



## Short communication

## Effects of instantaneous compression pressure on electrical resistance of carbon black filled silicone rubber composite during compressive stress relaxation

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## ARTICLE INFO

## Article history:

Received 13 May 2008

Received in revised form 22 August 2008

Accepted 26 August 2008

Available online 30 August 2008

## Keywords:

A. Polymer–matrix composites

B. Electrical properties

C. Stress relaxation

## ABSTRACT

The compressive resistance relaxation of carbon black filled silicone rubber composite with different instantaneous compression pressure was studied. The experimental results show that the sudden increment of composite resistance increases with the increase of the instantaneous compression pressure. The experimental data for the compressive resistance relaxation and the compressive stress relaxation were fitted. The coefficients (exponentials) of the fitted functions increase (remain constant) with the increase of the instantaneous compression pressure.

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## 1. Introduction

Conductive polymer composite can be used as the sensing element of flexible force sensor [1–7]. In many engineering applications this kind of sensor is required to have the ability to measure the compressive stress relaxation. Therefore it is necessary to research on the change in the composite resistance when the sample strain is kept constant.

Voet et al. [8] and Sircar et al. [9] researched on the resistance relaxation of carbon black filled styrene butadiene rubber (SBR) at minute shear strains. They found that the electrical resistance of the composite decreases with time. They fitted the resistivity data of the composite by the power function:

$$R_t = R_1 \times t^{-m} \quad (1)$$

where  $R_t$  and  $R_1$  indicate the composite resistivity at the time  $t$  and 1, respectively.  $m$  is a constant defined as the relaxation rate of the electrical resistivity. Kost et al. [10] studied the electrical resistance relaxation of carbon black filled silicone rubber composite in tension tests. They drew a conclusion that the electrical resistivity of the composite changes with time in a fashion very similar to the changes in stress. However, the samples in the researches mentioned above are all in tension, which do not satisfy the requirement of the engineering applications.

After 2000, there are many researches on the time-dependent uniaxial piezoresistive behavior of conductive polymer composite [11–15], including resistance creep, resistance relaxation and the

changes in the resistance under the zero-pressure. Following the Burgers equation used for describing the creep behaviors of amorphous polymers, Zheng et al. [11] proposed the expression for the resistance relaxation of high-density polyethylene/short carbon fiber conductive composite:

$$\frac{R(t) - R_0}{R_0} = a + b \times \exp(-t/\tau_R) + c \times t \quad (2)$$

where  $R(t)$  and  $R_0$  are the composite resistance at the time  $t$  and 0, respectively.  $\tau_R$  is mean relaxation time.  $a$ ,  $b$  and  $c$  are constants depending on pressure. However, it is more appropriate to use the function mode for the stress relaxation to fit the resistance relaxation.

In our previous research [13] the electrical resistance of carbon black filled silicone rubber composite during the compressive stress relaxation was studied. The sample was compressed to a given pressure and the strain remains constant for 4500 s. After that the sample was compressed to another pressure. The pressure range is from 0 to 0.25 MPa. The experimental results show that the composite resistance decreases over time. The composite resistance was fitted by:

$$r_i(t) = A \times e^{B \times t} + C \times e^{D \times t}, \quad i = 1, 2, 3, 4 \quad (3)$$

where  $r_i(t)$  is the fitted function for the relative resistance over time after the  $i$ th compression,  $B$  and  $D$  are the exponential parameters,  $A$  and  $C$  are the corresponding coefficient parameters, respectively. However, the changes in the stress and the resistance after the  $i$ th compression include not only the response to the  $i$ th compression, but also the response to the previous excitations. Moreover the effects of the external excitations on the fitted function were not

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studied quantitatively. Furthermore the fitting effect of the Eq. (3) is not good enough. Therefore, in this paper the changes in the electrical resistance of carbon black filled silicone rubber composite at different applied load pressures were studied. The fitted function was ameliorated. The influences of the instantaneous compression pressure on the coefficients and the exponentials of the fitted functions were also researched.

## 2. Experimental

Carbon black powder (SL30, Carbon Black R&D Institute, China) was used as conductive phase. Room temperature vulcanized liquid silicone rubber (107, Beijing Chem. Plant, China) was used as insulating matrix. The properties of carbon black and silicone rubber are shown in Table 1. The mass ratio of carbon black to silicone rubber is 0.07:1. Hexane was used as solvent to mix the fillers with the rubber. Mechanical stirring along with ultrasonic vibration was also used for better particle dispersion. After 3 h of vigorous mixing, the solvent was evaporated. The viscous mixture was molded into thin plates (4 mm × 6 mm × 0.08 mm) at 30 °C for 60 h. Fig. 1 is the SEM micrograph of the fractured surface for the composite, showing a good dispersion of carbon black particles in silicone rubber matrix.

