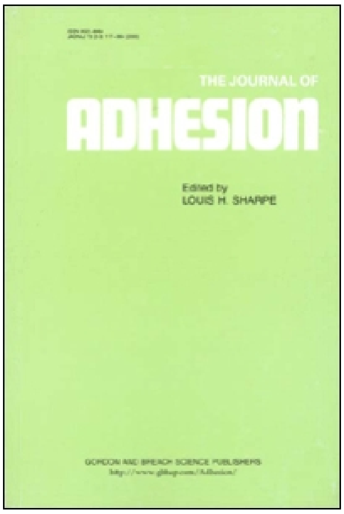

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Interfacial Friction in Filled Polymers Initiated by Adhesive Debonding. II. Relaxation Studies

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NOTE
Interfacial Friction in Filled Polymers Initiated by Adhesive Debonding.
II. Relaxation Studies

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KEY WORDS mechanical model of filled rubbers; structural damage; dissipative processes; stress relaxation; pressure influence; particulate filled composite; Mullins effect; hysteresis; matrix-filler detachment.

1. INTRODUCTION

A number of dissipative mechanisms exhibited by particulate polymeric composites are known. The viscoelasticity of matrices as a repeatable phenomenon and the Mullins effect as an irreversible one have been investigated by many researchers over a long period of time.

In the meantime, other dissipative manifestations have not yet been given sufficient attention although their influence on the mechanical properties may also be significant. In particular, we are concerned with the behavior of composite systems in which active interfacial phenomena come into play after adhesive debonding has set in.

The significant role of the interfacial friction caused by structural damage has been demonstrated in Reference 1. The importance of this factor has been corroborated through hysteretic tests. Specifically, the influence of superimposed pressure on frictional resistance has been investigated. However, in this set of experiments plausible time-dependent effects were not examined, all the tests having been made at a fixed rate of extension and retraction.

The present paper is the extension of the investigation reported in Reference 1 towards the elucidation of the temporal effects in an elastomeric particulate composite in which the degree of adhesive debonding is kept constant. Stress relaxation measurements were chosen as a research tool in performing this study.

2. THEORETICAL DESCRIPTION

A speculative examination of the behavior of a composite structural element helps one to discover an appropriate test approach. The changes in a structural element exposed to extension may be imagined as follows. Let us assume this element to have the form of an elastic cylinder (matrix) containing a solid sphere (filler particle) (Fig. 1).

Initially, matrix and sphere are bonded to one another. The ends of the cylinder are fastened to the clamps. No internal and external stresses exist in the element in the initial state (Fig. 1a). When the clamps are pulled apart (Fig. 1b) the detachment of the matrix from the sphere occurs at a certain elongation, and almost instantly a vacuole forms around the sphere. This primary form of microdamage may be thought of as the representation of elementary Mullins damage.

The detachment of the matrix from the sphere occurs in its polar parts whereas the equatorial site of the sphere retains contact with the elastomer. On further elongation the matrix is thought to slide over the sphere's surface in the region of contact creating Coulomb friction. This friction is now the only remaining source of time-dependent energy dissipation, provided that the matrix is regarded to be a purely elastic material.

The commonly-observed relaxation of the virgin specimens reflects the combined effects of both Mullins irreversible damage accumulation and newly-created and enhancing frictional dissipative processes. Therefore, if one intends to study frictional relaxation as such, time-dependent Mullins softening of the specimens must be somehow eliminated.

The preparation of specimens free of Mullins effect interference, for instance, may be attained by their previous cycling to some strain amplitude until the narrowing hysteresis loops become repeatable. The deformation values imposed on the specimen in subsequent relaxation tests must, of course, be lower than the amplitudes in preparatory cyclings in order not to trigger new Mullins damage.

