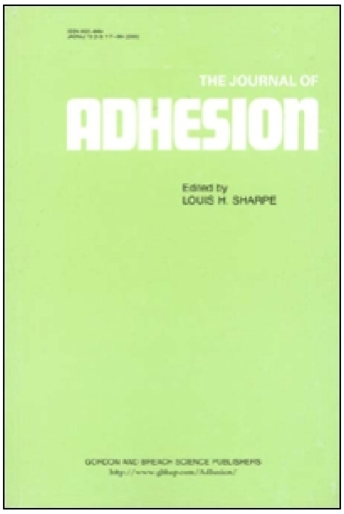

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Interfacial Friction in Filled Polymers Initiated by Adhesive Debonding

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NOTE

Interfacial Friction in Filled Polymers Initiated by Adhesive Debonding

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KEY WORDS Mechanical models of filled rubber; structural damage; hysteresis; dissipative processes; Mullins effect; energy loss mechanisms.

1. INTRODUCTION

When a rubber is subjected to stress-strain cycles, a part of the energy of deformation is dissipated as heat. This effect is called hysteresis. It has been recognized for years that hysteresis in filled rubbers is a complicated phenomenon comprising different mechanisms. The overall loss of energy in the strain cycle has been shown to be the sum of irrecoverable and recoverable (repeatable) dissipative processes.[1] The first process, known as the Mullins effect, represents a total action of several irreversible damage mechanisms, rubber-filler detachment being possibly the most important one in highly loaded elastomers.[2-5] The second process accounts for polymeric internal viscosity and some interfacial phenomena that probably could be treated as interfacial friction of damaged microstructural elements. There are also other dissipative sources of minor importance that will not be considered in this paper.

While the Mullins effect and the polymeric viscous dissipation have been objects of thorough investigation over a long period of time, the interfacial processes until now have not been given sufficient attention although their influence on the bulk properties of filled rubbers is, in some instances, more important than others.

The goal of this paper is to demonstrate experimentally the significance of interfacial dissipation and its influence on the overall behavior of filled rubbers.

2. THEORETICAL DESCRIPTION

The interfacial dissipation appears to be related to a number of structural and operational factors such as filler volume loading, surface of the filler particles, degree of structural failure, chemical nature of rubber and filler, superimposed pressure, rate of deformation and some others. The surface friction between the damaged structural elements is assumed to be the main source of this dissipation.

Its detailed theoretical description is at present hardly possible mainly because of the lack of experimental evidence. The present-day task is to get some new experimental data on the subject in order to provide more insight into the nature of the phenomenon.

In this respect, the first step to be done is to separate the interfacial dissipative component from the whole of dissipation. This can be achieved by preliminary specimen cycling to eliminate the Mullins effect and by using a highly plasticized rubber matrix to make molecular viscosity negligibly small.

If that is so, then interfacial effects remain the only source of recoverable hysteresis, and one may proceed to the systemic experimental study of the phenomenon.

The present investigation is aimed at establishing the influence of filler content on the interfacial energy dissipation.

3. EXPERIMENTAL DETAILS

Highly plasticized (50% oil by weight) polybutadiene rubber and potassium chloride were used as matrix and filler. Volume loadings investigated were 0, 0.1, 0.35 and 0.55 percent with particle sizes of 200 micrometers.

Solid and rubber ingredients were mixed on laboratory rolls, formed into sheets and cured. Dumbbell standard specimens were cut out of the sheets with an effective gauge length about 50 mm and a cross-section area of about 0.28 sq. cm. The experiments were carried out on a tensile tester at room temperature with a crosshead speed of 3 mm per minute under atmospheric and elevated pressures. The surface of the highly loaded specimens was protected by a thin film of soft, highly elastic SBS rubber against air or liquid penetration into pores generated by adhesive debonding.

4. RESULTS AND DISCUSSION

A typical set of stress-strain cycles is shown in Fig. 1 for the specimen containing 55% by volume filler particles. Progressive diminution of the initial large Mullins loop to a minor recoverable one is observed, whereas the cycling of unfilled rubber (Fig. 2) shows no discernible dissipative effect. Therefore, the unfilled rubber matrix studied may be regarded as the non-dissipative element of the composite system, and the recoverable hysteresis is to be ascribed to interfacial dissipation only.

