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TECHNICAL PAPER

Strength of hierarchical materials

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Abstract Many biological materials exhibit a hierarchical structure over more than one length scale. Understanding how hierarchy affects their mechanical properties emerges as a primary concern, since it can guide the synthesis of new materials to be tailored for specific applications. In this paper the strength and stiffness of hierarchical materials are investigated by means of a fractal approach. A new model is proposed, based both on geometric and material considerations and involving simple recursive formulas.

1 Introduction

Many biological materials are structured in a hierarchical way over more than one length scale. Multiple examples can be given such as bones (Currey 1984), teeth (Warshawsky 1989) and shells (Currey 1977): bones, for instance, have seven levels of hierarchy, while sea shells present two or three orders of lamellar structures. These materials, at the most elementary level of structural hierarchy, are composed by hard and strong mineral structures embedded in a soft and tough matrix. In bone the mineral platelets are $\sim$3 nm thick, in shell their thickness is of $\sim$300 nm, while in tooth crystals are $\sim$15–20 nm thick, with a very high slenderness (Gao 2006). Understanding how these structures are related to their mechanical properties emerges, hence, as a primary concern, since it may provide guidance on the development of novel materials with unique properties (Fratzl and Wienkamer 2007).

Hierarchical structures of biomaterials have been recognized to exhibit self-similarity and to be fractal-like (Lakes 1993). Size effects on apparent mechanical properties due to the fractal nature of material microstructure have been extensively studied (Carpinteri 1994a, b) (see also Carpinteri and Pugno 2005). If infinite levels of hierarchy are considered, new universal properties (i.e. scale-invariant quantities) having non-conventional or anomalous physical dimensions must be defined (Carpinteri 1994a, b). On the other hand, if the hierarchical character is exhibited only over a finite range of scales, as for biological materials, new physical considerations can be drawn.

In the present study, the strength and stiffness of hierarchical biomaterials are investigated by means of a fractal approach. The rules of mixture which let estimate the nominal strength of hierarchical materials are firstly presented (Sect. 2). By exploiting the self-similarity character of the structure, these rules can be condensed in a unique and synthetic manner by means of a fractal approach (Sect. 3) (Pugno 2006, Pugno and Carpinteri 2008). Finally, a new fractal model based on a multiplicative process (Halsey et al. 1986), which takes into account both geometry and material features, is proposed (Sect. 4).

2 Prediction of strength in hierarchical materials

Let us consider a tensile test on N-hierarchical fibre-reinforced bar. Its cross-section is composed by hard inclusions embedded in a soft matrix and it is represented in Fig. 1. The nominal stress σ_0 could be evaluated by means of a recursive scheme of rules of mixture as (Pugno and Carpinteri 2008):

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$$\begin{aligned}
\sigma_0 &= v_1\sigma_1 + (1-v_1)\sigma_{m,1}\\
\sigma_1 &= v_2\sigma_2 + (1-v_2)\sigma_{m,2}\\
&\cdots\\
\sigma_i &= v_{i+1}\sigma_{i+1} + (1-v_{i+1})\sigma_{m,i+1}\\
&\cdots\\
\sigma_{N-1} &= v_N\sigma_N + (1-v_N)\sigma_{m,N},
\end{aligned} \tag{1}$$

where σ_i and $\sigma_{m,i}$ denote the stresses in the hard and soft phases, respectively, and v_i is the volumetric fraction of inclusions at the i-level (the nanostructure of bones, for instance, shows a mineral to matrix volume ratio in the order of 1 to 2). The values of the material properties at each level hence depend on those of the preceding levels.

In the case of round (or square)-shaped inclusions at each level, v_i could be expressed in the following form:

$$v_i = n_{h_i}\left(\frac{R_i}{R_{i-1}}\right)^2, \tag{2}$$

n_{h_i} being the number of inclusions and $R_i \sim \sqrt{A_i}$ their mean average radius. Henceforth, the area of the cross-section of the bar A_0 and its characteristic dimension R_0 will be denoted simply by A and R, respectively.

Note that the rules of mixture (Eq. 1) involve, at each step, only the hard phase (i.e., the inclusions, Fig. 1). Although experimental data (Lee and Gurland 1978) show that the properties of the matrix may vary at each level, it is often assumed, for the sake of simplicity, that they do not change, i.e., $\sigma_{m,i} = \sigma_m$. In such a case Eq. 1 can be rewritten via a bottom-up approach as:

$$\begin{aligned}
\sigma_0 &= \left(\prod_{i=1}^{N} v_i\right)\sigma_N + \left(1 - \prod_{i=1}^{N} v_i\right)\sigma_m\\
&= v_r\sigma_N + (1-v_r)\sigma_m,
\end{aligned} \tag{3}$$

where v_r is the total volumetric fraction of inclusions.

3 Fractal model: constant matrix properties

Natural optimization suggests self-similar structures (Brown and West 1999), for which $R_i/R_{i-1} = 1/\delta$ and $n_{h_i} = n_h$; thus,

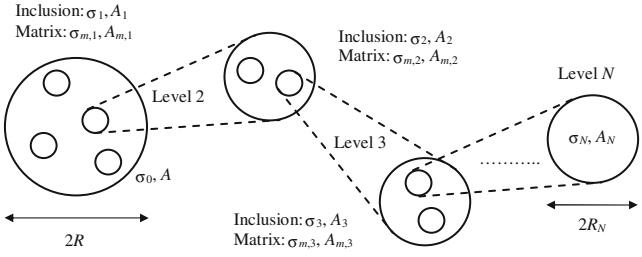

Fig. 1 The cross-section of a hierarchical bar

$v_i = v$ and $v_r = v^N$. Thanks to self-similarity, further considerations based on fractal geometry naturally rise up (Pugno 2006). Particularly, if the hierarchical levels were infinite ($N \to \infty$), the domain of inclusions would result into a fractal set of dimension D:

$$D = \frac{2\ln n_h}{\ln n_h - \ln v} = \frac{\ln n_h}{\ln \delta}, \tag{4}$$

where $0 < D < 2$. See, for instance, the Sierpinski carpet displayed in Fig. 2.

