Stress softening of carbon black filled SBRs submitted to various large strain uniaxial tension cycles

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Abstract

Two carbon black filled SBRs were submitted to various uniaxial tension cyclic tests, including stretch-control tests and stress-control tests. Stress relaxation and stretch creep were measured according to the number of cycles. Results show a strong dependence of the material softening to the maximum loading applied and the number of cycles. After a few cycles (around 40), the softening evolves linearly according to the logarithm of the number of cycles. The cyclic softening is also shown to be locked by a low number (5) of overload cycles. More interestingly, the material softening appears to be unaffected by the loading cycle amplitude. This study aims at defining the parameters to include in a subsequent cyclic stress-strain model.

Keywords: Filled rubber; cyclic softening; cycle amplitude; ratcheting

1. Introduction

Applications of filled rubbers include cyclic loading conditions. These conditions induce a material softening followed by the appearance of a crack and a possible catastrophic failure of the material. Many contributions have focused on proposing a lifetime criterion (see [1] for a review) but fewer addressed the problem of the stress-softening [2-5]. In this study, we have submitted two carbon black filled SBRs to various cyclic loading conditions in order to decide on the main parameters controlling the stress-softening of such materials.

Filled rubbers undergo a large stress-softening during the first load. This softening is well known as the Mullins effect [6, 7]. During the following cycles, the softening evolves slowly and therefore is often neglected. Nonetheless, [8] showed that the cumulative stress-softening may be significant after a moderate number of cycles. Moreover, [3] produced experimental evidences of the dependence of failure lifetime to the material stress-softening history.

Previous studies [2,5] on rubber softening, proved a strong dependence of the stress-softening to the maximum stretch applied. Also, [2,4] observed a stress amplitude dependence of the stress softening. This effect is more questionable, since at least one of these studies was conducted on crystallizing rubber and it is well known that...
crystallization delay fatigue failure. Also, test strain rates in both studies [2,4] were high enough to introduce a viscous effect. In consequence, SBR gum, which does not crystallize was chosen and tests were conducted at a strain rate of 10^−3 s^−1, which is low according to fatigue standard and which will limit viscous effects. We applied various loading conditions in stretch-control mode and in stress-control mode in order to understand the consequences of the loading history on the cyclic softening.

2. Material and experiments

2.1. Material

Two carbon-black filled SBRs were supplied by MICHELIN. Compositions of the materials appear in Table 1. Dumbbell samples of dimension, 25mm long and 4mm wide, were cut from plates of 2mm thickness.

<table>
<thead>
<tr>
<th>Ingredients</th>
<th>Material A (Phr)</th>
<th>Material B (Phr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SBR gum</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>N347</td>
<td>30</td>
<td>60</td>
</tr>
<tr>
<td>6PPD</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Stearic Acid</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>ZnO</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>CBS</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>Sulfur</td>
<td>1.2</td>
<td>1.2</td>
</tr>
</tbody>
</table>

2.2. Experiments

Cyclic tests were conducted at room temperature on an Instron 5802 uniaxial tension machine operated in displacement control mode at a constant crosshead speed of 500 mm/min. Local strains were measured by video extensometry.

Several loading conditions, which are summarized in Fig. 1, were applied. Test (a) studies the softening vs. stretch level and number of cycles. Test (b) studies the effect of stretch amplitude on the material softening. Test (c) is designed to show a possible overload effect. Test (d) studies the effect of a preloading at lower stretch. Tests (e) and (f) are equivalent to tests (a) and (b) respectively, but are conducted in stress control mode. In the following, we will refer to these tests by the letter (a), (b), (c), (d), (e) and (f). Specimens were loaded from to 500 to 2000 cycles and maximum stretch ranged between 100% and 300%.

Fig. 1. Loading conditions
3. Strain controlled tests

Material 1 and 2 were submitted to various stretch control mode cyclic uniaxial tensile tests. In this section we will present the results obtained on material 2 only. Similar results were obtained on material 1

3.1. Maximum strain dependence

First, the material was submitted to a test (a) with several values of the maximum stretch. During the first cycle, the material undergoes a large softening, known as the Mullins effect. During the following cycles, a slow stress-softening is observed and we characterized it by the following quantity:

\[ R(\lambda_{\text{max}}) = \frac{\sigma_{\text{max}}^N}{\sigma_{\text{max}}^{N-1}} \]  

where \( \tau \) is the engineering stress, \( \lambda_{\text{max}} \) the maximum stretch and \( N \) is the number of cycles. Fig. 2 (a) shows that the stress-softening increases with the maximum of stretch applied. Fig. 2 (b) shows that after a fairly low number of cycles (~40), the stress softening decreases linearly with \( ln(N) \). [3, 8] obtained a similar dependence of the stretch relaxation according to \( ln(N) \). Fig. 2 (b) shows also that the stress softening rate is increasing with the value of the stretch peak.

