Improving the mechanical and ceramifiable properties of low temperature prepared silicone rubber composites

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Abstract
Silicone rubber has been widely used in electronic appliances, nuclear power cable, aerospace and so on. However, silicone rubber will continue to burn when exposed to flame, so flame retardant modification is required. In this work, aluminum hypophosphate (AHP) and Mica were used fluxing agent to make ceramizable silicone rubber material (CSR). The ceramization mechanism was investigated based on microstructure, mechanical and flame retardation properties. The scanning electron microscope result shows that the addition of AHP reduce the ceramization temperature effectively. AHP/Mica modified CSR have better flame retardation properties than pure silicone rubber, the addition of AHP increase LOI to 33.9%, and the LOI is 34.8% and the UL-94 is FV1 as 25 parts of Mica are added. When the 10 phr aluminum hypophosphate replaces Mica, the compression strength reach 14.46 MPa, increased by 42.5%. Moreover, the phenomenon of expansion into ceramics was founded and studied.

1. Introduction
In order to protect public fire safety, measures to improve the fire resistance of refractories are taken to reduce the destructive cost of fire [1–3]. By comparing the heat release rate of silicone resin with that of most organic polymers, it is discovered that the heat release rate of silicone resin is relatively low [4–6]. As a result, the silicone resin is less sensitive to external heat flow, so silicone is used as the raw material for cable production. The thermal resistance of organosilicon has been proved in the research of pyrolytic properties of organosilicon [7–11]. Although organosilicon has excellent fire resistance, the application of organosilicon is limited due to the poor mechanical properties of the pyrolysis products.

Ceramization is an efficient way used to improve the fire resistance of polymers which can obtain a hard, durable, and porous ceramic structure during the process of burning at high temperature [12]. Plasticizable ceramics are expected to have polymer properties at room temperature and ceramic properties with flame at high temperatures. The inner copper conductor layer of the cable is protected by the ceramic layer formed by the ceramizable polymer at high temperature. Experimental research found that the ceramic layer formed can resist the flame at high temperature of more than 1000 °C. In order to get ceramic able polymer, ceramic fillers, fluxes, and other additives [13–15] are usually added to improve the ceramization. With the development of ceramizable polymers, ceramic polymers in some field replaced other materials in fire protection fields, such as steel structure and high melting point fireproof cable [16,17] and sealant [18]. Many kinds of inorganic material can be used as modified substance for polymer matrix, while polydimethylsiloxane (PDMS) is usually used as matrix due to PDMS has excellent fireproof ability [19] and the amorphous silica formed on the surface getting good oxygen and heat insulation properties [1,13].

The mechanical strength of silicone rubber composites before pyrolysis with the strength and integrity of ceramic residues determines the fire-resistant application. Through experimental study, it is found that both the two properties can be improved. Hanu et al [7] reported that silicone polymer composites filled with Mica, glass frit, ferric oxide and/or a combination of these can develop as a part of a ceramifiable polymer as electrical power
The lower the ceramic temperature, the more widely CSR can be used. Low ceramization temperature, good expansion into ceramics was founded and discussed, the property expansion into porcelain has potential application value.

2. Experiment

2.1. Materials

A high-temperature vulcanized silicone rubber purchase from Yangzhou Dongjue CheMical New Material Co., Ltd was used as the silicone matrix and bis2,4-dichlorobenzoyl peroxide (DCBP) purchased from Jiangsu Tianchen New Material Co., Ltd as the curing agent in this work. CaCO$_3$ was purchased from Lianzhou Chaowei new materials Co. Ltd SiO$_2$ was purchased from Langfang Tongmao CheMical Co., Ltd. Aluminium hypophosphite was supplied by Shandong xiucheng cheMical. Mica was from huayuan Mica co., Ltd, Hebei, China, with a mean particle size of 15 $\mu$m. The source of frits is the product of the conventional melt-quenching method.

