Polymer Bulletin 12, 209-214 (1985)

Polymer Bulletin

Steady-State Creep Characteristics of Carbon Black Loaded Butadiene Rubber

S. S. Hamza and H. Osman

Physics Department, Faculty of Science, Cairo University, Cairo, Egypt

Summary

The temperature dependence of tensile creep of butadiene rubber (BR) loaded with different concentrations of carbon black ISAF was tested under different applied stresses. The steady creep rate showed a dependence on stress, temperature and carbon black content. The energy-activating steady creep was of order 0.15 eV for samples containing 20 phr of ISAF carbon black and reached a value of 0.37 eV for samples containing 80 phr of carbon black. This was attributed to the blocking of motion of the molecular segments by the carbon black. This view point was confirmed by the activation volume which was found to decrease with the increase of carbon black content.

The stress sensitivity parameter was found decrease from 3.4 for samples containing 20 phr to 1.8 for samples containing 80 phr of carbon black.

1. Introduction

The mixing of rubber with carbon black improves mechanical as well as electrical properties [1,2]. This is due to the formation of carbon aggregates which restrict the plastic flow in polymer. Analogous to the case of metal [3], the temperature and stress dependence of the creep rate, $\dot{\varepsilon}$, in polymers might be approximated by:

$$\dot{\varepsilon} = A \exp\left(- \frac{E - q\sigma}{kT}\right) \qquad (1)$$

where, E , is the activation energy of the flow process, q is the activation volume, k is the Boltzmann's constant, T is the absolute temperature and σ is the applied stress. Besides, according to Cannon and Sherby [4], the applied stress sensitivity parameter, m', was defined by:

$$m' = \left.\frac{\partial \ln \dot{\varepsilon}}{\partial \ln \sigma}\right)_T \tag{2}$$

In the present work we are concerned with throwing more light on the role of molecular movement and the mechanisms responsible for the steady creep of BR loaded with different amounts of carbon black. Pure BR samples free of carbon black are difficult to be tested, since, they have poor mechanical properties.

2. Experimental technique

Cis - 1,4 butadiene rubber loaded with different amounts of intermediate super abarasion furnace black ISAF were prepared [2]. The composition of our samples is formed 100 phr BR rubber mixed with 1.5 stearic acid, 10 processing oil, 20 ISAF, 1.0 PBN, 3.0 TMTD and 5.0 phr ZnO and different concentrations of carbon black 20, 40, 60, 80 phr. The test pieces were prepared in the form of strips with dimensions of 2 x 5 x 30 mm. The tensile creep measurement was carried out on a locally manufactured creep tester [5]. The tensile strain was measured manually with an optical microscope of sensitivity 10^{-3} cm. The temperature stability of the furnace was within $\pm$ 0.5 ^{o}C.

3. Results and discussion

Creep runs on BR samples containing 20, 40, 60 and 80 phr of ISAF carbon black were carried out at room temperature.

The steady state creep rate, $\dot{\varepsilon}$, was found to increase by increasing the applied stress and decreased with increasing the carbon black concentration in BR matrix as shown in Fig. 1.

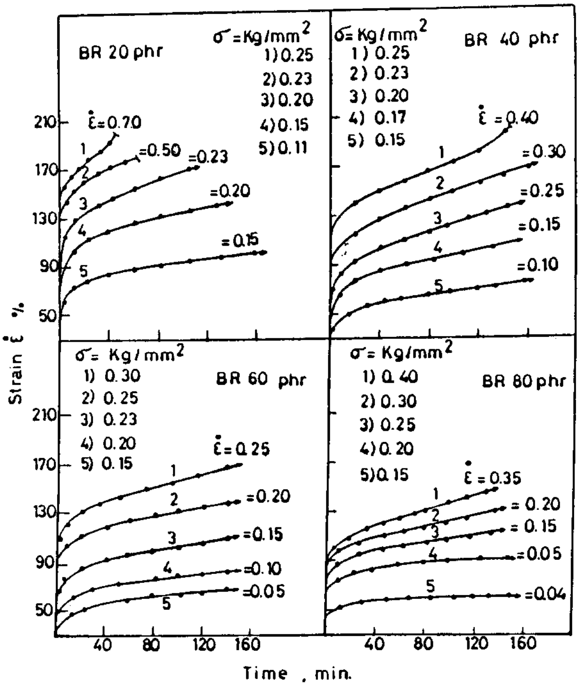


Fig. 1. Creep curves at 20 °C under different values of applied stresses for BR loaded with different concentrations of carbon black.

