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Investigating the role of hierarchy on the strength of composite materials: evidence of a crucial synergy between hierarchy and material mixing.

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Abstract

Natural materials are often organized in complex hierarchical architectures to optimize mechanical properties. Artificial bio-inspired materials, however, have thus far failed to successfully mimic how these architectures improve material characteristics, for example strength. Here, a method is proposed for evaluating the role of hierarchy on structural strength. To do this, we consider different hierarchical architectures of fiber bundles through analytical multiscale calculations based on a fiber bundle model at each hierarchical level. In general, we find that an increase in the number of hierarchy levels leads to a decrease in the strength of material. However, when a composite bundle with two different types of fibers is considered, an improvement in the mean strength is obtained for some specific hierarchical architectures, indicating that both hierarchy and material "mixing" are necessary ingredients to obtain improved mechanical properties. Results are promising for the improvement and "tuning" of the strength of bio-inspired materials.

1. Introduction

The vast majority of biological materials is hierarchically structured, beginning at the smallest scale with mineral particles, nano-fibers or platelets, which are typically embedded within a protein matrix[1]. For example, up to 7 levels of hierarchy can be found in bone and dentin[2], where the largest structural elements reach length scales of millimeters. Detailed descriptions of the hierarchical structures of several biological materials, such as shells, bone, teeth, sponge and spicules, can be found in recently published review articles[3-5].

Given a hierarchical organization, a variety of designs are possible, by changing the type and arrangement of the components at different hierarchical levels[6]. In the case of bone, for example, the variability at the nanometer level is in the shape and size of mineral particles, at the micron level in the arrangement of mineralized collagen fibers into lamellar structures and beyond in the inner architecture, the porosity and the shape of the bone. The mechanical properties of bone are well known to strongly depend on all these parameters [7-11]. The same behavior is found in other natural materials, e.g. wood [12], nacre[13, 14], spider silk[15], etc.

Biological materials differ fundamentally from most man-made materials, in being inherently structurally hierarchical. For example, as shown in Fig. 1a), the structure of tendons can be divided into six major hierarchical levels, from collagen fibrils (groups of interconnected collagen strands), to collagen fibers (bundles of fibrils), to bundles of collagen fibers, to secondary bundles of fiber bundles, to "fascicles" of bundles, to groups of fascicles which constitute the tendon itself. At all hierarchical levels, bundles are bound together by sheaths of stabilizing endotenon and the tendon also has an exterior sheath of connective tissue called epitenon. Hierarchy and functional grading implies that the mechanical properties of natural materials are also different at different length scales, i.e. the overall mechanical properties of a structure rarely reflect the bulk

properties of the materials constituting it, and rather they depend on the hierarchical and functional grading architecture[1, 16].

Virtually all stiff biological materials are composites with the smallest components mostly in the size-range of nanometers[17]. In some cases (plants or insect cuticles, for example), a polymeric matrix is reinforced by stiff polymer fibers, such as cellulose or keratin[12]. Even stiffer structures are obtained when a (fibrous) polymeric matrix is reinforced by hard particles, such as carbonated hydroxylapatite in the case of bone or dentin[18]. The general mechanical performance of these composites is quite remarkable. In particular, they combine two properties which are usually quite contradictory, but essential for the function of these materials, i.e. strength and toughness. Bones, for example, need to be stiff to prevent bending and buckling (or strong to prevent crushing), but they must also be tough, since they should not break catastrophically even when the load exceeds the normal range. This is achieved using proteins (collagen in the case of bone and dentin) are tough but not very stiff, whilst mineral is stiff but not very tough[8, 19]. The combination of these two material types in a hierarchical architecture is responsible for the exceptional properties of bone.

At nanoscale, studies have been carried out on intermediate filaments in cell cytoskeletons, ranging from atomic to cellular ranges, showing that a multi-scale structure is crucial for their characteristic mechanical properties, in particular their ability to undergo large deformations[20]. Atomistic modeling has also recently been employed to derive hierarchy-related increased crack-propagation resistance in silica-based composite structures[21].

All of these observations lead to the hypothesis that the exceptional mechanical behavior of biological materials is due to two essential elements: hierarchy and material heterogeneity. Thus, in fracture mechanics it is necessary to model these materials as hierarchical heterogeneous structures in order to correctly capture the observed behavior. Surprisingly, the complexity involved in these fracture processes can often be suitably treated by grossly simplified models. However, only few engineering models explicitly consider the role of complex hierarchical structures in fracture processes [16-19, 22]. A very important class of models of material failure is the fiber bundle model

(FBM) which has been extensively studied during the past years (see the review[23] and references therein). This model consists of a set of parallel fibers having statistically distributed strengths. The sample is loaded parallel to the fiber direction, and the fibers fail if the load exceeds their threshold value, with the load carried by the broken fiber being redistributed among the intact ones. Among the several theoretical approaches, one simplification that makes the problem analytically tractable is the assumption of global load transfer, which means that after each fiber breaking the stress is equally distributed on the intact fibers, neglecting stress enhancement in the vicinity of failed regions (Equal Load Sharing, ELS). A wealth of analytical and numerical results are available in the literature to derive predictions on fibrous materials and fiber-based composites. For example, Bosia et al. studied the strength and toughness of nanotube-based composites, starting from the properties and volume fractions of the fragile and ductile constituents[24]. Also, a numerical study of damage evolution in hierarchical FBMs was recently carried out by Mishnaevsky[25]. The relevance of FBM is manifold: in spite of their simplicity, these models capture the most important aspects of material damage and due to the analytic solutions they provide a deeper understanding of the fracture process. Furthermore, they serve as a basis for more realistic damage models also having practical importance.

In this paper we try to give an answer to the following question: "how does hierarchy and/or material heterogeneity affect the strength of a structure?", or, in other words, "is it possible by varying the hierarchical structure and mixing different material components to optimize the mechanical behavior of a material/structure?". More specifically, we wish to evaluate the pure role of hierarchy on multi-component fiber-based materials, without addressing issues like the effect of the staggered reinforcements, the effect of matrix shear[26] and other geometry-related issues. To answer the above questions, we introduce an analytical theory for hierarchical composite FBMs with different fiber types in the case of ELS.

