THERMOFLUCTUATIONAL APPROACH TO FRACTURE OF FILLED ELASTOMERS

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The hypothesis for statistical space-time cause of every new damage initiation in a material is used to analyze the dependence of elastomer rupture strength on the size of filler particles and the rate of material loading. The occurrence of destructive thermofluctuations in a medium is the key supposition that forms the basis of this hypothesis. The accumulation process of matrix debondings from inclusions has been calculated in the framework of the proposed model. The peculiarities of material response under cyclic loading have been considered. It is shown that matrix adhesion to filler particles may cause the observed hysteresis losses.

INTRODUCTION

According to numerous experimental investigations, damage in materials with large solid particles is initiated earlier and accumulated faster than in materials with small particles. Damage accumulation depends on the rate of specimen loading and temperature.

Different hypotheses have been advanced to explain this phenomena. Our treatment is based on the hypothesis that the probability of damage initiation at given time and point depends only on the stresses acting exactly at the same time and point (1, 2). Damage initiation can be attributed to destructive thermofluctuations, which have an extremely small probability of occurence but are certain to appear in large volumes and at long time intervals.

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DAMAGE INITIATION PROBABILITY

In the present paper, we consider particulate composite materials with elastomeric matrix. The probability P that at least one new damage occurs in the elastomer matrix at time $[t_1, t_2]$ is defined by the formula

$$P = 1 - \exp\left(\int_{t_1}^{t_2} \iiint_V F_k(\sigma_k)\ dV\ dt + \int_{t_1}^{t_2} \iint_S F_a(\sigma_a)\ dS\ dt \right) , \qquad (1)$$

where V is the matrix volume, S is the surface of all undebonded inclusions, σ_k is the invariant of tensor stress, and σ_a is the scalar characteristic of the debonding stress on the surface S. This formula may be used to describe a random process of damage accumulation in a medium. The functions F_k, F_a involved in the formula are expressed as

$$F_a(\sigma_a) \ = \ - \ B_a\ {\sigma_a}^{n_a}\ H(\sigma_a - \sigma_a^{min}) \ ,$$

$$F_k(\sigma_k) \ = \ - \ B_k\ {\sigma_k}^{n_k}\ H(\sigma_k - \sigma_k^{min}) \ ,$$

where B_a, B_k, n_a, n_k, σ_a^{min}, σ_k^{min} are the material strength constants, and $H(...)$ is the Heaviside function.

As shown in (1 - 4), the space-time probability hypothesis for damage initiation offers a number of advantages: (i) it allows modeling the scale phenomena of elastomer failure; (ii) contains all necessary prerequisites for evaluating the probability of damage cluster formation in a material; (iii) predicts the situation when the strength of a particulate elastomer can be higher than that of a pure binder. Here we analyze the probability of relation that might exist between the adhesive strength and hysteresis phenomena in filled elastomers.

ADHESIVE STRENGTH AND HYSTERESIS LOSSES

Consider fibrous composite with a periodic cell, the quarter of which is shown in Fig. 1a. Using this cell we can calculate the ratio of Young's composite module to that of the matrix $c = E_c/E_m$ (in the plane perpendicular to the direction of fibers) and the integral over the surface of undebonded inclusions

$$I(\varphi) = \frac{1}{{\sigma_c}^3} \iint_S {\sigma_r}^3 H(\sigma_r)\ dS,$$

where φ=0, 1/6, 1/3, 2/3, 5/6, 1 are the fractions of debonded fibers, σ_c is tensile macrostress, σ_r is debonding tensile stress, and S is the surface of undebonded inclusions. The relations $E(\varphi)$ and $I(\varphi)$ approximated for the above values of φ have been used to calculate the kinetics of adhesive damage accumulation in a grain material with randomly distributed inclusions. A replacement of three-dimensional problem by a two-dimensional analog and elimination of nonlinear elastomer properties must naturally lead to serious quantitative errors. With such approach it is impossible to exactly define the macroscopic constants of a grain composite. On the other hand, in the context of a two-dimensional and, hence, a more simple model we can readily identify the basic mechanisms of fracture in a material and demonstrate that the time characteristics of the adhesive strength are of crucial importance for damage accumulation. These seem to be good reason for using the "plane" approach in qualitative analysis of the governing relationships.