![Fig. 2. Material 2: (a) Dependence of stress softening vs. maximum stretch (b) Logarithmic dependence of stress softening vs. number of cycles.](image)

![Fig. 3. Material 2: (a) Effect of an overloading: Comparison of stress softening vs. cycle number at maximum stretch for a material submitted to an overloading and the same material without overloading history (b) Effect of preloading at low stretch: Comparison of the stress softening rate calculated for cycles reaching the maximum stretch of 3.5 only, during tests (a) and (d).](image)
In order to study the effect of a preloading, we compared two samples submitted to 100% peak stretch cyclic tensile test. The first one was submitted to a preloading of 5 cycles at 350% (test (c)) while the second one was not previously overloads (test (a)). The stress relaxations for both samples are presented in Fig. 3 (a). The overloading induced a high stress softening of the material (stress corresponding to the peak stretch is low). Moreover, the overloading seems to freeze the subsequent damage. Therefore, the material damage evolving slowly during the cyclic loading at a given stretch and evidenced by the continuous stress relaxation, may happen in a very few cycles (here 5) at a higher stretch. Let us note that we observed the same damage freezing for an overload stretch less than 10% higher than the peak stretch of the cycles (overload stretch=3.5, cyclic peak stretch=3.25).

Fig. 3 (b) illustrates the effect of low stretch cycles inserted during cycles at higher stretch. The first sample was submitted to a test (a) of 1000 cycles at a peak stretch of \( \lambda_{\text{max}} = 3.5 \), while the second sample was submitted to the same number of high stretch cycles but added of 1000 cycles of peak stretch \( \lambda_{\text{max}} = 2.0 \) inserted after the fifth high stretch cycle (test (d)). In Fig. 3(b), we reported for both samples the stress softening rate, \( R(\lambda_{\text{max}}) \) eq. (1), at \( \lambda_{\text{max}}=3.5 \) vs. the number of cycles reaching this stretch level only. Both curves superpose, evidencing that a preload at a lower stretch does not affect the stress softening at higher stretch.

In this section we showed that the stress relaxation is strongly dependent to the level of stretch during the cyclic loading. We also observed that an overload induce a damage that stabilizes the cyclic softening at lower stretch. Finally, a preloading at a lower stretch does not affect the cyclic softening. In the next section, we will study the effect of loading amplitude, by applying cyclic tensile tests with identical maximum stretch and various cycle amplitudes.

3.2. Strain amplitude dependence

Samples were submitted to test (a) or test (b) at both maximum stretch \( \lambda_{\text{max}} = 3 \) and \( \lambda_{\text{max}} = 3.5 \). Fig. 4 shows the softening rate, \( R(\lambda_{\text{max}}) \) eq. (1), plotted vs. the number of cycles. Curves from tests (a) and (b) coincide when maximum stretch is identical for both tests. Therefore, the cycle amplitude had no effect on the material stress softening. This results differs from results previously reported in the literature [2,4]. Nonetheless, in the case of NR [4], crystallization may be the source of the observed cycle amplitude effect. When the cycle amplitude is low, crystallization remains reducing the effect of damage. As a consequence of the present results, stress-softening of filled SBR cannot be characterized by an energy parameter, even though it seems as a relevant parameter for fatigue failure [2,11].

![Fig. 4. Comparison of the stress-softening of material 2 for uniaxial tension cyclic loadings of various stretch amplitudes and identical maximum stretch.](image)

In the next section, we present equivalent results obtained in stress control mode.
4. Ratcheting tests

4.1. Maximum stress dependence

While submitted to a cyclic uniaxial tension tests at a constant peak force, filled rubber show a ratchet effect, as illustrated by Fig. 5 (a). This effect is evidence by an increasing peak stretch and an increasing permanent stretch (stretch at zero force) with increasing number of cycles. This ratchet effect is dual from the stress softening effect described in section 3. Samples were submitted to a ratcheting at various maximum peak forces. Fig. 5 (b) shows a linear dependence vs. ln(N) of the stretch at peak stress after a few cycle (~20). Fig. 5 (b) shows also that the stretch at peak stress evolves faster with the increase of the peak stress.

Other tests, not presented here, showed an effect of loading history on ratcheting equivalent to the one observed in section 3. A pre-overload, stabilizes the ratcheting and pre-underload do not affect it.

4.2. Stress amplitude dependence

The cycle amplitude effect on ratcheting was also studied. Samples were submitted to test (e) or test (f) with an identical peak stress of 2.6 MPa. The stretches at peak stress were monitored vs. the number of cycle for 3 cycle amplitudes. Fig. 6 shows the evolution of the quantity. Fig. 6 shows a stretch at peak stress evolving independently of the cycle amplitude. This results corroborates the result obtained in stretch control mode.
5. Conclusions

This work explored the stress-softening of filled SBR under cyclic uniaxial loading conditions. Both stretch control mode and ratcheting tests were performed in order to monitor the stress relaxation and the peak stretch evolution according to the number of cycle and the loading conditions. We observed a strong dependence of the material softening according to the maximum stretch or equivalently the maximum stress applied. It was shown to depend linearly on the logarithm of the number of cycles, with a slope increasing with the increasing level of loading. Preloading was proved to affect the material softening only when exceeding the cyclic peak.

The cycle amplitude was shown to have no effect on the material softening. Therefore any parameter characterizing the cycle amplitude, as the cyclic energy, appears to be an irrelevant parameter to feature the material cyclic softening.

References