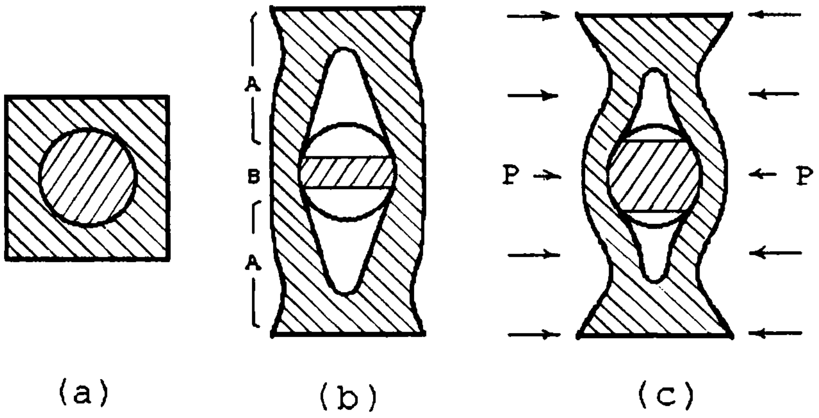

FIGURE 1 Structural element states under extension: (a)—the initial state, (b)—the state after detachment has occurred, (c)—the state after the lateral pressure, P, has been imposed.

One may suppose that the external pressure, P, imposed on the debonded, stretched element must lead to the increase of the contact surface between the matrix and the sphere (Fig. 1c), thus contributing to enhancement of the frictional manifestation. An idea suggests itself that the relaxation of the debonded composites might be a rather sensible function of the external pressure.

The present investigation is aimed at experimental corroboration of the above theoretical predictions.

3. EXPERIMENTAL DETAILS

Highly plasticized (50% oil by weight) polybutadiene rubber and potassium chloride (particle sizes about 200 micrometers) were used as matrix and filler with a volume loading of 35%. 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 of about 50 mm and a cross-section area of about 0.28 cm^2.

The cycling and relaxation experiments were carried out on a tensile tester at room temperature under reduced, atmospheric and elevated air pressures, the surface of specimens being protected by a thin film of soft, highly elastic SBS rubber against gas penetration into the pores generated by adhesive debonding.

4. RESULTS AND DISCUSSION

Figure 2 depicts the joint manifestation of both Mullins damage and friction mechanisms on the stress relaxation of a virgin specimen extended to 40% (curve 1). The

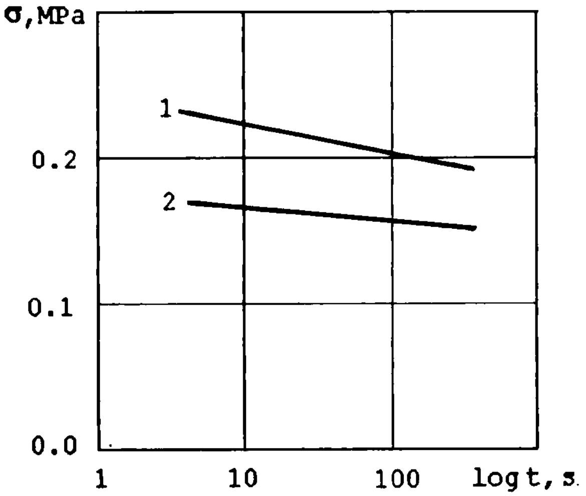

FIGURE 2 Comparison of the stress relaxation, σ (MPa), *vs.* log t (sec) of the virgin specimen with that in which Mullins relaxation has been eliminated: 1—the virgin specimen, 2—the specimen after previous treatment by cycling.

same specimen was further cycled at an amplitude of 50% to bring the Mullins damage to its limiting value for this elongation. The hysteresis loops became repeatable after 5 cycles. This provided evidence of the stabilization of Mullins damage. For brevity the specimens so treated subsequently will be referred to as "trained" specimens.

