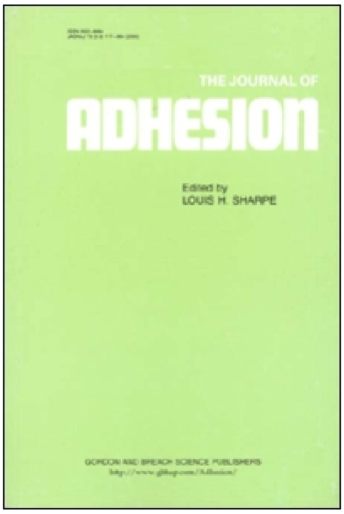

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Structure-Property Relations in Plane Particulate Polymeric Composites under Simple Extension and Compression

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An approach is offered for evaluation of effective stress-strain curves of particulate composites under plane strain conditions based on a solution of the boundary-value problem providing simultaneously the data on the corresponding structural stress and strain fields. The influence of randomness in the particle arrangements and the filler concentration on the effective behavior is established. It is demonstrated that the observed plateaus on the tensile and compressive curves are caused to a degree by the geometrical non-uniformity of the composite structures. The difference between tensile and compressive curves is examined and explained from the structural point of view.

Keywords: Polymeric particulate composites; matrix-filler debonding; plane boundary value problem

1. INTRODUCTION

The relationship between structural and effective characteristics of composite materials remains a permanent area of interest among material scientists. This problem applies in full measure to particulate composites, whose behavior, in some cases, is still obscure in spite a huge body of experimental work.

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It is well known that highly-filled polymeric matrix composites inevitably soften under deformation. This process is well evidenced in tensile testing, where the initial effective modulus of specimens always diminishes with extension, providing nonlinear stress-strain curves of various shapes depending on the structural features of the composite system tested. Experience demonstrates that this softening is provoked mostly (at least at the initial stages) by progressive debonding of the matrix from the filler surface [1–4].

A recent development of an external field method [5] for elastic plates, apparently, has opened a way for directly correlating structural changes in composites with their effective behavior. It allows an understanding of how some structural mechanisms influence macroscopic stress-strain relations. This paper represents the results of theoretical research in this domain.

2. THEORETICAL BACKGROUND

2.1. Problem Statements

The system to be analyzed represents an ensemble of circular, uniform, rigid inclusions embedded into an infinite elastic matrix plate. Initially, inclusions are assumed to be bonded to the matrix. When the matrix plate is extended or compressed under plane strain, local damage within the composite having the form of matrix detachments appears around some inclusions increasing in number with the matrix plate deformation until all of them detached from the matrix.

From the general considerations, it is clear that the resistance of an initially cohesive system characterized by the modulus, E_1, drops continuously in the course of deformation to some lower level representing a completely debonded system characterized by the modulus, E_2. A question arises of how the shape of the transition zone between the initial bonded state and the final debonded one is generated.

To simplify the problem, let us first assume that the debond strength is the same for all the inclusions and that we have an infinite regular system. In this case, the passage from E_1 to E_2 looks quite clear: the detachment of the matrix from all the inclusions occurs instantaneously, when the local critical condition is reached, due to uniformity of local interface stresses. On the stress-strain curves, this event must look like a vertical drop of the stress from some upper level characteristic of the bonded state of the system to the lower one characteristic of its completely debonded state.

Let us now reconsider the problem having assumed that the regular arrangement of inclusions is substituted for a random one, the condition of constant local inclusion-matrix interface strength being kept unaltered. The new statement of the problem immediately cancels the assumption of the uniformity of local critical stresses and leads to a conclusion that matrix separation cannot now be instantaneous. A more smooth transition from the initial maximum to the final minimum rigidity on the stress-strain curves is now to be expected. Obviously, its shape must depend somehow on the degree of disorder in the inclusion arrangement. How the degree of irregularity in the arrangement of inclusions influences the stress-strain curves is the first point to be examined in the paper.