Fig. 3 represents recoverable hysteresis losses as functions of strain amplitudes and filler content. It is seen that hysteresis increases with the rise of strain and filler content. This relationship is in accordance with the theoretical anticipation that

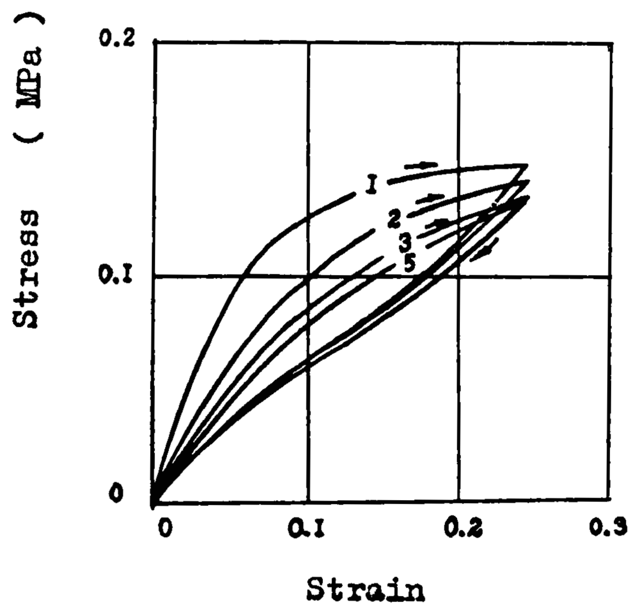

FIGURE 1 The stress-strain cycles for a specimen filled 55% by volume at 24% strain amplitude.

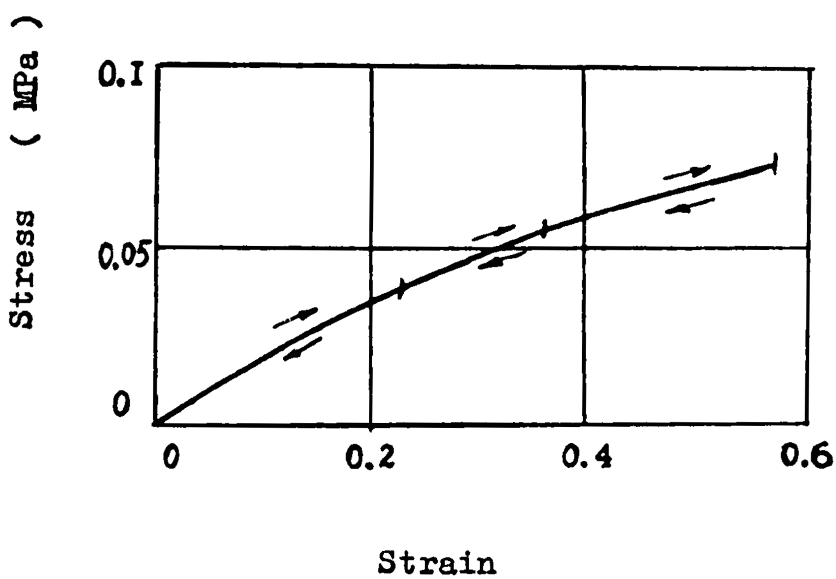

FIGURE 2 The stress-strain cycles for an unfilled rubber specimen. Dashes on the line signify strain amplitudes.

more strain and more filler content produce more structural damage and more recoverable interface dissipation.

The results obtained confirm the assumption that surface friction is really the cause of the interfacial dissipation. Furthermore, it is clear that frictional losses can be produced only by those portions of the total interface area that are in direct

