

Experiment Research on the Mechanical Property of Micron Particles Filled Silicone Rubber Material

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Abstract: Six kinds of different content of micron solid particles of silicone rubber materials were prepared by mixing micron Al_2O_3 , SiO_2 , cross-linking agent, and catalyst, etc. into the matrix material of micron-sized hydroxyl-terminated polydimethylsiloxane. The effect of micron solid particles on the mechanical property of silicone rubber was examined by the uniaxial tensile tests. Non-contact strain test system was adopted to measure the strain response of the samples in the experiments. The results show that, the mechanical property of silicone rubber material is rate-dependent, presenting a hyper-elastic property, which is similar to rubber-like material. The strength of the granular filled material increased with the content of the solid particles, moreover, the enhancement effect caused by SiO_2 was stronger than that caused by Al_2O_3 .

Keywords: Silicone rubber, mechanical property, uniaxial tension, rate-dependence, breaking strength.

1. INTRODUCTION

Silicone rubber is one of the little smoke thermal protection materials which has been generally researched in the field of aeronautics and astronautics because of favorable properties of thermal stability, high temperature resistance and aging resistance [1]. In the 28th joint propulsion conference held by AIAA, it was mentioned that polydimethylsiloxane is likely to be the next generation of thermal protection material [2]. Recently, studies on silicone rubber mainly focused on the material prescription, ablation property and aging property. Research results show that the mechanical property of the thermal protection material is worth studying under the environmental loading and high temperature gas brush [3]. However, there are still many deficiencies in the studies analyzing mechanical property of silicone rubber, especially the micron particles filled silicone rubber. Therefore, it is necessary to conduct a detailed study on the mechanical property of silicone rubber material.

To lucubrate the mechanical property of silicone rubber material, Kwan [4] studied the rubber reinforcing effect of SiO_2 produced by sol-gel method, and the results show that different elastic modulus and hardness of silicone rubber material can be obtained through adjusting the proportion of ethylene content and solid particle content. Meunier [5] conducted a series of tensile tests, pure shear tests and compression tests on silicone rubber material without any fillers in it. Five kinds of hyper-elastic models were used to compare the experimental results, and the Mullins effect, hysteresis and rate-dependence were ignored in the

study. Xu [6] studied the mechanical property of silicone rubber material with different contents of ethylene and found that the material has viscoelastic properties. The glass transition temperature of the material was obtained by dynamic mechanical analysis (DMA) technology. Korochkina [7] adopted experimental and numerical methods to study the applicability of constitutive models of silicone rubber. Yingwen Zhou *et al.* [8] discussed the influence of micron-sized particle with a diameter of Al_2O_3 and the filled level on the mechanical property of silicone rubber, where the mechanical property was used to compare with the nano-sized Al_2O_3 filled material. Liu Jun *et al.* [9] studied the effect of different kinds and amount of coupling agents on the mechanical property of silicone rubber material, and observed the microscopic structure of colloid section by using the electron microscopy. Wenyong Zhou *et al.* [10] studied the effects of the amount, particle size, and mixing mass ratio of the hybrid alumina filler particles on the thermal conductivity and mechanical properties of silicone rubber. Luheng Wang [11] studied the compressive resistance relaxation of black carbon filled with silicone rubber composite, and the results show that at the stage of the compressive stress relaxation, the composite resistance and the stress both can be adjusted by the linear combination of three negative exponential functions. Yoshimura *et al.* [12] researched the electrical and mechanical properties of the silicone-rubber composites formed by spring-shaped carbon microcoils (CMCs) embedded in silicone-rubber. Results show that with a compressive or tensile strain, the resistivity increased more greatly than any of the other composites.

According to the above review, researchers have studied the effect of content and the kind of filler on the mechanical property of silicone rubber material, and further studied the constitutive model of this kind of material, which lay a

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theoretical foundation for the application of the material. However, the theoretical study was still in a fledging period, and needs to be further developed. Here, hydroxyl-terminated polydimethylsiloxane (silicone rubber) was taken as the matrix material, and the influence of micron SiO_2 and Al_2O_3 particles on the mechanical property were studied, laying the foundation for the constitutive model of particle-filled silicone rubber material.