The extent of the modulus drop caused by debonding must depend on the filler concentration. When the filler content is low, the matrix detachment from rarely-encountered filler particles cannot reduce appreciably the initial effective modulus of the material. On the contrary, at high filler concentration, the modulus drop becomes considerable, since the detached matrix turns into a kind of soft cellular structure containing filler particles inside opened pores. How the shape of this transition changes with the filler concentration is the second point to be examined in the paper.

It is well known that the behavior of composites under tension differs from that under compression due to the significant distinction in the magnitudes of local stresses at the same effective deformations. How this structural peculiarity is reflected on the macroscopic behavior of composites is the third point to be examined in the paper.

2.2. System Geometry and Loading Conditions

The object to be investigated represents a nearly-circular ensemble of identical rigid discs (filler particles) embedded into an infinite elastic matrix plate (Fig. 1). The arrangement of discs in the ensemble may be varied from regular (traingular grid) to completely- disordered. The

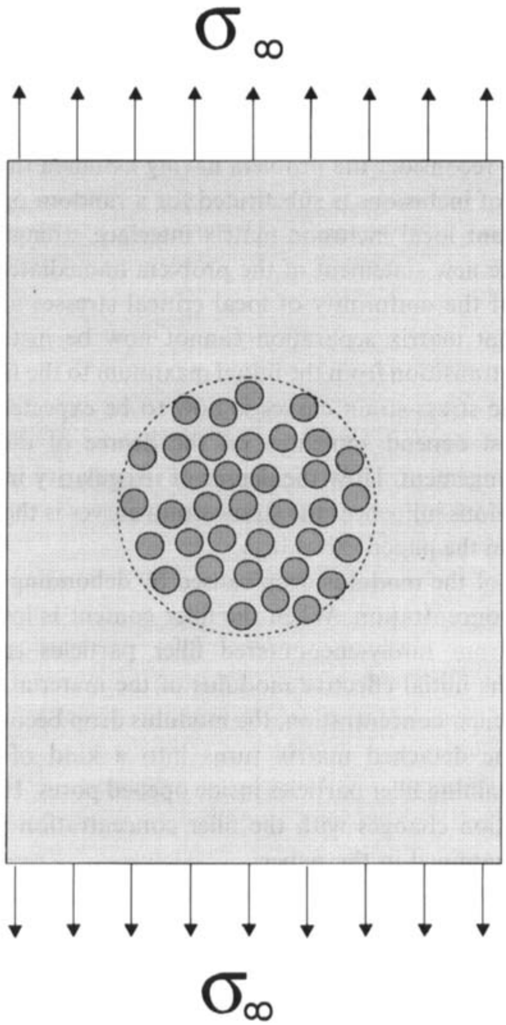

FIGURE 1 Scheme of the particulate ensemble under examination.

random structure later on referred to as completely disordered is generated by randomly placing discs into a previously imposed circular area. The filler concentration is specified by the number and radius of inclusions within the prescribed circular area. The matrix is loaded at infinity by tensile or compressive stress under plane strain conditions.

2.3. Mechanical Properties of Constituent Elements and Matrix Debonding Condition

Constituent elements are assumed to be elastic materials. The Young's modulus of the matrix, E_m, is taken to be 1 MPa and the Poisson's ratio 0.5. The Young's modulus of discs is taken to be 10^4 MPa with the same Poisson's ratio.

Debonding of inclusions is controlled by the stress state at the inclusion-matrix interface. The adhesive bond is to be higher than the cohesive strength of the matrix material. Hence, the matrix detachment from a disc, according to Gent [6], is assumed to originate from a microscopic cavitation inside the matrix material, when the hydrostatic tensile stress, s_o, somewhere along the contour of the inclusion, reaches the magnitude equal to the Young's modulus of the matrix. The detachment is assumed to be an instantaneous event provoked by cavitation. This condition of the detachment defines the ultimately attainable bond strength.