The paper is structured as follows: in Section 2, we present the analytical procedure to calculate the strength of hierarchical fiber bundle architectures, both in the case of single-phase and

composite materials; in Section 3, we present results of calculations, together with comparisons with numerical simulations to validate the procedure; finally, conclusions and outlook are given.

2. Theory

2.1 Hierarchical fiber bundle theory

Many fibrous biological materials can be seen as a hierarchical ensemble of fibers, much like a rope. Each can be seen to correspond to a different hierarchical level, starting from single fibers (level 0), a bundle of fibers (level1), a bundle of bundle of fibers (level 2), and so on. This hierarchical arrangement suggests the use of a hierarchical procedure to determine higher-level properties from level 0 constituent fiber properties only.

The strength distribution of a single element composing a fiber bundle is assumed to be described by means of a two-parameter Weibull distribution[27, 28] $W(\sigma)$ as:

$$W(\sigma) = 1 - e^{-(\frac{\sigma}{\sigma_0})^m} \tag{1}$$

where σ is the stress applied in the longitudinal direction, and σ_0 and m are the scale and shape parameters, respectively. Weibull statistics are widely used in deterministic linear elastic fracture mechanics for the strength distribution of solids, and can be applied to natural biological or artificial polymer fibers such as those considered in this paper. However, the mathematical treatment outlined below can also be extended to the nanoscale (e.g. carbon nanotubes), with appropriate modifications (e.g. nanoscale Weibull statistics[29]). The mean strength $< \sigma_W >$ is given by[26]:

$$< \sigma_w > = \sigma_0 \Gamma \left(1 + \frac{1}{m} \right) \tag{2}$$

and the standard deviation is[26]:

$$\theta_w = <\sigma_w> \sqrt{\frac{\Gamma(1+\frac{2}{m})}{\Gamma^2(1+\frac{1}{m})} - 1} \qquad (3)$$

The shape parameter, m, represents the dispersion of the strength. A greater m value indicates a small strength variation and when m tends to infinity the strength becomes deterministic.

The case of a bundle made of a very large number N of parallel elements of Weibull type was first tackled by Daniels. Based on his analysis, the mean bundle strength is[30]:

$$<\sigma_D> = \sigma_0 (m)^{-1/m} e^{-1/m} \qquad (4)$$

with standard deviation[30]:

$$\theta_D = \sqrt{\frac{<\sigma_D>^2 \left(1 - e^{-1/m}\right)}{Ne^{-1/m}}} \qquad (5)$$

Again for m tending to infinity a deterministic strength is predicted.

The previous relations can be exploited to derive strength distributions for hierarchical structures such as a tendon shown in Fig 1a. To do this, we assume that each hierarchical level can be represented as a bundle of N_n fibers, in which each constituent fiber can in turn be represented by a bundle of lower level fibers, and so on, as shown in Fig.1b. It is reasonable to assume that at each level n in the structure the strength of the constituent fibers is Weibull distributed, i.e. is described by eqs. (1)-(3) with scale and shape parameters σ_n and m_n. We now exploit the fact that analytical results show a transition of the strength distribution function for a fiber bundle from a Weibull to a Gaussian form for large values of the number of fibers N_n. Therefore, the mean strength $<\sigma_{Wn}>$ and

standard deviation θ_{Wn} of the fibers at level n should coincide with those calculated using Daniels'

theory (eqs. (4) and (5)) applied at level (n-1). Thus, the Weibull parameters of the constituent

fibers at each hierarchical level can be determined from those at the lower level, down to level 0

(single fiber), where the distribution parameters are usually known or can be inferred. Accordingly,

we impose:

$$< \sigma_{Wn+1} >=< \sigma_{Dn} > \tag{6}$$

$$\theta_{Wn+1} = \theta_{Dn} \tag{7}$$

thus linking two adjacent hierarchical levels and extending Daniels' theory to hierarchical materials.

The two equations lead to

$$\frac{\Gamma\left(1+\dfrac{2}{m_{n+1}}\right)}{\Gamma^2\left(1+\dfrac{1}{m_{n+1}}\right)} = \left(\frac{\theta_{Dn}}{< \sigma_{Dn} >}\right)^2 + 1 \tag{8}$$

$$\sigma_{(n+1)} = \frac{< \sigma_{Dn} >}{\Gamma\left(1+\dfrac{1}{m_{n+1}}\right)} \tag{9}$$

The shape factor m_{n+1} for level ($n+1$) can be easily numerically calculated from equation (8), and

the scale factor $\sigma_{(n+1)}$ can be obtained from equation (9). This procedure can be repeated for each

hierarchical level, i.e. starting from the Weibull distribution at level 0, Daniels' theory can be

applied to derive the strength at the first hierarchical level, and so on up to level n. Notice that this

hierarchical procedure amounts to relaxing the equal load sharing hypothesis, because load sharing applies only to single fiber bundles. This provides more realistic strength distribution estimations than "single level" estimations, because in real materials some form of "local load sharing" always takes place.

In the case of small bundles, i.e. structures composed of a limited number of fibers in parallel, the previous equations need to be modified because the "large N" hypothesis in Daniels' theory is no longer justified. Thus, correction factors f_N and g_N need to be introduced to account for the discrepancy between Daniels' normal approximation and the real Gaussian distributions for relatively small bundles: $<\widehat{\sigma}_n> = f_n <\sigma_{Dn}>$ and $\widehat{\theta}_n = g_n \theta_{Dn}$ where $<\widehat{\sigma}_n>$ and $\widehat{\theta}_n$ are the corrected mean strength and standard deviation. As discussed in detail elsewhere[31], we find the expressions given in the literature[32] for f_N and g_N to be inadequate (due to the non self-consistency in the trivial case of $N=1$) for very small bundles, i.e. for typical values in hierarchical structures. Comparing results with a recently-introduced numerical Hierarchical Fibre Bundle Model (HFBM)[33], our improved and self-consistent expressions for the correction factors are:

$$f_n = 1 + \left(\frac{\Gamma(1 + \frac{1}{m_n})}{m_n^{-1/m_n} e^{-1/m_n}} - 1 \right) N^{-2/3} \tag{10}$$

$$g_n = \sqrt{\frac{\Gamma\left(1 + \frac{2}{m_n}\right) - \Gamma^2\left(1 + \frac{1}{m_n}\right)}{e^{-1/m_n}\left(1 - e^{-1/m_n}\right)}} N^{(am_n + b)} \tag{11}$$

where $a=0.01$, $b=-0.05$ are numerically derived coefficients.

