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URN: urn:nbn:de:gbv:ilm1-2012100142-125-0
URL: http://nbn-resolving.de/urn:nbn:de:gbv:ilm1-2012100142-125-0

Erschienen in:

Mechanismentechnik in Ilmenau, Budapest und Niš : Technische Universität Ilmenau, 29. - 31. August 2012. - Ilmenau : Univ.-Verl. Ilmenau, 2012. - S. 125-134. (Berichte der Ilmenauer Mechanismentechnik ; 1)

ISSN: 2194-9476
ISBN: 978-3-86360-034-1 [Druckausgabe]
URN: urn:nbn:de:gbv:ilm1-2012100142
URL: http://nbn-resolving.de/urn:nbn:de:gbv:ilm1-2012100142

ELECTRICAL AND MECHANICAL TESTING OF CONDUCTIVE SILICONE RUBBER FILLED BY CARBON BLACK NANOPARTICLES

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Abstract

The objective of this investigation is to determine the electrical and mechanical properties of the conductive silicone elastomers and to examine the usability of these materials for mechanical sensor (e.g. force, strain).

The tested silicone elastomers were filled with conductive nanoparticles (Carbon Black, Silver and Nickel). In order to determine the electrical and mechanical properties of the elastomers the resistance was measured at the same time of the mechanical measurements (e.g. tensile, compression and relaxation tests). The complex impedance was also measured as a function of the frequency of the applied voltage. Scanning electron microscopy was performed to view the structure of the conductive particle net.

1. Introduction

There are more and more research on the application of the silicone rubber as a piezoresistive sensor. The hyperelastic strain gauges and the compression force sensors are the two main types [1, 2].

The hyperelastic strain gauges use the large deformation capability of the material. These can be used as a biomechanical stretch sensor, a dress built biometric sensors or sensing element in telemanipulating gloves, etc. [3, 4]

Simple compression force sensors can be built from conductive silicon rubber in varying structure. Some type of tactile sensors (touch-sensible artificial skin, flexible robot grippers) and flexible "sensor carpet" for pressure and force distribution measurement can be built with it [5].

It is possible to build integrated *sensors - actuator* structures (e.g. force or deformation sensing of flexible silicon rubber actuators operated by Internal pressure) from both type of piezoresistive silicone sensors.[4, 6] Some of the integrated sensors work by bending or more complex deformation of the sensor element.

The reason of this investigation is that only specific sensor designs are studied in the literature without examining its general material properties. and the information supplied by raw material producers is not sufficient. Some special mechanical properties of the elastomers (hysteresis, relaxation, creep), are unfavorable in terms of the measurement technology. In case of the electrical properties similar effects can be observed. The question is the applicability of the materials for reproducible measurements.

The conductivity of the polymers filled with conductive particles depends on:

- the dielectric constant of the matrix polymer
- the conductivity of the aggregate,
- concentration,
- spatial distribution and,
- the shape of the particles.

The main external factors influencing the electrical conductivity are:

- the temperature,
- the applied electrical frequency,
- and the deformation state

[7, 8]

2. Materials and Specimens

The specimens were made by transfer moulding and press moulding from conductive silicone rubbers. The applied materials can shown in the tab. 1. All of these silicone rubbers contain pre-mixed conductive particles.

The ELASTOSIL® R 570/50 and R 570/70 one-part, HTV silicones, were press-moulded at 175 °C. In order to obtain the optimal properties they had to be post cured at 200 °C for 4 hours.

The Nusil® R-2631 and R-2637 RTV-2 (two-part) silicones were press moulded at 150°C to accelerate the curing. In this case, the post curing was not necessary.

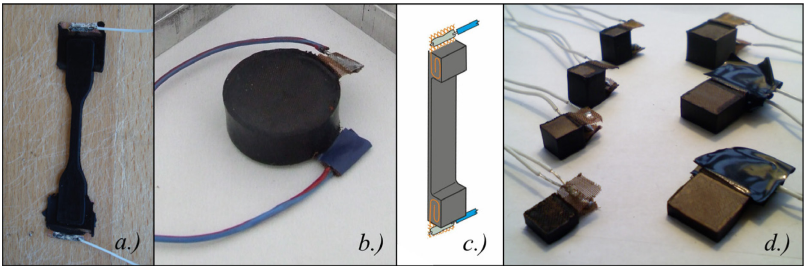

Fig. 1: Test specimens.