2.2. Sample preparation and pyrolysis

AHP was dried at 60 °C for 12 h before use. The preparation method of composites is a conventional two-roll mill at room temperature. Mix the filler well with the raw material, when the filler is added. At last, the composite material and curing agent are mixed, and the two are mixed evenly. Silicone compounds were molded and cured into flat sheets by compression molding at 160 °C for 12 min at 15 MPa pressure. The formulations of

| Sample code | SiR | Mica | AHP | SiO$_2$ | CaCO$_3$ | Frits A | Frits B |
|-------------|-----|------|-----|---------|----------|---------|---------|
| SAM0        | 100 | 0    | 0   | 10      | 20       | 10      | 10      |
| SAM1        | 100 | 15   | 0   | 10      | 20       | 10      | 10      |
| SAM2        | 100 | 5    | 10  | 10      | 20       | 10      | 10      |

It can be seen that many scholars committed to the research work on improving the thermal stability, flame retardant, mechanical properties by changing the SR matrixes and filler [3–6, 9, 13]. However, in actual research, it is found that CSR can’t meet the good mechanical characters of a rubber and ceramic state at the same time. The lower the ceramic temperature, the more widely CSR can be used. Low ceramization temperature, good flame retardancy and excellent mechanical CSR composites are very much needed in the field of nuclear power cable and aviation thermal protection system, none of the three properties is indispensable, however there are few researches on these three at the same time. Study on ceramifiable mechanism is urgent and necessary too.

In this work, CRS was prepared with improved mechanical and flame-resistant properties. The microstructure, flame retardancy and mechanical properties has been discussed, moreover, the structure-properties relationships and ceramifiable mechanism are investigated systematically. The phenomenon of expansion into ceramics was founded and discussed, the property expansion into porcelain has potential application value.
compounds were given in Table 1. The different softening point temperatures were used with the ratio of 1:1 to make sure the formation of the liquid phase gradually.

The prepared composites were slowly heated from room temperature to target temperature in a muffle furnace in an air atmosphere. The solid residue was achieved when heated to 500 °C, 650 °C, 850 °C, 950 °C, 1000 °C and kept at the temperature for 30 min.

2.3. Characterization of the composites
The composite of CheMical composites polymer is revealed by x-ray Fluorescence (XRF) using a PANalytical B.V Axios advanced XRF spectrometer. And composition is Mica and frits. Mechanical properties of the vulcanized composites before pyrolysis were tested by the Chinese standard GB/T 528–2009.

Zwick Instrument was used to compress the pyrolysis residue in a 20 mm high and 20 mm diameter cylinder. When a cylindrical sensor is used for compression experiments, the compression speed of the sensor is 1 mm min⁻¹ and the value of the compression force of the pyrolysis residue is recorded. For the accuracy of the experiment, each group of experiments was conducted more than five times, and the average value was taken as the final result.

Thermogravimetric analysis (TGA) was used to conduct pyrolysis experiments on the pyrolysis residues. The parameters are that the experiments ranged from 30 to 800 °C, the heating rate was 10 °C min⁻¹, the experimental environment was flowing nitrogen atmosphere, and the sample mass was 5 mg.

X-ray diffraction spectra of the crystal phases of the pyrolysis residues were performed from 5° to 90° at a scan rate of 10° min⁻¹ carried out on Rigaku D/max-2500 diffract meter with Cu-Kα radiation source (λ = 0.154 nm).

The morphology of the residues who was covered the pyrolysis surface was observed by scanning electronic microscopy (SEM) whose accelerating voltage is 20 kV and whose manufacturer is HITACHI S4700 instrument. The samples that will be measured were covered with Pt, and energy dispersive x-ray spectroscopy (EDS) was performed for elemental composition determination.

By measuring the height change of residues in the cylindrical instrument, the volume change in the pyrolysis process was obtained. The calculation formula of the volume change is as follows:

\[
\text{volume variation(\%) =} \frac{(\text{Volume residue} - \text{(Volume) sample})}{\text{(Volume) sample}} \times 100\%
\]

(\text{Volume}) residue represents the volume of residue calculated by the diameter and high of the residues. (\text{Volume}) sample represents the volume of original samples.

| Sample | T onset | T max | Yield at 800 °C (%) |
|--------|---------|-------|---------------------|
| SAM0   | 426     | 489   | 47.84               |
| SAM1   | 440     | 499   | 51.21               |
| SAM2   | 374     | 532   | 52.04               |

![Figure 1. TGA and DTG curves of SAM0, SAM1, and SAM2.](image)
3. Results and discussion

