furnace. The linear shrinkage of the ceramic residue was calculated according to Eq. (2) where L is the linear shrinkage, and l_1 and l_2 are the length of the samples before and after sintering, respectively.

$$L = (l_1 - l_2)/l_1 \times 100\% \quad (2)$$

Thermal gravity analysis (TGA) was performed by using TG 309 F1 instrument (Netzsch, Germany). Samples (weight 8–10 mg) were heated at a heating rate of 10 °C·min⁻¹ from 35 to 800 °C under nitrogen atmosphere. The surfaces of the ceramizable polysiloxane elastomers and ceramic residues were observed by a digital camera (Canon 600D, Japan). The surface morphology of the glass frit and the impact fracture surface of the ceramic residues were sprayed with gold and then observed by scanning electron microscopy (SEM, Zeiss EVO18, Germany). The element composition of the glass frit was detected using an energy dispersive X-ray spectroscopy (EDS, Oxford X-Max). X-ray diffraction spectra of the mica, glass frit and ceramic residue were obtained using an X-ray diffractometer (XRD, PANalytical X'pert). Each scan was conducted from a 2 θ angle of 5°–90° at a scan rate of 1°·min⁻¹. The Fourier transform infrared spectroscopy (ATR-FTIR) spectra were recorded by a TENSOR27 spectrometer (Bruker, Germany) and the wave number range was set from 4000 to 500 cm⁻¹.

3. Results and discussion

3.1. SEM and EDS analysis of the glass frit

The microscopic morphology and EDS analysis of the glass frit were shown in Fig. 1. The glass frit was found to be irregular and rough with a particle size of less than 60 μm . The glass frit contains eight elements, with an oxygen content of 51.86%, a silicon content of 18.55%, and a fluorine content of 6.07%.

3.2. Curing characteristics of polysiloxane elastomer-based ceramizable composites

The curing characteristics of polysiloxane elastomer-based ceramizable composites with different contents of the glass frit were shown in Table 2. With the increased content of the glass frit from 0 to 30 phr, the scorch time (T_{10}) and the optimum curing time (T_{90}) decreased due to the decreased amount of the rubber components. However, the difference of the maximum torque and the minimum torque ($M_H - M_L$) did not show the obvious variation, indicating no reinforcing effect of the glass frit on the polysiloxane elastomer.

3.3. Mechanical properties of polysiloxane elastomer-based ceramizable composites

Effect of the glass frit content on the mechanical properties of

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