Standard, "dumbbell" shaped tensile test specimens (*a.*: ISO 37, L_0=20±0,1 mm; t=2±0,2 mm; w=4±0,1 mm) and cylindrical compression test specimens (*b.*: ASTM D 575, H_0=12,5±0,5 mm; D=28,6 ±0,1 mm) were manufactured for the standard mechanical material tests. Non standard strip shaped tensile test specimens, (*c.*: L_0=20 mm; t=0,5 mm; w=vary) and “box” shaped compression test specimens (*d.*: H_0=2; 4; 8; 12 mm; w=8x8; 13x13 mm) were also made with varying dimensions for strain gauge and compression force sensor test samples.

To measure the resistance, cooper electrode meshes were vulcanized into the end of the specimens to decrease the contact resistance.

Tab. 1: Tested materials

Product Name	Type	Hardness [Shore A]	Aggregate	Volume res. [Ωcm]
Elastosil® R570/70	HTV1	70	carbon black	2.8
Elastosil® R570/50	HTV1	50	carbon black	5.2
Nusil® R-2631	RTV2	40	carbon black	50
Nusil® R-2637	RTV2	60	Ag / Ni	0.006

3. Mechanical and electrical tests

3.1. Test Methods

For the electrical and mechanical characterisation, the resistance was measured during the mechanical measurements (e.g. tensile, compression and relaxation tests). The resistance was recorded with Velleman PC-SCOPE and with NI USB-6008 data acquisition device. Additional interface circuit had to be used to extend the measuring range, because the wide resistance range of the used specimens.

Tensile, compressive and short-term relaxation tests were performed on a Zwick Z020/TN2S tensile test machine. For longer-term relaxation tests, a precision mechanical positioning device was constructed with a load cell.

Fig. 2: Test setup.

3.2. Results

The complete test data is not presented in this paper, only the typical behaviour of the tested materials are shown below.

3.2.1. Tensile tests to failure

The tensile force (F [N]), the elongation (ΔL [mm]) and the resistance (R [Ω]) of the “dumbbell” specimens were measured at constant tensile velocity (10, 100, 250, 500 mm/min). The specimens were stretched until failure in one step. The test were performed at room temperature (22 °C). According to the test results, the resistant characteristic properly follows the stress characteristic (Fig. 3).

Both the initial value and the gradient of the resistance of Nusil® R-2631 are higher than the ones of the Elastosil® R570/70. These are important for sensor developing; because their sensitivity depend on it.

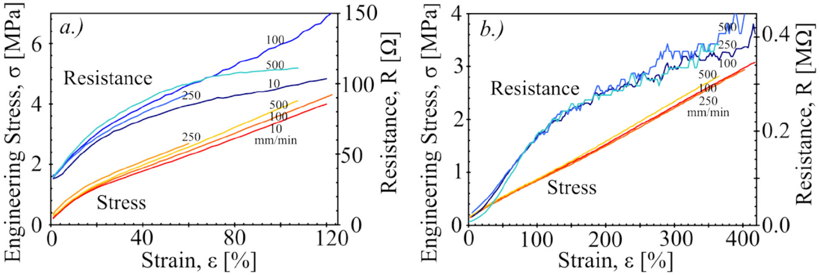

Fig. 3: Tensile tests of *a.)* R570/70 and *b.)* R-2631.

3.2.2. *Cyclic Tensile tests*

Cyclic tensile tests are also required in case of a sensor application. After several extractions, both the mechanical and electrical characteristics were significantly different from the initial state. The characteristic depends on the amplitude and the number of load cycles. On the Fig. 4 can be seen that the stress characteristic has the Mullin's effect. The single step characteristic (stretch to failure) is the enveloping curve of the cyclic loops. These effects have also arisen in case of the resistance characteristic. The single step resistance curve is the lower envelope of the cyclic loops, which also show hysteresis and a Mullin's like effect. The resistance curve of the cyclic test show a "8" shape, because it is increasing at the beginning of the unloading stage (after the maximum point of the stretch). This can be better observed on the relaxation curves.