2. PREPARATION OF MATERIALS AND SPECIMENS

The silicon rubber with thermal protection material used in this experiment was made up of silicone rubber (107, Beijing Chem. Plant, China) as the matrix [11], Al_2O_3 with 5 microns particle size, SiO_2 with 3 microns particle size, cross-linking agent and catalyst. The diameters of all the micron solid particles were around 20 micron. The equipment used in the preparation of the material included: vacuum chamber, mixer, beaker, medical syringe and homemade mould. Raw materials required include: matrix material (silicone rubber), catalyst (dibutyltin dilaurate), cross-linking agent (tetraethoxysilane), mold release agent (methyl silicone oil) and filler materials (micron Al_2O_3 and SiO_2 particles). The process of material preparation can be divided into the following five steps:

- (1) Mix fillers into matrix (silicone rubber) in a beaker and stir with a speed of 300-400 rpm for 10 minutes, to obtain a mixed rubber that has uniformity and stable performance
- (2) Add some cross-linking agent and catalyst in the mixed rubber, and continue stirring with the speed of 300-400 rpm for 15 minutes;
- (3) Put the mixture into the vacuum chamber for 20 minutes;
- (4) Inject the cure rubber into the mold, and then keep the mixed material in the vacuum chamber for three hours again (methyl silicone oil is used as mold release agent);
- (5) Keep the mixed material for seven days at room temperature after the inner bubbles disappear.

Micron particles filled silicone rubber materials with different mechanical property were obtained based on different content of micron particles in matrix, while keeping the technological parameters consistent. Different silicone rubber materials were used including Mass proportion of micron Al_2O_3 particles filled in the matrix was 10%, 20% and 30%. These materials were used to study the effect of micron Al_2O_3 particles on the mechanical property of silicone rubber material. (2) The proportion of micron SiO_2 particles filled in the mixture with 10% micron Al_2O_3 particles was 2%, 4% and 6%. These materials were used to study the effect of micron SiO_2 particles on the mechanical property of micron particles-filled silicone rubber material.

3. EXPERIMENT STUDY

3.1. Experiment

QJ-211B electronic universal testing machine (EUTM) and incubator were used to conduct the experiment; the

temperature was $20^\circ\text{C}\pm 2^\circ\text{C}$, and humidity was $50\%\pm 2\%$, due to the limited range of the EUTM's force sensor of 20000 N, which is significantly larger than the tensile strength of silicone rubber. In order to avoid large system error induced by the sensor, a sensor with a range of 200 N was used to measure the mechanical response.

Table 1. Silicone rubber materials with different content of micron solid particles.

Material	Al_2O_3	SiO_2
silicone rubber 1	10%	0%
silicone rubber 2	20%	0%
silicone rubber 3	30%	0%
silicone rubber 4	10%	2%
silicone rubber 5	10%	4%
silicone rubber 6	10%	6%

The design of specimens was considered according to the slab construction characteristic of the prepared silicone rubber. The gauge length of the specimen was 50 mm. Glue was used on each end of the specimen to adhere with aluminum joints, to construct the dumbbell tensile specimen [12] according to the aerospace industry standard of PRC, QJ 924-85. This kind of design can avoid relative slip between the specimens and chucks of testing machine and ensure the gauge length of the specimen is 50 mm (Fig. 1).

