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# HIERARCHICAL COMPOSITES: Analysis of damage evolution based on fiber bundle model

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**Abstract:** A computational model of multiscale composites is developed on the basis of the fiber bundle model with the hierarchical load sharing rule, and employed to study the effect of the microstructures of hierarchical composites on their damage resistance. Two types of hierarchical materials were considered: “hierarchical tree” (bundles-of-bundles of fibers) and self-similar particle and fiber reinforced composite (in which reinforcements at each scale level represents composites in turn consisting of lower level reinforcements and matrix). For the case of the hierarchical tree (“bundle-of-bundles” material), it was observed that the increase in the amount of hierarchy levels leads to the lower strength of material. In the self-similar fiber reinforced matrix materials, as differed from the hierarchical trees, the damage resistance of the hierarchical materials increases with increasing the amount of hierarchy levels. The effect of mixed fiber and particle reinforcement on the damage resistance of the hierarchical composites is investigated as well.

**Keywords:** C: Modeling; B: Strength; C: Probabilistic methods; C: Stress transfer; C: Damage mechanics

## 1. Introduction

In recent years, the interest to the hierarchical, multiscaled composites and to the methods of their modeling has increased. This is related to several factors. First, the investigations of microstructures-properties relationships of natural materials (wood, bones, etc.) suggested that these materials represent hierarchical composites with fibrous reinforcement, and that their extraordinary properties (high strength, fracture toughness, etc.) are determined to a large degree by the hierarchical architectures of the materials [1-4]. Further, the reserves of the optimization of composite properties by varying their structures at the microscale level, first of all, volume content and properties of reinforcement are approaching their limits. While some properties (e.g., stiffness) are improved by increasing the volume content of hard reinforcement in composites, other properties (fracture toughness) degrade in this case. The idea to create a new family of materials, by tailoring material properties using the “simultaneous control of different structural elements, such as shape and size, at plural scale levels” has been studied in the framework of the Japanese “Synergy Ceramics Projects” [5]. Kanzaki et al. [5] presented an example of the improved material which has both high strength and toughness achieved by combination of aligned anisotropic grains (at microlevel) with the intragranular dispersion of nanoparticles (at nanolevel). Another example of a material with a hierarchical microstructure, and excellent properties (extremely high compressive yield strength) is a “trimodal” Al-composite developed by Ye et al. [5]. In this composite, coarse-grained Al is introduced into the nanocrystalline Al reinforced with B<sub>4</sub>C particles in order to achieve both improved strength and acceptable ductility.

The multiscale composite design allowed to improve different mechanical properties of different groups of composites: 80% improvement of fracture toughness in a carbon fiber reinforced epoxy composites achieved as a result of carbon nanotubes (CNTs) engineered matrix [8], drastic improvements in elastic modulus, compressive strength and interlaminar strength of carbon fiber/polymer composites caused by dispersed carbon nanofibers [9], 85% increased fracture toughness of carbon fiber reinforced epoxy/clay nanocomposites CFRENCs with the introduction of 4 phr nanoclay in epoxy [7], 38% increased flexural strength due to small amount of nanoclay (2 phr) added into the epoxy of carbon/ epoxy composites [7]. Thus, tailoring of material properties at different scale level makes it possible to improve qualitatively mechanical properties of composites, including their competing properties.

In order to to analyze the effect of the microstructures of hierarchical materials on the strength and mechanical properties, a number of mathematical models of the materials have been developed. Carpinteri and Paggi [10] developed a model of hierarchical, fractal grained composite, in which mesograins are recursively composed of smaller and smaller micrograins. Using top down approach, rule of the mixture and generalized Hall-Petch relationship (for hardness), and the authors analyzed the effect of the multiple hierarchy levels on the material hardness and toughness. They demonstrated that “a hierarchical material is tougher than its conventional counterpart”, and that the material hardness increases with increasing the amount of hierarchy levels. Joshi and Ramesh [10] developed a micromechanical model of particle reinforced, multiscale composites, “where at least one phase is itself a composite at a finer scale”. They used the multiscale secant Mori-Tanaka method and added subscale terms, in particular, grain size, particle size and dispersoid strengthening, and computed overall response of the material. Yao and Gao [12] developed self-similar models of hierarchical materials, one corresponding to gecko attachment system and one to the microstructure of bone. They demonstrated that “a hierarchical material with multi-scale cohesive law can be designed from bottom up to achieve flaw insensitivity”. A series of analytical models of hierarchical materials, based on fracture mechanics approach, has been presented by Gao [2]. Using the “fractal bone” model (multiple level self-similar composite structure), Gao demonstrated that a hierarchical material with different properties at different length scales “can be designed to tolerate crack-like flaws”.

While the general conclusion of these works is that the hierarchical materials have higher strength and damage resistance than common composites, this conclusion is in contrast with the numerical results from [24, 25], and with experimental and numerical observations showing that clustering and bundling of reinforcement in composites leads to the lower damage resistance [Chapter 8 from 4, 28, 29]. (It should be noted that the term “clustering” is used here in more materials science sense, as the close arrangement, grouping of reinforcing elements, and should not be mixed with, lets say, the clustering of failed fibers in the fiber bundle models).

Another group of models are based on the fiber bundle model and the renormalization methods, and seek to analyze the asymptotic strength distributions of the fiber bundles [13-17], taking into account the effects of time dependent behavior of fibers and different load sharing rules. A more detailed overview of various fiber bundle models can be found in the works by Newman, Phoenix, Hansen, Herrmann, Bazant and colleagues. Some

references can be found in [4, Chapter 10].

In this paper, we seek to analyze which aspects of the hierarchical microstructures of composites ensure higher damage resistance of the material. In order to solve this problem, a program code for the damage analysis based on the fiber bundle model with the hierarchical load sharing rule is developed. This code is applied for the analysis of the effect of the microstructure, amount of hierarchy levels, type of reinforcement, degree of clustering of elements on the damage resistance of the hierarchical composite. Due to the space limitation, we do not discuss strength distributions, or the time dependent fiber behavior in this paper.

## 2. Computational Model: Multiscale Fiber Bundle Model with Hierarchical Load Sharing

In this section, we develop numerical tools for the computational analysis of the microstructure-damage resistance relationships of hierarchical materials. A program code for the simulation of damage evolution in materials was developed on FORTRAN, on the basis of multiscale fiber bundle model and hierarchical load transfer rule.

Let us consider a multiscale fiber bundle model (see Figure 1). The system, built as a hierarchical tree, is subject to a tensile mechanical loading (Figure 2). In the following, we will use the following designations:  $N$  – total amount of fibers on the lowest hierarchy level (actual fibers),  $M$  – amount of hierarchy levels,  $z$  – branching number (i.e., the amount of lower level bundles in an upper level bundle, or the amount of fibers in the  $2^{\text{nd}}$  level bundle).

In the hierarchical load sharing rule (HLS), the load is transferred from the upper elements of the hierarchical “tree” (“roots”, corresponding to the “bundles-of-bundles-of-bundles-of-fibers in a 3 level material) to the lower (“branches”) and down to the lowest elements of the material (fibers, in this case). The load is shared equally amount all the sub-elements of a given higher level element (as long as they are intact) or to remaining intact sub-elements after some of them fail. In the easiest version, the amount of sub-elements in each higher level element (hereafter called branching number or simply branching) is supposed to be constant at all the levels. More complex versions when the branching number is varied at different scale levels or in different phases was implemented as well and is discussed in the section 3.3.

At the lowest level (which consist not of bundles, but of actual fibers), the elements are assigned the strength, using to the Weibull probability law random number generator. The fibers are taken as glass fibers in our model (in some special case, we model also carbon fibers, epoxy matrix or glass particle/matrix elements). The properties of the glass fiber are: Young modulus  $E_P=72$  GPa, and Poisson’s ratio 0.26. The failure strength of glass fibers follows the Weibull probability law, with parameters  $\sigma_0=1649$  MPa and  $m=3.09$  [19, 20, 21].

The failure condition of fibers is checked for each fiber, in the increasing order from the lowest strength/stress ratios to the highest ones. If the strength of a given fiber is less than the applied load, the fiber fails and the load is redistributed on the remaining fibers belonging to *the same bundle/branch*. After all the fibers in the branch fail, the higher level element is considered as failed, and the load is distributed among all the remaining elements belonging to the same higher level branch (“bundle of bundles”), and so on.

The program<sup>1</sup> calculates the damage (fraction of failed lowest level fibers) versus the applied load curves. Special cases included into the program code are mixed fibers (carbon and glass fibers, randomly mixed), particle reinforced composites (implemented on the basis of the model described in the section 5.1), mixed fiber and particle reinforcement. For the case of fiber, matrix and particle reinforced composites as well as mixed carbon/glass fibers, the program output includes the damage parameters separately for matrix, carbon fibers and glass fibers, particles as well as overall damage.

As a parameter of strength/damage resistance of the composites, we used the “critical load”, i.e. an applied stress at which the damage parameter (fraction of failed elements, either in the composite, or in separate phases) exceeds 90%. This value was determined from the damage-stress curves. Since the damage versus strain curves are monotonous, the choice of the critical damage level does not influence the qualitative relationships between arrangement of fibers and the damage resistance of the composite. The 90% level was chosen as a almost full failure value.