3.1. Thermogravimetric analysis
The thermal behavior of composites can be divided into thermal stability and thermal decomposition behavior at different temperatures. The thermal behavior parameters of the composites can be obtained by TGA. The obtained data are TGA curve and DGA curve, as shown in Figure 1. and relevant data are listed in Table 2. $T_{\text{onset}}$ and $T_{\text{max}}$ represent the temperature of 5% weight loss and the temperature of peak degradation, respectively. It can be seen that SAM0 begins to decompose at about 400 °C and there are two steps in its thermal degradation process. The first degradation step occurs in the temperature range of 380 °C–580 °C, indicates the decomposition of the silicone rubber matrix, the second degradation step occurs in the temperature range of 600 °C–700 °C represents the decomposition of CaCO$_3$. It can be seen that with the addition of Mica into the composite material, the initial decomposition temperature is not change. However, compared with SAM0, when 10phr of AHP is used instead of Mica, the thermal degradation of SAM2 systems is similarly composed of three steps. The temperature of peak degradation is higher than SMA0 and SAM1, which indicates the presence of AHP retarded the decomposition of the silicone rubber matrix. The first step happens from 374 °C, which belongs to the degradation of AHP. The degradation of AHP produced phosphine and aluminum phosphite. The second step from 500 °C–600 °C is ascribed to the depolymerization of the siloxane chains by producing low molecular weight cyclic siloxane [4, 10, 23, 24] and aluminum phosphate elimination of water to produce aluminum polyphosphate. It can be seen that the pyrolysis yield of SAM0 at 800 °C is 47.84%, when adding porcelain filler, the yield has a certain upgrade, what’s more, when aluminum hypophosphite replaced the Mica, the yield of residue improved indicating the role of aluminum hypophosphite silicate of promoting the formation of the residue.

3.2. LOI and UL-94
The LOI and UL-94 results are shown in Table 3. it can see that the oxygen index (LOI) of unmodified samples is 30.6%, and there is no grade for vertical combustion (UL-94) due to the completely burning. When AHP was added, the oxygen index increased to 33.9%. It is well known that AHP is a phosphorus flame retardant which can generate a glass partition layer to cover the material surface to prevent the heat transfer in the air to achieve the flame retardant when firing [1, 6]. However, the samples still burned completely cause having no UL-94
grade either. With the addition of Mica, the change of oxygen index is very small. When 25 parts of Mica are added, the LOI is 34.8% and the UL-94 is FV1, Mica powder is layered mineral with excellent insulation and heat insulation. When the temperature is low, the structure of Mica powder is relatively stable and can only play the role of heat insulation, therefore, it does not greatly improve the flame retardant performance of the CSR matrixes. with the increase of Mica added in, ceramization occur when firing [7–10, 14].

3.3. Macroscopic morphology and volume change

Figure 2 shows the macroscopic morphology of composites after fired at various temperatures. The residues of the composites after pyrolysis at 650 °C showed good regular shape and a few cracks appear on the surface, all the residues are much bigger than the initial sample. Ceramization occurred at high temperature when the filler calcined to produce a hard solid. When the temperature exceeds 950 °C, CSR produces an obvious volume shrinkage, and there are no obvious cracks on the surface of the composites after pyrolysis, but there are many pockets that can be found on the surface of the residue. These results are accord with the previous study [25].

Volume variations of the residues of the composites at different temperatures were measured and presents in table 4. It shows that before 750 °C, the volume varies in expansion because the silicone rubber matrix decomposes to produce gas resulting in expansion. When AHP replaced the Mica, the volume has shrinkage because of the melting of glass frits and the replacement of glass frit by ammonium polyphosphate with aluminum hydroxide results in the decrease of the ceramization temperature of silicone rubber and the improvement of the mechanical properties of silicone rubber at low temperatures. Moreover, eutectic products of silicone rubber and Mica can enhance the mechanical properties at high temperatures above 800 °C [14] because of the reactions with the phosphates.

### Table 4. Volume variations of the residues of the composites at different temperatures.

| Sample | 650 °C | 750 °C | 850 °C | 950 °C | 1000 °C |
|--------|--------|--------|--------|--------|---------|
| SAM0   | 48     | 31     | 10     | −4     | −7      |
| SAM1   | 52     | 16     | −3     | −51    | −53     |
| SAM2   | 49     | −7     | −23    | −46    | −36     |

### Table 5. Mechanical properties for CSR.

| Sample | Tensile strength (MPa) | Elongation at break (%) |
|--------|------------------------|-------------------------|
| SAM0   | 3.23                   | 167                     |
| SAM1   | 3.75                   | 256                     |
| SAM2   | 3.88                   | 315                     |

3.4. Mechanical properties

3.4.1. The tensile strength and elongation at break

The tensile strength of SAM0, SAM1 and SAM1 samples have been measured by the tensile test and the elongation at break test. The results of the tensile strength are listed in table 5. It can be seen that SAM1 shows the tensile strength of 3.75 MPa and the elongation at a break of 256%. The silicone matrix filled with SiO2 and CaCO3 produce an acceptable tensile strength, the tensile strength increased by 0.52 MPa and elongation at break increase by 89% when Mica was added, it is because the original crosslinking equilibrium is disturbed by the addition. SAM2 has the best mechanical properties of 3.88 MPa for tensile strength and 256% for elongation.