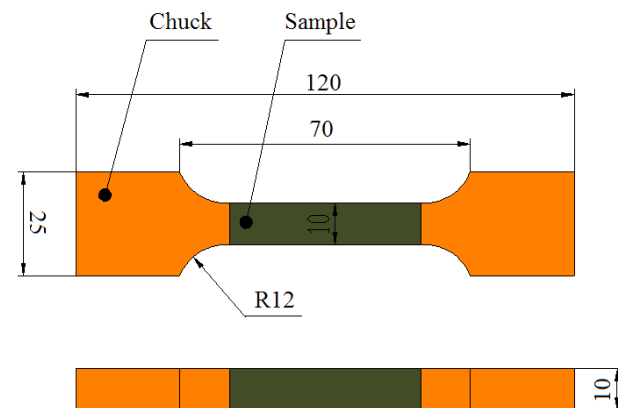


Fig. (1). Tensile specimen of micron particles filled silicone rubber material.

Non-contact test method was used to measure the strain response in the experiments. The system was mainly used to analyze the specklegram of sample surface filmed by CCD and automatically and quickly analyze the displacement and strain of the complete field, as shown in Fig. (2).

In order to study the rate-dependent mechanical property of the micron particles-filled materials, the stretching speeds of EUTM were set as 5 mm/min, 50 mm/min and 500 mm/min.

3.2. Data Processing

Experiments under each loading condition were repeated many times, and 3 effective results were selected to obtain

the average results as the final experimental curve. Taking micron particles-filled silicone rubber 1 for example, three $\epsilon-t$ effective curves for 50 mm/min tensile speed can be obtained, and the strain response measured by non-contact test system is shown in Fig. (3).

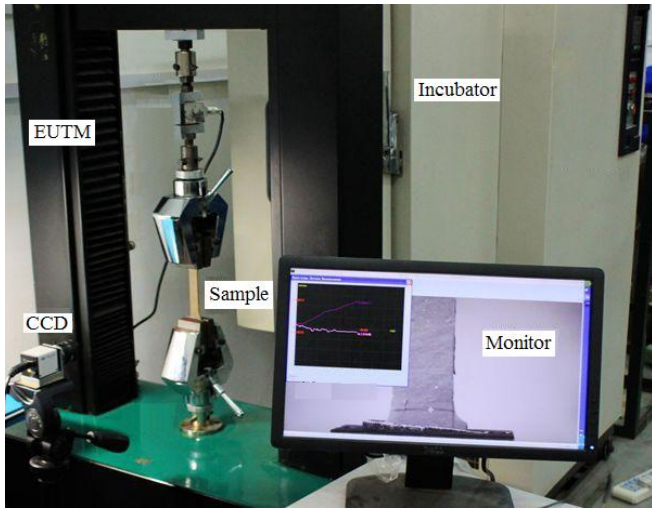


Fig. (2). Progress of non-contact strain test.

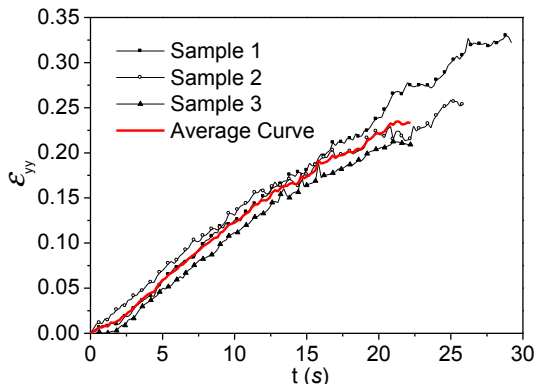


Fig. (3). Strain response measured by non-contact test system (50 mm/min).

As shown in Fig. (3), the strain rate of the specimens mainly obeyed the linear characteristic which is similar to the constant tensile speed (50 mm/min), which illustrates that the stress-strain response of the silicone rubber 1 under constant strain rate can be obtained, and constant stretching rate can be calculated by fitting the $\epsilon-t$ average curve. The strain rate is $0.011s^{-1}$ (50 mm/min). Thus, the $\sigma-\epsilon$ response can also be obtained by combining the constant strain rate value with the $\sigma-t$ curves measured by force sensor. The actual constant strain rate for 5 mm/min and 500 mm/min are $0.00146s^{-1}$ and $0.1s^{-1}$ obtained by the same method, respectively.