It is of interest that the “hierarchical distribution” of deformation and damage (i.e., when some subregions of the material deform as a whole and the total deformation of the material is controlled by interplay of these subregions or clusters of the subregions in a self-similar manner) has been reported not only for classical hierarchical materials, but also common “homogeneous” materials [22]. In the framework of physical mesomechanics of solids [22], Panin and colleagues described several “structural levels of deformation”, related to different rotational modes and the self-organization of local translational and rotational local modes of deformation. At each structural level, the deformation is controlled by the interplay of rotation and shear of lower level regions.

### **3. Numerical experiments: Damage evolution in “hierarchical trees” (bundle of bundles systems)**

In this paper, we analyze two groups of hierarchical materials: “hierarchical tree” (“bundle-of-bundles” system) and multiple level self-similar composites. A self-similar composite (similar to “fractal bone” model by Gao [2]) consists of both strong and weak elements at each level. The strong elements at the different levels are self-similar: they consist of some amount of stronger lower level elements and weak lower level element (matrix). Physically, the “bundle of bundles” system can be represented as clustered (or clustered at several scale levels) fiber reinforced composites with weak matrix. The multiscale self similar composites correspond physically to the case when reinforcing elements in the composite are replaced by the cluster/bundle of lower level elements (or again clusters at even lower scale level). An example of such material is the tool steel with “double dispersion structure” and extraordinary properties, developed by Berns et al. [22]. Thus, these two groups of hierarchical materials represent physically different microstructures.

In this section, we consider a “hierarchical tree” (bundle of bundles system), in which the load is shared only between fibers (the matrix is assumed to be too weak, and do not bear any loading).

<sup>1</sup> The program is available from the author on request free of charge

### 3.1. *Effect of the amount of hierarchical levels on damage in hierarchical tree system*

Let us consider the influence of the amount of the hierarchy levels on the damage resistance of the composite consisting of bundles (or bundles of bundles) of fibers. The total amount of the glass fibers at the lowest level of hierarchy is kept constant. We consider the following cases: (a) total amount of glass fibers  $N=15625$ , branching number (i.e., the amount of lower level bundles in an upper level bundle, or the amount of fibers in the 2<sup>nd</sup> level bundle)  $z=5$ , and the number of hierarchy levels  $M$  varied from 1 to 6, (b)  $N=16807(=7^5)$ ,  $z=7$  and  $M=1\dots5$ , and (c)  $N=1000$ ,  $z=10$  and  $M=1\dots3$ . In this case, we keep  $N$  (the amount of lowest level fibers/actual fibers) constant, but just vary the amount of hierarchy levels, thus, seeking to analyze the effect of amount of hierarchy levels on the damage resistance.

Figure 3 shows the damage (fraction of failed fibers in lowest level) versus applied stress for these cases. One can see that the damage resistance of the material decreases with increasing the amount of hierarchy levels. The data on Figure 3 were averaged over 5 realizations.

Figure 4 shows the critical force plotted versus the amount of hierarchy levels, for the case  $N=15625$  and  $z=5$ .

The conclusion that the increase in the amount of hierarchy levels leads to the lower strength of material corresponds to the results of Gomez and Pacheco [24] and Newman and Gabrielov [25]. Further, this conclusion corresponds also to the observations of the effect of clustering of reinforcing elements on the damage resistance of composites: clustering of reinforcement (which is the main mechanisms of realization of the hierarchical load transfer) has a negative effect on the damage resistance of composites [4, 28, 29]. However, it is in contrast with the observations that many natural materials, built as hierarchical fibrous composites, have extra-ordinary high strength.

Apparently, with increasing the hierarchy levels  $M$  the variability of the strengths increases as well, and that influences the strength [17, 18].

### 3.2. *Effect of the mixed fibers on damage in hierarchical tree system*

Let us consider the effect of mixed fibers on the damage resistance of hierarchical tree system. We seek to explore what will be the effect of hierarchy if we mix the relatively weak (glass) fibers with strong carbon fibers. The tensile strength of carbon fibers follows the Weibull distribution as well, with the parameters  $\sigma_0=2322$  MPa and  $m=4.4$  [26, 27]. The Young modulus is 276 GPa. The total volume content of fibers is 30%.  $N=16807$ .  $M$  was varied from 1 to 4,  $z=7$ .

Figure 5 shows the ratio of critical stresses (at which the damage reaches 0.9) for the model materials with different carbon/glass mixes (5% carbon fibers+25% glass, 15%/15%, 25% carbon+5% glass) to the critical stress for the material with 100% carbon fibers plotted versus the number of hierarchy levels.

As expected, the addition of stronger fibers leads to the lower damage at all the stages of damage evolution. It is of interest that effect of the addition of weaker glass fibers (leading to the reducing of the critical load) is higher for the hierarchical composites, than for non-hierarchical composites. Thus, the hierarchical tree microstructures lead to the higher sensitivity to the heterogeneity of the material.

### 3.3 *Branching and its variations at different hierarchy levels*

Let us consider the effect of the branching number (i.e., the amount of lower level elements in each higher level element) on the damage resistance of the material. The simulations were carried for the  $N(M)$  (total number of fibers at the lowest level) 16384 ( $=4 \cdot 8^4$ ),  $M$  (amount of hierarchy levels) = 4, branching was constant at all the levels and varied from 1 to 16 for different cases (1, 2, 4, 8, 16). Figure 6 shows the critical stress (i.e., the stress at which the damage in the composite exceeds 0.9) as a function of the branching number in the model.

One can see that the critical stress decreases when going from the case of the composite without clustering to the cases with clustering ( $z > 2$ ). Again, we can see that the increasing the degree of fiber clustering (amount of elements per cluster) leads to the lower damage resistance. This was observed also in analytical and computational (finite element) studies presented in [4, 28]: the more elements are in a cluster, the higher the likelihood of reinforcement failure. Above, we considered hierarchical systems with constant branching over several scale levels. It means that upper bundle at  $i$ -th hierarchy level contained constant amount of lower bundles at  $i-1$ -th level, for  $i=1 \dots M$ . Now, let us study the effect of varied branching at the different scale levels.

Figure 7 shows a schema of hierarchical bundle systems with varied branching: the cases of “few roots, many branches”, and “many roots, few branches”.

The systems with 4096 lowest level fibers (all glass fibers), 4 hierarchy levels and varied branching from 8 to 2, from 2 to 8 and constant (8) constant, decreasing and increasing branching numbers, and  $N(M)=4000$ , (the variation is from 5 via 10 to 20) were considered. Figure 8 shows some typical curves of damage parameter (fraction of failed lowest level fibers) plotted versus applied stress, for the system with varied branching. The amount of elements (bundles) on each hierarchical level for the considered cases is shown on the Figure 8.

Here, we denote the microstructure “C” as those with lower level, close to fibers branches, and less higher level, close to roots branches. The microstructure “B” is that with small amount of lower level, close to fibers branches, but with bigger amount of higher level, close to roots branches. In the simulations, it could be observed that while the microstructure “C” gives sometimes more and sometimes less damage resistance, than the constant branching number case, the microstructure “B” gives always higher damage resistance. Apparently, the maximum branching at the lowest level (and the minimum amount of elements at the lowest+1 level) allows localizing the damage propagation and slowing down the damage growth. Varied branching (over several scale levels), however, has a positive effect on the damage resistance of the composite.

## 4. Numerical experiments: Damage evolution in glass fiber/epoxy matrix hierarchical system

In this section, we consider a case of a self-similar composite, consisting of reinforcement (fiber, particles or mixed) and matrix. As differed from the case of the “hierarchical tree”, considered above, the self-similar composite consists of both strong and weak elements at each level. The strong elements at the different levels are self-similar: they consist of some amount of stronger lower level elements and weak lower level element (matrix).

As noted above, the “bundle of bundles” system corresponds to clustered (or clustered at several scale levels) fiber reinforced composites with weak matrix, while the self-similar composites correspond physically to the case when reinforcing elements in the composite are replaced by the clusters of lower level elements (or again clusters at even lower scale level), of somewhat bigger size and with correspondingly lower stiffness.

#### 4.1. Hierarchical self-similar fiber/matrix microstructures

In this part, we consider the case of hierarchical self-similar fiber/matrix composites. The microstructure consists of elements which are either pure matrix or reinforcement at each level. The reinforcing elements, in turn, consist of pure matrix and the lower level reinforcing elements. Figure 9 shows a schema of the hierarchical self-similar fiber/matrix composites. If the volume content of the reinforcing elements at each level is constant and designated as  $vc$ , the global volume content of lowest level fibers in the material is given by

$$vc_{\text{glob}} = vc^M$$

where  $M$  – the amount of hierarchy levels. Thus, if we define the total volume content of glass in the composite, the volume content of stronger phase at each scale level is calculated as an  $M$ -degree root from this number. It should be noted here that while it looks like when the number of hierarchy levels  $M$  becomes large enough, the global volume fraction of the strong elements will approach almost zero, it is not realistic situation: hierarchical material with  $M$  larger than 5 is a theoretical idealization.

If we determine the Young modulus of the material at each level using the rule of mixture, and, again, assuming that volume content is constant at all the levels, we have the Young module at the  $j$ -th level as:

$$E = vc^j E_f + (1 - vc) E_M \left( 1 + \sum_{i=1, j-1} vc^i \right)$$

where  $E_f$ ,  $E_m$  are the Young moduli of (lowest level) fibers and pure matrix respectively.