2.2 Composite fiber bundles

We now consider a fiber bundle composed of two types of fibers ("composite bundle") with a well-defined percentage of each type and apply the classical Daniels' theory. The probability that this structure will fail when subjected to a stress σ is

$$W(\sigma) = x\left[1 - e^{-\left(\frac{\sigma}{\sigma_{01}}\right)^{m_{01}}}\right] + (1-x)\left[1 - e^{-\left(\frac{\sigma}{\sigma_{02}}\right)^{m_{02}}}\right] \qquad (12)$$

where x is the mixing parameter and m_{01}, σ_{01}, m_{02} and σ_{02} are the shape and scale parameters of the fibres of first and second type. The subscript "0" is an indication of the hierarchical level (0 in this example) and subscripts 1 and 2 are indications of the fiber types. By applying Daniels' theory, the mean bundle stress is obtained:

$$\sigma = P\left[xe^{-\left(\frac{P}{\sigma_{01}}\right)^{m_{01}}} + (1-x)e^{-\left(\frac{P}{\sigma_{02}}\right)^{m_{02}}}\right] \qquad (13)$$

where P is the stress sustained by surviving elements. The standard deviation is:

$$\theta_D = \sqrt{\frac{\sigma^2\left[1 - \left(xe^{-\left(\frac{P}{\sigma_{01}}\right)^{m_{01}}} + (1-x)e^{-\left(\frac{P}{\sigma_{02}}\right)^{m_{02}}}\right)\right]}{N_x\left(xe^{-\left(\frac{P}{\sigma_{01}}\right)^{m_{01}}} + (1-x)e^{-\left(\frac{P}{\sigma_{02}}\right)^{m_{02}}}\right)}} \qquad (14)$$

where N_x is the number of fibers in the bundle, and subscript "D" indicates that Daniel's theory has been used. By deriving eq. (13), it is possible to numerically determine the stress P_f that maximizes

σ and hence obtain the strength of the composite bundle as well as the standard deviation of the strength by substituting P_f in eqs. (13) and (14).

When considering fiber bundles with two fiber types, it is also necessary to consider the case in which they have different stiffnesses E_1 and E_2, as well as different Weibull-distributed strengths. In this case, one must calculate the maximal load sustained by the bundle to calculate the overall strength of the structure. For simplicity, a linear elastic relationship $\sigma = E_i \varepsilon$ is assumed up to fracture and a displacement controlled experiment is considered. Equation (1) for fiber type i may be written in alternative form:

$$W(\varepsilon) = 1 - e^{-(\frac{\varepsilon}{\varepsilon_{0i}})^{m_{0i}}} \tag{15}$$

where $W(\varepsilon)$ is the failure probability of a single fiber under a strain ε and $\varepsilon_{0i} = \sigma_{0i}/E_i$ is the scale parameter of the Weibull distribution for strain. The tensile load acting on a bundle constituted by two types of fibers ($i=1,2$) is given by

$$F(\varepsilon) = \varepsilon \left(A_1 E_1 N_1 e^{-(\frac{\varepsilon}{\varepsilon_{01}})^{m_{01}}} + A_2 E_2 N_2 e^{-(\frac{\varepsilon}{\varepsilon_{02}})^{m_{02}}} \right) \tag{16}$$

where A_1 and A_2 are the cross-sections of type 1 and type 2 fibers, and N_1 and N_2 their total number in the bundle, respectively. The maximal load F_{max} sustained by the bundle can again be obtained by setting to zero the derivative of eq. (16), and the mean strength of the bundle can be expressed as $\langle \sigma \rangle = F_{max}/(N_{01}A_1 + N_{02}A_2)$.

Clearly, the hierarchical Daniels' theory outlined previously can also be applied to a composite bundle of mixed fibers. In this work we will consider two different fiber types, but the procedure can be extended to any number of components. Thus, in the general case, the nth level will be constituted by 2 types of bundles of mixed fibers, each having characteristic shape and scale parameters m_{ni} and σ_{ni}, which can be calculated, from level $(n-1)$ parameters. Equations (13) and (14) can thus be expressed in general hierarchical form as:

$$\langle \sigma_{Dn} \rangle = \sigma_{nf} \left[x_n e^{-\left(\frac{\sigma_{nf}}{\sigma_{n1}}\right)^{m_{n1}}} + (1-x_n) e^{-\left(\frac{\sigma_{nf}}{\sigma_{n2}}\right)^{m_{n2}}} \right] \tag{17}$$

$$\theta_{Dn} = \sqrt{\frac{\langle \sigma_{Dn} \rangle^2 \left[1 - \left(x_n e^{-\left(\frac{\sigma_{nf}}{\sigma_{n1}}\right)^{m_{n1}}} + (1-x_n) e^{-\left(\frac{\sigma_{nf}}{\sigma_{n2}}\right)^{m_{n2}}} \right) \right]}{N_n \left(x_n e^{-\left(\frac{\sigma_{nf}}{\sigma_{n1}}\right)^{m_{n1}}} + (1-x_n) e^{-\left(\frac{\sigma_{nf}}{\sigma_{n2}}\right)^{m_{n2}}} \right)}} \tag{18}$$

where σ_{nf} is the value of the stress that maximizes the load sustained by the bundle at hierarchical level n and N_n is the corresponding total number of bundles.

