# The influence of silica on the properties of addition-type liquid silicone rubber

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**Abstract:** Addition-type liquid silicone rubber has a wide range of applications and has replaced some traditional compounding silicone rubber. This article explores the influence of silica content on the static and dynamic properties of addition-type liquid silicone rubber. Comparative experimental results show that the silica filler network has a significant impact on the properties of silicone rubber. An appropriate amount of silica reinforcing agent can improve the material strength. At the same elongation rate, with the increase of silica filler content, the stress relaxation attenuation of the material intensifies, the relaxation time shortens, the internal friction in the dynamic tensile retraction cycle increases, and the crack propagation rate of the material increases. In practical engineering applications, materials should be selected reasonably.

**Key words:** addition-type liquid silicone rubber; silica; stress relaxation; fatigue

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Addition-type liquid silicone rubber, due to the theoretically non-production of low molecular weight by-products during the hydrosilylation crosslinking process and its high conversion rate, exhibits no shrinkage during vulcanization, is non-toxic, possesses good low compression deformation, and allows for easy control of crosslinking density. The physical and mechanical properties and electrical properties of addition-type liquid silicone rubber after vulcanization can reach or exceed the level of compound silicone rubber. Due to its ability to be injection molded and compression molded, processes such as compounding, tablet pressing, and cutting are not required. It has the advantages of convenient processing, high production efficiency, low cost, and energy saving. Therefore, some traditional compound silicone rubber products have been replaced by liquid silicone rubber. Silicone rubber has excellent high and low temperature resistance, but its tensile strength is lower than that of some modified natural rubbers.

This article prepared a series of liquid silicone rubber materials with different filling amounts by controlling the amount of reinforcing filler silica in the formula, and studied the influence of reinforcing filler silica on the static and dynamic properties of addition-type liquid silicone rubber through tensile property tests, stress relaxation tests, and fatigue tests.

## 1 Experimental part

### 1.1 Raw materials

Liquid silicone rubber uses addition-type liquid silicone rubber, which is a type of organic material that can be crosslinked and vulcanized at room temperature or under heating in the presence of platinum catalysts. It is made from linear polyorganosiloxane containing vinyl groups as the base polymer and oligomeric organosiloxane containing Si-H bonds as the crosslinking agent. The reaction equation is shown as follows:

$$\equiv Si—CH=CH_2+\equiv Si—H\rightarrow\equiv SiCH_2CH_2Si$$

The main components of addition-type liquid silicone rubber include base polymer, crosslinking agent, catalyst,

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reaction inhibitor, reinforcing filler, and additives. Four types of liquid silicone rubber materials were prepared by controlling the amount of reinforcing filler, silica aerogel. Due to the high purity of fumed silica, high reinforcing efficiency, excellent heat resistance, and dynamic fatigue performance of vulcanized rubber, fumed silica was selected. The experimental formula design is shown in Table 1.

**Table 1 Design of liquid silicone rubber test formula**

| Plan | Filling level of fumed silica | Viscosity (Pa·S) |
|---|---|---|
| 1# | Low | 3 000 |
| 2# | Middle | 3 500 |
| 3# | High | 4 000 |
| 4# | Higher | 4 000 |

### 1.2 Apparatus and equipment

Flat curing press; blast high-temperature oven. Rubber hardness tester; tensile machine complies with ISO5893, with a force measurement accuracy of Level 2. The accuracy of the extensometer used in the testing machine is Level D for Type I, Type II, and Type IA dumbbell-shaped samples, and Type A ring samples, and Level E for Type III and Type IV dumbbell-shaped samples, and Type B samples. The testing machine can operate at a minimum speed of 100 mm/min ± 10 mm/min, 200 mm/min ± 20 mm/min, and 500 mm/min ± 50 mm/min; fatigue dynamic testing machine.

### 1.3 Preparation of test samples

Prepare test specimens for the addition-type liquid silicone rubber from schemes 1# to 4# using a flat curing press through molding curing. Conduct a first-stage curing on the flat curing press at 120℃ for 10 minutes, and then proceed with a second-stage curing in a high-temperature blast oven at 200℃ for 4 hours. Cut the specimens into standard test samples using a cutter.

### 1.4 Measurement of performance

The Shore A hardness is tested according to GB/T531.1, with the test sample being measured after being stacked in three layers.