The load distribution between the fiber (or strong elements at the given hierarchy level, which represent composites, in turn consisting of fibers and matrix) and matrix take place in the following way. At each hierarchy level, the load is distributed among all the elements, including the fibers and the matrix. The strain on the fibers and matrix in each element is constant, while stresses are different. Thus, we determine the stress on each element (either matrix or stronger reinforcement) by the following formula:

$$\sigma = \frac{\text{Force}}{\text{Area}} \frac{E_{\text{phase1}}}{E_{\text{phase1}}vc_1 + E_{\text{phase2}}vc_2}$$

where 1, 2 – two phases,  $E$  and  $vc$  – Young modules and volume contents of the corresponding phases.

While the load distribution between matrix and fibers follows the displacement/strain controlled boundary condition, the load distribution between fibers (main bearers of load) is the same as in the case of “dry” hierarchical tree - “global” load distribution inside each cluster at the cluster level.

The properties of the epoxy matrix are: Young modulus 3790 GPa, failure stress  $s_{\text{matr}}=88$  MPa [30]. The properties of fibers are the same as in the previous section. Only “full failure” of the matrix or “full failure” of a fiber are possible. If an element (fiber or matrix) inside a bundle fails, the load is redistributed only to other elements inside the bundle.

The properties of lowest level fibers were kept constant, at the level, corresponding to the properties of glass fibers in polymer/epoxy glass composites (the experimentally determined properties, see subsection ). The global volume fraction was, again, kept constant in all the compared cases.

Let us look at the damage behavior of a non-hierarchical system. Figure 10 shows the fiber, matrix and overall damage plotted versus stress for the non-hierarchical material ( $M=z=1$ ). It can be seen that the fiber cracking begins rather early and follows smooth curve, while the matrix cracking starts much later and goes very quickly. For comparison, it is of interest to look at the damage-strain curves obtained with the use of 3D finite-element micromechanical models [20] (see Figure 10b). One can see that the more exact finite element simulations lead to the qualitatively similar results: early and slow fiber damage accumulation, and late and quick damage growth in the matrix.

Now, we consider the effect of hierarchization of the self-similar materials on the damage resistance. Figure 11 shows the critical stress (at which the damage exceeds 0.9) plotted versus the amount of hierarchy levels for the total damage, and separately for fibers and matrix, for glass (a) and carbon (b) fiber reinforced composites.

The important observation is that the damage resistance of the multiscale self-similar fiber reinforced composites increases with increasing the amount of hierarchy levels in the material. This is clearly different from the case of “hierarchical tree” considered in the Section 3. While the strength of hierarchical structures, built via grouping/clustering/bundling of strong elements, reduces with increased clustering/hierarchization, the strength of fractal, self-similar composites increases with increasing the amount of hierarchical levels.

Apparently, the reason for such a strong difference is the availability of matrix. With increasing the amount of hierarchy levels, Young modules of the reinforcing elements at higher levels become lower, and, thus, bigger part of the load is born by the matrix (since the load is distributed proportionally to the stiffness of a given element). This effect, combined with the localized damage evolution (the damage in one bundle begins to be felt in other bundles only after this first bundle fails completely) leads to the situation when increasing the number of hierarchy levels increases the damage resistance. Similar conclusions about the positive effect of the amount of the hierarchy levels on the damage resistance of multiple level self-similar composite structure (“fractal bone”) model has been obtained by Gao [2].

#### 4.2. Hierarchical self-similar fiber reinforced composites with mixed fibers

In this part, we consider the case of self-similar fiber reinforced composites with mixed (weak/glass and strong/carbon) fibers. The properties of fibers are given in sections 2 and 3.2.

The simulations for the self-similar fiber reinforced composites with different amounts of hierarchy levels, different fractions of carbon and glass fibers (20/80, 80/20, 50/50, 0/100, 100/0 of totally 30% reinforcement volume content) and 16807 lowest level elements were carried out.  $z=7$ .

Figure 12 shows the critical stress (at which the damage exceeds 0.9) plotted versus the amount of hierarchy levels for the cases of 100% glass reinforcement, 100% carbon reinforcement, 50/50% carbon/glass reinforcement and 80/20% carbon glass reinforcement.

Looking at the relative numbers (ratio of the critical stress for a material with a given  $M$  to that with  $M=1$ ) we can see that the positive effect of the material hierarchization is observed most clearly in the material with strong or mainly strong reinforcement (pure carbon or 80% carbon). For pure carbon, the ratio of the critical stress for a material with  $M=4$  to that with  $M=1$  is more than 2, while for pure glass reinforcement, it is 1.4.

Figure 13 shows the ratio of the critical stress for different mixed reinforcement materials divided by that for 10% carbon and plotted versus the fraction of carbon fibers in the reinforcement. It is of interest that even relatively small addition of carbon fibers (20%) in a non-hierarchical composite leads to the drastic increase in the material strength, even making it close to maximal one (at 100% carbon fibers). For hierarchical composites, the strength is increasing roughly proportional to the volume content of strong carbon fibers.

Further, we sought to investigate the effect of pre-damage on the strength of the composite [31]. In the following simulations, we assumed that 20% of all lowest level fibers are failed before we started the loading. We assume the glass fibers, 16807 lowest level fibers,  $z=7$ . The fixed fraction (20%) of pre-damaged fibers was randomly chosen. Figure 14 shows the ratio of critical stresses for composites with 20% pre-damaged lowest level element to that of composites with all intact elements, plotted versus the amount of hierarchy levels. One can see that the composites with amount of hierarchy levels more than 2 have almost the same strength independently whether the composite is pre-damaged or not. Thus, the effect of pre-damage is important in the case of non-hierarchical material, but becomes almost negligible for the hierarchical material. This corresponds to the results by Gao and colleagues who observed that hierarchical materials are insensitive to damage in some cases [2].

## 5. Modeling hierarchical particle reinforced and mixed fiber/particle reinforced composites

In this section, we develop a model of particle reinforced continuum to be introduced into the hierarchical fiber bundle model, and use this approach to analyze the effect of hierarchization of particle reinforced composites and combining fiber and particle reinforcement on the damage evolution in the material.

### 5.1. “Embedded equivalent fiber” model of particle reinforced composites

Here, we consider the case of self-similar particle/matrix composites. There are a lot of very efficient analytical and computational models of strength of particle reinforced composites (see the reviews in [4, Chapter 4]). However, in order to remain in the framework of the assumptions of fiber bundle model (and not to compare apples and oranges), we opted to adapt/generalize the fiber bundle model to the case of the particle reinforced composites.

Figure 15 shows the schema of the model: from an intricate, real shape of a particle embedded into a cubic matrix, to an equivalent model with a vertical fiber with quadratic section and equivalent properties, embedded into a series of fibers with the pure matrix properties.

In this model, the particle is represented as a cube embedded into the polymer matrix. This system (glass cube in a polymer cube or a parallelepiped) is in turn divided into vertical elements, one of them containing the particle and the others, consisting of pure matrix material (following the logics of the Voigt/Reuss model of composites presented in [32]). The cubic particle sandwiched between two parallelepipeds with matrix properties is represented as an element of the hierarchical system (“equivalent fiber”) which properties are averaged out (following the Reuss rule). The Young

modulus of the “equivalent fiber” containing the particle lie between the those of fiber and matrix materials, and is calculated by the Reuss formula [4], as  $\left[vc_{particle,linear} / E_{particle} + (1 - vc_{particle,linear}) / E_{matrix}\right]^{-1}$ . Here  $vc_{particle,linear}$  (“vertical volume content”) means the height of the cube representing the particle, divided by the total height of the element. The vertical volume content of the particle material in this element is calculated as

$$vc_{particle,linear} \approx (vc_{particle})^{1/3}$$

The “equivalent fibers” are embedded into the matrix. The volume content of the vertical elements containing the particles as a cubic layer (“equivalent fibers”) in all the material is calculated as  $(vc_{particle,linear})^2 \approx (vc_{particle})^{2/3}$ .

The load is transferred as discussed in the subsection 4.1, with the “equivalent” fiber subject to the stress proportional to its Young modulus, and the global load distribution inside each cluster. This model of the particle reinforced composite was implemented into the program code of the section 2, and the computational studies of the damage behavior of the self-similar, hierarchical, particle reinforced systems (similar to the system studied in the previous section, but with particle reinforcement) were carried out.

## 5.2. Hierarchical particle reinforced composites

Here, we consider the effect of the hierarchization (replacement of the reinforcing particles by clusters or even multilevel clusters of even smaller particles) on the damage resistance of the material. The simulations were carried out for the case of 16807 particles) lowest level element), the global volume content of the glass particle 30%.

Figure 16 shows the critical stress (at which the damage exceeds 0.9 either in each phase or totally in the material) plotted versus the amount of the levels in the material. One can see that the particle reinforced composites are much more sensitive to the clustering effect than the fiber reinforced composites: when the system goes over from non-hierarchical ( $M=1$ ) to hierarchical structure, its damage resistance reduces considerably. Still, for  $M>1$ , the damage resistance increases with increasing the amount of hierarchy levels.

It is of interest that the damage resistance of the matrix monotonically increases with increasing the amount of the hierarchy levels, beginning from  $M=1$  (non-hierarchical material). Since the damage resistance of the reinforcement is most relevant for the stiffness, and that of the matrix for the toughness and lifetime of the composite, we can conclude that increasing the amount of the hierarchy levels might lead to the improvement of the fracture toughness and lifetime of the composite, while the stiffness (after the begin of the damage evolution) might suffer.