3.4.2. Compression strength of the residues

The compression strengths of the pyrolysis at different calcined temperatures are shown in table 6. In the compression test, the maximum compressive stress applied to the specimen until it ruptures has been recorded and averaged to evaluate the mechanical strength of the residues. With the temperature rising, the compression strength is promoted to a higher level. Compared with the result at 650 °C, the compression strength has an enormous increase to 1000 °C. The compression strength of the composites without porcelain filler is in the range of 0.07 to 1.91 MPa, indicating that there is a close correlation with temperature. When added in 15 phr Mica, the compression strength is in the range of 0.09 to 8.31 MPa. At the same temperature, the addition of Mica silicone rubber composite material has been significantly improved. Also, when 10 phr of hypophosphite instead of Mica, the compressive strength has been more improved. What’s more, it can reach 14.5 MPa in 1000 °C
mainly for the generation of aluminosilicate and phosphates. It can be seen that when the temperature is about 650 °C, their compressive strength is no different, mainly for the temperature is low, mainly by the melting of the physical bonding of glass powder, with the temperature rise, the filler began to react, the intensity started to increase. It can be seen that the phase compositions, microstructural features, and the calcined temperature have a great influence on the compression strength.

In summary, SAM2 has highest mechanical properties before and after calcination due to the bonding of glass powder at low temperature and the adhesive phosphate generated by the reactions in the process of firing at high temperature [9, 13]. The thermal decomposition produces at low and medium temperature, as the eutectic material generates at high temperatures.

### 3.5. XRD analysis

XRD patterns of the ceramic residues were shown in figure 3. XRD patterns of SAM0 sintered at different temperatures of 650 °C, 850 °C, and 950 °C are illustrated in figure 3(a). As displayed in figure 3(a), it can be seen that a large hump appears at around 2θ = 20°–30°, confirming the presence of pyrolysis product of amorphous SiO2, the peak of the calcite appeared at 2θ = 28.9°, indicating the silicone rubber decomposition had taken place and calcium carbonate has not decomposed. As the temperature arising to 850 °C, the peak of calcite disappeared along with a new peak at 25.7° and 30.3°, which proved to the formation of wollastonite (CaSiO3). This suggests the decomposition of calcium carbonate and the reaction with SiO2. After heated to 950 °C, a new peak appearing at 21.7° became a major phase that the amorphous SiO2 phase gradually transforms into the crystalline state as the temperature increases, what’s more, the peak of fluorapatite (Ca5(PO4)3F) increased, for the reaction of frits with fillers.

Figure 3(b) and (c) show the XRD patterns of SAM1 and SAM2 at the temperature of 650 °C, 850 °C, and 950 °C. At 650°C, it is clearly showing that 2θ = 27.3° is the muscovite diffraction peak, indicating the presence of the Mica. With the calcined temperature increased, the contents of the peak of fluorapatite increased, meanwhile, the peak of muscovite at 850 °C can be seen, which indicating the part of the presence of muscovite. It could be observed that the XRD spectra were significantly similar and the characteristic peak of muscovite became weaker and the peak of berlinite (AlPO4) appeared at 2θ = 21.3° and 2θ = 26.1° as the calcined temperature increased from 650 °C to 850 °C and 950 °C which was attributed to the cristobalite crystalline phase appearance [26] as shown in figure 3(b). From figure 3(c) when 10 phr of aluminum hypophosphite replaced Mica, in addition to the peak of muscovite and the peak of fluorapatite peak intensity is more pronounced, the contents of the amorphous phosphate increased at 650 °C. When heated to 850 °C, the peak of muscovite disappeared showing the muscovite reacted with SiO2 or other fillers completely. Compared with SAM1, at a temperature up to 950 °C, the peak of berlinite disappeared, explaining that a new reaction between

| Sample | 650 °C | 750 °C | 850 °C | 950 °C | 1000 °C |
|--------|-------|-------|-------|-------|--------|
| SAM0   | 0.07  | 0.14  | 0.17  | 0.39  | 1.91   |
| SAM1   | 0.09  | 0.77  | 1.16  | 6.77  | 8.31   |
| SAM2   | 0.11  | 1.23  | 1.93  | 8.56  | 14.46  |

![Figure 3. XRD patterns of the ceramic residue at different temperatures. (a) SAM0, (b) SAM1, (c) SAM2.](image-url)
Mica, AlPO₄, SiO₂, and CaO mainly exchanged fluorapatite, and some peak of Ca₃(PO₄)₂ has been detected. The formation of these compounds might then indicate the chemical interactions between active Mica and phosphates. These changes indicated that the reactions have taken place in Mica, aluminum hypophosphite,

Figure 4. SEM images of the ceramics of SAM0 at 650 °C(A1), 850 °C(A2), 950 °C(A3), SAM1 at 650 °C(B1), 850 °C(B2), 950 °C(B3), SAM2 at 650 °C(C1), 850 °C(C2), 950 °C(C3).