It is here thought that the high values of stress applied to the samples enhanced the flow mobility of the chain segments. On the other hand the values of steady creep rate, $\dot{\varepsilon}$, decreased as the carbon black content increased. It is well known that the carbon black particles of ISAF form aggregates between polymeric chains [1], which might lead to the diminishing of the mobility of chain segments.

Fig. 2 shows the effect of temperature on the creep curves of BR loaded with different concentration of carbon black.

In order to use equation (1) for the determination of the energy activating the creep process a plot was made relating $\ln \dot{\varepsilon}$ vs 1000/T (K) at constant stress (see Fig. 3b). From the linear relation thus obtained the activation energy, E , was found. Its value amounted 0.15 eV for BR containing 20 phr of carbon black and increased to a value 0.37 eV for samples containing 80 phr carbon black. It was thus concluded that carbon black represented abstacles for the motion of molecular segments,

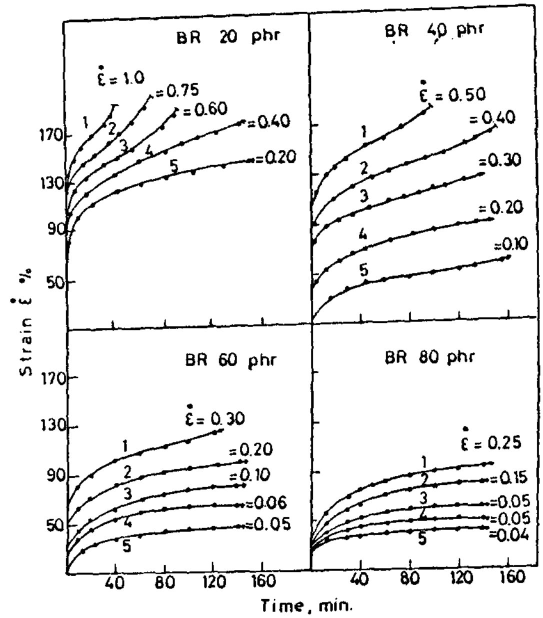


Fig. 2. Creep curves of carbon black loaded BR at σ = 0.15 Kg/mm^2 at different temperatures 1,90; 2, 80; 3,70; 4,50 and 5,20 ^{o}C.

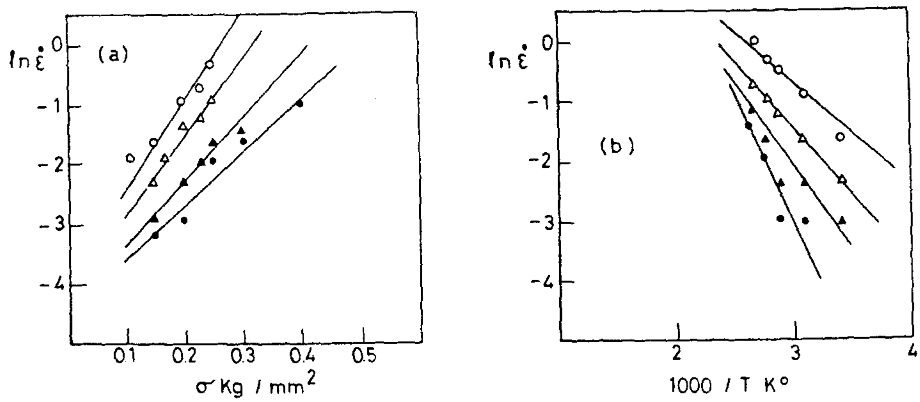


Fig. 3. a- Relation between $\ln \dot{\varepsilon}$ and σ at temperature 20 ^{o}C, and
b- The temperature dependence of $\ln \dot{\varepsilon}$ under stress 0.15 Kg/mm^2 for BR loaded with ○, 20 ; △, 40 ; ▲, 60 and ●, 80 phr carbon black.

thus requiring higher activation energies when the concentration of carbon black increased in BR matrix as shown in Fig. 4a.