Let us compare these results with the observations of Ye et al. [5] on the tri-modal Al-based composite, and the results of the modeling by Joshi and Ramesh [10]. We did not observe the monotonous increase in the strength when comparing non-hierarchical, bimodal and trimodal composites. Apparently, the effects observed in [5, 10] are caused not only by the hierarchization of the material, but also by the small scale effects and different properties of low level and high level reinforcements. However, the hierarchization of a material as such do contribute to the improvement of damage resistance of the composite as long as the stronger elements bear most of the load (as fibers do), or when we compare hierarchical materials with low amount of hierarchical levels with a material with higher amount of hierarchical levels.

Comparing the non-hierarchical system ( $M=1$ ) of fiber and particle reinforcement, one can observe that the matrix in particle reinforced composites (PRC) fails at much lower load than that in the fiber reinforced composites (900 MPa versus 1300 MPa). The reason for this is that the main load in PRC is not born by fibers, but rather by matrix, and, thus, the load at the matrix in PRC is much higher than in FRC. The particle failure (again, taking 90% damage in all the reinforcement) takes place at somewhat higher load than the fibers failure (2000 MPa versus 1800 MPa), again, due to the higher load born by fibers. When we go over to the hierarchical systems (i.e., clustering the particles), the damage resistance of the particles goes down significantly. This observation corresponds to the results from [28, 29] and Chapter 8 of [4]: while the clustering of particles leads to the 3...6 times higher likelihood of particle failure, the fiber clustering causes much more limited increase of the likelihood of fiber failure, of the order of 30% (compare [27]). That is why the transition from non-hierarchical to hierarchical composites leads to much stronger negative effect for the particle, than for the fiber reinforced composites.

We can conclude that the homogeneous distribution of reinforcing particles (no clustering,  $M=1$ ) ensures the highest damage resistance. However, if some particle clustering is unavoidable (as is the case in real world), more hierarchy means more damage resistance.

### 5.3. Composites with mixed reinforcement (fibers + particles)

Now, let us consider the composite with mixed reinforcement: fibers and particles (Figure 18a). Figure 17 shows the damage in fibers, particles, matrix and overall composite plotted versus stress for the non-hierarchical self-similar particle reinforced material ( $M=z=1$ ). The volume contents of the particles and fibers are both 15% (totally 30% reinforcement content). The reinforcement (both fibers and particles) have the properties and strength of glass. One can see that the damage in particles develops much more slowly than in fibers. While 40% fibers fail at around 1100 MPa, 40% of particles fail at 1400 MPa (27% higher).

Let us consider now the self-similar structure consisting of matrix reinforced by both fibers and particles, with equal volume content. The total volume content of reinforcement is still 30%.

Figure 18 shows the critical stress (at which the damage exceeds 0.9) plotted versus the amount of the hierarchy levels in the material. One can see, again, that the damage in particles, and to a lesser degree, in fibers increases when the material become hierarchical, but decreases with the increasing the amount of the hierarchy levels. Again, damage resistance of the matrix monotonically increases with increasing the amount of the hierarchy levels, beginning from the transition from non-hierarchical ( $M=1$ ) to the hierarchical ( $M>1$ ) micro.

Figure 19 shows the critical stress (at which the damage exceeds 0.9) plotted versus versus the fraction of particles in the total (30%) reinforcement (fiber+particles). It is of interest that the dependency of the strength of composite with mixed fiber/particle reinforcement of the fraction of particle reinforcement is very different for hierarchical and non-hierarchical materials.

For non-hierarchical composites, the strength of the composite with mixed reinforcement is higher than the strength of purely fiber or purely particle reinforced composites. The dependency of the strength of the composite on the fraction of particles has the shape of  $\Pi$  curve. For hierarchical composites with mixed reinforcements, the strength decreases monotonically with increasing the fraction of the particles and reducing the fraction of fibers. One should notice that we consider here the particles and fibers having the same radius, and made of the same material (glass).

It is of interest to look how the addition of particles to the matrix influences the strength and damage resistance of the composite. In this case, we keep the volume content of fibers constant, at the level of 30%.  $N=16807$ ,  $z=7$ ,  $M=3$ . The volume content of particles in the matrix increases, from 0% (pure, non-reinforced matrix) to 70% (thus, making the total volume content of reinforcement  $30\%+70\%*(100-30)=79\%$ ). Figure 20 shows the critical force (at which 90% of fibers fail) plotted versus the volume content of particles in the matrix. One can see that the higher the volume content of particles in the matrix, the higher the damage resistance of fibers. Apparently, the stronger and stiffer matrix bears bigger load and thus slows down the damage evolution in the fibers.

## 5. Conclusions

The computational model of multiscale composites is developed on the basis of the fiber bundle model with hierarchical load sharing rule, and used to study the effect of the microstructures of hierarchical composites on their damage resistance. Two types of hierarchical materials were considered: “hierarchical tree” (bundles-of-bundles of fibers) and self-similar composite (in which each reinforcement at each scale level represents a composite consisting in turn of reinforcements and matrix). For the case of the hierarchical tree (“bundle-of-bundles” material), it was observed that the increase in the amount of hierarchy levels leads to the lower strength of material.

In the self-similar fiber reinforced matrix materials, as differed from hierarchical trees, the damage resistance increases with increasing the amount of hierarchy levels. Apparently, the strength of hierarchical structures, built via grouping/clustering/bundling of strong elements, reduces with increased clustering/hierarchization, while the strength of fractal, self-similar composites increases with increasing the amount of hierarchical levels.

The particle reinforced composites are much more sensitive to the clustering effect than the fiber reinforced composites: when the system goes over from non-hierarchical ( $M=1$ ) to hierarchical structure, its damage resistance reduces considerably. Still, for  $M>1$ , the damage resistance increases with increasing the amount of hierarchy levels. One can conclude that the homogeneous distribution of reinforcing particles (no clustering,  $M=1$ ) ensures the highest damage resistance. However, if some particle clustering is unavoidable (as is the case in real world), more hierarchy means more damage resistance. The damage resistance of the matrix monotonically increases with increasing the amount of the hierarchy levels, beginning from  $M=1$ .

Assuming that the damage resistance of the reinforcement is most relevant for the stiffness, and that of the matrix for the toughness and lifetime of the composite, one can speculate that increasing the amount of the hierarchy levels (in self-similar materials) might lead to the improvement of the fracture toughness and lifetime of the composite, while the stiffness (after the begin of the damage evolution) might suffer. The strength of the composite with mixed reinforcement is higher than the strength of purely fiber or purely particle reinforced composites for non-hierarchical composites. For hierarchical composites with mixed reinforcements, the strength decreases monotonically with increasing the fraction of the particles and reducing the fraction of fibers.

It should be noted that the properties of reinforcing elements depend on their sizes (“small is strong”), and the truly correct model of nanoreinforced materials should take this factor into account. However, at this stage, we sought to analyze only the effect of structure/ architecture/ hierarchization of the material, separately from other effects, and left out the important size effects.

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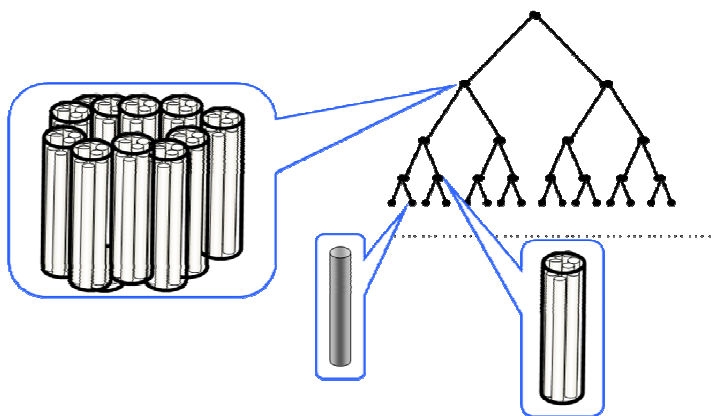


Figure 21. Schema: Multiscale (5 level) fiber bundle model. For the sake of easier presentation, the branching number in the hierarchical tree shown is two (and not eight as the depicted amount of fibers in the cluster)

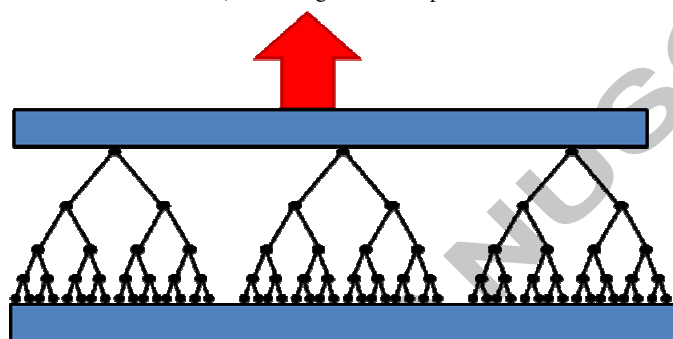


Figure 22. Hierarchical tree subject to the tensile loading. The branching number is 2, and the schema contains 5 levels of hierarchy