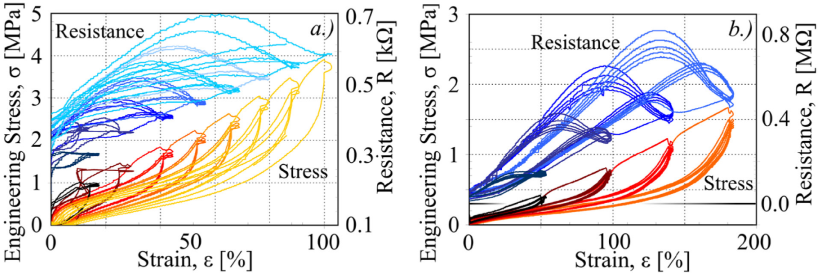

Fig. 4: Cyclic Tensile tests of *a.)* R570/70 and *b.)* R-2631.

The resistance curves show a shift in its maximum value. On the Fig. 5 can be seen that this shifting is decreasing by an exponential function of the load numbers. After enough number of load cycle the properties obtain a stationary condition. The minimum level of the resistant is almost constant.

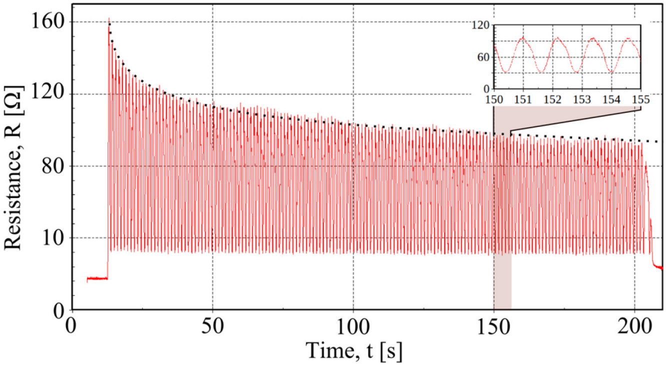

Fig. 5: Cyclic Tensile tests of a never stretched R570/70 sensor specimen.

3.2.3. *Cyclic Compression tests*

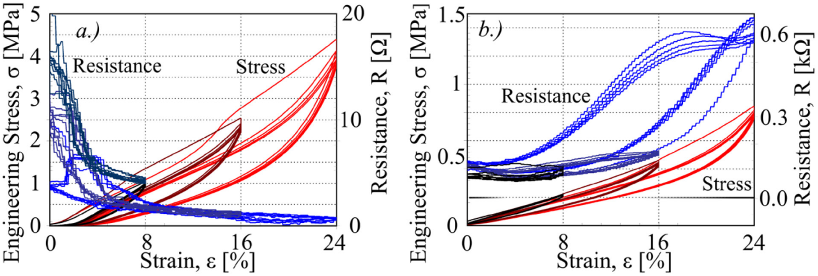

Fig. 6: Cyclic Compresion tests of *a.)* R-570/70 and *b.)* R-2631 conductive silicone rubber.

Electrically conductive silicone rubber can be used as tactile or compressive force sensor. To understand the relation between compression force, deformation and the resistance, compression test were executed. During the compression tests, the compression force (F[N]), the displacement of the yaws (ΔL [mm]), and the resistance (R[Ω]) were measured with constant pression velocity (12, 20 mm/min) at room temperature (22 °C).

The compression tests show similar effects to the tensile tests. The electrical characteristic also has “8” shaped hysteresis loops, remaining part, shifting and Mullin’s effect, etc. But in case of the compression tests the directions of the resistance characteristic of the R-570/70 and the R-2631 were opposite. The resistance of the R-570/70, as the expected, is decreased under the load, but in case of the R-2631 it was increased. (This opposite behaviour is also observable on the relaxation under compression.)

3.2.4. Relaxation tests

The time domain behavior of the sensor materials is very important because the rubbers show viscoelastic properties which appear as stress relaxation or creeping. Relaxation tests were performed to examine the time dependency of the electrical resistance.

During the test, the specimen was stretched and kept this elongation for 300 second then unloaded it. On the diagram (Fig. 7) we can see that the relaxation of the resistance is similar to the stress relaxation. Higher strain cause higher stress and resistance relaxation.

The “post-increasing” of the resistance is better observable on the relaxation diagram than on the tensile one. When unloaded the resistance increased sharply. It can be seen as a little “hill” on the end of the loaded section. Its height depends on the value of the deformation. Greater strain causes higher resistance “hill”.