2.3 Chains of composite fiber bundles

Finally, it is possible to extend the model at any level n to a chain of N_{yn} statistically independent bundles with N_{xn} fibers in each bundle, as shown in Figure 2. To do this, weakest link theory[34] can be adopted. The strength distribution of the chain of first-type bundles at the first hierarchical level will thus be given by:

$$W_1(\sigma) = 1 - \exp(-N_{y1}(\frac{\sigma}{\sigma_{11}})^{m_{11}}) \tag{19}$$

where N_{y1} is the number of bundles in the chain. For each bundle at the second hierarchical level Daniels' theory can be applied to calculate the Weibull parameters of the corresponding bundles, and so on.

3. Model application and results

3.1 Multi scale fiber bundles with hierarchical load sharing

The first issue we wish to address using the developed approach is the effect of hierarchy alone on the strength of structures. Thus, we first consider a multiscale hierarchical fiber bundle model such as that described in Section 2.1. In the following, we will adopt the following assumptions:

N: total number of fibers;

n: hierarchical level ($n=1, 2 \ldots, M$);

$k=k(n)$: number of elements at the n-th hierarchical level (i.e., the number of lower level bundles in an upper level bundle, or the number of fibers in the 2nd level bundle).

To illustrate the adopted hierarchical load sharing rule, we consider a 3-level structure as an example. In this case, the load is transferred from the upper-level elements of the hierarchical structure (corresponding to the ''bundles-of-bundles-of-bundles-of-fibers in a three level structure) to all lower elements of the material (fibers, ultimately). The load is shared equally among all the sub-elements of a given higher level element (as long as they are intact) or among all remaining intact sub-elements after some of them fail. For example, when one fiber breaks its load will be redistributed to all fibers in the same bundle but not to all fibers in the whole structure. Also, when a bundle fails, the load will be redistributed among bundles at the same level. In other words, there is equal load sharing at each hierarchical level. This we define as "hierarchical load sharing".

We start with a very simple example. In Fig 3a, the strengths of 4 different hierarchical structures made up of $N=8$ fibers are compared, with one to three hierarchical levels.

- The single level structure is made of eight parallel fibers (indicated as "8").

- Two different double-level structures are considered: two bundles of four fibers (indicated as "4,2"), and four bundles of two fibers (indicated as "2,4).

- The third level structure is composed by two bundles made of two bundles of two fibers (indicated as "2,2,2").

These structures are schematically shown in Fig. 3a. The level 0 fibers are assigned random Weibull distributed strengths, using carbon nanotube (CNT) properties: $\sigma_0 = 34$ GPa and $m = 2.74$[35]. Results in Fig 3a show that the lowest hierarchy level structure has the highest strength. Also, the strongest of the two double-level structures is that with the highest number of fibers in parallel (highest lower-level k). The latter would therefore seem to be the required condition for optimizing strength, i.e. the highest possible number of lower level elements set in parallel. This is confirmed by results in Fig. 3b relative to various 128-fiber arrangements, ranging from single-level (128 fibers in parallel) to 4-level structures. Again the highest strength is achieved by 128 fibers in parallel, then with two 64-fiber bundles, and so on. It is important to stress how the use of correction factors (eqs.10 and 11) are essential in these calculations whenever a structure with a small number of parallel fibers or bundles is considered. Neglecting these corrections would lead to significant differences in the ordering according to strength of the considered structures.

The influence of the number of hierarchical levels on the mean strength is next evaluated again for structures with the same total number of fibers N. In Fig. 4, structures having the same number of elements (fibers) at the lowest level are compared, i.e. $k(1)=8$, with $N=160$, as well as with the corresponding level 1 structure ($k(1)=160$) for reference. Once again a strength decrease is found from 1st level to 4th level structures, indicating that increasing hierarchy leads to decreasing strength.

The same tendency is found when keeping constant the number of elements at the highest hierarchical level, as shown in Fig. 5, for N=320. In Fig. 5a the comparison is between four different structures with $k(M)$=4, whilst in Fig. 5b the mean strength is plotted vs. number of hierarchical levels for three different values of $k(M)$.

The observation that the increase in the number of hierarchical levels leads to a lower material strength is consistent with other results in the literature[25, 36]. However, it is in contrast with the observations that many natural materials, built as hierarchical fibrous composites, have extraordinarily high strength. This leads to the conclusion that hierarchy alone is insufficient to justify the strength of natural biomaterials.

3.2 Composite fiber bundle

Next, we wish to apply the theory outlined in Section 2.2 and evaluate the influence on the mean strength of composite fiber bundles of the chosen Weibull parameters for the two types of fibers involved.

In the first example, shown in Fig. 6a, the mean composite bundle strength is calculated for varying σ_{01}, m_{01} and m_{02} values, setting σ_{02} =0.01 GPa, and a linear dependence is highlighted. As expected, the variation of m_1 has an effect on the results in a manner which is proportional to the mixture ratio x, i.e. its effect increases with x, and increasing m_1 values yield an increase in mean strength. A linear behavior is also found between mean strength and mixture ratio as shown in Fig. 6b.

Another issue of interest is the comparison between the results obtained with the present model (application of Daniels' theory to a composite bundle) and those obtained using a rule of mixtures approach. In the latter, the mean composite bundle strength $\langle \sigma_{RM} \rangle$ is calculated by using Daniels' theory to separately obtain the strengths $\langle \sigma_{1,D} \rangle$ and $\langle \sigma_{2,D} \rangle$ relative to bundles composed

of 100% of first and second types of fibres, respectively, and then combining the two values using the relation[34]:

$$\langle \sigma_{RM} \rangle = x \langle \sigma_{1,D} \rangle + (1-x)\langle \sigma_{2,D} \rangle \tag{20}$$

where x is the volume fraction of the first bundle. Figure 7a illustrates the discrepancy $\Delta = |\langle \sigma_D \rangle - \langle \sigma_{RM} \rangle|$ between the mean bundle strengths calculated using the two approaches for σ_{02}=0.1 GPa, m_{01}=4, m_{02}=2 and various values of σ_{01}. Clearly, Δ is zero for x=0 and x=1, but the discrepancy is not negligible for intermediate x values. This is due to the fact that when adopting a classical rule of mixture approach an unrealistic load redistribution is assumed among the different types of fibers as damage progresses in the composite bundle. This justifies the adoption of the approach outlined in Section 2.2.