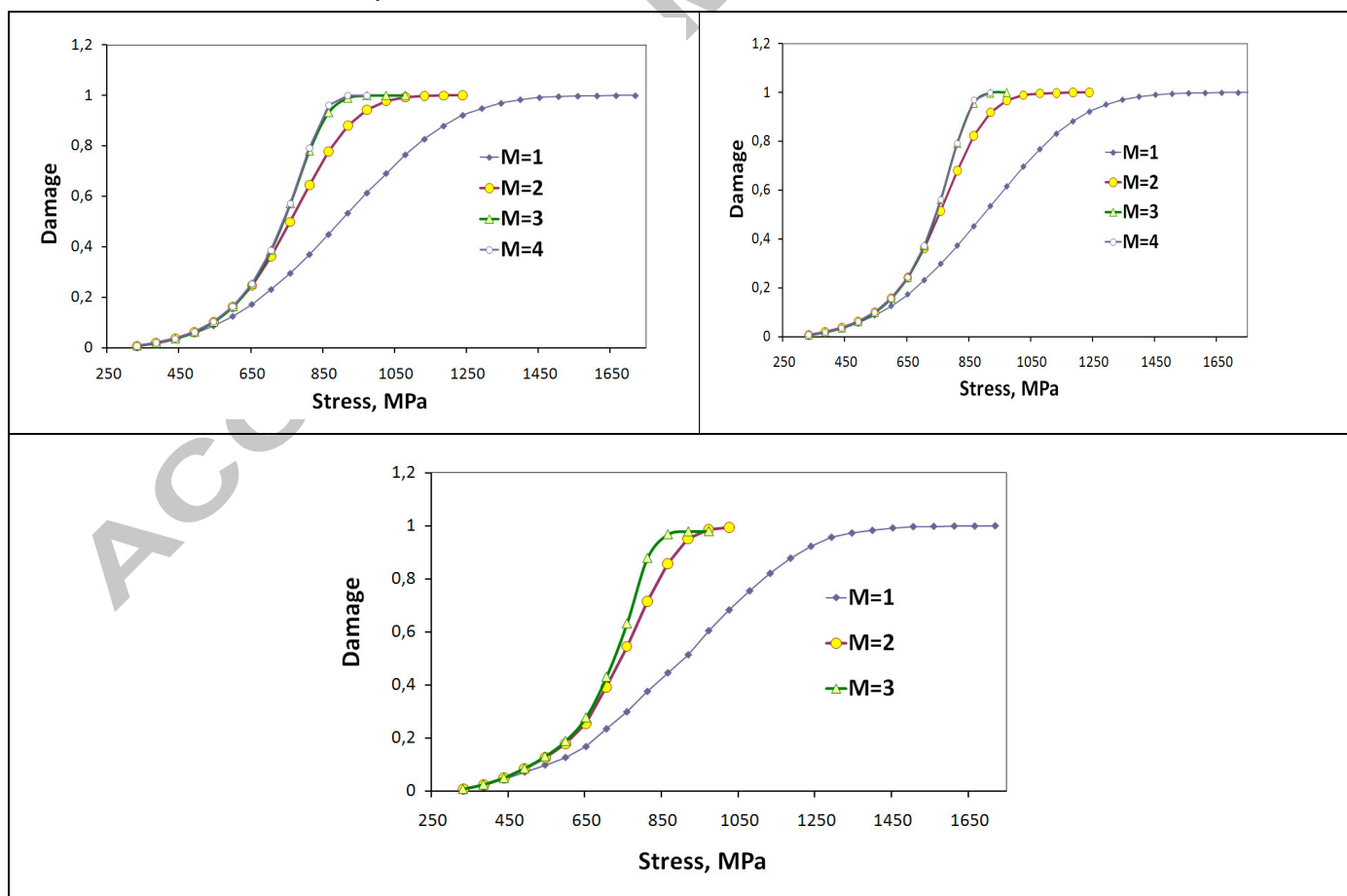


Figure 23. Damage (fraction of failed fibers) versus applied stress, for different branching numbers and hierarchy levels numbers. (a) Amount of lowest level fibers  $N=15625 (=5^6)$ , branching number  $z=5$ , and amount of hierarchy levels  $M$  is varied from 1 to 4, (b)  $N=16807 (=7^4)$ ,  $z=7$  and  $M=1 \dots 4$ , and (c)  $N=1000$ ,  $z=10$  and  $M=1 \dots 3$ .  $N$  (the amount of lowest level fibers/actual fibers) is kept constant in each case

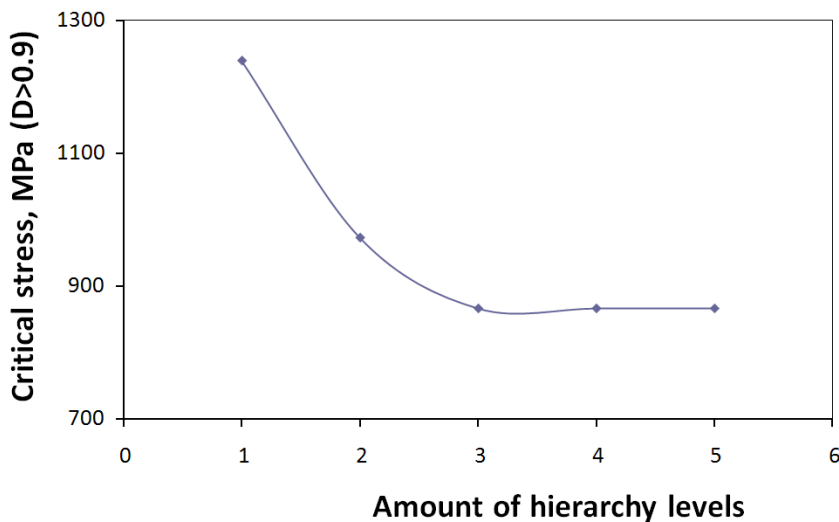


Figure 24. Critical force plotted versus the amount of hierarchy levels, for  $N=15625$ ,  $z=5$ ,  $M$  varied from 1 to 6

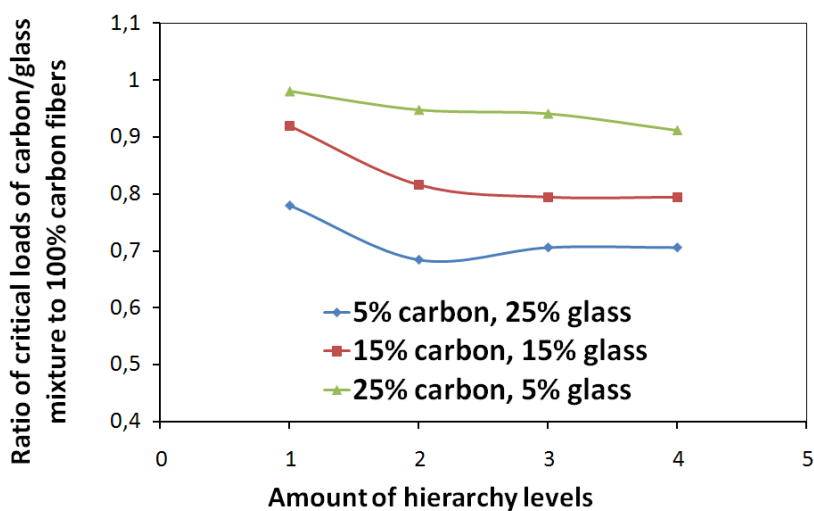


Figure 25. Ratio of damage for carbon/glass mix to 100% carbon fibers plotted versus the number of hierarchy levels.  $N=16807$ .  $M$  was varied from 1 to 4,  $z=7$ .

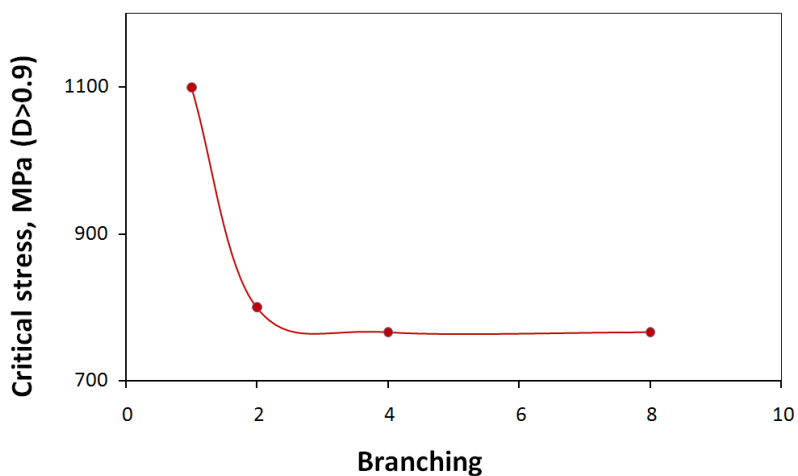


Figure 26. Effect of branching number on the damage evolution ( $M=4$ ).  $N=4096$ .  $Z=1\dots 8$