Five $\sigma-\epsilon$ curves of silicone rubber 1 with a tension of 50 mm/min are shown in Fig. (4). It can be found that the stress-strain characteristics of the material are similar to each other, but the scattered error of elongations is larger

. The cause of these problems includes:

- a) Debonding between the particles and the matrix occur in the particle-filled materials under certain loading, as shown in Fig. (5), with different scattered errors in

the mechanical property due to different particle shape, size and orientation.

- b) Cure conditions were different with the different location of cure mold during the preparation of the material, leading to the different mechanical property of the silicone rubber material.

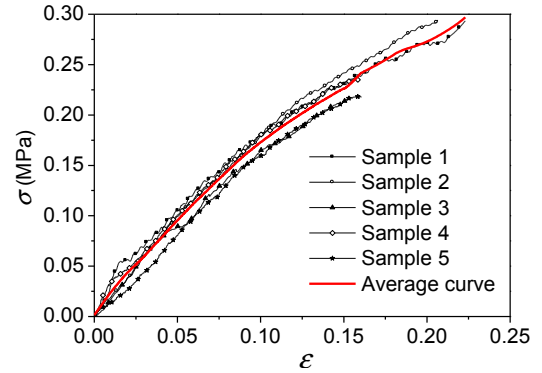


Fig. (4). $\sigma-\epsilon$ Curves (50 mm/min).

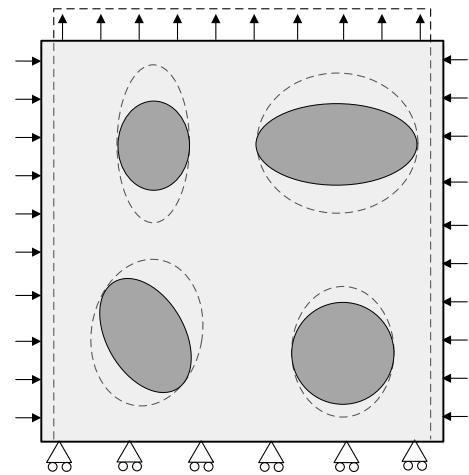


Fig. (5). Matrix filled with particles.

4. RESULTS ANALYSIS AND DISCUSSION

4.1. Effect of Strain Rate on Mechanical Property

According to the data processing method mentioned above, stress-strain responses of micron particles-filled silicone rubber material were obtained under all loading conditions. Here, the influence of strain rate, micron Al_2O_3 content and micron SiO_2 content on the mechanical property of the material was analyzed.

The test results of silicone rubber 1 and silicone rubber 4 are given in Fig. (6). It can be found that the mechanical property of micron solid particles-filled silicone rubber was similar to the matrix material, which is rate-dependent, and depends on the time and loading history, belonging to a typical viscoelastic mechanic property. Tensile strength significantly increased with the increased tensile rate, which is associated to viscosity characteristics of the matrix material. Elongation also ultimately increased with increased strain rate. However, the inverse change law shown in Fig. (6b), was observed due to the scattered errors in the mechanical property induced by micron solid particles.

Micron solid particles-filled silicone rubber materials in Table 1 have good elastic property and their maximum elongation is more than 25%. Furthermore, the materials showed hysteresis characteristics during unloading, as the strain returned to the initial state gradually after 30 minutes, which can be described by a visco-hyper-elastic constitutive model as the rubber-like material. Thus, micron solid particles-filled silicone rubber belongs to the polymer having both hyper-elastic and viscoelastic properties. In few micron particles-filled materials, a similar behavior has been reported [13-15]. For better application in the field of engineering, the mechanical property of the material should be researched in detail.

