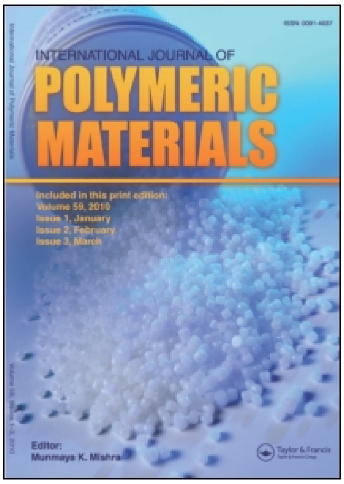

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# Micromechanical Features of Highly-Filled Composites are Most Distinctly Defined by Mechanical Heterogeneity

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# Micromechanical Features of Highly-Filled Composites are Most Distinctly Defined by Mechanical Heterogeneity†

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Theoretical calculations of the stress fields in highly-filled composites consisting of a soft rubbery binder and a hard inclusions have been carried out.

Two types of stress concentrations (hydrostatic and distortional) have been found in the binder phase. The hydrostatic tensions do not disappear even under rather high superimposed pressures.

Thin stiffened layers of the "bound" rubber on the surface of inclusions do not influence considerably the field of stresses around inclusions.

The shape of the binder stress-strain curve strongly influences the strength and strain capabilities of composites, final steep rise on the stress-strain curve of the binder resulting in a more uniform stress and strain distributions in the composite.

## INTRODUCTION

There is a large body of literature devoted to micromechanics of composite materials. Nonetheless, the important problem of predicting the behavior of a composite on the basis of the analysis of its structure remains substantially unsolved. The stress analysis, as an important step in solving this problem, is hindered by the complicated character of the nonhomogeneous field of stresses and strains in structural elements of these materials. The common practice of considering the separate components of the stress field[1] is not

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well adapted for a general overview of the problem. For that reason one of the objectives of the present investigation is to represent the stress fields in a more conventional form, two scalar characteristics only being examined: hydrostatic and distortional ones.

The most appropriate materials for such an analysis are those in which rigid inclusions are embedded into the soft binder. The inclusions being undeformable, the binder becomes the only phase which contributes to the overall strain capability of the composite, and the stress and strain non-uniformity of the binder phase is highest possibly manifested.

The main point for consideration in this paper is stress fields in the binder phase of highly-loaded composites. Other important features of the binder influencing its stress state, such as stiffened polymer layers around the inclusions and non-linearity of the binder stress-strain relations, will also be considered.

## STRESS STATE OF THE BINDER PHASE IN HIGH-LOADED COMPOSITES

The distances between the inclusions in a high-loaded composite being a small fraction of the inclusion diameter, pairwise interactions become important. So two circular inclusions embedded in an infinite binder plate were chosen for the investigation, as a first approximation representing the cell of the composite (Figure 1).

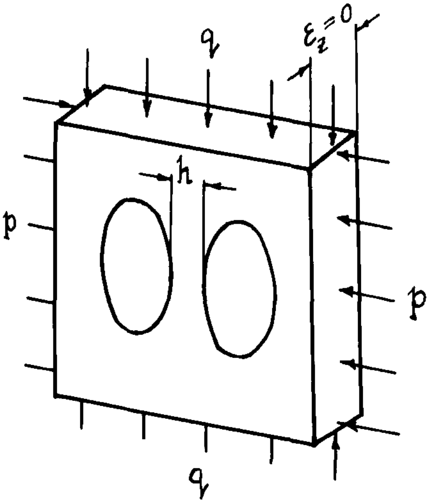

FIGURE 1 The elementary two-inclusion-cell of a high loaded composite. Inclusion diameters are taken to be unity.

The assumptions accepted are: (1) The inclusions are nondeformable solids; (2) The binder is an incompressible Hoockean elastic material; (3) The adhesion between the inclusions and the binder is perfect; (4) The deformation is effectuated under plane strain conditions with loading stresses being applied at infinity.

Calculations of stresses and strains have been carried out on the basis of the well known mathematical theory of analytical functions.[2] The calculation method having been described elsewhere,[3] only the results of calculation are given below.