The sample with two electrodes is placed between the lift platform and the sensing element of the digital force gauge. The sample was compressed from the zero-pressure to a certain pressure by the upward move of the lift platform. Then the displacement was kept invariant for about 1800 s. The pressure values were recorded by the digital force gauge (HF-50) with the precision of 0.003 MPa. The electrical resistance was measured through the digital data acquiring system at room temperature. The experimental steps aforementioned were repeated for five times. The instantaneous compression pressures of the five measurements were 0.1, 0.2, 0.3, 0.4 and 0.5 MPa, respectively.

## 3. Results and discussion

The electrical resistance of the composite under the zero-pressure is 1.5 MΩ. As can be seen in Fig. 2, the compression induces sudden increase of resistance at the moment immediately after the compression. The sudden increments of the composite resistance and the strain both increase with the increase of the instantaneous compression pressure. The experimental phenomena aforementioned can be explained qualitatively as follows. The composite is a three-dimensional conductive network composed of carbon black particles and the rubber macromolecule [16]. The conductive network is destructed by the compression, leading to the sudden increase of the composite resistance. The destruction degree of the conductive network increases with the increase of the instantaneous pressure, resulting in the increase of the increment degree for the composite resistance.

As shown in Fig. 3, the change tendency of the compressive stress relaxation is similar to that of the compressive resistance relaxation. Therefore the fitted functions for the stress and the

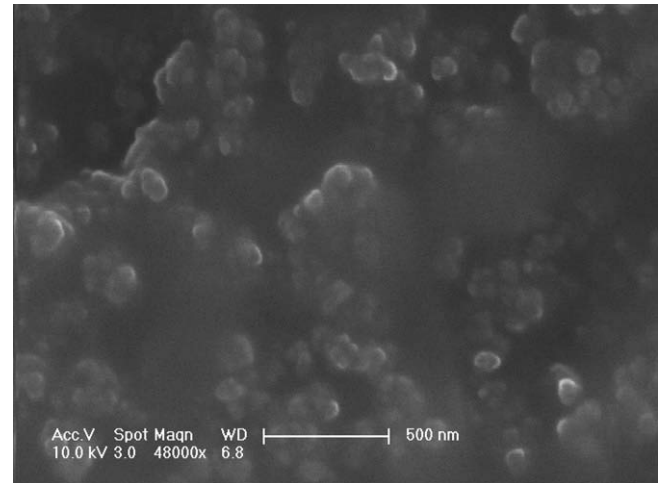


Fig. 1. SEM micrograph of the fractured surface for the composite.

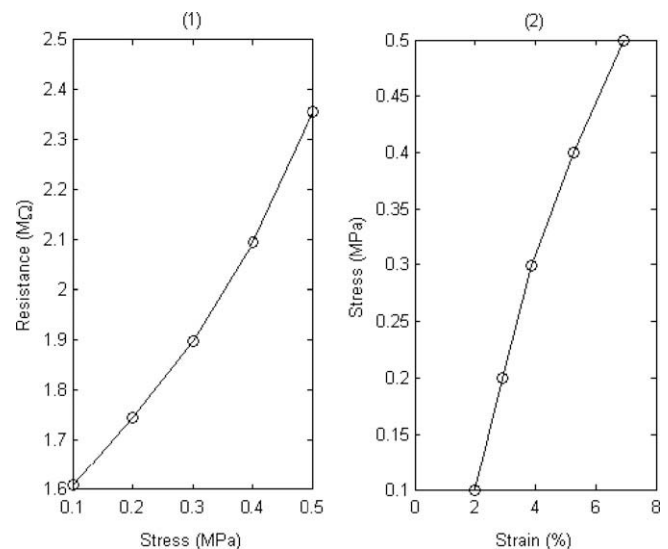


Fig. 2. Relation between the stress and the strain (resistance) at the moment immediately after the compression.