3.4 Comparison with numerical results

To check the validity of the proposed approach, we now compare some analytical calculations with numerical results obtained with the Hierarchical Fiber Bundle Model (HFBM)[33]. First, we wish to analyze the mean strength of various bundles composed of different types of fibers. As an example, we consider a mean strength calculation for a varying mixture ratio x and Weibull parameter $\sigma_{01,}$ for σ_{02}=10 GPa, m_1=2, m_2=2. The mean strength of all composite bundles is calculated analytically using the procedure described in Section 2.2 and compared to values obtained through numerical simulations. Results are shown in Fig. 7b and display considerable agreement between analytical and numerical calculations.

The comparison between analytical and numerical results is extended to various different cases of composite bundles composed by fibers with different Weibull parameters and for various mixture ratios. Results in Table 1 are relative to bundles composed of fibers with the same elastic

modulus, whilst those in Table 2 are composed by fibers with different elastic moduli. Again good agreement is found in all cases, thus proving the validity of the approach.

3.5 Hierarchical composite bundle

To illustrate the possible variations in the mechanical behavior of a hierarchical composite bundle, we consider some specific examples. First, let us analyze the case of a bundle with two types of fibers and a mixture ratio of x=0.5, with σ_{01}=10 GPa, σ_{02}=0.01 GPa , m_{01}=2, m_{02}=3 and N=480. In the non-hierarchical case, i.e. in the case of a level 1 bundle with all 480 fibers in parallel, the expected mean strength, according to the calculation procedure in section 2.2, is $\langle\sigma\rangle$=2.14 GPa.

One possibility for creating hierarchical architectures with this set of fibers is to form single-fiber bundles at level 1 and mixed bundle types at level 2. For example, we can build two types of level 1 bundles, the first one consisting of two fibers of the first type (σ_{01}=10 GPa and m_{01}=2), the second of 5 fibers of the second type (σ_{02}=0.01 GPa and m_{02}=3), and create a level 2 structure composed of the resulting 120 bundles of the first type and 48 of the second type. The chosen nomenclature for this type of structure is [(2,5);(120,48)]. From the application of our hierarchical fiber bundle model we get: σ_{11}=8 MPa, σ_{12}=0.007 MPa, m_{01}=2.4, m_{02}=5.2, and a mean strength for the 2nd level bundle of 2.56 GPa, which is larger than the above non-hierarchical level 1 bundle.

This strength increase is obtained through various other configurations, as documented in Fig. 8. All of these are 2nd level structures, where the total number of the two types of fibers remaining constants, and only their hierarchical configuration in bundles changing in the various considered cases. The general tendency is that the greatest strength increase is obtained by grouping strong fibers in small bundles and weak fibers in large bundles at level 1. Clearly, the load redistribution during specimen failure in this type of configuration favours an enhancement of the resistance to damage progression. Clearly, this is only a first example of how mechanical properties of composite structures can be tailored by an appropriate choice of the components and their hierarchical arrangement. Further future work requires extensive parametric studies to highlight in

greater detail the necessary strategies to obtain the desired structures with superior mechanical behavior.

4. Conclusions

We have presented an altogether general and self-consistent analytical procedure to calculate the strength of hierarchical fiber bundles constituted by two (or more) types of fibers. We have demonstrated how hierarchy alone is insufficient to yield strength enhancement, and how this increase in strength can be obtained through a suitable choice of fiber distributions at different hierarchical levels. In other words, the key to an improvement in the strength and mechanical performance in general of multiscale materials would seem to lie in hierarchical structuring of multi-components. These results can be of great interest, first as a means to interpret and further investigate the exceptional mechanical performance of biomaterials, and secondly as a strategy to design and fabricate new bio-inspired materials with desired tailor-made properties. In future, a possible continuation of this work could be to extend its application to the molecular level by integrating results from molecular dynamics simulations (e.g. on spider silk[37]) into the described procedure, which at present relies on experimentally determined level 0 Weibull parameters. Therefore, the role of hierarchy and material mixing could truly be evaluated from nano- to macroscale, ideally with no free model parameters. Future HFBM numerical simulations could also include shear effects into a 2-D formulation, thus improving the evaluation of the mechanical performance of hierarchical composite materials. In any case, the theory and analytical procedures outlined in this work can provide a useful tool to help in understanding the underlying mechanisms in the mechanical behavior of natural materials and in designing bio-inspired materials with tailor made properties.

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List of Figure captions

Fig. 1: a) Hierarchical tendon structure (from[38]); b) Corresponding schematic representation of the present hierarchical fiber bundle model. The Weibull strength distribution at hierarchical level n is determined from Daniels' theory applied to the fiber bundle at level (n-1).

Fig. 2: Schematic representation of the composite fiber bundle model at the 1st level of the hierarchical chain of bundles structure.

Fig. 3: Strength versus hierarchical structure for (a) N=8, (b) N=128.

Fig. 4: Mean strength plotted versus different hierarchical structures for a) $k(1)$=8; b) $k(M)$ =4.

Fig. 5: a) Mean strength of different hierarchical structures for k(M)=4; b) Mean strength vs. number of hierarchical levels for structures having the same number of elements at the highest hierarchical level $k(M)$.

Fig. 6: Mean bundle strength versus a) scale parameter of the first type of fibers. b) mixture ratio x of first type of fibers with m_1=2.

Fig. 7: a) Discrepancy between mean strength values for composite bundle calculated using Daniels' theory and rule of mixtures (σ_{02}=0.1 GPa, m_{01}=4, m_{02}=2); b) Comparison between mean strength predictions for a mixed fiber bundle: values are calculated using theory ("Theo"), and HFBM numerical simulations ("HFBM") for σ_{02}=10 GPa, m_1=2, m_2=2.

Fig. 8: Mean bundle strength versus hierarchical structure for composite hierarchical 2nd level bundles. The staggered line indicates the strength of the corresponding non-hierarchical level 1 bundle.