The stress state is expressed in the form of hydrostatic $\sigma_h$ and mean distortional $\sigma_d$ stresses, calculated as

$$\sigma_h = (\sigma_x+\sigma_y+\sigma_z)/3$$

$$\sigma_d = 2^{-1/2}[(\sigma_x-\sigma_y)^2+(\sigma_y-\sigma_z)^2+(\sigma_z-\sigma_x)^2+6\tau_{xy}^2]^{1/2}$$

in which $\sigma_x$, $\sigma_y$, $\sigma_z$, $\tau_{xy}$ are the stress components under the plain strain conditions.

Hydrostatic and distortional fields are depicted as lines of constant levels of a given quantity. A typical picture of these fields is shown in Figures 2 and 3, for the loading by a vertical unit compressive stress, $q = -1$.

Each inclusion acts as a stress concentrator, and the stress state between the inclusion is a result of the interaction of the two fields produced by the inclusions.

From Figures 2 and 3 one may reveal two kinds of stress concentrations (hydrostatic and distortional) which do not coincide spacely.

It may be concluded from Figure 2 that the hydrostatic tension between

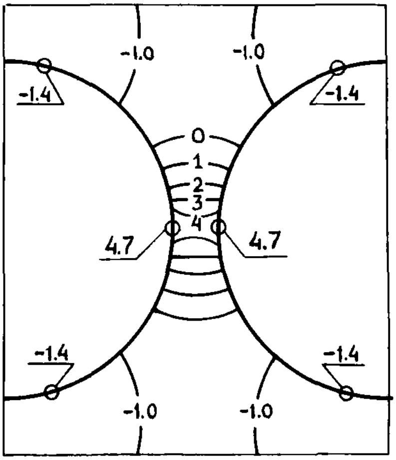

FIGURE 2 Typical hydrostatic stress field in the binder between two inclusions ($h = 0.1$).

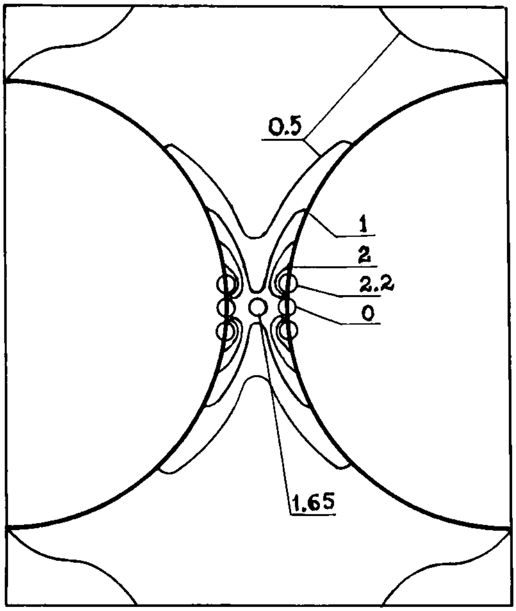

FIGURE 3 Typical distortional stress field in the binder between two inclusions ($h = 0.1$).

the inclusions is produced despite the externally applied hydrostatic compression (−0.5). The region of hydrostatic tension occupies a substantial part of the space between the inclusions, the hydrostatic maximum 4.7 being located on the surface of the inclusions in the narrowest place of the gap. Since the distortional component is zero at this point, the binder is subjected to the pure triaxial tension. Decreasing the gap between the inclusions leads to sharp increase in the hydrostatic tensile maximum (Figure 4).

Increasing both $p$ and $q$ compressive stresses (Figure 1), while the difference $(p-q)$ is unaltered, tends to decrease the hydrostatic tension region and ultimately to eliminate it at a certain critical value $P_c$ of the external hydrostatic compression (Figure 5).

On the other hand, hydrostatic tensions in the gap are enhanced by increasing the difference $(p-q)$ under the condition of a constant superimposed pressure which directly follows from the linearity of the system being investigated.