It can be shown from equation (1) that during the first cycle of loading the growth of damage in the material is described by the equation

$$\frac{d\varphi}{dt} = (1-\varphi)4\pi R^2 B_a {\sigma_c}^3 I(\varphi).$$

After unloading the matrix readheres to the inclusion and its debonding requires application of a certain force. However this force will be less than in the first loading. The strength constant B_a^* used in our calculations for repeated loadings was a factor of 10 greater, than the constant B_a. In the N-th cycle of loading the total amount of debonded particles

$$\varphi = \varphi_1 + \varphi_2$$

is made up of the particles debonded during previous cycles and redebonded in the current cycle φ_1, plus the particles first debonded in the current cycle φ_2. To calculate these fractions we use the following formulae

$$\frac{d\varphi_1}{dt} = (\varphi_{max} - \varphi_1)\, 4\pi R^2 B_a^* {\sigma_c}^3 I(\varphi),$$

$$\frac{d\varphi_2}{dt} = (1 - \varphi_{max} - \varphi_2)\, 4\pi R^2 B_a {\sigma_c}^3 I(\varphi),$$

where φ_{max} is the maximum fraction of the particles deboned during previous loading cycles.

NUMERICAL SIMULATION

The predicted values of composite stresses σ_c are shown in Fig. 1c as the functions of composite strains ε. The following strength constants were used in calculation: n_a=3, σ_a^{min}=0. The theoretical curves clearly demonstrate the dependence of debond accumulation rate on the strain rate. Similar dependencies have been obtained from the experiments (5), (6). The same qualitative behavior is expected in the case of microdamage accumulation in the matrix.

Consider the material response under loading-unloading conditions. A relative reproducibility of the results occurs in the fifth cycle (Fig. 1c), though the situation may vary. It means that depending on the values of material constants a relative reproducibility can be observed at later cycles.

The hysteresis losses in elastomeric composites are frequently related to viscoelastic properties of the matrix and the effect of its friction on the filler surfaces. The calculations show that matrix debonding from the inclusions can also be one of the mechanisms for dissipative losses in materials. Moreover, the dependence of losses on the strain rate can be explained by thermofluctuation mechanism of debond.

THE RELATION BETWEEN ADHESIVE AND COHESIVE FRACTURE

The proposed approach can be used to examine the relation between adhesive and cohesive mechanisms of damage accumulation. Fig. 2a gives a schematic representation of the simplest possible system involving identical structural elements. Each element consists of the three elastic, uniformly loaded parts A, B, C and one adhesive joint D. Such system represents the key features of the response of high-particulate composite materials: a nonuniform loading of the material (each part of the structural element is under different stresses), redistribution of loads after the matrix debonds from the inclusion (when the joint D fails). This model has been used to qualitatively describe the relation of adhesive and cohesive damages in the system.

In a real composite damage accumulation is similar to the process of damage growth in the considered model. In computational procedure we have used the elastic elements A, B and C with the following lengths and cross-sections: 0.12 mm, 0.02 mm, 0.01 mm and 0.001 mm^2, 0.009 mm^2, 0.01 mm^2. The contact area in the adhesive is equal to 0.009 mm^2. The strength constants used to model a strong and weak adhesion are, respectively, B_a=8.67$\cdot$10^4 MPa^{-3}m^{-2}c^{-1} and B_a=8.67$\cdot$10^5 MPa^{-3}m^{-2}c^{-1}, B_k=7.17$\cdot$10^7 MPa$^{-3.3}$m^{-3}c^{-1}, $n_a = 3$, $n_b =$ 3.3, $\sigma_a^{min} = 0$, $\sigma_k^{min} = 1.7$ MPa. The structural elements incorporated in calculation were 1000 in number.