The modulus of the detached discs is taken to be nearly zero (10^{-4} MPa) and Poisson's ratio 0.25 to reflect the compressibility due to the vacuole appearance around the inclusion. Mochida and Tohgo [7] showed that the restraints due to the debonded particle for lateral contraction do not appreciably affect the overall properties of the composite, even if the site of the inclusion is regarded as a void after debonding [7, 8].

2.4. Calculation Procedure

From the above it follows that two distinct boundary value problems are to be solved. The first one is the calculation of microscopic stresses in the matrix inside the assembly, while the other is the calculation of effective (averaged) properties of the assembly.

2.4.1. Structural Stresses and Strains

Taking into account that the stress state of the matrix between the closely-spaced particles is strongly inhomogeneous, a complex variable approach developed by Muskhelishvili [9] seemed most appropriate for the quantitative evaluation of the high gradient stress field. The general solution was sought in the form of the Laurent series expansion. A special iterative procedure has been developed by Svistkov and Evlampieva [10] for calculating stress and strain distribution through summation of the disturbances from all the particles at each point of the matrix inside and outside of the assembly. A similar approach has been recently described in Ref. [11].

2.4.2. Effective Properties of the Assembly

The effective behavior, in contrast to the common approach, is calculated not by averaging stresses and strains over the assembly area, but by analyzing the external stress and strain field in the matrix around the circular assembly according the scheme developed by Evlampieva and Moshev [12]. The simple calculation allows one to evaluate effective Young's modulus, Poisson's ratio and the effective radius of the assembly.

2.4.3. Stress-Strain Behavior of the Assembly

Calculation of the stress-strain curves consists in the small incremental increases of the stress, σ_∞, at infinity. At each step, the boundary value problem is solved and the mean stresses, s_o, on the boundary of each inclusion are analyzed. For some time the ensemble with characteristic initial modulus, E_1, is deformed non-damaged. Then a critical situation ($s_o = E_m$ or $s_o > E_m$) around some inclusion occurs. This effect is regarded as structural damage requiring changing the mechanical characteristics of the bonded inclusion into those characteristic of a debonded one as is adopted above. The calculation cycle is repeated at the same level of σ_∞ until no new debond that might be provoked by the preceding ones comes about. Then, the current effective modulus of the damaged ensemble is calculated from the analysis of the stress state in the matrix around the ensemble and the effective strain and stress within the ensemble.

In the course of deformation, the number of the debonded inclusions increases until all of them become detached from the matrix. The effective modulus, having reached its lower limit, becomes stable again.

It was established earlier [5] that ensembles consisting of 37 inclusions, as is shown in Figure 1, are sufficiently representative for evaluating the effective behavior of composite systems. The following results have been obtained in testing ensembles of such a size.

3. RESULTS OF CALCULATION

3.1. Tensile Curves *Versus* Degree of Irregularity

The influence of the structural irregularity on the macroscopic behavior is illustrated in Figure 2 for a system having 0.4 volume fraction of particles. In this figure the dashed line represents a tensile relation of the hypothetical regular infinite structure. A curve for a regular hexagonal ensemble containing 37 inclusions (the number needed to complete this object) is characterized by a pronounced hump on a tensile curve, while