Tables

Table. 1: Comparison between theory and HFBM code for different cases of composite bundles composed by fibers with the same elastic modulus.

Case	σ_{01} (GPa)	σ_{02} (GPa)	m_1	m_2	x	Mean strength (GPa, Theory)	Mean strength (GPa, HFBM code)
1	50	10	2	4	1	21.440	21.672
2	50	10	2	4	0.5	10.780	10.917
3	50	10	2	4	0	5.506	5.533
4	20	10	2	4	1	8.577	8.676
5	20	10	2	4	0.5	6.070	6.133
6	20	10	2	4	0	5.506	5.523
7	10	0.01	2	4	1	4.300	4.329
8	10	0.01	2	4	0.7	3.002	3.021
9	10	0.01	2	4	0.5	2.140	2.173
10	10	0.01	2	4	0	0.006	0.005

Table. 2: Comparison between theory and HFBM code for bundles composed of fibers elastic with different elastic moduli (x=0.5).

N_1, N_2	m_1	m_2	σ_{01} (GPa)	σ_{02} (GPa)	E_1 (GPa)	E_2 (GPa)	Mean strength (GPa, Theory)	Mean strength (GPa, HFBM code)
500	2	3	4	4	10	20	1.642	1.510
500	3	6	50	400	300	800	125.400	122.940
500	2	4	40	20	110	200	9.506	8.857

