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Stress relaxation, creep and set recovery of elastomers

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ABSTRACT

The stress relaxation, creep and recovery behaviour of a cross-linked unfilled natural rubber has been investigated at moderate stresses in tension. The aim being to extend the idea, initially developed by Alan Gent in his seminal 1962 paper on the relaxation behaviour of rubber, in order to understand and examine the time dependent mechanisms that are present in elastomers under strain. A method based upon the Boltzmann superposition principle was used to compare the creep compliance with a measurement of its recovery after release from a range of constant loads held for different times. The creep behaviour was seen to exhibit the usual linear dependence on the logarithm of time. The recovery data was also seen to reduce onto a single recovery curve for any given applied tensile stress for a range of loading times using the Boltzmann superposition principle. The differences between the relative rates of the creep and the recovery behaviour can in part be attributed to the non-linearity in the stress–strain behaviour exhibited in tension of the elastomer.

1. Introduction

Creep is when a material extends gradually with time under a constant load. A similar phenomenon known as stress relaxation takes place when a material is held at a constant extension and the stresses gradually decrease with time [2]. Both behaviours can lead to significant problems when using elastomer components in engineering applications. A further time dependent behaviour is observed once the applied loading is removed, as the unstrained state is not achieved immediately. This residual deformation is known as set which then typically decreases with time.

The relationship between stress relaxation, creep and the recovery of set after a period of constant loading are examined both theoretically and experimentally in this paper. It is to be expected that the behaviour of filled elastomers will be more complex than that of unfilled; for example the Mullins effect, involving stress softening in filled elastomers, is a well-known source of complications in the elastic behaviour of such materials. As a result of these anticipated complexities this investigation is only concerned with unfilled elastomers.

Alan [3] examined the relationships between various manifestations of viscoelastic behaviour, such as creep, stress relaxation and set recovery. He showed that the stress relaxation rate was substantially independent of the amount or the type of the deformation for moderate deformations and that this rate was simply related to the hysteresis. Gent also suggested that the rates of creep and recovery are in good agreement with values calculated from the form of the load–deformation relationship and the constant value of the relaxation rate. His treatment of the kinetics of the recovery of the set was, however, approximate and limited in scope, and a more comprehensive approach is proposed below.

The relation between creep and recovery can be derived for a linear viscoelastic material using the Boltzmann superposition principle [1] which can be considered to be a fundamental theorem of linear viscoelasticity. The approach adopted below is to consider the creep and recovery behaviour using the Boltzmann principle modified to account for the non-linear elastic behaviour of the rubber.

2. Theory

The Boltzmann superposition principle, valid for linear viscoelastic materials, gives the strain $e(t)$ at time t after loading with a stress σ as

$$e(t) = \int_{-\infty}^{t} J(t-t_1)\dot{\sigma}(t_1)dt_1 \quad (1)$$

where $\dot{\sigma}$ is $d\sigma/dt$ and $J(t)$ is the creep compliance. In a creep experiment where a constant stress σ is applied to the material for the time period from $t=0$ to $t=t_1$ and is then removed, the subsequent strain (or set) can be considered to arise from the

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superimposed effect of $+\sigma$ applied at $t=0$ and a superimposed stress of $-\sigma$ applied at time $t=t_1$. The rate of change of deformation changes sign at time t_1 and the body returns slowly to its original state. The remaining set after unloading at time t can be calculated from Eq. (1) as

$$e(t)=\sigma[J(t)-J(t-t_1)] \tag{2}$$

This can be rewritten in terms of the time after unloading, x giving

$$e(t)=\sigma[J(t_1+x)-J(x)] \tag{3}$$

The creep compliance of elastomers is often found to have an approximately linear dependence upon the logarithm of time, whereby

$$J(t)=J_0\left(1+A\log\frac{t}{t_0}\right) \tag{4}$$

where J_0 is the creep compliance at a reference time t_0 and A is a parameter indicative of the rate of increase of $J(t)$.

An equation analogous to Eq. (4) may be written for the stress relaxation in terms of the logarithm of the time, viz.

$$\sigma(t)=\sigma_0(1+A\log(t/t_0)) \tag{5}$$

where σ_0 is the stress relaxation at a reference time t_0 and A is a parameter indicative of the rate of decrease of $\sigma(t)$.

Using Eqs. (3) and (4), the set, $\gamma(t)$ at time x after unloading is given by

$$\gamma(t)=\sigma AJ_0\left(\log\frac{t_1+x}{t_0}-\log\frac{x}{t_0}\right) \tag{6}$$

$$\gamma(t)=\sigma AJ_0\log\left(\frac{t_1}{x}+1\right) \tag{7}$$

Thus, provided the linear dependence of the creep compliance with the logarithm of time, shown in Eq. (7) applies, a plot of $\gamma(t)/\sigma J_0$ against $\log[(t_1/x)+1]$ should produce a straight line through the origin of slope A.