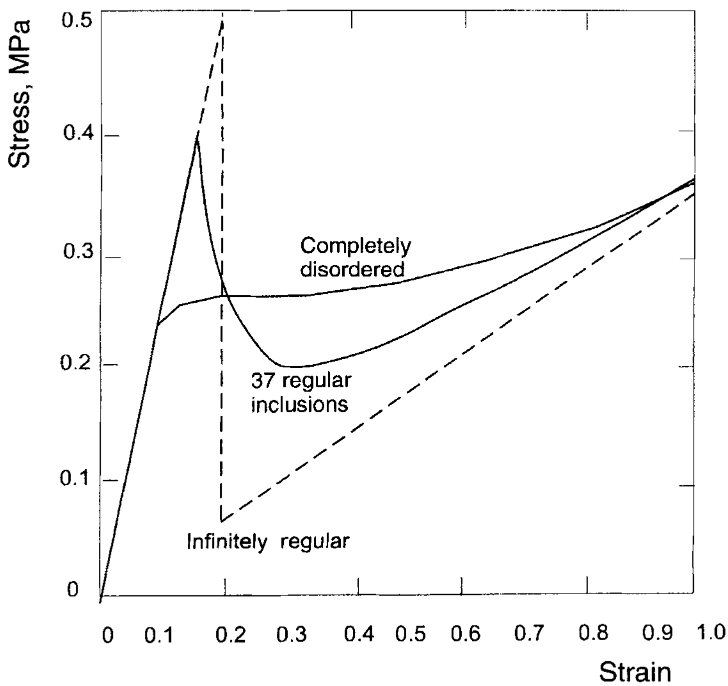

FIGURE 2 Tensile curves of ensembles with various regularities of the inclusion arrangement. Volume fraction of particles = 0.4.

completely-disordered ensembles provide wide plateau curves without humps extending from strains of about 10% to about 70%. Similar relations are typical for other filler volume concentrations above 0.3.

This demonstration reveals that the shape of the tensile curves is strongly influenced by the degree of irregularity in the arrangement of inclusions. Remembering that regular structures are hardly possible in real composite materials allows us to make a conclusion that the contribution of the local geometrical non-uniformity in plateau formation, among other favoring mechanisms, is considerable.

3.2. Tensile Curves *Versus* Filler Concentration

Increasing filler concentration leads to enhancing two counteracting effects: the first is the growth of the resistance due to the initial modulus increase, the second is the most pronounced softening from detached inclusions due to lower final modulus. For disordered ensembles, this is clearly exemplified by Figure 3, where a set of tensile curves for filler concentrations, φ, varying from 0.1 to 0.5 is depicted. The curves are means of five repeated calculations with different random inclusion arrangements. The results of the preceding section are well corroborated as regards plateau zone formation. At lower filler concentrations of about $0.1-0.2$, the transition zone is small, starts at higher stresses and slightly rises. At higher filler fractions of about $0.4-0.5$, the transition zones tend to flatten and are positioned at lower stresses as rather extended plateaus.

3.3. Tensile Curves under Extension and Compression

Figure 4, where tensile and compressive curves are depicted together, reveals that they are qualitatively similar. However, under compression structural damage originates at higher absolute stresses and strains with more extended transition zones. This result agrees qualitatively with known experimental data on the reinforcing action of pressurization [2, 3].

Structural explanation of this peculiarity is quite obvious. Let us examine a single disc in an infinite matrix under unit tensile stress (Fig. 5(a)). Elementary analysis shows that the maximum extensive mean stress provoking debond is located in point A and is equal $+1.5$,