Calculations made for different values of relative gaps $h$ and external stress differences $(p-q)$, show that the relation of critical external pressure $P_c$ to these quantities may be given by

$$P_c = (p-q)/1.6h$$

One can easily conclude that for small gaps rather high external pressures

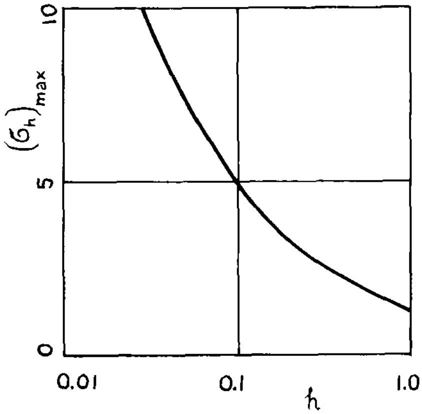

FIGURE 4 Dependence of maximum hydrostatic tensile stress on the relative gap between the inclusions.

are needed to eliminate local hydrostatic tensions. For example, if the gap is 0.1 and $(p-q)$ is 5, the critical pressure required becomes equal to about 30.

These results quantitatively confirm the known experience that straining the highly-filled composites even at rather high external pressure promotes, nevertheless, local hydrostatic tensions in the binder phase.

Returning to the distortional field (Figure 3), one can see that there are several maxima in the gap, global ones being located on the surface of the inclusion, aside from the line of centres, and the local maximum in the middle of the gap.

Both maximum hydrostatic and distortional components increase with the decreasing gap. However, the rise of the former component is much faster than that of the latter one (Figure 6).

The present results strongly suggest that the two revealed types of stress concentration can become two independent sources of binder failure, adhesion type failure being set aside.

Gent and Lindley[4] have shown that hydrostatic failure is usually initiated at moderate hydrostatic stresses roughly equal to Young's modulus of the binder. Taking into account their results and those stated above in Figure 6, one may conclude that it is the hydrostatic stress concentration that is most probably the primary cause of the binder phase failure.

It is obvious, however, that under high superimposed pressure hydrostatic tensions can be excluded. Failure point is then displaced to the distortional maximum region, the distortion remaining the only mode of failure.

From the above analysis it is also apparent, that microfailures due to the high hydrostatic and distortional stress concentrations, are practically

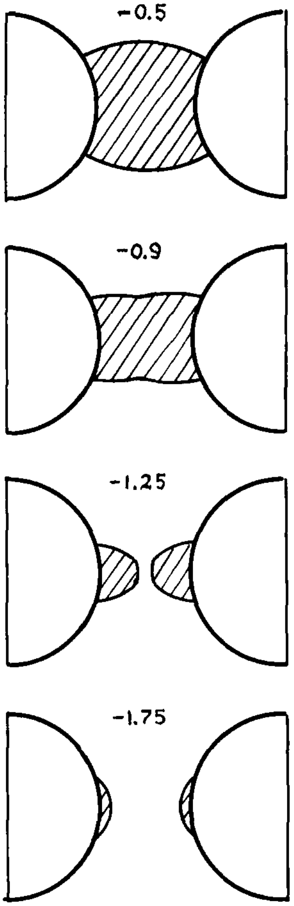

FIGURE 5 Regions of hydrostatic tension (shaded area) under different superimposed pressures (indicated above each picture) for $h = 0.5$ and $(p-q) = -1$.

inevitable at the very beginning of straining high-loaded elastomeric composites.

With the present understanding of the binder stress state it is important to investigate the possible ways of impeding the failure initiation. Two possibilities are discussed below.