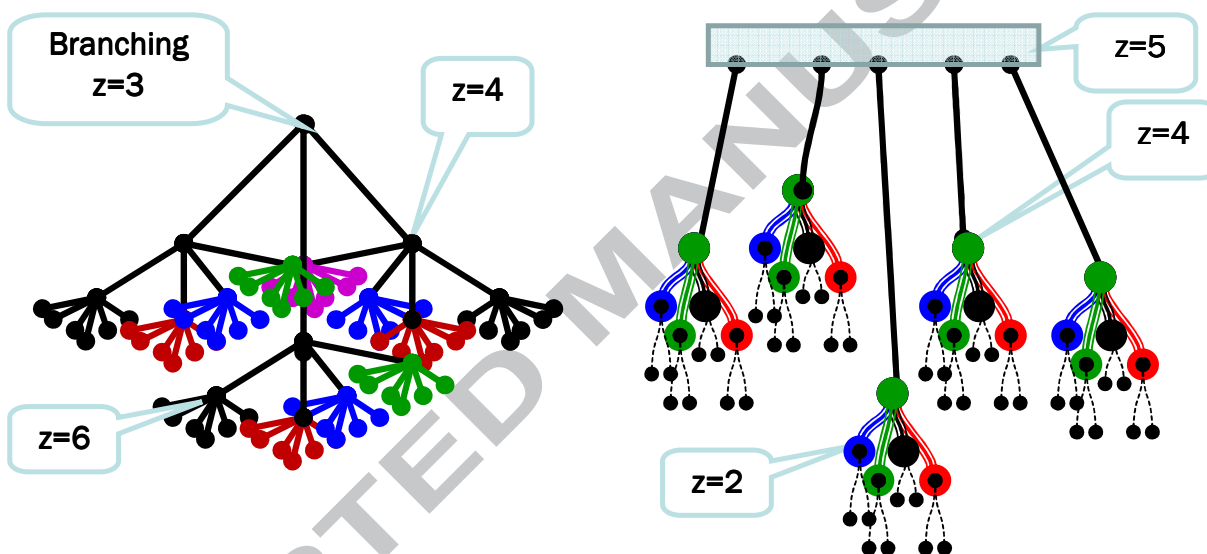
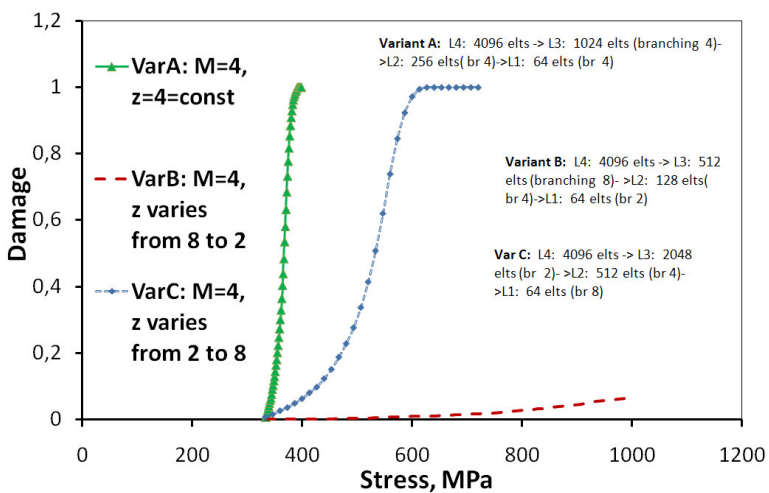


Figure 27. Schema: Hierarchical bundle system with varied branching: left, branching is highest at the lower level of hierarchy (few roots, many branches), and right, branching is highest at the upper level of hierarchy (many roots, few branches).



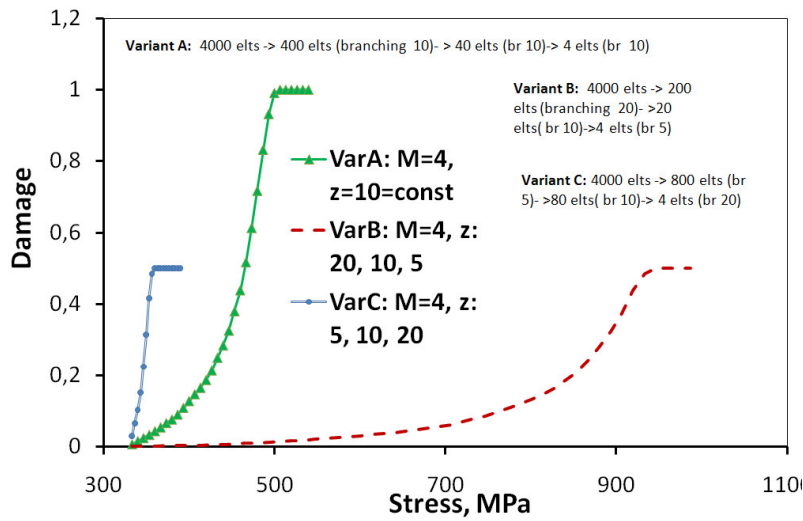
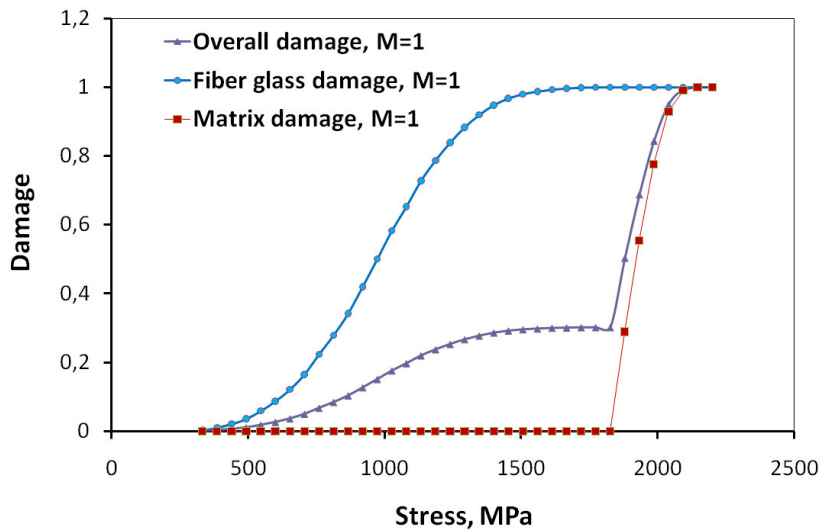


Figure 28. Damage versus applied stress: constant, decreasing and increasing branching numbers considered,  $M=4$ . (a) The average branching  $z=4$  (the variation is from 2 via 4 to 8) (b) The average branching 10, the variation is from 5 via 10 to 20



(a)

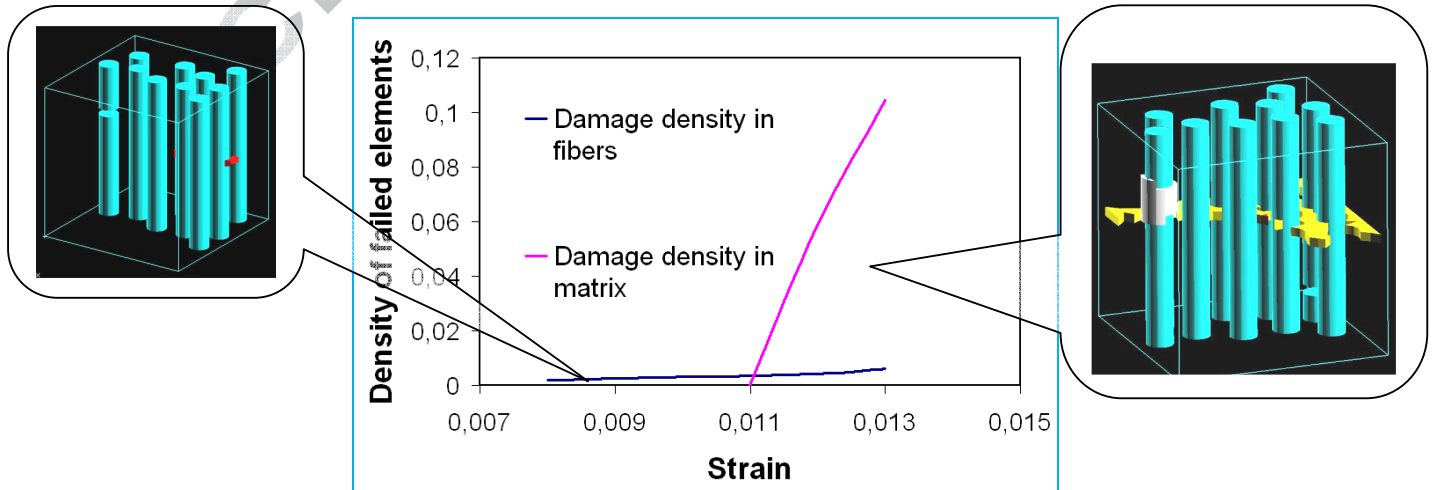


Figure 29. Damage evolution: overall damage, fibers and matrix.  $M=z=1$  (non-hierarchical material) (a). Right (b), for

comparison: finite element results from [14] (damage-strain curves and damaged elements distribution)