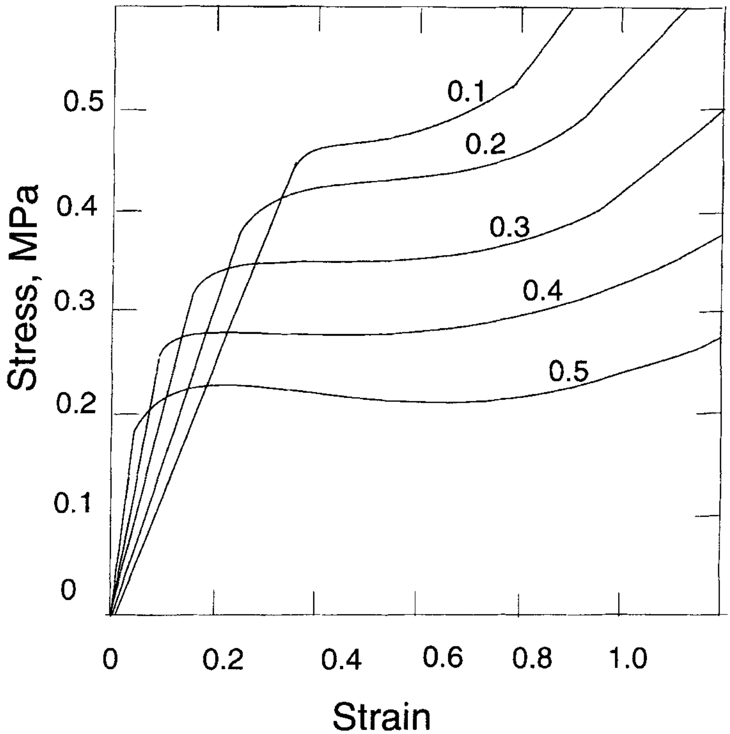

FIGURE 3 Tensile curves for various filler volume contents.

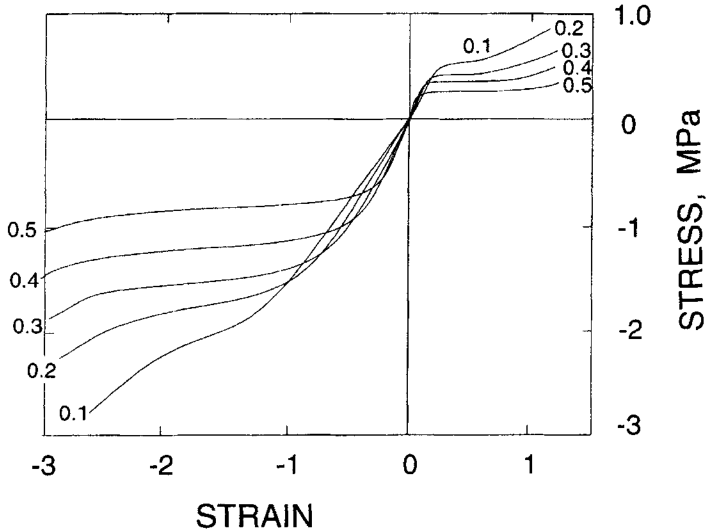

FIGURE 4 Tensile and compressive curves for various filler volume contents (indicated near the curves).

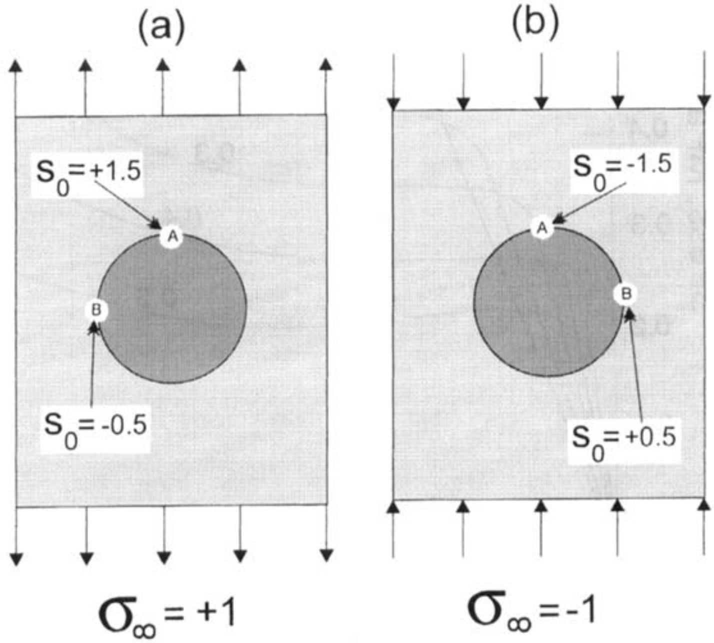

FIGURE 5 Features of the matrix stress state under extension and compression.

while in point B a compressive maximum mean stress is originated equal to -0.5. If a negative unit stress is applied at infinity (Fig. 5(b)), then an inverse contour situation occurs: the maximum extensive mean stress shifts to point B and is equal $+0.5$. In order to raise the mean stress at this locality to a critical level of $+1.5$, the external load must be raised by a factor of three. Indeed, approximately threefold increase in plateau stresses for compression curves is observed in Figure 5 for stochastic inclusion arrangements.