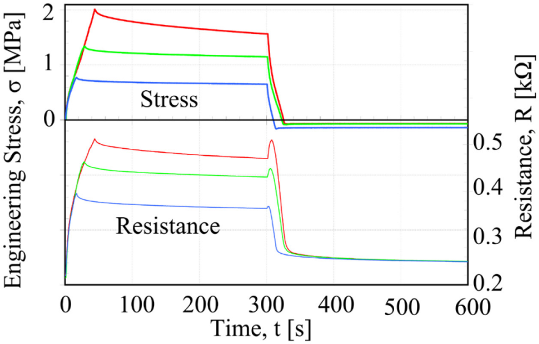

Fig. 7: Relaxation of the electrical resistance and the mechanical stress.

4. Impedance spectrum analysis

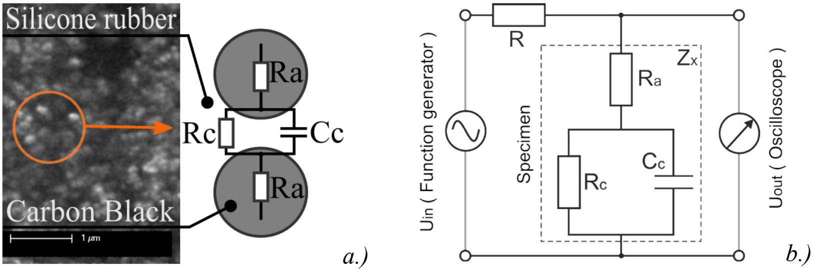

Fig. 8: *a.)* The model of the electrical conductivity of the silicone rubber filled with conductive nanoparticles. *b.)* The test setup of the impedance analysis.

To understand the conduction mechanism of the conductive silicone rubber, frequency analysis were made (with the box shaped specimens). The reason of this is that the capacity between the conductive nanoparticles causes the frequency dependency of the impedance. According to the literature [7, 8] the resulting impedance of the material consists of the resistance of conductive particles, the contact resistance, and the capacitance between the particles (Fig. 8/*a.*).

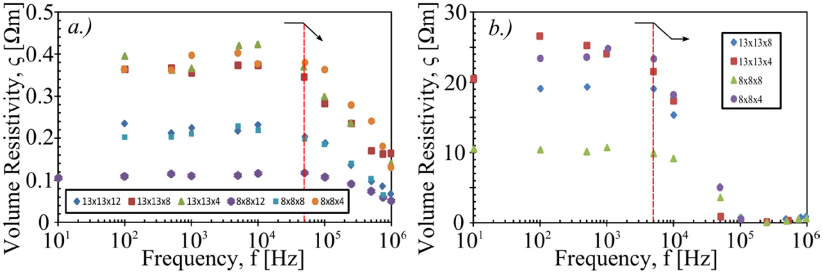

Fig. 9: The impedance of R-570/70 and R-2631 conductive silicone rubbers as a function of the frequency.

Between the conductive particles, in the thin isolating silicone rubber layer the thermally activated hoping or the tunnel effect causes the conductivity.

The conductive particles are present in the silicone substrate as continuous chains (R570/50, R570/70, R-2631) or as well-separated particles (R-2637);

The electrical model of the entire sample can be described by a lumped parameter model, (Zx on Fig. 8*/b*.) [7, 8]

The tests were made with the "black box" method. The model of the "black box" was determined from the relation of the input and output signal. The input signal was sine (generated by a Signal Generator). The output was measured with a Digital Oscilloscope. The frequency range of the measurement was 1Hz - 10 MHz.

The results are shown on Fig. 9. The average value of the cut-off frequency and the time constants are: $f_{R\text{-}570/70}=5*10^3$ Hz, $T_{R\text{-}570/70}=2*10^{-4}$ s; $f_{R\text{-}2631}=5*10^4$ Hz, $T_{R\text{-}2631}=2*10^{-5}$ s.

5. Summary

The electrical resistance of the silicone rubber shows relaxation and hysteresis.

When unloading the resistance increases sharply. The degree of this resistance „hill” depends on the amount of the deformation.

In case of cyclic load, the resistance decreases by an exponential function of the load number. The electrical properties of the silicone depend on the pre loads history. For this reason, mechanical conditioning is necessary for a sensor development.

The above properties show that the electrical conductive silicon rubber is not suitable for precise analogue measurement, but it is applicable for discrete sensing like a digital or FUZZY type sensor.

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