## EFFECT OF SURFACE LAYERS ON THE STRESS STATE AROUND INCLUSIONS

The immobilization of the rubber near the surface of the solids and its

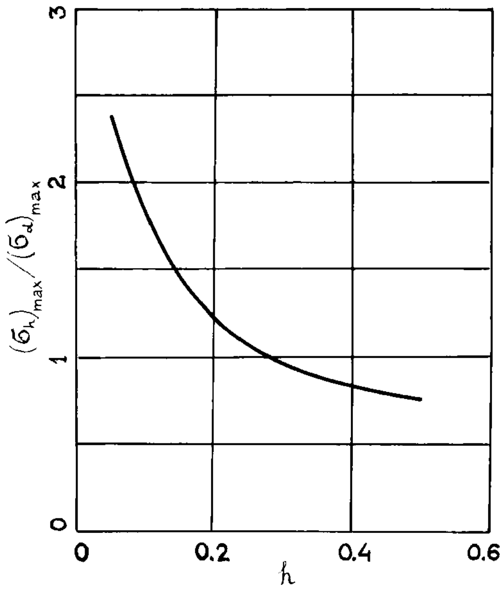

FIGURE 6 Hydrostatic to distortional stress concentration ratio versus relative gap.

influence on the mechanical properties of composites has long been recognized. The mechanism of this phenomenon is, however, complicated and not well understood. Our concern in this problem is the investigation of the effect of the immobilized rubber layer with higher Young's modulus on the stress and strain distributions around the inclusion.

A model chosen for the investigation, and the loading conditions are depicted in Figure 7. Perfect adhesion between the inclusion, layer, and binder has been assumed.

Figure 8 shows, as an example, the distortional strain field for a layer thickness of 0.1 inclusion diameter and for layer to binder moduli ratio 3.

We have calculated the maximum hydrostatic stresses and maximum distortional strains for a number of layer thicknesses and layer to binder moduli ratios. Layers with moduli higher than those of binder tend to decrease both hydrostatic and distortional stress and strain concentrations as compared with the no-layer case. The maximum reduction of the strain concentration of 0.6 takes place at relative thickness of 0.4 and moduli ratio of 2.1 (Figure 9). A similar relationship holds for hydrostatic stress field.

Taking into account that real relative thicknesses are essentially less than 0.1 (at least for 10 to 100 $\mu$m diameter inclusions), the layer influence on the stress distributions around such inclusions may be considered as negligible, as follows from Figure 9.

So it seems likely that the favourable effect of real layers is not related to

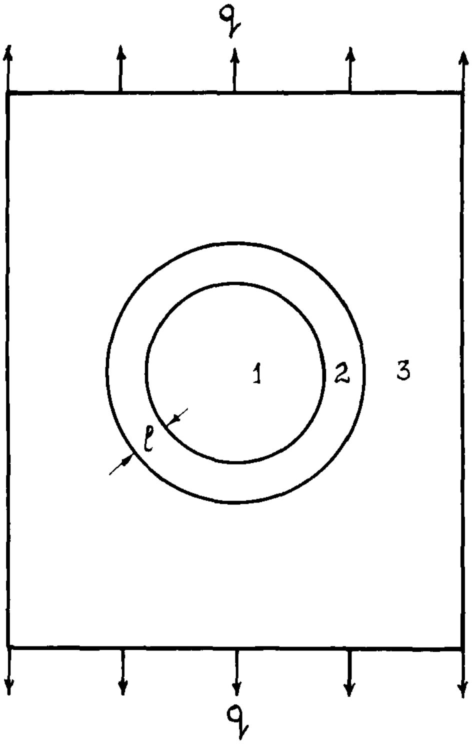

FIGURE 7 The model of an inclusion with a layer: 1 rigid inclusion of a unit length diameter; 2 layer of $l$ thickness; 3 infinite binder phase; $q$ is a unit tensile stress at infinity.

substantial stress changes in the binder phase. The cause of composite reinforcement by surface layers, probably, might be sought in the stress state variation after the separation of binder from inclusion has occurred. The surface layer then turns into a sort of a stiff peel, impeding the stress concentration on the new born free surface.

## BINDER PROPERTIES—COMPOSITE PROPERTIES RELATIONSHIP

Above linear solutions for the stress and strain fields are based on Hookean stress-strain law, and so they must be considered as a first approximation to the real stress state.