Figure 1
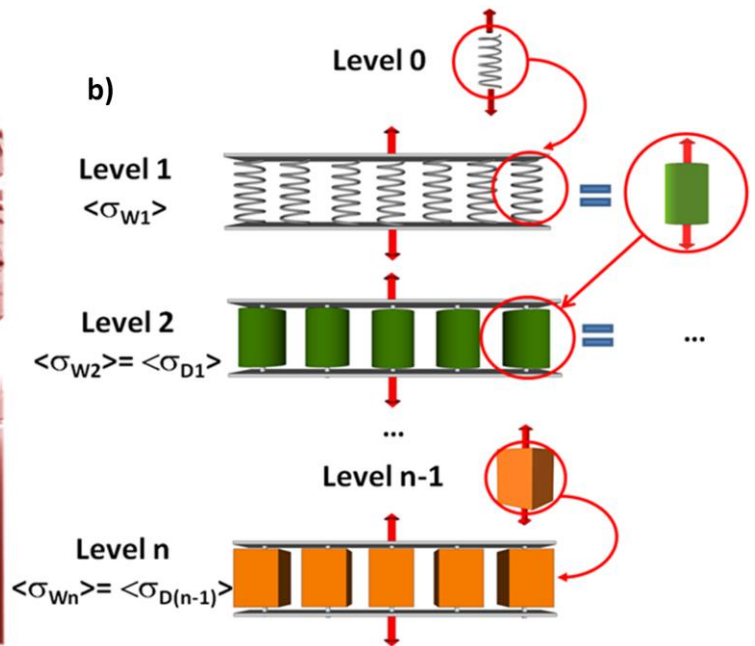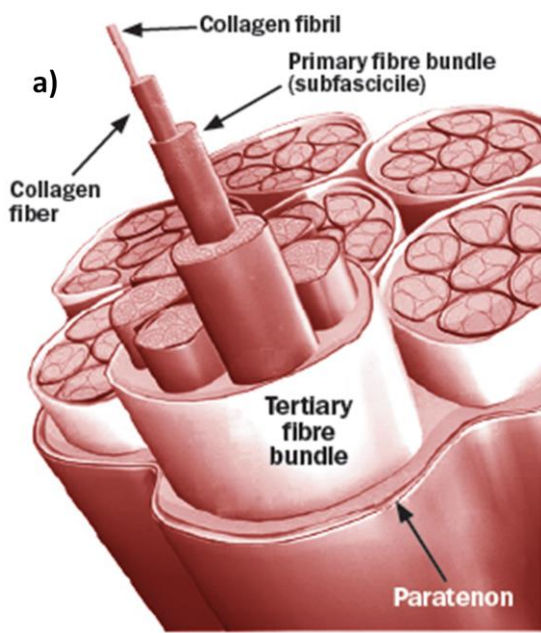
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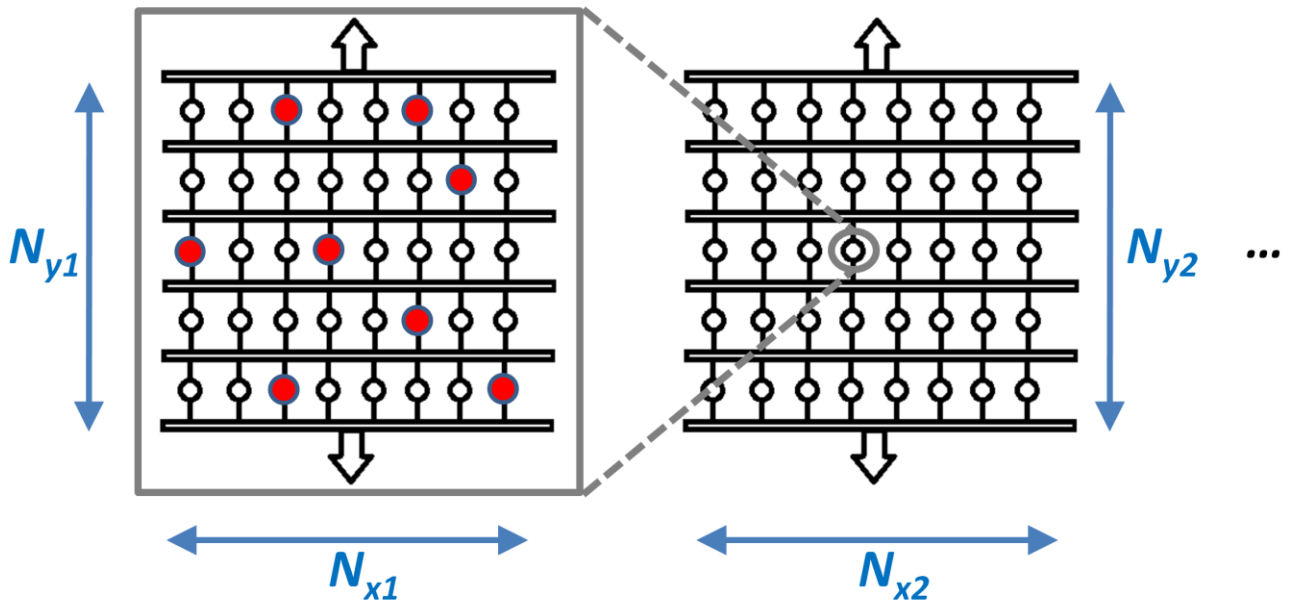
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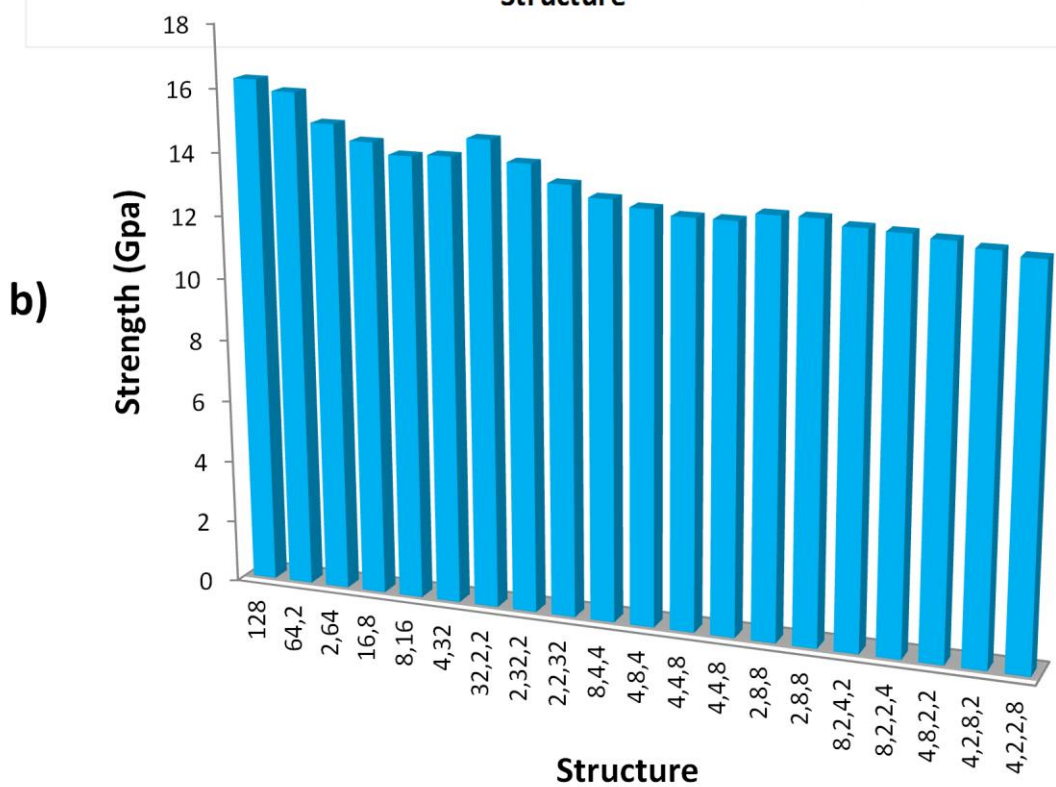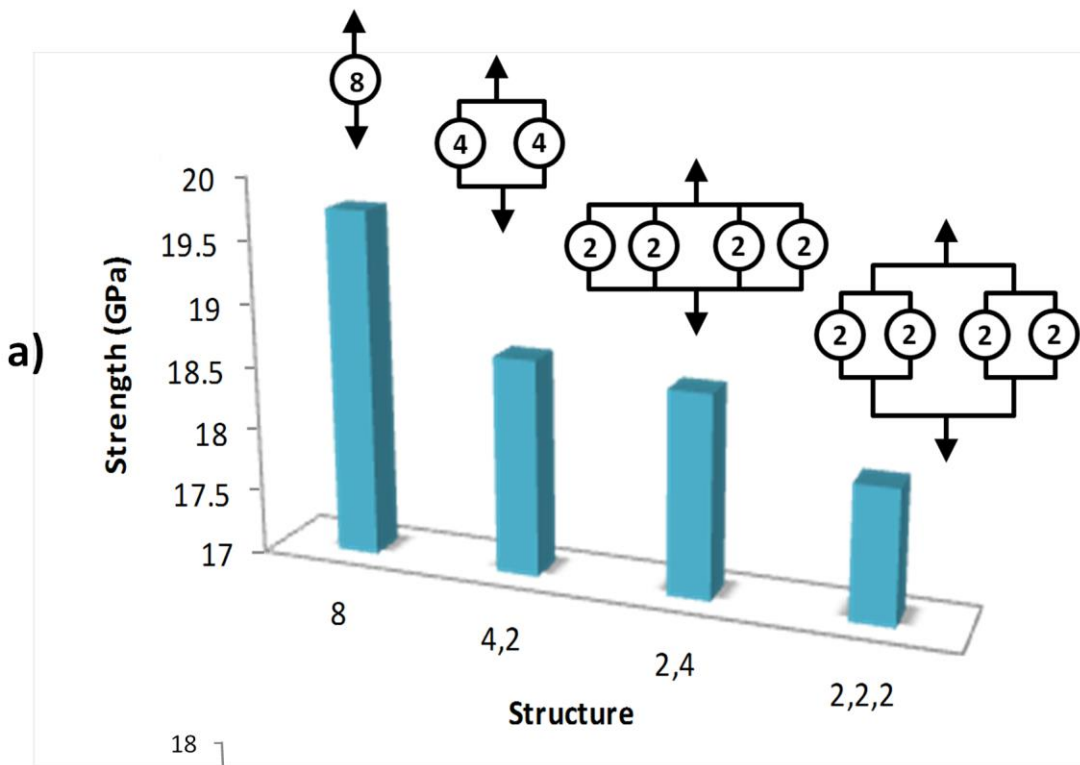