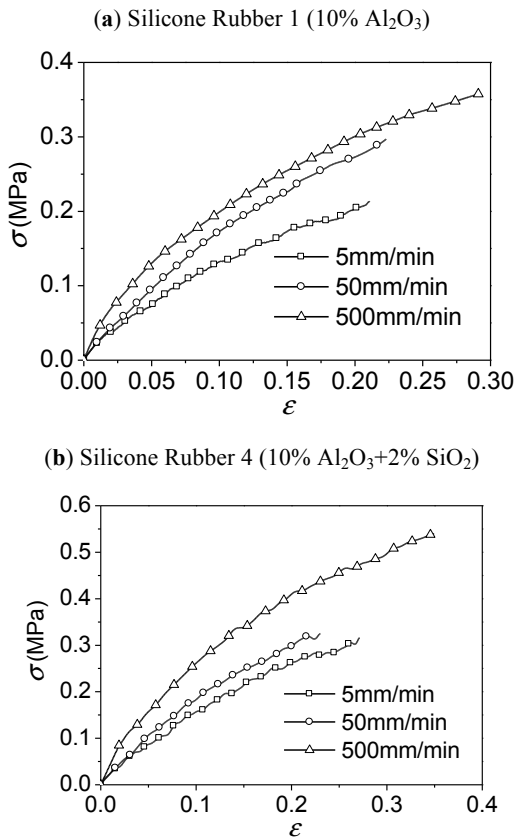


Fig. (6). σ - ε curves for different loading conditions.

4.2. Effect of Micron Solid Particles Proportion on Mechanical Property

The comparisons of mechanical property induced by different contents of micron Al_2O_3 and SiO_2 particles are shown in Fig. (7). The tensile strength of the material increased with the content of micron solid particles with any tensile speed. However, the maximum elongation showed large scattered errors and lack of regularity. This was due to lots of solid particles with high strength distributed in the silicone rubber matrix. The matrix was divided into many discontinuous matrix segments along the loading direction, as shown in Fig. (8), to increase the modulus of the matrix segments, resulting in increasing the materials' tensile strength along with the content of solid particles. This mechanical property is similar to the property of a spring, and it can be said that the modulus of a spring increased with the decrease in the spring's length.

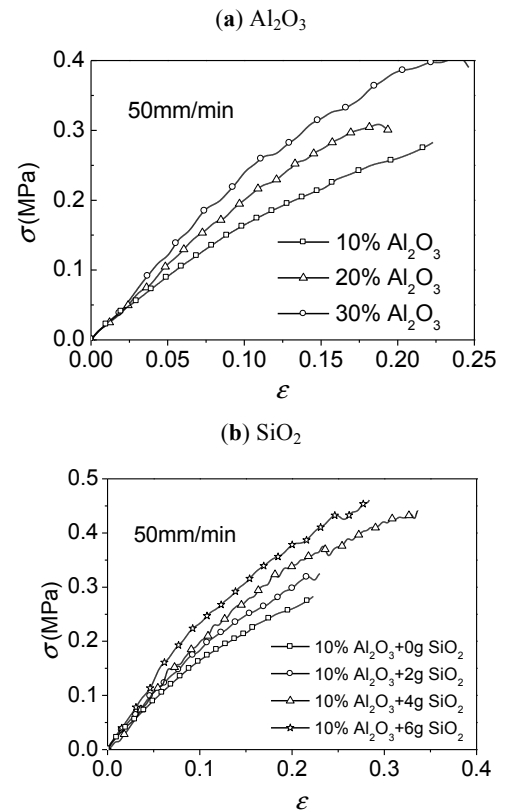


Fig. (7). Effect of micron solid particles' content on the mechanical property.

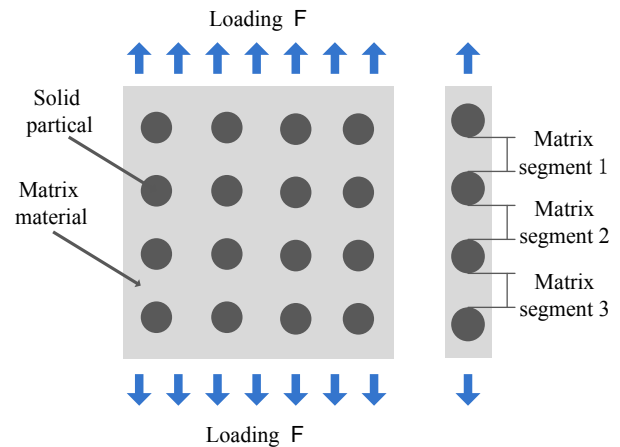


Fig. (8). Microstructure of micron particle filled material.