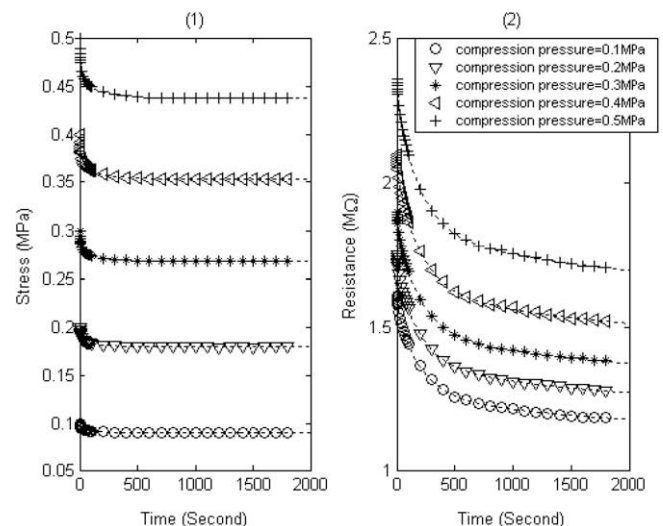


Fig. 3. Experimental data of stress and resistance during the compressive stress relaxation with different instantaneous compression pressure.

Table 1  
Properties of carbon black and silicone rubber

	Specific surface area (m <sup>2</sup> /g)	pH	Heating loss (%)	Light transmittance of toluene (%)
Carbon black	320	6.8	2.0	100
	Dielectric constant	Hardness (shore)	Tearing resistance (kg/cm)	Dielectric strength (kV/mm)
Silicone rubber	3.0	35	4	18

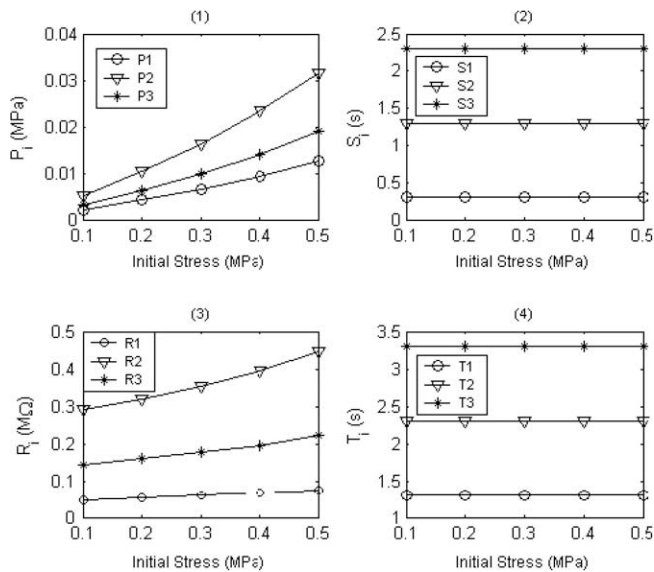


Fig. 4. Relations between the instantaneous compression pressure and the coefficients (exponentials) of the fitted functions for the compressive stress relaxation and the compressive resistance relaxation.

resistance during the compressive stress relaxation should also have the similar form.

According to the theory on the viscoelastic material, the stress relaxation can be simulated by:

$$\sigma(t) = \sigma(\infty) + P_1 \times e^{-t/\tau_{\sigma 1}} + P_2 \times e^{-t/\tau_{\sigma 2}} + P_3 e^{-t/\tau_{\sigma 3}} \quad (4)$$

where  $\sigma(t)$  and  $\sigma(\infty)$  represent the value of the stress at the moment  $t$  and  $\infty$ , respectively.  $\tau_{\sigma i}$  is the inner time scale of the viscoelastic material.  $P_i$  is the corresponding coefficient,  $i = 1, 2, 3$ .

According to the Eq. (4), the fitted function for the resistance data during the compressive stress relaxation can be given by:

$$r(t) = r(\infty) + R_1 \times e^{-t/\tau_{r1}} + R_2 \times e^{-t/\tau_{r2}} + R_3 e^{-t/\tau_{r3}} \quad (5)$$

where  $r(t)$  and  $r(\infty)$  represent the composite resistance at the moment  $t$  and  $\infty$ , respectively.  $\tau_{ri}$  and  $R_i$  correspond to  $\tau_{\sigma i}$  and  $P_i$  in the Eq. (4), respectively.

The fitted curves are shown in Fig. 3 (the dotted curves). The exponentials and the coefficients of the fitted functions with different compression pressure are shown in Fig. 4. For the conversion of the comparison, the logarithm of the exponential parameters were defined as  $S_i = \lg(\tau_{\sigma i})$  and  $T_i = \lg(\tau_{ri})$ ,  $i = 1, 2, 3$ . Based on the analysis on the results, the following conclusions can be drawn:

(1)  $P_i$  and  $R_i$  both increase with the increase of the instantaneous compression pressure.  $P_i$  represents the maximum attenuation value of the stress. Similarly,  $R_i$  represents the maximum attenuation value of the composite resistance.  $P_i$  and  $R_i$  are both directly related to the external excitation.

