Crack Growth and Strain Induced Anisotropy in Carbon Black Filled Natural Rubber†

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The work discussed outlines the development of a technique to predict failure of rubber components under repeated complex stressing. A fracture mechanics approach is adopted, which has been shown to work with simple plane stress type geometry in the past. Real components with three dimensional loading and geometry can be examined with the use of finite element analysis techniques. These now allow us to investigate real fracture type problems to large strain deformations.

To predict fatigue behaviour in components it is necessary to derive the energy release rate for that particular component, which in general involves FEA techniques. Also it is important to know the crack growth characteristics of the material. This is usually expressed in terms of the rate of crack growth as a function of the tearing energy (energy release rate). Conventional tearing energy, crack growth rate tests are performed in relatively simple deformation modes, using for example edge crack or pure shear test pieces. In practice, components used in engineering undergo more complicated strain histories. In order to study these potential complexities in the crack growth characteristics, the effect of pre-strain on cyclic crack growth behaviour using a predominantly pure shear test piece with an applied pre-strain has been studied. For the filled compounds studied so far there do appear to be complications and the observed crack growth rates may be substantially increased by these pre-strains. Some of these results are presented and their consequences discussed.

Catastrophic tearing studies¹² show that highly pre-stretched rubber sheets, are significantly weaker in the direction of pre-straining than in the direction running at a right angle to it. This strain induced anisotropy is shown most clearly in materials such as natural rubber that crystallise on straining. It is also much more obvious for typical engineering compounds filled with reinforcing carbon blacks.

Indeed carbon-black filled natural rubber compounds show a pre-strain induced anisotropy in strength even after the removal of the prior extension¹.

These differences in strength in different directions, induced either by a previous deformation or by the present one are of considerable interest. They may be responsible for the phenomenon known as knotty tearing, when the advancing crack profile spontaneously deviates from the anticipated path which is at right angles to the principle tensile stress field and turns instead in the direction of the stressed...
direction. This causes an effective blunting of the crack tip and arrests the crack growth until the applied stress exceeds a new threshold value. This 'stick slip' behaviour is a characteristic of filled materials and appears to be one of the principle mechanisms of carbon black reinforcement.

This change of strength properties caused by a pre-straining has many practical applications, for example in predicting the failure behaviour of tyres or other inflated devices. This is even the case with more general engineering components such as those encountered in suspension systems, when a complex loading is encountered. Measures of strength taken from uni-axial deformations are likely therefore to be misleading.

In order to formulate a more scientific approach, the derivation of a fundamental measure of strength is necessary. The approach adopted most commonly is due originally to Griffith. This leads to the assumption that the elastic energy available to drive a crack, $T$, determines the rate of crack growth. Where the tearing energy, $T$, may be defined as:

$$ T = - \left( \frac{\partial U}{\partial A} \right) $$

$U$ is the total strain energy stored in a specimen containing a crack and $A$ the area of one of the cracks fracture surfaces. The differentiation is done over a constant deformation so that the external forces do not work.

Rapid crack growth tearing in specimens loaded or strained at a continuous rate usually utilise the trouser test. Many rubber components experience repeated cyclic loading in service sometimes causing cyclic crack growth to occur at much slower rates than those normally associated with the catastrophic single cycle tear test.

![Figure 1 Pure shear test piece](image)

Cyclic crack growth tests can be investigated using a variety of different sample test geometries such as the trouser test piece, the pure shear test piece shown schematically in Figure 1, and the edge cut tensile strip. Of these, the edge cut tensile strip tests result in continuously increasing crack growth rate per cycle while the pure shear geometry test results in constant crack growth per cycle, for a specified applied stress or a given tearing energy. The pure shear test piece will be used throughout our study, because of this effect.

The applicability of this approach has been verified by a number of researchers using various test pieces cut from thin rubber sheets. This approach shows that a crack growth rate, $dc/dN$, versus tearing energy, $T$, relationship exists that is normally shown on a double logarithmic plot. Here, $c$ is the crack length and, $N$, the number of cycles completed. This relationship is a basic material property that is independent of the test piece geometry for uni-axial load cases. Failure under repeated stressing appears to be initiated from small...
naturally occurring defects or flaws in the object. The exact nature of these flaws is often unclear, however filler agglomerates or dirt particles are probably two of the major contributors.

In order to use the fracture mechanics approach the size, shape and orientation of the flaw relative to the applied stress field must be taken into account. Most rubber components have to be considered as three dimensional, and in order to calculate energy release rates large strain finite element techniques are required. Finite element techniques\textsuperscript{7-9} have been used before to derive solutions to fracture problems at large strains in elastomers. However the application of this methodology to predict crack growth behaviour in real components has been slow to be realised.