According to the GB/T528 standard, the tensile mechanical properties of the test samples are tested using a tensile machine. The tensile strength is measured using dumbbell-shaped samples at a test speed of 100 mm/min, with a gauge length of 25mm. The test environment temperature is 23±2℃, and the humidity is 50±5%. According to the GB/T529 standard, the tear strength of the test samples is tested using a tensile machine.

The dimensions of the sample for static mechanical relaxation performance testing are 40 mm × 10 mm × 2 mm. Each sample is tested individually. Once the sample reaches a constant extension, the initial stress is immediately recorded. Subsequently, the change in stress is continuously recorded over time. Each protocol is tested three times and the average value is taken.

The dynamic mechanical relaxation and fatigue properties are evaluated in combination with the actual usage of the product as follows: Cut a test sample with a width of 10 mm and a length of 50 mm from a 2 mm thick test piece, with a maximum strain value of 100%. Record the stress-displacement curve of the test sample after the initial 2,000 cycles. Afterwards, apply a 2 mm deep cut on one side of the test sample at its midpoint, and conduct a fatigue test on the sample. The crack propagation rate during the fatigue process of silicone rubber is studied.

## 2 Results and Discussion

### 2.1 Basic mechanical properties

The influence of the amount of white carbon black on the mechanical properties of addition-type liquid silicone rubber is shown in Table 2.

**Table 2 Basic mechanical properties of liquid silicone rubber with different filling levels**

| Number | Shore A hardness | 100% modulus/MPa | 200% modulus/MPa | Tensile strength/MPa | Elongation at break, % | Tear strength / (KN.m-1) |
|---|---|---|---|---|---|---|
| 1# | 40 | 1.04 | 1.84 | 9 | 722 | 25 |
| 2# | 50 | 1.74 | 3.16 | 10 | 593 | 35 |
| 3# | 60 | 3.05 | 4.70 | 10 | 485 | 35 |
| 4# | 70 | 3.42 | 4.89 | 9.5 | 350 | 30 |

As can be seen from Table 2, in addition-type liquid silicone rubber, with the increase in the amount of reinforcing filler silica aerogel, the material hardness and modulus increase, while the tensile elongation decreases. The tensile strength and tear strength first increase and then decrease. The mechanical properties of liquid silicone rubber are related

to the amount of silica aerogel added. The reinforcing effect of silica aerogel on silicone rubber can be explained by the surface effect of the filler particles, that is, the active surface of the filling reinforcing agent particles strongly adsorbs the macromolecular chains of silicone rubber. When silica aerogel is filled into silicone rubber, several molecular chains can be attached to the surface of the silica aerogel particles, forming chemical and physical cross-linking between chains. These particles, which have adsorbed molecular chains, can distribute the load evenly. A large number of chemical interfaces are formed between the silica aerogel particles and the matrix. The presence of these interfaces prevents the expansion of cracks, increasing the energy required to form a unit crack area, that is, the surface energy of the material increases after being reinforced by silica aerogel particles, thereby enhancing the tensile strength of the material. Therefore, the introduction of reinforcing agents increases the surface energy of the material, thus playing a reinforcing role. However, when the filling content of silica aerogel reaches a certain level, silica aerogel becomes difficult to disperse and may undergo agglomeration, leading to a decrease in the reinforcing effect of silicone rubber.

Due to the presence of various sizes of internal microcracks or microvoids during material preparation and loading processes, stress concentration occurs at the internal voids of the material under uniaxial tension. When the stress at the void exceeds a critical value, the microcracks expand to form macroscopic cracks. According to Eshelby's hybrid theory and Griffith's theory of fracture mechanics, when the strain energy released per unit area during crack propagation exceeds the surface energy required to form a unit of free surface, the crack begins to propagate. Assuming that a cylindrical silicone rubber sample is subjected to uniaxial tensile stress, and the radius of the microcracks inside the sample is a, the material surface energy of the silicone rubber is $\gamma_0$, the shear modulus is $\mu_0$, and the Poisson's ratio is$\sigma_0$. The relationship between the tensile strength and surface energy of the silicone rubber can be expressed as follows:

$$\sigma_0=\sqrt{\frac{\pi\mu_0\gamma_0}{(1-v_0)a}} \quad (1)$$

After filling with reinforcing agents, the relationship between the tensile strength and surface energy of reinforced silicone rubber is as follows:

$$\sigma_{re}=\sqrt{\frac{\pi\mu_{re}\gamma_{re}}{(1-v_0)a}} \quad (2)$$

Among them, μre represents the shear modulus of the reinforced silicone rubber, and νre denotes its Poisson's ratio. Silicone rubber is approximately incompressible, and the Poisson's ratios of reinforced and unreinforced silicone rubber are essentially identical. The shear modulus of silica is significantly greater than that of silicone rubber, and the shear modulus of reinforced silicone rubber exceeds that of unreinforced silicone rubber. This demonstrates the reinforcing effect of silica on silicone rubber. However, when the silica filling reaches a certain level and agglomeration occurs, the tensile strength of silicone rubber tends to decrease.