(2)  $\tau_{\sigma i}$  and  $\tau_{ri}$  almost remain constant with the increase of the instantaneous compression pressure.  $\tau_{\sigma i}$  and  $\tau_{ri}$  represent the inner time scale of the composite. They both have no relationship with the external excitation.

(3) Three exponentials of the fitted function for the compressive stress (resistance) relaxation are in three different orders of magnitudes. This experimental results indicate that the composite mainly have three different kinds of movement elements with

different stress attenuation speeds, which correspond to  $\tau_{\sigma 1}$ ,  $\tau_{\sigma 2}$  and  $\tau_{\sigma 3}$ , respectively. Similarly, there are three kinds of attenuation speeds for the composite resistance, which correspond to  $\tau_{r1}$ ,  $\tau_{r2}$  and  $\tau_{r3}$ , respectively.

The similarity between the fitted function form of the composite resistance and that of the stress is the macroscopic representation of the microscopic relation between the conductive network and the rubber macromolecule.

#### 4. Conclusion

The Sudden increment of composite resistance increases with the increase of the instantaneous compression pressure. At the stage of the compressive stress relaxation, the composite resistance and the stress both can be fitted by the linear combination of three negative exponential functions. The coefficient (exponential) of each term increases (almost remains constant) with the increase of the instantaneous compression pressure.

#### Acknowledgements

The authors wish to express their gratitude to “Specialized Research Fund for the Doctoral Program of Higher Education” and “Laboratory Fund of Tsinghua University”.

#### References

- [1] Knite M, Teteris V, Kiploka A, Kaupuzs J. Polyisoprene-carbon black nanocomposites as tensile strain and pressure sensor materials. *Sens Actuat A Phys* 2004;110:142–9.
- [2] Qu SY, Wong SC. Piezoresistive behavior of polymer reinforced by expanded graphite. *Compos Sci Technol* 2007;67:231–7.
- [3] Wang LH, Ding TH, Wang P. Effects of compression cycles and precompression pressure on the repeatability of piezoresistivity for carbon black-filled silicone rubber composite. *J Polym Sci B Polym Phys* 2008;46:1050–61.
- [4] Mahmoud WE, El-Lawindy AMY, El Eraki MH, Hassan HH. Butadiene acrylonitrile rubber loaded fast extrusion furnace black as a compressive strain and pressure sensors. *Sens Actuat A Phys* 2007;136:229–33.
- [5] Job AE, Oliveira FA, Alves N, Giacometti JA, Mattoso LHC. Conductive composites of natural rubber and carbon black for pressure sensors. *Synth Met* 2003;135:99–100.
- [6] Flandin L, Bréchet Y, Cavaillé JY. Electrically conductive polymer nanocomposites as deformation sensors. *Compos Sci Technol* 2001;61:895–901.
- [7] Wang LH, Ding TH, Wang P. Effects of conductive phase content on critical pressure of carbon black filled silicone rubber composite. *Sens Actuat A Phys* 2007;135:587–92.
- [8] Voet A, Cook FR, Sircar AK. Relaxation of stress and electrical resistivity in carbon-filled vulcanizates at minute shear strains. *Rubber Chem Technol* 1971;44:175–84.
- [9] Sircar AK, Voet A, Cook FR. Relaxation of stress and electrical resistivity in carbon-filled vulcanizates at moderate and high extensions. *Rubber Chem Technol* 1971;44:185–98.
- [10] Kost J, Foux A, Narkis M. Quantitative model relating electrical resistance, strain, and time for carbon black loaded silicone rubber. *Polym Eng Sci* 1994;34:1628–34.
- [11] Zheng Q, Zhou JF, Song YH. Time-dependent uniaxial piezoresistive behavior of high-density polyethylene/short carbon fiber conductive composites. *J Mater Res* 2004;19(9):2625–34.
- [12] Tao XL, Pan Y, Zheng Q, et al. Study on time-dependent resistance of carbon black loaded high-density polyethylene composites during the isothermal course. *J Appl Polym Sci* 2001;79:2258–63.
- [13] Ding TH, Wang LH, Wang P. Changes in electrical resistance of carbon black filled silicone rubber composite during compression. *J Polym Sci B Polym Phys* 2007;45:2700–6.
- [14] Yi XS. Function principle of filled conductive polymer composites. Beijing: National Defence Industry Press; 2004.
- [15] Zhang XW, Pan Y, Zheng Q, Yi XS. Time dependence of piezoresistance for the conductor-filled polymer composites. *J Polym Sci B Polym Phys* 2000;38(21):2739–49.
- [16] Zhu YJ. Mechanical modification of elastomers—filler reinforcement and blending. Beijing: Science and Technology Press; 1992.