This specimen was further tested once again for relaxation at 40% elongation (curve 2 from Fig. 2). It is seen that in spite of removing time-dependent Mullins softening some residual stress relaxation is still retained. It appears that this relaxation may be attributed solely to the internal friction of the Coulomb type. Proper matrix viscosity interference is hardly possible.[1] This is also corroborated by the hysteresis tests obtained with trained specimens under various external pressures. In Figure 3, the strong influence of the external pressure (within a narrow range from 0.002 to 0.4 MPa) on the recoverable hysteresis is shown. Obviously, such high pressure sensitivity cannot be attributed to the viscous properties of the matrix alone.

It is interesting to note that at the very low external pressure of 0.002 MPa the dissipation becomes negligibly small. This suggests that only a small part of the interface friction exists under zero external pressure due to unconstrained pore increase. It implies, as well, the importance of taking into account the atmospheric pressure influence when soft rubber composites are to be tested.

These considerations also suggest that interface relaxation phenomena must be rather sensitive functions of superimposed pressures. Figure 4 depicts the stress relaxation curves of the trained specimen under different superimposed pressures. The marked influence of the pressure on the stress level as well as on the stress relaxation rate caused by internal Coulomb friction is clearly seen.

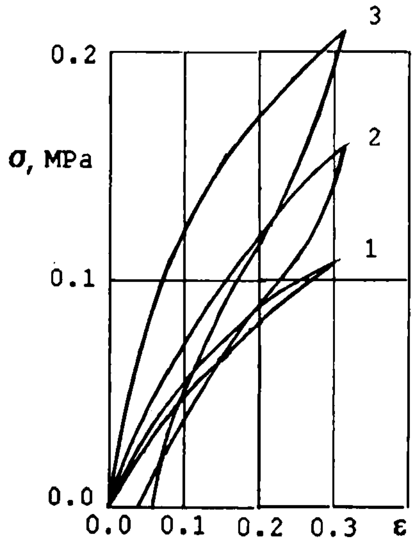

FIGURE 3 Hysteresis of the "trained" specimen under various external pressures: 1—0.002, 2—0.1, 3—0.4 (MPa).

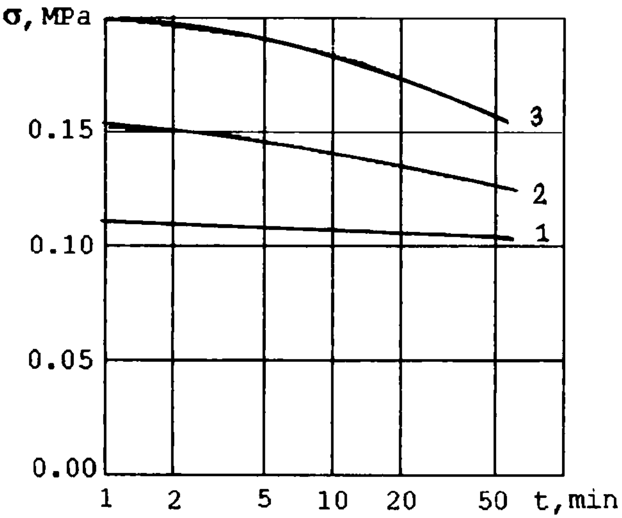

FIGURE 4 Relaxation (σ) curves under different pressures of the specimen freed of Mullins relaxation: 1—0.002, 2—0.1, 3—0.4 (MPa).

Obviously, such unusually high pressure sensitivity of the stress relaxation is characteristic only of low modulus composites using soft matrices. This effect is thought to decrease progressively with increasing matrix modulus.

5. CONCLUSION

The existence of the stress relaxation mechanism caused by the Coulomb internal friction induced by the appearance and accumulation of damage has been demonstrated experimentally.

The marked influence of moderate pressures on the rate of frictional relaxation has been predicted and corroborated experimentally.

Closer examination of the relaxation features triggered by matrix-filler detachment seems to be of interest. This study should take into account mechanical properties of the constituent elements (matrix and filler modulus, adhesive resistance, friction behavior), formulation of the composite (filler volume content, particle size), and operative conditions (current degree of adhesive debonding, pressure, imposed elongation).

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