Figure 5. Ceramization process of SAM2 composites.
frits, and silicone rubber matrix, resulting in the formation of phosphates at low temperature and aluminosilicate at high-temperature, which lead to strengthening the ceramic residues [26] in accordance with the compression strength of the fired samples.

3.6. Microstructure analysis of the pyrolysis residues
The relationship between the microstructure and the compressive strength of the fired ceramic can be seen in figure 4, which shows the SEM images of the interior structure of residues at different temperatures filled with different fillers. (A1), (B1) and (C1) shows the residues of SAM0, SAM1, SAM2 at 650 °C, the matrix seems to form a compact powder with no or limited bling, seen from A1 and B1, but when AHP instead of Mica, it can see there is some blinder to form an adhesive the powder as observed in C1. As shown in figures 4 (A1), (A2), (A3), it is clear that without porcelain filler SAM0 at 650 °C, 850 °C, 950 °C, and the structure of the residue is very loose during the temperature change and does not form a distinct continuous phase. The SAM1 and SAM2, which are added into the porcelain filler, can observe the presence of the continuous phase during the temperature change process. For SAM1, due to the low liquid phase, it is clear that the Mica layer is separated and the Mica sheets bonded together, resulting in lower mechanical strength. At the same time, it can be seen that the bridge structure is bridged and the whole particle is wrapped to form a continuous and dense skeleton structure, which can be observed in figures 4 (B1)–(B3), (C1)–(C3). Compared with the SAM1, when 10 phr of AHP replaced Mica, SAM2 combustion after the formation of carbon layer is more continuous, especially in the low temperature, for there is no new phases are formed mainly by the adhesion of phosphate melting, with the less layered structure being formed with the addition of AHP liquid phase of the ceramic continuous phase for the adhesive surface enhance by the reaction between Mica and AHP. Thus, the compression strength of the residues became higher.

3.7. Ceramization mechanism
The ceramization process mechanism is proposed in figure 5. The calcium carbonate, Mica, AHP, and frits uniform distribution in the silicone rubber matrix, when the temperatures increase from 400 °C to 500 °C, the thermal degradation of the AHP begin, while the strength of residue is mainly caused by the melting of phosphate and parts of glass frits. With the temperature rise to 700 °C, the molten frits bonded the fillers and silica produced from pyrolysis of silicone, and the decomposition products of calcium carbonate, they react and produce calcium silicate. When the temperature is below 700 °C, the samples are in the state of volume expansion macroscopically. With the further increase of temperature over 800 °C, the fillers were melted completely, so the samples are in the state of volume contraction macroscopically. As the liquid phase contacts with the inner Mica sheet, the continuous phase gradually forms on the ceramic surface. This is why the boundary between the liquid phase and the Mica layer is fuzzy [26]. The new eutectic compound formatted by the reaction between the Mica and the filler can be seen from figure 3 resulting in compression strength increases, the volume shrinkage macroscopically.

4. Conclusion
The microstructure and mechanical properties with different content of AHP and Mica at different calcined temperatures has been investigated. The XRD pattern showed reactions have taken place between Mica, AHP, frits, and CSR matrix resulting in the formation of phosphates at low temperature and aluminosilicate at high temperature. The SEM showed the structure of the residue changed from powder to a continual matrix because of the generated liquid phase. The volume expands first before 850 °C and shrinks later when calcined temperature increased in accord with the ceramization process diagrammatic sketch. AHP/Mica added CSR have better mechanical properties and fire resistant than pure silicone rubber. AHP acts as a flux at low temperature and can effectively reduce the ceramization temperature, Mica powder is layered mineral with excellent insulation and heat insulation. AHP/Mica modified CSR have better flame retardation properties than pure silicone rubber, the addition of AHP increase LOI to 33.9%, the LOI is 34.8% and the UL-94 is FV1 as 25 parts of Mica are added. SAM2 has the best mechanical properties before and after calcination of 3.88 MPa for tensile strength, 256% for elongation and 14.46 MPa for compression strength calcinated at 1000 °C.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Conflicts of interest/Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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