Aided by equation (1) the activation volume, q , for the creep mechanism was found from the slopes of the straight lines relating $\ln \dot{\varepsilon}$ vs σ as in Fig. 3a. The activation volume was found to decrease with increasing the concentration of carbon black in the test samples as shown in Fig. 4b. This result might be attributed to the expected reduction in mobility of the chain segments by the carbon black particles.

Finally, the stress sensitivity parameter, m', as obtained from the slope of straight line relating $\ln \dot{\varepsilon}$ vs $\ln \sigma$ (see Fig. 5a), was found to decrease with increasing the carbon black content, due to the fact that the increasing carbon black particles in BR matrix hardened the butadiene rubber, thus causing the loss of its sensitivity forwards applied stresses.

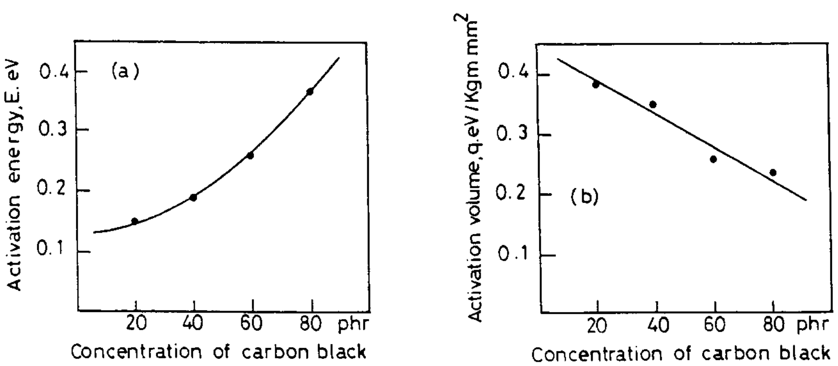
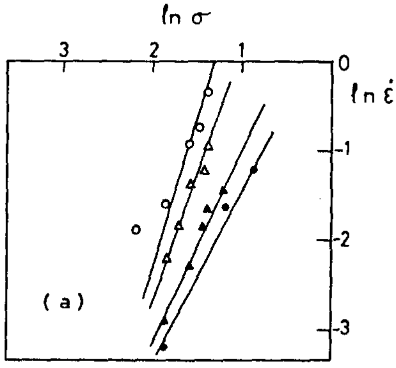


Fig. 4. The relation of the activation energy, E , and the activation volume, q , with carbon black concentration.



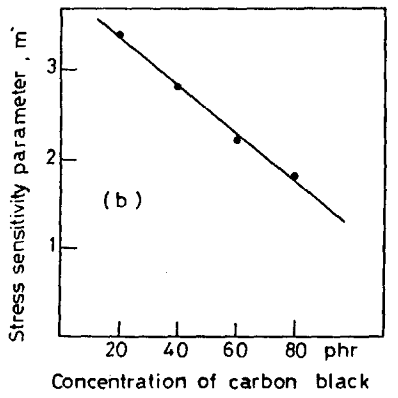


Fig. 5. a- The relation between $\ln \sigma$ and $\ln \dot{\varepsilon}$ for BR loaded with: ○, 20 ; △, 40 ; ▲, 60 and ●, 80 phr carbon black, and

b- Relation between stress sensitivity parameter m' and carbon black concentration.

Acknowledgement

The authors would like to thank Professor Dr. R. Kamel for his valuable discussions and comments.

References

[1] R.H. Norman, conductive Rubber and Plastics, London 1970.

[2] M. Amin, H. Osman and E.M. Abdelbary, Sonderdruk aus Kautschuk + Gummi, Kunststoffe 35, 1049-1052, Heft 12, 1982.

[3] D. Mclean, Trans. Met. Soc. AMIE, 242-1193, 1968.

[4] W.R. Cannon and O.D. Sherby, Metallurg. Trans. 1, 1031, 1970.

[5] R. Kamel and K. Halim, Phep-State-Sol., 15, 63, 1966.

Accepted July 3, 1984