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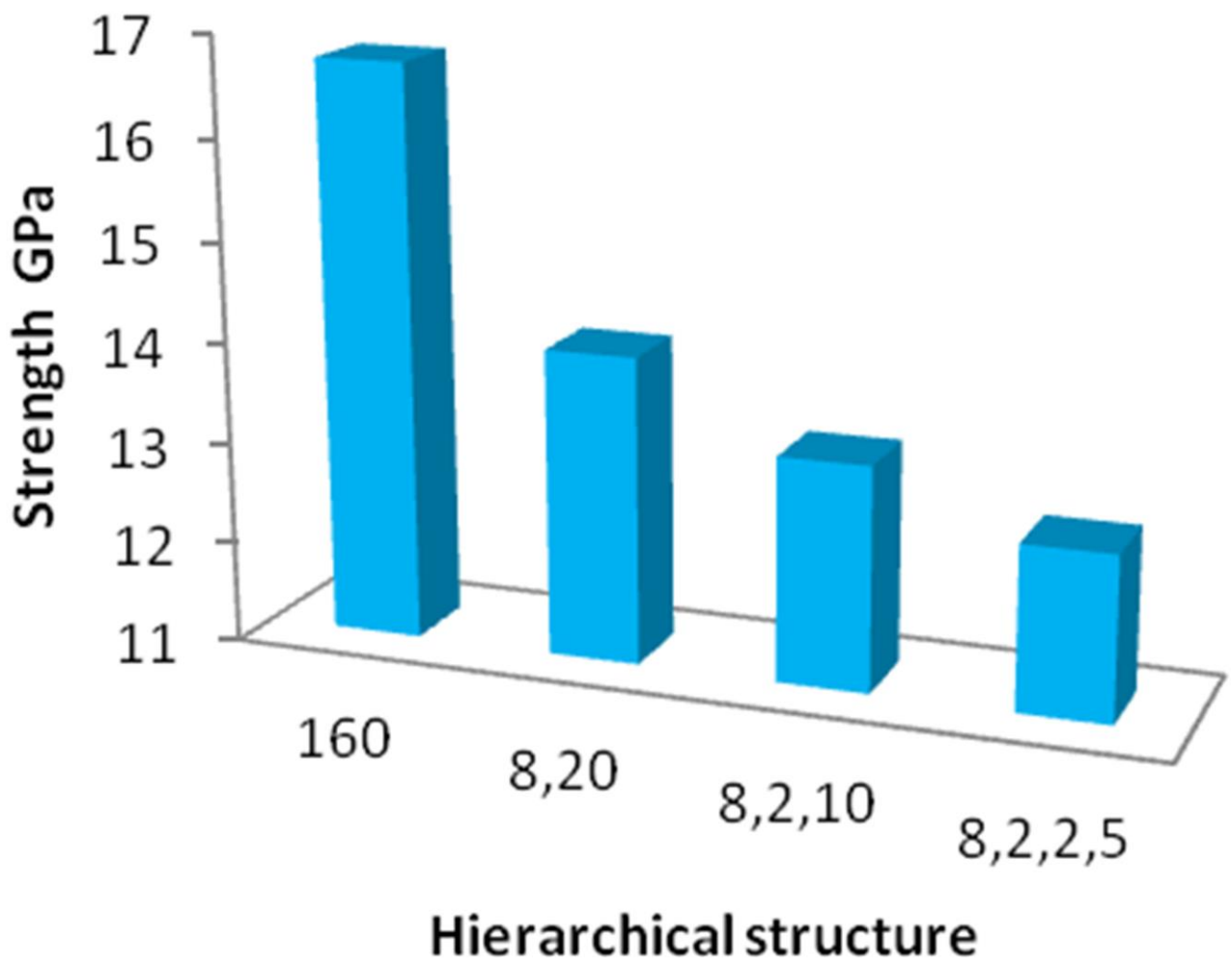
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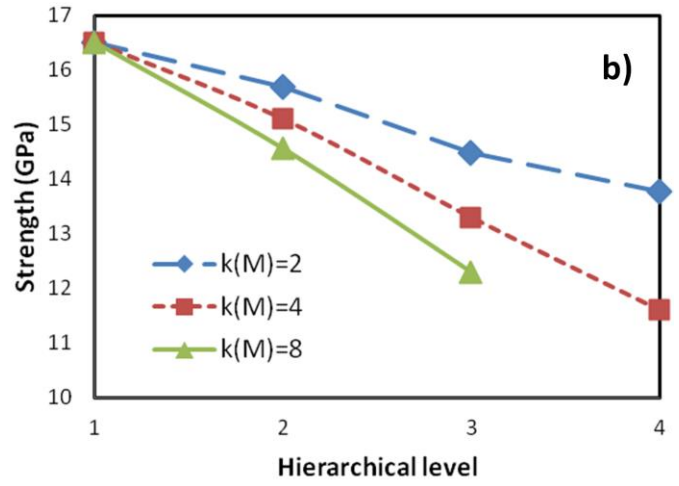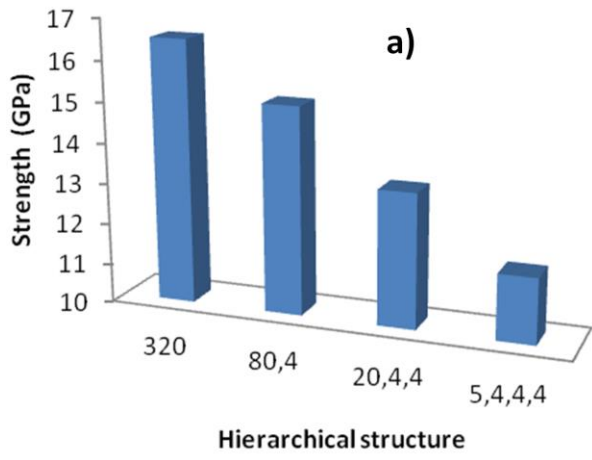
Figure 6

Figure 7

Figure 8