In Fig. (9), the tensile strength of six kinds of micron solid particles-filled silicone rubber was considered to make comparison with each other. It can be observed that micron Al_2O_3 and SiO_2 particles increased the elastic modulus of micron solid particles-filled silicone rubber at a certain degree, while the enhancement effect of micron Al_2O_3 was significantly less than that of micron SiO_2 . This was because the particle size of micron Al_2O_3 was greater than that of micron SiO_2 , then the content of micron SiO_2 particles was more than the content of micron Al_2O_3 per unit volume. According to the hypotheses represented in Fig. (8), the matrix segments were divided micron SiO_2 particles are more than matrix segments cut by micron Al_2O_3 particles, so the enhancement effect of micron SiO_2 particles is greater than that of micron Al_2O_3 particles.

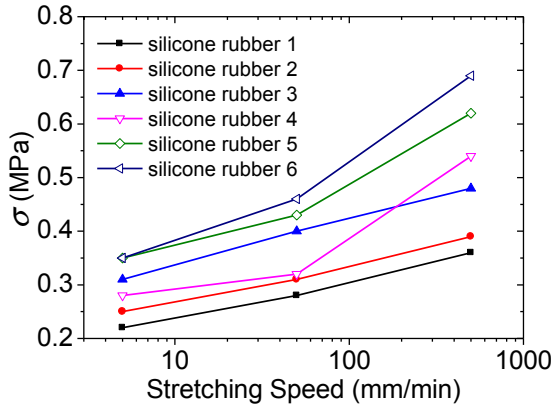


Fig. (9). Tensile strength of the materials with different content of micron solid particles.

To sum up, the mechanical property of micron solid particles-filled silicone rubber was observed to be largely rate-dependent, and the scattered errors in the elongations were larger. This phenomenon reflects that the rate-dependent mechanical property of micron solid particles-filled silicone rubber which is mainly due to the property of matrix material, and micron solid particles increased the modulus of the matrix. Moreover, the instability of bonding performance between micron particle surface and matrix led to the bigger difference in the elongation of the material.

5. RATE-DEPENDENT STRENGTH CRITERION

5.1. Strength Criterion

The octahedral shear strain failure criterion has been widely applied to polymers, i.e.

$$\gamma_8 \leq \frac{\gamma_{8m}}{n} \quad (1)$$

where γ_{8m} is the critical value, and n is the safety coefficient. The octahedral shear strain can be expressed as:

$$\gamma_8 = \frac{1}{3} \sqrt{(\varepsilon_1 - \varepsilon_2)^2 + (\varepsilon_2 - \varepsilon_3)^2 + (\varepsilon_3 - \varepsilon_1)^2} \quad (2)$$

where ε_1 , ε_2 and ε_3 are the principal strains. Under uniaxial tension, $\varepsilon_2 = \varepsilon_3 = -\mu\varepsilon_1$, μ is the poisson's ratio, then γ_{8m} can be expressed as the function of the largest elongation ε_m

$$\gamma_{8m} = \frac{\sqrt{2}}{3} (1 + \mu) \varepsilon_m \quad (3)$$

Mises equivalent strain can be described as:

$$\varepsilon_e = \frac{\sqrt{2}}{3} \sqrt{(\varepsilon_1 - \varepsilon_2)^2 + (\varepsilon_2 - \varepsilon_3)^2 + (\varepsilon_3 - \varepsilon_1)^2} \quad (4)$$