## 2.2 Static mechanical relaxation test

The stress relaxation curve and stress decay curve of the samples from Scheme 1# to Scheme 4# under a fixed extension of 40% are shown in Figures 1 and 2. In Figure 1, f(0) represents the stress of the sample at the beginning of relaxation, and f(t) represents the stress of the sample at time t. By plotting the natural logarithm of the relative stress, lnf(t)/f(0), against time, it can be observed from Figures 1 and 2 that the stress relaxation is fast initially and then slows down. Under the same fixed extension condition, as the amount of reinforcing filler silica increases, the stress decay intensifies.

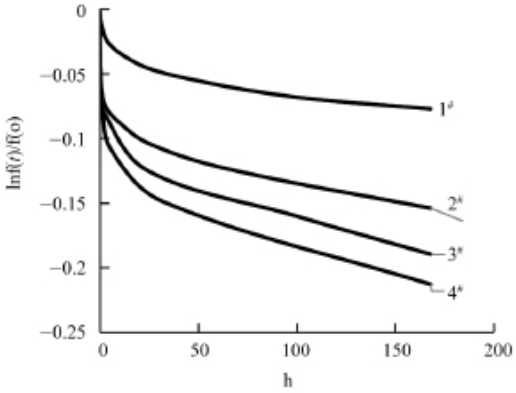


**Figure 1 Stress relaxation of silicone rubber with different fillers**

The reason for stress relaxation in silicone rubber is that under external force, after suddenly generating a constant strain, the entangled polymer segments are forced to straighten along the direction of the external force, generating an internal force that opposes the external force. As time

goes on, the thermal motion of the molecular chains causes the entanglement points to be released, leading to slippage between the molecular chains, adjustment of the molecular conformation, and gradual restoration of its original crimped state. The internal force gradually diminishes, and the external force gradually decays. Since relative displacement cannot occur between the vulcanized molecular chains, the stress can only decay to a certain equilibrium value.

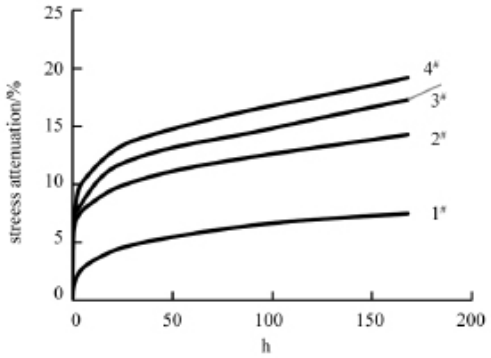


**Figure 2 Stress decay of silicone with different fillings**

In the Maxwell model, a spring with elastic modulus E and a viscous pot with viscosity η are connected in series to represent the viscoelastic deformation process. Based on this ideal model, the relationship between the tensile stress σ applied to the model at time t and the initial tensile stress $\sigma_0$ can be obtained as follows:

$$\sigma=\sigma_0^{e-t/\tau} \quad (3)$$

Where τ represents the relaxation time. Combining Formula 3 with Figures 1 and 2, it can be observed that under constant extension conditions, the relaxation time decreases as the amount of reinforcing filler silica increases.

## 2.3 Dynamic mechanical relaxation

After the initial 2,000 cycles, the recorded stress-strain curves for the samples from Scheme 1# to Scheme 4# are shown in Figure 3:

From the graph, it can be observed that the stiffness of silicone rubber increases with the increase in the amount of silica. The material exhibits viscoelasticity, and the stress-strain curve of the same material is non-linear. During the stretching and retraction process, due to the segmental motion being hindered by internal frictional resistance, the strain cannot keep up with the stress, resulting in the curves during stretching and retraction not coinciding. Under alternating