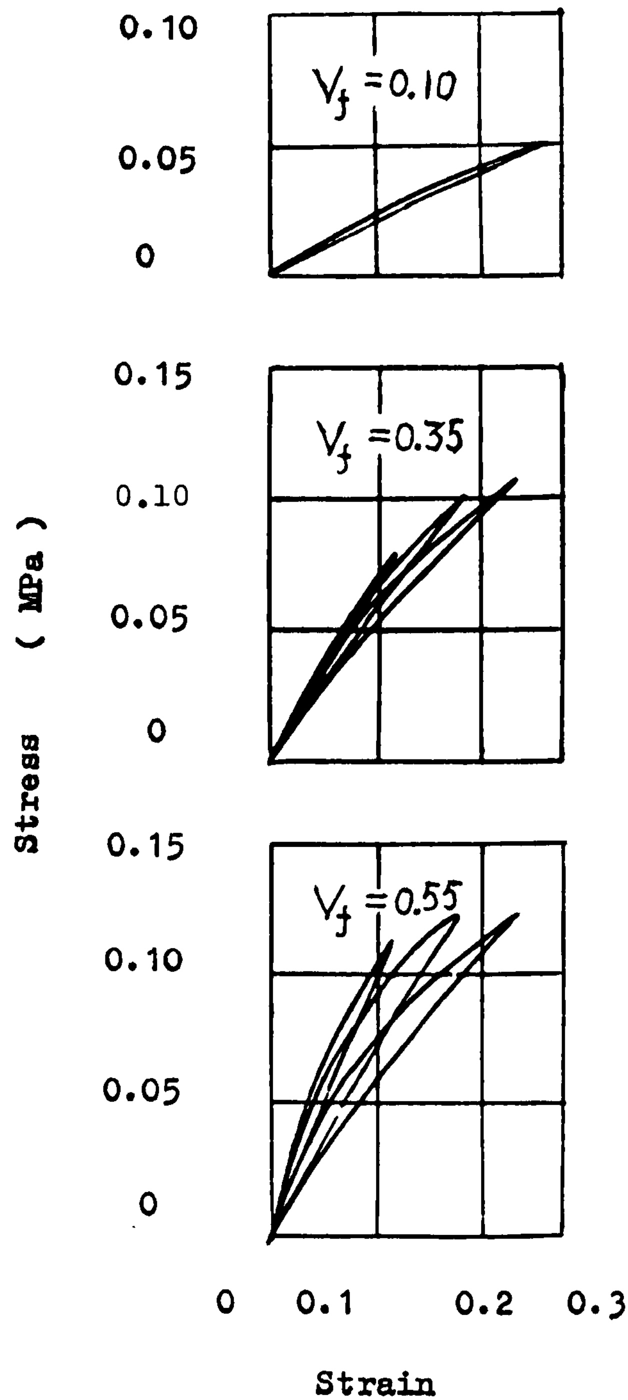

FIGURE 3 Recoverable hysteresis loop at various strain amplitudes and filler contents: 0.10, 0.35, 0.55%, by volume.

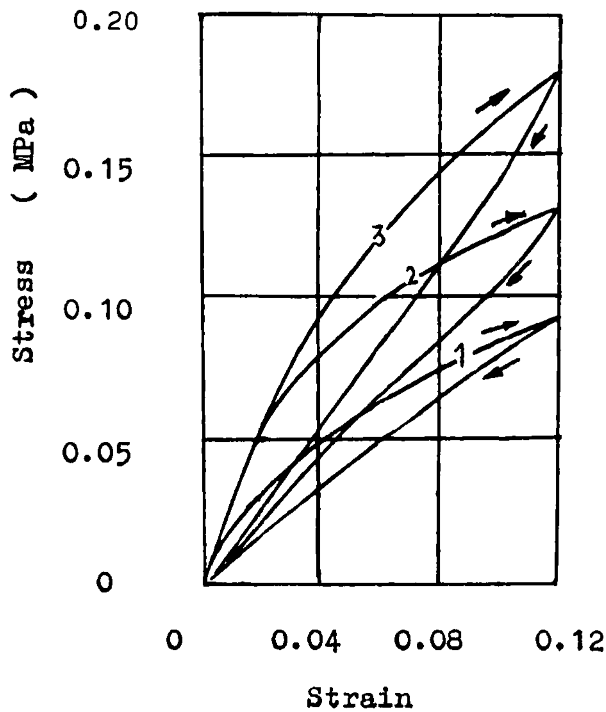

FIGURE 4 The hysteresis of a sample containing 55% by volume filler under various pressures: 1–0.10, 2–0.22, 3–0.58 (MPa).

contact with each other. Changing the contact area might influence recoverable hysteresis and possibly the overall behavior of the composite system. Strain cycling under the superimposed pressure seems to be the simplest way of ensuring action on the interfacial contact area. The influence of pressure on the recoverable hysteresis is shown in Fig. 4. Indeed, elevated pressure provides a marked rise of the hysteresis values whereas nonfilled rubber shows no pressure sensitivity.

The important point is that the frictional effect is not the only cause of pressure reinforcement of the specimens, the elastic contribution being also significant.

In the light of the data obtained, a more accurate interpretation of the stress-strain relations is needed. It becomes clear that any extension of filled rubber is accompanied not only by the stress-softening but also by some increase of resistance due to the interfacial friction resulting from the damage. This is to be taken into account in developing constitutive relations for filler-loaded vulcanizates.

5. CONCLUSIONS

1. Significant influence of the interfacial friction due to structural damage on the bulk behavior of filled vulcanizates is demonstrated.

2. The superimposed pressure is shown to enhance internal resistance partly through an increase of interfacial friction.

3. Constitutive relations and mechanical models for filled rubbers must take into account the interfacial friction produced by the structural damage.

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