where ε_e is the equivalent strain. Under uniaxial tension, the relationship between the critical value ε_{em} and the largest elongation ε_e can be described as:

$$\varepsilon_{em} = \frac{2}{3} (1 + \mu) \varepsilon_m \quad (5)$$

Thus, the octahedral shear strain γ_8 and Mises equivalent strain ε_e have the following relationship:

$$\varepsilon_e = \sqrt{2} \gamma_8 \quad (6)$$

It is shown that the Mises equivalent strain criterion and octahedral shear strain criterion were essentially equivalent. Micron solid particles-filled silicone rubber can be seen as incompressible materials, as the Poisson's ratio is usually between 0.49 ~ 0.499, thus making the maximum Mises strain approximately equal to the largest elongation ε_m according to Eq.(5). Therefore, the largest elongation was used as the failure criteria of micron solid particles-filled silicone rubber specimen in this paper.

Similarly, for micron solid particles-filled silicone rubber, Mises equivalent stress can be expressed as:

$$\sigma_e = \frac{\sqrt{2}}{2} \sqrt{(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2} \quad (7)$$

Under uniaxial loading condition, $\sigma_e = \sigma_1$.

Thus, under uniaxial loading condition, the largest elongation and the tensile strength of the specimen can be directly used as the material's failure criterion.

5.2. Rate-Related Strength Criterion

According to the mechanical test results of micron solid particles-filled silicone rubber. shown in Fig. (9), the tension strength of the micron solid particles-filled silicone rubber showed a nearly linear relationship with the logarithmic value of strain rate except in some special testing condition. Thus, the strength criterion of the material can be defined as:

$$\sigma_{\max} = \sigma_0 [1 + \phi(V) \log V / 5 + \phi(\text{Al}_2\text{O}_3)(\text{Al}_2\text{O}_3\% - 10\%) + \phi(\text{SiO}_2)\text{SiO}_2\%] \quad (8)$$

where $\sigma_0 = 0.22 \text{ MPa}$ is the tensile strength of micron solid particles-filled silicon rubber 1 under 5 mm/min tensile speed; $\phi(V)$ is the impact factor of tensile speed; $\phi(\text{Al}_2\text{O}_3)$ is the impact factor of micron Al_2O_3 particles; and $\phi(\text{SiO}_2)$ is the impact factor of micron SiO_2 particles.

The impacts of micron solid particles and strain rate on the mechanical property of materials were emphatically analyzed as shown below. The strength value (0.22 MPa) of micron solid particles-filled silicone rubber 1 was taken under 5 mm/min tensile speed as the standard, based on the un-dimensioned process of material strength to calculate the impact factors of particles and tensile speed. :

- (1) The strength values under all test conditions divided the value of 0.22 MPa to be non-dimensionalized.
- (2) For the silicone rubber 1, the increase in the ratio of strength with the increase of tensile speed (logarithmic value) was calculated;
- (3) Step (2) was repeated until all silicone rubbers were processed;
- (4) Similar to step (2), the increase in the ratio of strength with the increase in the particles' content was calculated.

The impact factors of tensile speed and micron particles' content influencing the mechanical property of the materials are shown in Fig. (10). Three impact factors $\phi(V)$, $\phi(\text{Al}_2\text{O}_3)$

and $\varphi(\text{SiO}_2)$ under different load conditions were obtained from Fig. (10); the average values of these parameters were taken as the final values as 0.25, 2.5 and 20.45, respectively. To verify the accuracy of the model given in Eq. (7), the predicting tensile strength of silicone rubber 1 and silicone rubber 3 was compared with the test results as shown in Fig. (11).