Steady state crack growth behaviour measured in cyclical deformations on filled elastomers, for example as shown by De\textsuperscript{10}, also show rough fracture surfaces. The roughness of these fracture surfaces are much smaller in scale than that observed from knotty tearing using the trouser tear test piece, and obviously the associated tearing energies are much smaller.

However the roughness may still be associated with small scale anisotropy, developed in the direction of the strain, normal to the direction of the crack growth. If this is the case then an applied pre-strain in the direction of the crack growth should reduce the effect and may, if the pre-strain is sufficiently large, produce an anisotropy in the crack growth direction, resulting in reduced tearing energies.

In order to investigate and understand this strain induced anisotropy phenomenon, a series of tests have been conducted on three natural rubbers of varying carbon black content.

In the pure shear test, the width of the specimen is much greater than the height (finite element studies have shown that this should be at least 8 times). When the clamps are pulled apart, the test piece undergoes a pure shear deformation because the clamps effectively prevent the rubber strip from decreasing in width.

The first experiment is designed to investigate the deviation of the stress versus strain behaviour of a normal isotropic material caused by different levels of perpendicular pre-straining. Secondly, measurements are made of the cyclic crack growth behaviour at various levels of pre-strain and tearing energy and finally, an investigation is made into the effect of an applied and then removed pre-strain on the cyclic crack growth behaviour.

It has been shown by Davies \textit{et al.}\textsuperscript{11} that the stress strain behaviour of a carbon black filled natural rubber material can be expressed in terms of a single stored energy function over a wide range of strains and deformations, even when that stored energy function is a term of the first strain invariant, \( I_1 \) only. Where:

\[
I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2 \quad \ldots 2
\]

It should be possible therefore to measure and fit the stress versus strain data, measured by a tensile test, and derive the functional relationship to the proposed stored energy function. This stored energy function could then be used to predict the behaviour of a more complex pre-strained pure shear test piece and comparison could be made with experimental results.
The Davies et al. function is designed to predict the behaviour of engineering materials from small strains to reasonable strains of about 200%. As the current experiments do not measure the behaviour accurately at the small strains, it is possible to adopt a simpler stored energy function, involving only a power series in $I_1$. For example the function proposed by Yeoh:

$$W = A_1(J_1 - 3) + A_2(J_1 - 3)^2 + A_3(J_1 - 3)^3$$

...3

Rivlin has showed that the general relationship for the engineering stress $\sigma_2$ measured in the perpendicular direction as the grips are pulled apart, for the bi-axial deformation mode is given by:

$$\frac{\sigma_2}{\lambda_2^2} - \left(\frac{1}{\lambda_1 \lambda_2}\right)^2 = 2 \left[ \frac{\partial W}{\partial I_1} \right]$$

...4

where,

$$I_1 = \left(\lambda_1^2 + \lambda_2^2 + \frac{1}{(\lambda_1 \lambda_2)^2}\right)$$

...5

In region B, around the cut tip, the stress field is complex and the strain energy $U_B$ is difficult to determine. Region D will be in pure shear and so the strain energy function in this region will be given by $U_D = W V_D$ where $W$ is the strain energy per unit volume of the material in pure shear at a specified strain and $V_D$ is the volume of region $D$. The region E is in a more complex state of stress because of the edge effects.

![Diagram](image)

Figure 2. A schematic of the various stresses regions in a pre-strained test pure shear test piece, with a cut of length, $c$.

The undeformed dimensions are defined.

When the crack length increases by $dc$, a simple transfer of material from region D to region A takes place and the total reduction in the stored energy is:

$$dU = W L_0 \, dc$$

...6

where $W$ is the strain energy density in pure shear and $L_0$ is the initial height of the specimen.

This leads to a simple expression for the tearing energy of:

$$T = W L_0$$

...7
However due to a cyclical stress softening effects, it is not easy to use this relationship to calculate the reduction in the tearing energy caused by the advancing crack. Therefore an alternative expression of the tearing energy is used in our calculations. By defining $U$ the elastic stored energy which is equal to the area under the force-deflection curve and $V$ the volume of the specimen contributing to the force-deflection curve, then $W = U/V$. By imposing an incompressible material constraint, the expression of $V$ as referred to the unstrained dimensions is:

$$V = L_0 t(w-c)$$

where $L_0$ is the unstrained height, $t$ is the thickness, $w$ the width and $c$ the crack length. This expression assumes that only the volume of the uncut specimen contributes to the force-deflection curve and the cut part of the specimen is totally zero energy; which is untrue. Finite element studies indicate that an equivalent strip of width, $x$, of the cut specimen is not free of energy, where $x$ had been determined by the analysis to be equal to 14% of $L_0$. Therefore a suitable expression for $T$ is:

$$T = \frac{U}{t(w-c+x)}$$

For the pre-strained case where the test piece is pulled to a strain of $\lambda_1$ in the orthogonal direction prior to being clamped in the test grips, this tearing energy relationship has to be modified. For definition purposes all the dimensions referred to in the pre-strained state are indicated by a prime sign, therefore; $t'$ for the thickness, $c'$ the crack length, $L_0'$ the height and $w'$ the width.