There are elastomers which are characterized by stress-strain curves with a sharp increase in slope at certain characteristic strains usually of about 300% (Figure 10). Such binder features might be expected to influence deeply the composite behavior. There is always some distribution of local extensions

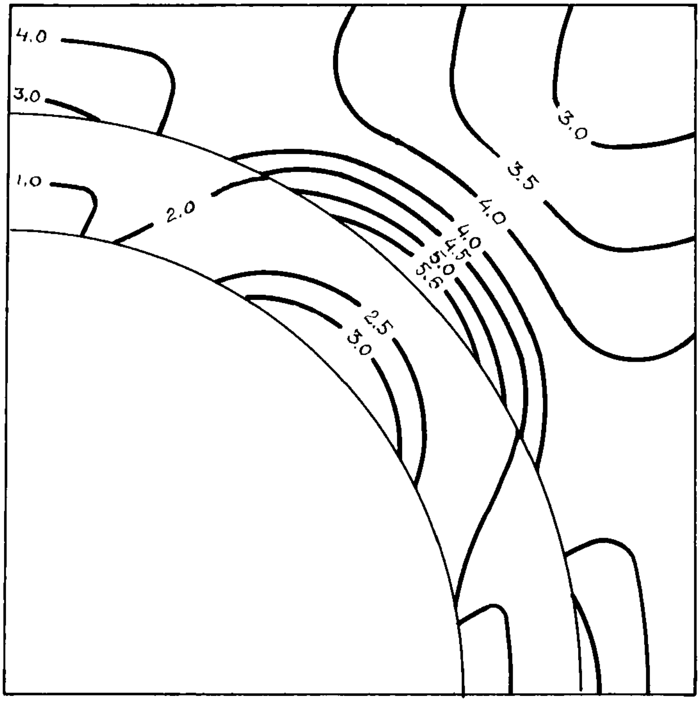

FIGURE 8 Typical distortional strain distribution (percents) in a layer and a binder.

in the non-uniform stress field of the loaded binder phase. In the course of stretching, places with higher initial extensions meet first the characteristic strain above which stiffening occurs. The rate of local elongations at these places is then impeded as compared with other ones which have not yet reached the state of stiffening. A more uniform distribution of strains is expected in the binder phase, while approaching the final stage of stretching. Hence there is also considerable favouring of the composite behavior to be observed.

If the binder stress-strain curve does not exhibit the final steep rising section (Figure 10), the opposite situation may be suggested. The initial local maximum elongations while stretching are progressively enhanced in comparison with the other less stressed regions, and the strain capability of the composite must be poor.

The above consideration can be well substantiated by the illustrative calculation that has been performed on a simple model. This one represents a specimen with a non-uniform cross-section as shown in Figure 11. The relative decrease of cross-section *a* and the relative length of the decreased cross-section portion *b* characterize the geometrical non-homogeneity.

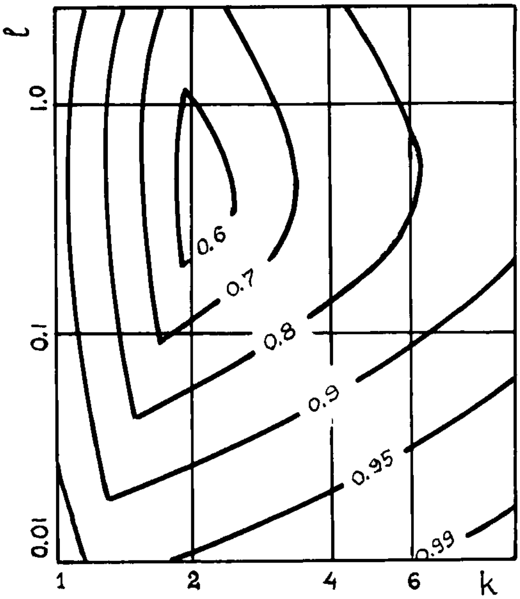

FIGURE 9 The relative reduction of distortional strain concentrations (comparing with no-layer case) versus relative layer thickness *l* and layer to binder moduli ratio *k*.