Eqs. (4), (5) and (7) are the initial basis for the analysis of the experimental creep, stress relaxation and set recovery results, respectively.

3. Experimental procedure

A natural rubber (SMR CV60) compound was used with the formulation shown in Table 1. The mixing was carried out using a two-roll mill. The stress relaxation, creep and set recovery test pieces were made from 1 mm thick sheets cured at 160 °C for approximately 15 min. A rectangular specimen of approximately 10 mm width, 1 mm thickness and 80 mm length was cut from the rubber sheets.

The stress relaxation behaviour was measured by deforming the specimen to different constant extensions, ranging from 24% to 200% extension. During the creep testing, the specimen was held by grips to which weights were attached. The length between two marks made along the length of the test piece was measured, after applying the load, using a travelling microscope. The length between the two marks was also measured after unloading the specimen during the long-term recovery. A load was applied to the lower clamp for a range of different periods of time, t_1, before it was removed. For each value of t_1 the creep and recovery were measured at various times during loading and unloading respectively.

Table 1
Formulation of the rubber material.

Ingredients	Parts per 100 rubber
NR (SMR CV60)	100
Stearic acid	2
Zinc oxide	5
Anitoxidant (HPPD)	3
Accelerator (CBS)	1.5
Sulphur	0.5

4. Results and discussion

The stress–strain behaviour for the unfilled material used in this study (NR1) was measured carefully and the Mooney constants for the material were determined from a Mooney plot to be $C_1=0.0735$ MPa and $C_2=0.0719$ MPa.

Fig. 1 shows the stress decay data for the different constant extensions of the unfilled compound (NR1). The stress–time data is non-linear as is expected for elastomers.

In order to determine the rate of stress relaxation, the non-linear stress–time curves were linearised through a semi-logarithmic plot following Eq. (5) as can be seen in Fig. 2 for 103% extension.

The experimental data was seen to reduce well to a straight line for the different extensions apart from a few initial data points which showed some discrepancy. This discrepancy can largely be attributed to the inherent difficulty in synchronising the stress–time data at the start of the measurement. Data points were measured at time scales of the order of seconds during the start of the experiment hence a time lapse of the order of fractions of a second, between the stress and the time measurements could have significant effect on the data points at the shortest time scales. Data points in later stages of the measurements fitted better because they were measured at time scales of the order of minutes rather than seconds; as a result, the effect of time lapse in the measurements becomes much less significant. It must be

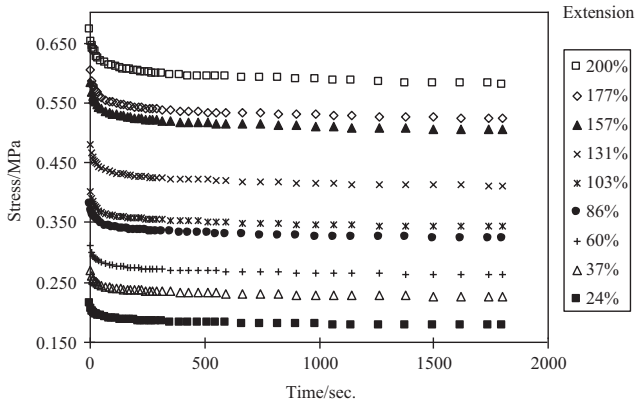

Fig. 1. Tensile stress relaxation of the rubber at different extensions.

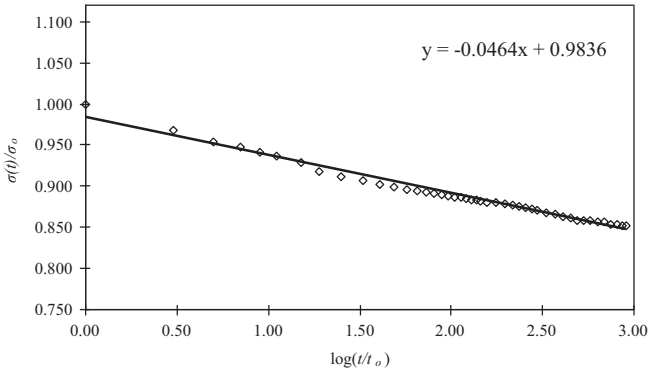

Fig. 2. A semi logarithmic plot for the stress decay data at 103% extension.