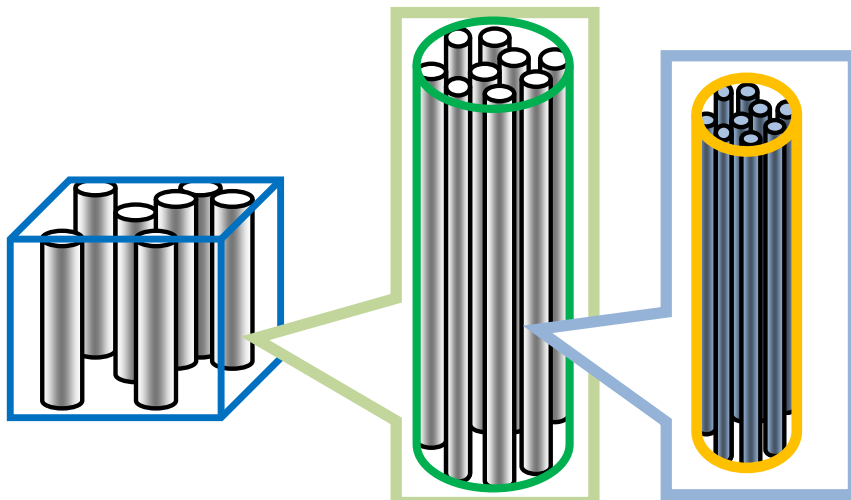


Figure 30. Schema: Hierarchical self-similar fiber/matrix composites

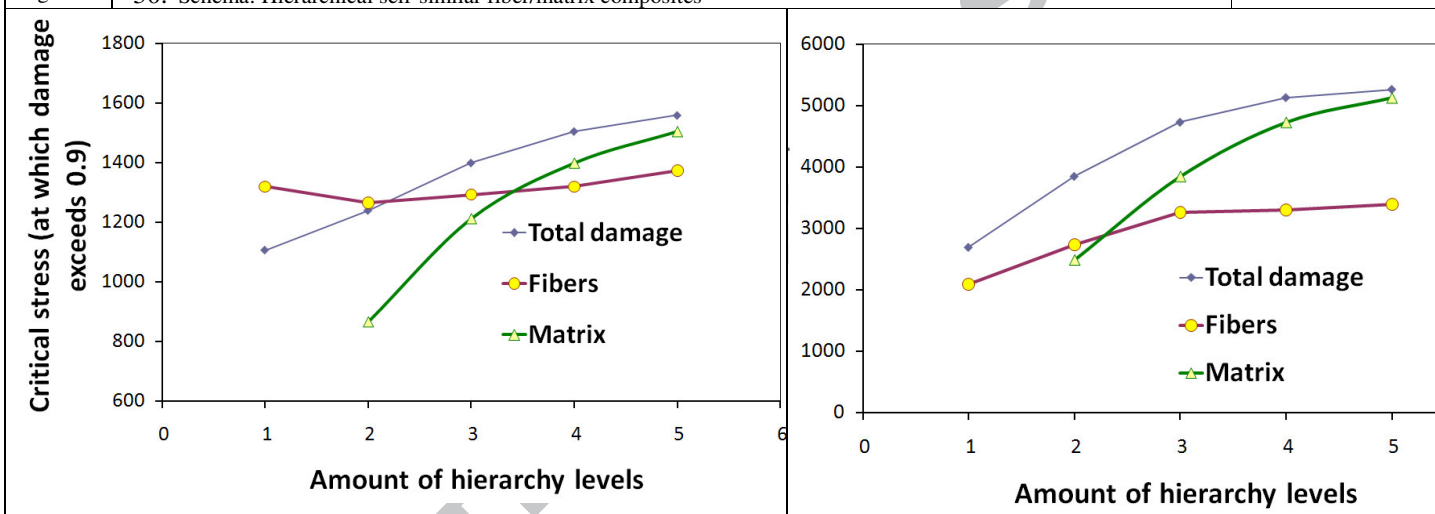


Figure 31. Critical stress (at which the damage parameter in a given phase reaches 0.9) plotted versus the amount of hierarchy levels in a system (for self-similar fiber reinforced composites) (a) Glass fiber composites, (b) Carbon fiber composites.  $N=16807$ ,  $z=7$ ,  $M=1\dots 5$

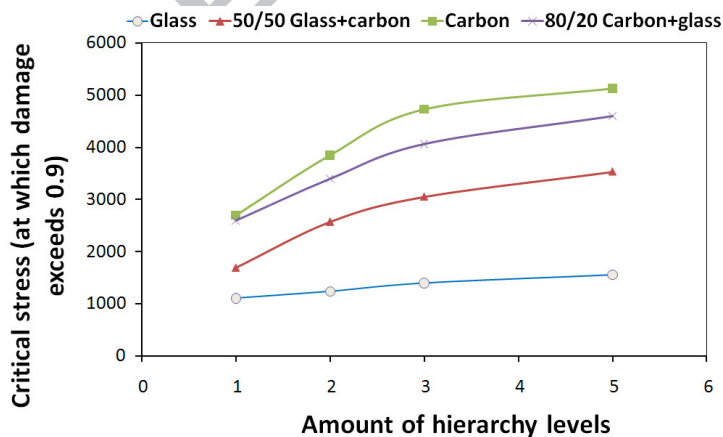


Figure 32. Critical load for hierarchical composites reinforced by mixed fibers (total volume content of fibers 30%, of them 0/100, 20/80, 50/50, 80/20 and 100/1 percents of carbon/glass fibers) of these composites plotted versus the amount of hierarchy levels.  $N=16807$ ,  $z=7$ ,  $M=1\dots 5$

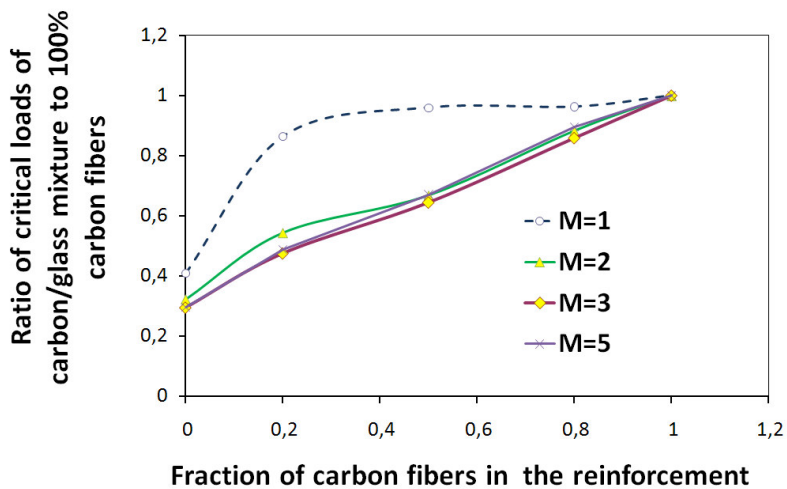


Figure 33. Ratio of critical stresses for non-hierarchical ( $M=1$ ) and hierarchical ( $M=2$  and  $3$ ) composites with mixed (carbon+glass) reinforcement divided by the critical stress for pure carbon composites plotted versus the fraction of the carbon fibers in the reinforcement.  $N=16807$ ,  $z=7$ ,  $M=1\dots5$

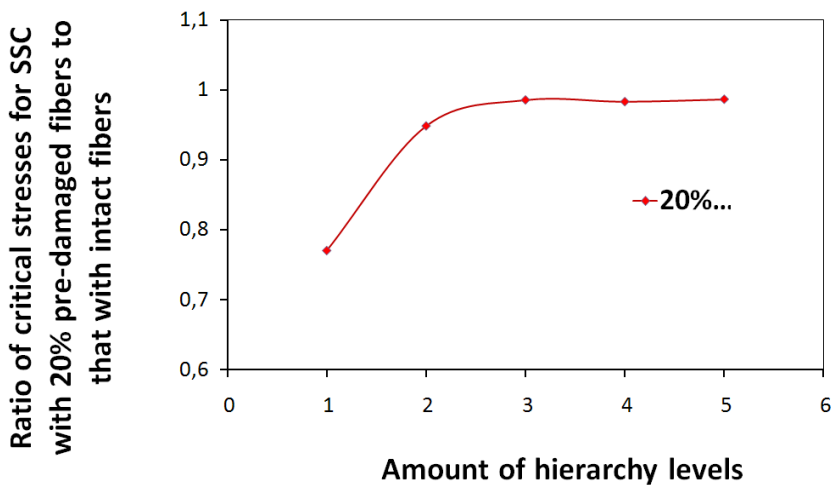


Figure 34. Ratio of critical stresses for composites with 20% pre-damaged lowest level element to that with all intact elements, plotted versus the amount of hierarchy levels.  $N=16807$ ,  $z=7$ ,  $M=1\dots5$

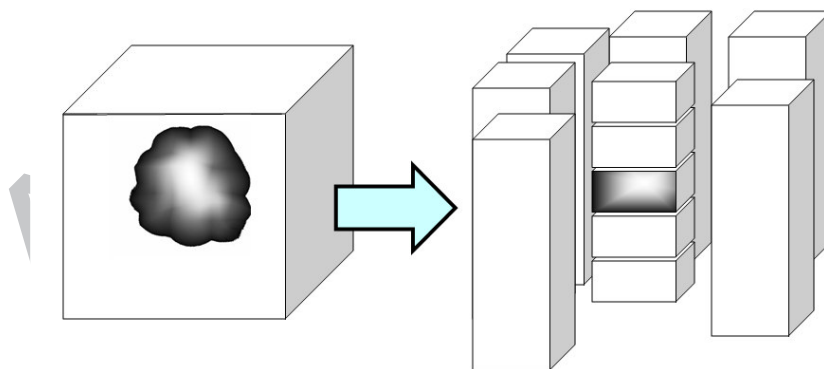


Figure 35. Schema: modeling of the particle reinforced composites

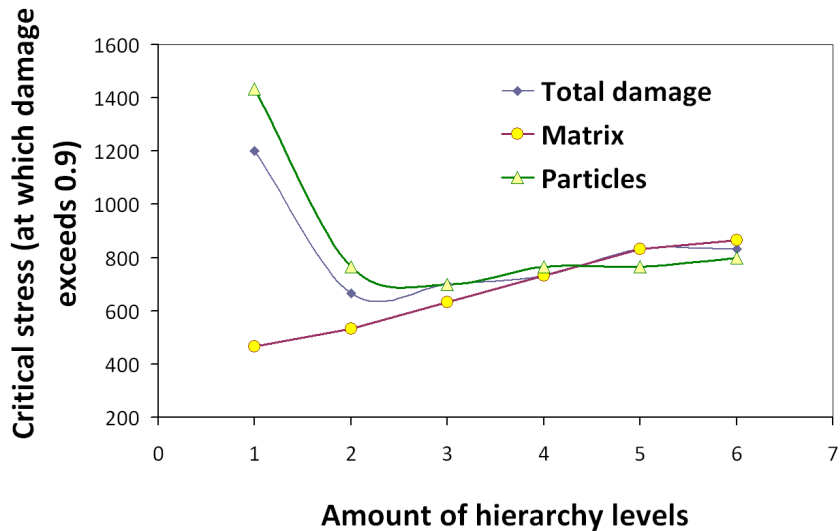


Figure 36. Critical stress plotted versus the amount of hierarchy levels in a system (for self-similar particle reinforced composites).  $N=16807$ ,  $z=7$ ,  $M=1\dots 5$