When the crack propagates a length of $\Delta c'$, only regions A and D have a change in their elastic energy. If $W_A$ and $W_D$ are the strain energy densities, respectively in regions A and D, then the tearing energy for the system can be shown to be given by:

$$T = L_0 (W_D - W_A)$$

As is known for the conventional pure shear test, $L_0$ is difficult to determine because of the stress-softening effects. Therefore another expression for deriving the tearing energy relationship is required.

Ignoring edge effects, $W_D - W_A$, which is now defined as the area under the force-deflection curve, $U$, divided by the volume of the specimen contributing to the force-deflection curve. This is the volume of the uncut specimen corrected by a term $x'$. This indicates that a vertical strip of width $x'$ of the cut specimen contributes to the force-deflection curve:

$$V = L_0 t'(w'-c'+x')$$

From a finite element analysis of a specimen pre-strained 100% the value for $x$ is calculated as being 19% of the un-strained height $L_0$. Therefore we can assume that:

$$x' = \lambda_1 x = \lambda_1 0.19L_0$$

This assumption that the original strip of width $x$ will increase by a factor of $\lambda_1$ if a pre-strain $\lambda_1$ is applied has also been verified analytically by using finite element analysis. We finally obtain:
TABLE 1. COMPOUNDS USED IN THE INVESTIGATION

<table>
<thead>
<tr>
<th>Material</th>
<th>HAF N330 Carbon Black content (parts per hundred rubber)</th>
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</thead>
<tbody>
<tr>
<td>A</td>
<td>21</td>
</tr>
<tr>
<td>B</td>
<td>32</td>
</tr>
<tr>
<td>C</td>
<td>50</td>
</tr>
</tbody>
</table>

All the materials use typical semi-EV curing systems. The square flat sheets (175 mm x 175 mm) were all cured for 10 min at 155°C strained to a maximum strain amplitude of between 5%–35% and relaxed to zero strain in each cycle.

A horizontal cut of 40 mm in length was introduced on the central axis of one edge of the test piece using a razor blade. This placed the crack/cut tip sufficiently far from the edge to avoid any edge effects.

The crack length was measured using a magnifying, time lapse video camera and recorder, and load-displacement curves were simultaneously recorded to measure the tearing energy.

Due to the initial sharp crack tip the crack grows rapidly; however, as the crack grows the normal blunting of the crack tip occurs and the behaviour becomes more typical of steady state behaviour. It is after this initial time that crack growth rates were measured.

The pre-strained test pieces were easy to prepare, a rubber strip that was 175 mm long was pulled to the required single pre-strain for five minutes. It was then relaxed for 2 h to allow the dimensions to stabilise. The sheet is then tested under the conventional cyclical conditions.

In tearing tests it has been observed that a prior pre-strain in the direction of the crack propagation causes a weakening of the material when an orthogonal tear test is performed. This suggests that some form of permanent damage or orientation has taken place. In order to investigate this phenomenon further it is proposed to conduct cyclic crack growth tests on our materials after the pre-strain is removed and compare the results with materials tested from a virgin state.

EXPERIMENTS

The three materials used are summarised in Table 1.

All the testing was performed using the pure shear test piece geometry. The pre-straining was achieved by performing a simple tensile test on a strip of rubber until the desired pre-strain extension was reached. The rubber was then gripped, in situ, along its free edges by the pure shear grips. It was then removed from the tensile test machine. The pre-strain application remained because of the shape induced constraints of the test piece geometry. From this test piece the stress strain behaviour can again be measured using an Instron test machine.

Cyclic crack growth tests were carried out using a servo-hydraulic testing machine operating at room temperature. A sinusoidal wave function of the correct displacement at a frequency of 5 Hz was imposed on the specimen. The pure shear test-piece was

\[ T = \frac{U_s \sqrt{\lambda_1}}{t(w' - c' + x')} \]

\[ = \frac{U_s \lambda_1}{t(w' - c' + x')} \]

... 13
RESULTS AND DISCUSSION

The curve fitting parameters for the stored energy function and predicted stress strain behaviour which are derived from simple tensile tests on the three materials, are tabulated in Table 2.

<table>
<thead>
<tr>
<th>Material</th>
<th>A1/MPa</th>
<th>A2/MPa</th>
<th>A3/MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.2630</td>
<td>-0.0006</td>
<td>0.0004</td>
</tr>
<tr>
<td>B</td>
<td>0.4629</td>
<td>-0.0009</td>
<td>0.0029</td>
</tr>
<tr>
<td>C</td>
<td>0.9643</td>
<td>-0.0013</td>
<td>0.0142</td>
</tr>
</tbody>
</table>

These parameters are all derived from tensile tests.