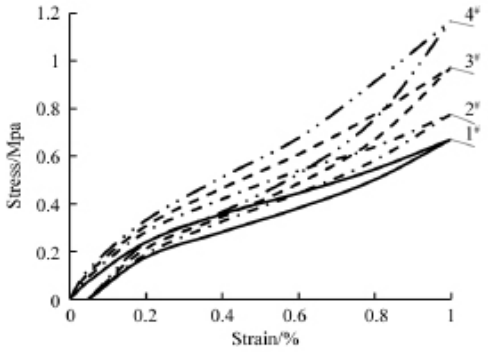


**Figure 3 The impact of different filler contents on the dynamic performance of silicone rubber**

stress, a hysteresis phenomenon occurs where deformation lags behind stress. When silicone rubber is stretched, external force does work on it, and the magnitude of this work is the area enclosed under the stretching curve, which means the silicone rubber absorbs external energy. When silicone rubber retracts, it will do work on the external force, and the value of this work is the area enclosed under the retraction curve, which means the silicone rubber releases energy to the outside world. The difference between these two parts of energy is the internal friction consumed in overcoming the intermolecular internal frictional force and converted into heat. The internal friction $\Delta W$ of silicone rubber in each stretching and retraction cycle can be calculated using formula (4):

$$\Delta W=\pi\sigma_0\varepsilon_0\sin\delta \quad (4)$$

Where $\sigma_0$ and $\varepsilon_0$ represent the amplitude values of stress and strain, respectively; $\delta$ denotes the angle at which strain lags behind stress.

As the amount of silica in silica gel increases, the internal friction in each stretching and retraction cycle correspondingly increases. Factors affecting dynamic relaxation include molecular structure, temperature, and external force frequency. With the increase of silica in silica gel, the internal frictional resistance of segmental motion increases, leading to an increase in internal friction during the stretching and retraction cycle.

## 2.4 Fatigue crack propagation

Under the sine wave loading method with a maximum strain value of 100%, the crack propagation length values of silicone rubber samples from Scheme 1# to Scheme 4# after 2000 cycles are shown in Table 3. Table 3 indicates that as the amount of silica filler in the liquid silicone rubber increases,

the crack propagation rate of the test specimens increases.

**Table 3 Crack propagation length values of liquid silicone rubber materials with different filling levels**

| Number | Frequency/Hz | Crack propagation length (mm) |
|---|---|---|
| 1# | 5 | 6.06 |
| 2# | 5 | 6.16 |
| 3# | 5 | 6..20 |
| 4# | 5 | 6.22 |

Fatigue failure originates from the gradual destruction of microscopic defects or weak spots within rubber materials under the influence of external factors. Generally speaking, the dynamic fatigue process of rubber materials can be divided into three stages: the first stage involves the rubber material softening under stress; the second stage involves the emergence of microcracks on the surface or within the rubber material under continuous external stress; and the third stage involves the development of microcracks into cracks, which continuously expand until the rubber material completely fractures. Since rubber can concentrate stress relaxation when receiving external energy and continuously absorbs it in this form, the fatigue failure of rubber is related to fatigue conditions. Legorju K et al. found that fatigue failure depends on three basic factors: chemical (composition, crystallization), environmental (oxygen), and mechanical (tension, triaxial stress) factors. Based on the crack propagation method, the calculation of rubber fatigue life is as shown in Equation (5):

$$da/dN = f(T, R) \quad (5)$$

In the formula: da/dN represents the crack propagation rate, T denotes the tear energy on a specific plane, and R signifies the ratio of the load valley to the load peak.

Based on the life formula of elastomers estimated from the "theoretical" S-N curve and the relationship between the periodic crack growth rate and the maximum reciprocating tear energy, it can be concluded that the fatigue life of silicone rubber is approximately inversely proportional to its strain energy density. In fatigue tests under the same elongation rate, as the filling amount of silica increases, the strain energy density increases, and the crack propagation rate of the material increases.

## 3 Conclusion

The introduction of silica into addition-type liquid silicone rubber enhances the surface energy of the material, thereby providing reinforcement. However, when the silica content reaches a certain level, it becomes difficult for the silica to disperse, potentially leading to agglomeration and a decrease in the reinforcing effect of the silicone rubber.

During the static mechanical relaxation process of addition-type liquid silicone rubber, the stress changes rapidly in the initial stage and then tends to stabilize. Under constant extension conditions, as the amount of reinforcing filler silica increases, the stress decay of the material increases, and the relaxation time becomes shorter.

During the dynamic mechanical relaxation process of addition-type liquid silicone rubber, as the amount of silica in the silicone rubber increases, the internal friction in each stretching-retraction cycle under the same strain conditions correspondingly increases, leading to an increase in the crack propagation rate of the material.