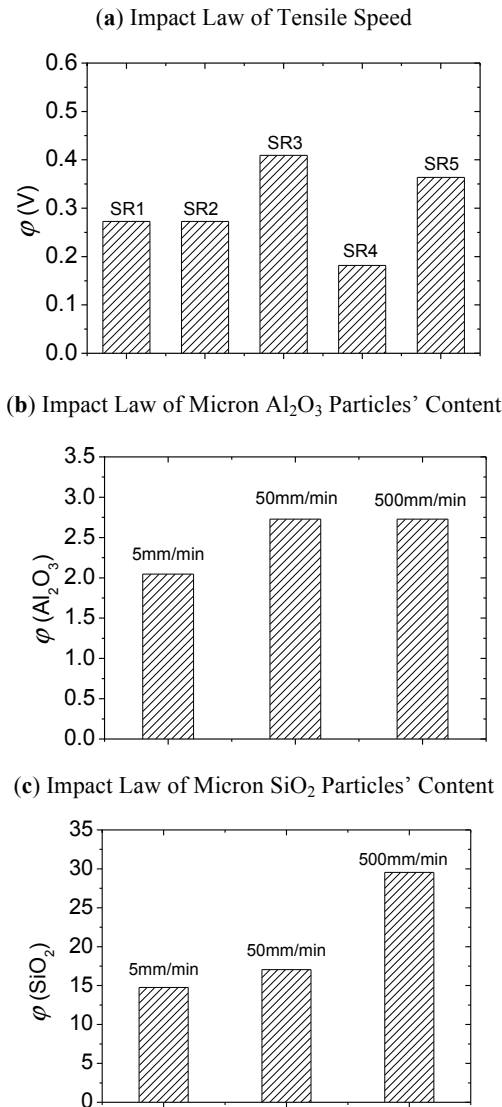


Fig. (10). Impact factor of tensile speed and micron particles' content.

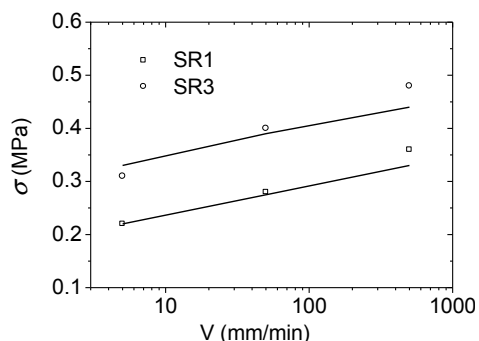


Fig. (11). Tensile strength verification of silicone rubber 1 and silicone rubber 3.

In Fig. (11), the model qualitatively describes the influence of the speed of tensile and micron particles' content on the tensile strength of the micron particles-filled silicone rubber materials, but there are still some limitations. In future, the experimental conditions of micron particles-filled rubber material should be extended to reduce the experimental errors by increasing the repetition number for the same loading condition.

CONCLUSION

Six kinds of micron solid particles-filled silicone rubber materials were prepared by mixing different content of micron Al_2O_3 and SiO_2 particles, cross-linking agent and catalyst into Hydroxyl-terminated polydimethylsiloxane (used as matrix). The effect of micron solid particles and tensile speed on the mechanical property of micron particles-filled silicone rubber was researched by uniaxial tensile test, and the following conclusions were obtained as follows:

- The mechanical property of micron particles-filled silicone rubber is rate-dependent which is similar to the rubber-like material, and the property has a hyper-elastic character. Thus, the silicone rubber belongs to the polymer materials that have both hyper-elastic and viscoelastic properties.
- The tensile strength of micron particles-filled silicone rubber increases with the content of the micron solid particles, but the elongation lack of the change rules due to the fact that lots of micron solid particles with high modulus are distributed in the silicone rubber matrix. (AUTHOR: The highlighted is vague and must be re-phrased) The matrix was cut into many discontinuous matrix segments along the loading direction. the increase in the matrix segments resulted in increase in the materials' strength with the increase in solid particles' content;
- The rate-dependent mechanical property of micron particles-filled silicone rubber is mainly due to the property of matrix. Micron solid particles increased the modulus of the material and the instability of adhesive properties between micron solid particles and matrix, leading to large differences in the materials' elongation.

CONFLICT OF INTEREST

The authors confirm that this article content has no conflicts of interest.

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