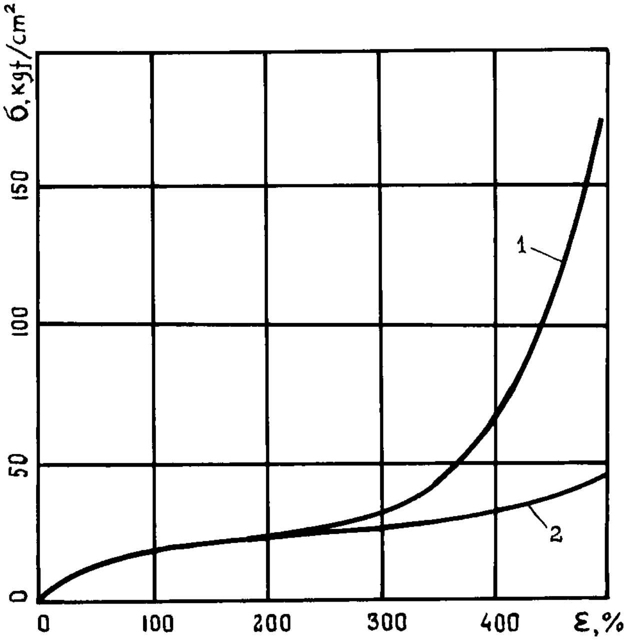

FIGURE 10 Typical stress-strain curves for the stiffening (1) and non-stiffening (2) elastomers.

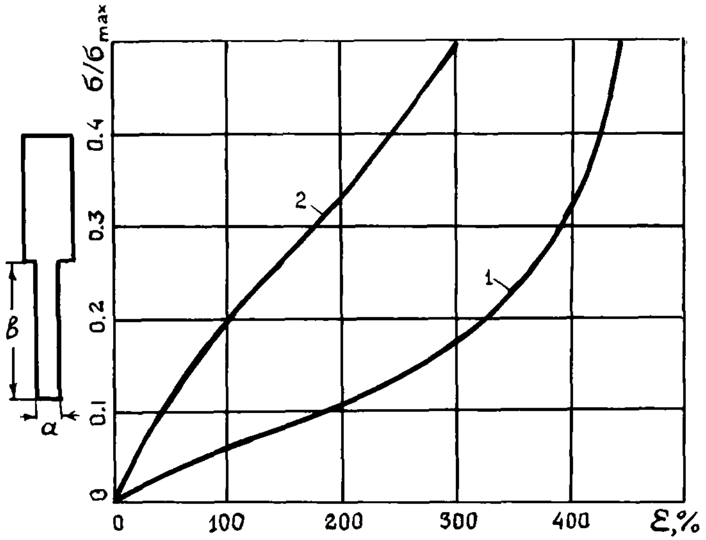

FIGURE 11 Specimen with non-uniform cross-section and the calculated stress-strain curves for the stiffening (1) and non-stiffening (2) elastomers.

Figure 11 shows the calculated stress-strain curves of this specimen for two elastomers, indicated in Figure 10 *a* and *b* both being equal to 0.5. It is seen that a stiffening elastomer preserves at about 90% ultimate strain, whereas a non-stiffening one only 60%.

Thus, the character of the binder stress-strain relation must control to a large degree the strength and strain capabilities of composites so that essential improvement by using strain-stiffening elastomers is to be expected.

## CONCLUSIONS

A theoretical analysis has been carried out aimed at investigation of the stress field in the binder of the highly loaded elastomeric composites. From the study the following conclusions may be drawn:

Two types of stress concentrations (hydrostatic and distortional) have been found in the binder after hydrostatic and distortional components of the stress field having been examined separately.

Quantitative confirmation is given that high loaded composites cannot be deformed significantly without producing local hydrostatic tensions in the binder phase high enough to initiate crazing, even under rather high superimposed pressure.

As far as good adhesion between the phases exists, stress field and failure conditions around the filler particles are not influenced considerably by the presence of the stiffened binder layers on the surface of the particles. These stiffened layers are believed to improve strength capabilities of composites only after separation of the binder from inclusion have taken place.

Characteristic shape of the binder stress-strain curve might deeply influence the macroscopic strength of the composite. Final steep rise on the curve results in a more uniform distribution of strain in the binder phase and, consequently, provides the increase in the strength and elongation of the composite.

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