*Figure 3* shows a sample of the typical results displaying the agreement between the measured pre-strained results and those predicted if the material was exhibiting perfectly elastic isotropic behaviour.

In general for material B at the 50% pre-strains the predictions up to extensions of 100% are quite good. For higher pre-strains, the experimental data lies somewhat above the predictions. For the more highly filled material at a rather lower pre-strain, this is also the case.

This may be accounted for by the recovery of the black structure that may have broken down during the pre-straining process. This leads to a corresponding increase in the stiffness that can not be accounted for by the use of a stored energy function that assumes perfect elasticity.

*Figure 3*. Three typical sets of data curves are shown for material B at both 50% and 150% pre-strains and material C at 100% pre-strain. The points represent the measured data and the lines represent the prediction from the theoretical relationship.

*Figures 4, 5 and 6* show the tearing energy versus crack growth relationships for three materials.

At the lower tearing energies, the effect of pre-strain is broadly similar. A pre-strain of 100% reduces the $T$ value for the material A and B by a factor of 3.5 and for material C by a factor of 8.5.

For the highly filled compound C, at the higher tearing energies and at high pre-strains the crack growth rate increases dramatically.
This can be contrasted with the tearing results reported by Gent and Kim\textsuperscript{1} and A.B. Samsuri\textsuperscript{2} where pre-strain showed a dramatic drop in the tear strength, especially in the case of filled rubbers.

Tear measurements have been made on the highly filled material C at a pre-strain of 75\% with $T$ values in the range of 1600 Jm\textsuperscript{-2}. The rate at which the crack propagates is broadly similar in time to the cyclic crack growth rate. This suggests that the behaviour in the upsweep region is dominated by a time dependent phenomenon.

Pre-stressing by 100\% and 200\% were carried out and the specimens were allowed to relax. The tearing energy versus crack growth results are plotted in Figure 7. These data are compared with the data from an un-strained specimen.

No significant differences are observed between the 3 sets of data. This indicates that the pre-stressing in the direction of the crack does not affect the strength of the specimen in that direction. These results are in contradiction with the tear measurements found in the literature. A possible explanation could be the
The essential difference between these two experiments. Namely, the cyclic tests are not performed under continuous loading. This cyclic loading could to some extent reduce or even erase the anisotropy of strength in the direction of the crack. According to Gent and Kim, the pre-stressing caused the formation of an oriented region in the direction of the crack free of carbon black. This is due to segregation of rubber and carbon black when the rubber strain crystallises under the pre-stressing. One can reasonably say that the cyclic loading in the vertical direction would somehow produce the same effect and would probably re-establish an isotropy of strength, in a state close to the virgin state.

The fracture surfaces are investigated using SEM. The samples are sectioned perpendicular to the fracture surface, so that the crack profiles can be readily seen. The fractured surface is the upper surface shown on the photographs. The two photographs shown in Figure 8 are both for compound C with a crack growth at 75% pre-strains. The two different tests correspond to different tearing energies. With

Figure 6 The crack growth rate versus the tearing energy relationship for a variety of different pre-strains for material C

Figure 7 The effects of an orthogonal and relaxed pre-strain on the tearing energy versus crack growth rate behaviour. Data is shown for tests performed without a pre-strain and with specimens that have had a previous pre-strain history of 100% and 200% strain applied.
The pre-straining in the direction of the crack growth thus appears to inhibit the anisotropy development necessary to produce knotty tearing; thus under these circumstances the material is weak.

CONCLUSION

We have examined the effect of pre-straining on both the stress strain relationships and the tearing properties of filled natural rubber engineering compounds.

If it is assumed that \( W \) is a function of \( I_1 \) only then the stress strain behaviour for the pre-stressed sample can be predicted from the measured simple extension stress strain relationship. In general these predictions are in good accord with the measurements. A small discrepancy is observed for the higher pre-strains which is probably due to the breakdown and the reformation of the reinforcing filler structure. This will manifest itself as imperfect elastic behaviour.

The effect of pre-straining on the crack growth characteristics is to reduce the strength in the direction of the pre-strain by a factor ranging from three to eight depending on the amount of carbon black filler at a pre-strain of 100%. This behaviour is modified at higher tearing energies for the filler materials at pre-strains greater than about 75% when the time dependent crack growth rate increases by several orders of magnitude and then dominates the cyclic crack growth behaviour. This correlates with a marked change in the surface appearance from rough at the low rates of crack growth rate to smooth at the higher rates.
ACKNOWLEDGEMENTS

The authors are very grateful to BTR (AVS) for continued and generous support of this research.

REFERENCES