The computational experiments have shown that the process of debond accumulation taking place in the system with a weak adhesive joint (Fig. 2b, curves 2 and 4) results in material softening due to a break of bonds D. The load is redistributed from the more rigid elements B to the less rigid A. Such material has essentially less strength and may be more deformable than a material with strong adhesive bonds (Fig. 2b, curves 1 and 3). A 1000-fold increase of the structural element size (with a ten-fold decrease of their number) accelerates the fracture processes in the medium (Fig. 2b, curves 1 and 2).

There are two ways of increasing the strength of the material: (i) to essential reduce the matrix volumes counteracting loading (the volumes, in which destructive fluctuations may occur), and (ii) to increase the stress growth rates in the overloaded regions, and consequently, to shorten the time interval within which one may expect the onset of fluctuations.

The calculations show that materials with strong adhesive bonds (active particulate materials) can withstand greater loads, than materials with weak adhesive bonds (filled with inactive particulate). In the framework of the proposed approach it is possible to qualitative simulate the regularities observed in the experiments without consideration in the model the layers surrounding the inclusions.

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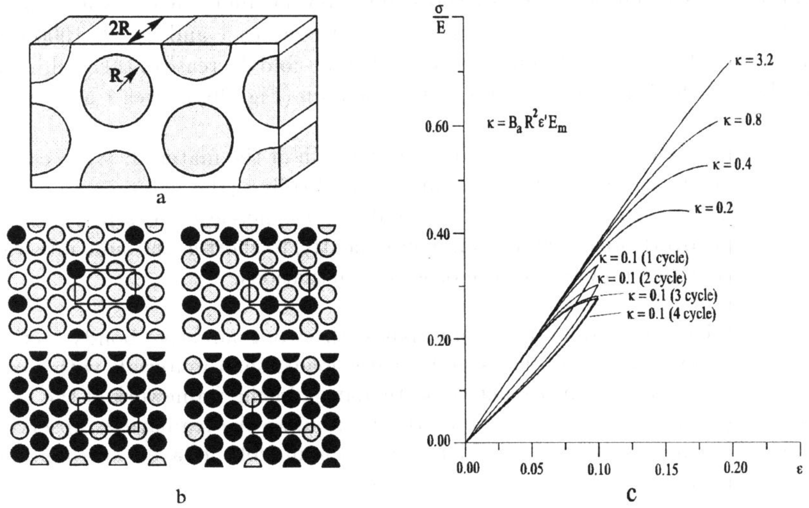

Figure 1 Schematic quarter of a periodical cell (a) being used to calculate damage evolution (b) and loading curves (c) for an elastomer composite.

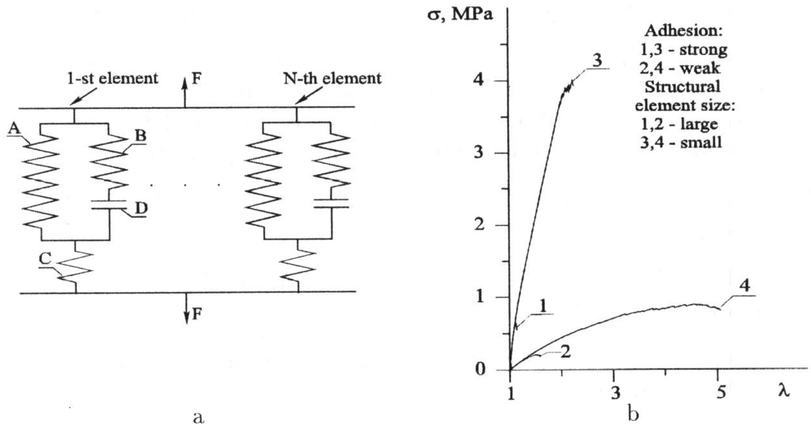

Figure 2 A set of structural elements (a) employing to qualitatively model the loading curves of an elastomer composite (b).