4. DISCUSSION

The calculations of structural stresses and strains have been performed in the framework of the theory of small deformations. Hence, the results of calculations going beyond the limits of the infinitesimal theory are to be regarded as semiquantitative. However, qualitatively, they look quite realistic.

It is demonstrated that the ever-existing geometrical non-uniformity of the structure alone is able to explain plateau zone appearance on the stress-strain curves of elastic, highly-filled particulate composites. Humps on the stress-strain curves can also have the same origin for systems with high particulate content, where close mutual arrangement of inclusions makes their geometry more regular.

If a plateau stress, σ_p, is regarded as a certain critical characteristic of a composite material, then its dependence on the filler concentration, represented through a dimensional ratio $\psi = \sigma_p/E_m$, may become a useful guide in the development of new composites. This characteristic, different for extension and compression, is depicted in Figure 6.

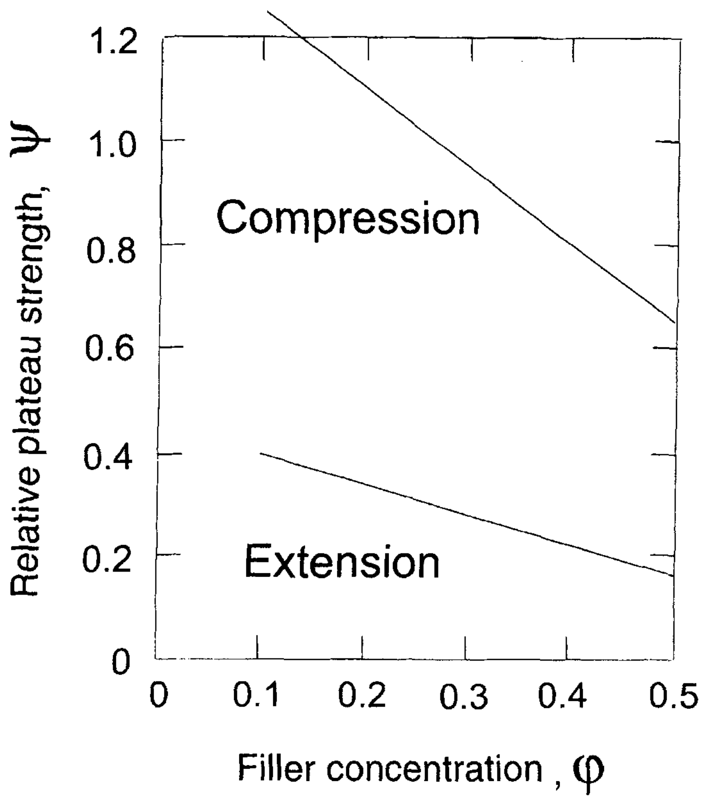

FIGURE 6 Relative plateau strength *versus* filler volume content.

It must be recalled that the results discussed above have been obtained under assumption that the adhesive strength is higher than the cohesive strength of the matrix. Therefore, the ψ-value should be considered as a maximum attainable strength characteristic for a given filler concentration.

It may be supposed that, in reality, some filler particles in composite materials are initially debonded or weakly bonded. In such cases, the decrease of the initial modulus will start at somewhat lower stresses than those indicated in Figure 6.

5. CONCLUSIONS

Tensile and compressive curves of elastic particulate composites under plane strain are investigated through an external stress field estimation.

It is demonstrated that the random character of filler particle arrangement alone (among other plateau formative mechanisms) provides rather extended plateau zones.

The effect of filler concentration on the shape of tensile and compressive curves is established. The ultimate plateau stress dependence on the filler content and the matrix modulus is established.

A microstructural explanation of the higher compressive strength as compared with the tensile one is presented.

Though the above results cannot be regarded as strictly quantitative, they give new information that might be useful for researchers in the materials science field.

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