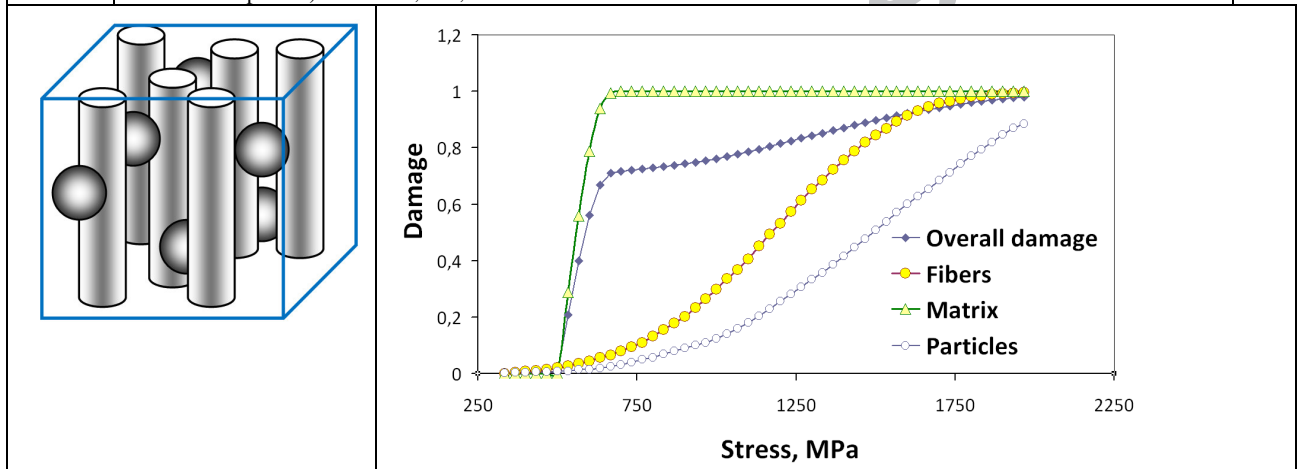


Figure 37. Schema of the non-hierarchical material reinforced by 15% glass fibers and 15% glass particles (a) and the damage growth curves for matrix, particles and fibers for the material (b).  $N=16807$ ,  $z=7$ ,  $M=1$ .

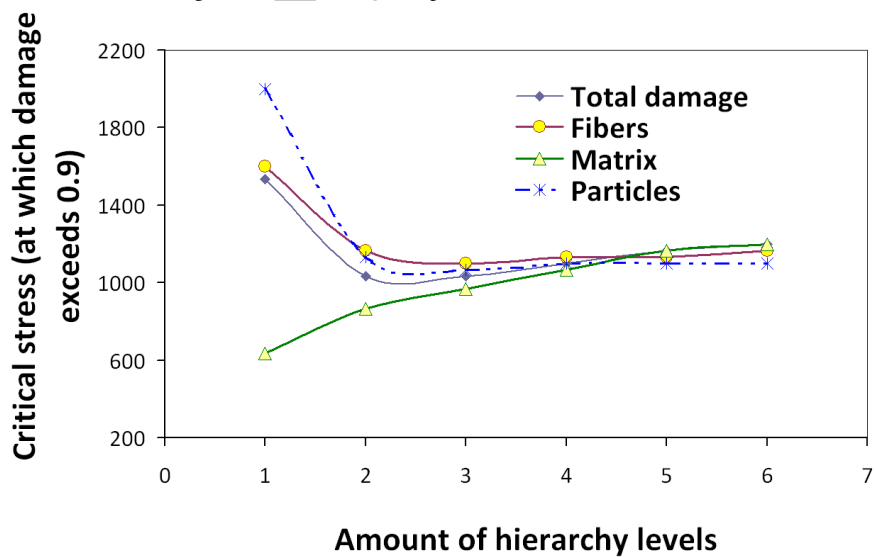


Figure 38. Critical stress plotted versus the amount of hierarchy levels in a system (for self-similar fiber + particle reinforced composites).  $N=16807$ ,  $z=7$ ,  $M=1\dots 5$

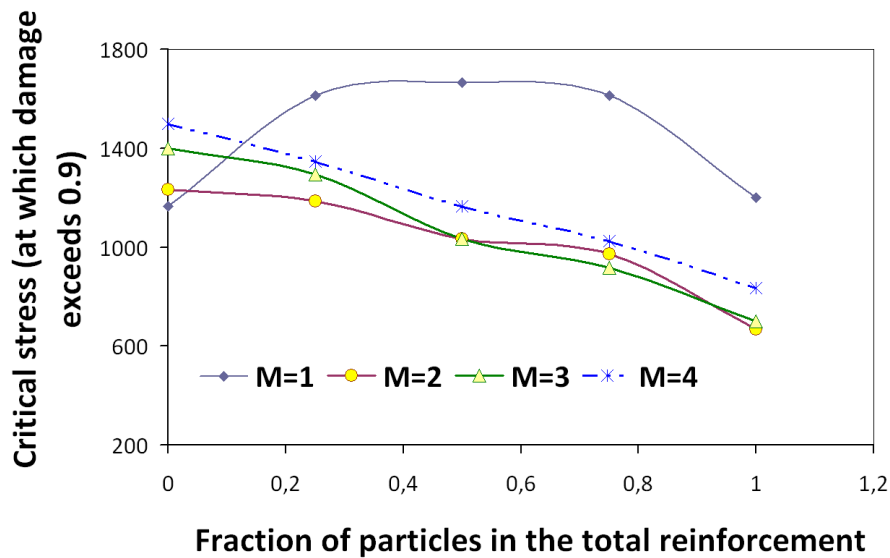


Figure 39. Critical stress plotted versus the fraction of particles in the total (30%) reinforcement (fiber+particles),  $N=16807$ ,  $z=7$ ,  $N=16807$ ,  $z=7$ ,  $M=1...5$

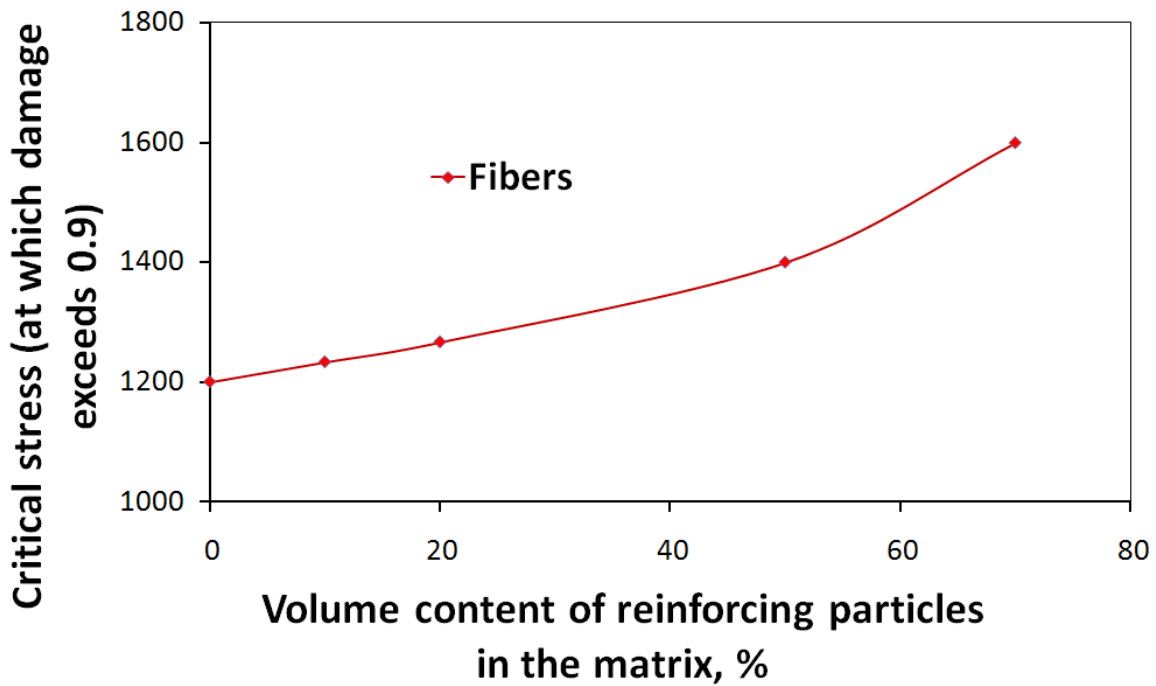


Figure 40. Critical force of the fiber failure in fiber reinforced composite plotted versus the volume content of particles in the matrix.  $N=16807$ ,  $z=7$ ,  $M=3$