Since a finite range of scales is taken into account, the total volumetric content of inclusions v_r can be modelled as (Pugno and Carpinteri 2008):

$$v_{\mathrm{r}} = v^N = \left(\frac{R_N}{R}\right)^{2-D}. \tag{5}$$

Thus, by substituting Eq. 5 into Eq. 3:

$$\begin{aligned}
\sigma_0 &= \left(\frac{R_N}{R}\right)^{2-D}\sigma_N + \left(1 - \left(\frac{R_N}{R}\right)^{2-D}\right)\sigma_m\\
&\approx \left(\frac{R_N}{R}\right)^{2-D}\sigma_N,
\end{aligned} \tag{6}$$

which predicts that the nominal strength σ_0 decreases as the size increases ($R \to \infty$) i.e. "smaller is stronger". The approximation in Eq. 6 keeps true as long as $\sigma_N \gg \sigma_m$, a condition which is usually satisfied.

Note that, according to Eq. 6, the stress levels always remain two, whatever is the hierarchical level of the material. The same scaling law is valid for stiffness.

4 Fractal model: variable matrix properties

In the previous section a scaling law for strength in hierarchical materials has been derived, involving the fractal dimension related to the distribution of inclusions (Eq. 4). This model is based on the simplifying hypothesis of identical matrix properties at each scale level (Eq. 3). On the other hand, as pointed out in Sect. 1, different properties of the matrix are often noticed at different levels. In order to consider also this feature, a new model is proposed, based on a multiplicative process (Halsey et al. 1986), which will be shown to provide a simple and natural extension of Eq. 6.

Let us suppose that the ligament, which is apparently homogeneous at the macro-scale, is divided into n equal parts: the stiffness is allocated such that n_h cells are enriched by a factor Φ, and n_m cells are depleted by a factor φ. Naturally, $n_h + n_m = n = R^2/{R_1}^2$. If the condition of perfect bonding between inclusions and matrix is satisfied, the same repartition occurs for stresses:

Fig. 2 The Sierpinski carpet (D = 1.89) at different levels of observation; it corresponds to the cross section of a deterministic hierarchical bar in which the white zones represent the soft matrix and the black ones are the hard inclusions

$$A\sigma_0 = A_1\sigma_1 + A_{m,1}\sigma_{m,1} = \left(\frac{n_h}{n}\Phi + \frac{n_m}{n}\varphi\right)A\sigma_0, \tag{7}$$

where A_i now denotes the total area of inclusions at the level i.

The relationship between Φ and φ can be obtained by imposing the condition that the critical applied force must be the same; from Eq. 7, in formulae:

$$\frac{n_h}{n}\Phi + \frac{n_m}{n}\varphi = 1, \tag{8a}$$

and thus

$$\varphi = \frac{n - n_h\Phi}{n_m}. \tag{8b}$$

Since negative values of parameters Φ and φ lack a physical meaning, it can be deduced from Eq. 8b that $\Phi \in (1, n/n_h)$ and $\varphi \in (0, 1)$.

At the following level, the problem is renormalized so that the "enriched" cells are structured exactly in the same way:

$$A_1\sigma_1 = A_2\sigma_2 + A_2\sigma_{m,2} = \left(\left(\frac{n_h}{n}\right)^2\Phi^2 + \frac{n_h n_m}{n^2}\Phi\varphi\right)A\sigma_0. \tag{9}$$

Note that this approach is consistent with Eq. 1. At the generic level N, iterating such a procedure yields:

$$F = A\sigma_0 = A_N\sigma_N + \sum_{j=1}^{N} A_{m,j}\sigma_{m,j}, \tag{10}$$

where

$$A_N = \left(\frac{n_h}{n}\right)^N A, \tag{11a}$$

$$\sigma_N = \Phi^N\sigma_0, \tag{11b}$$

and

$$A_{m,j} = \left(\frac{n_h}{n}\right)^j \frac{n_m}{n_h}A, \tag{11c}$$

$$\sigma_{m,j} = \varphi\Phi^{j-1}\sigma_0. \tag{11d}$$

The material properties are hence distributed in a non-homogeneous way to form a hierarchical structure with different characteristics at different length scales. The distribution process here proposed (Eq. 10) is equivalent to assuming that the ratio between the Young's moduli of matrix and inclusions remains constant at each level.

By expressing Eqs. 10a, b as a function of N and equalling the two expressions, the relationship between the nominal strength σ_0 and the strength of inclusions at level N can be obtained:

$$\sigma_0 = \left(\frac{A_N}{A}\right)^{\frac{\log\Phi}{\log(n/n_h)}}\sigma_N. \tag{12}$$

Finally, since:

$$\frac{A_n}{A} = \left(\frac{R_N}{R}\right)^{2^{\frac{\log(n/n_h)}{\log n}}}, \tag{13}$$

substituting Eq. 13 into Eq. 12, yields:

$$\sigma_0 = \left(\frac{R_N}{R}\right)^{D^*}\sigma_N, \tag{14}$$

where

$$D^* = \frac{2\log\Phi}{\log n} = D\frac{\log\Phi}{\log n_h}, \quad \Phi \in (1, n/n_h). \tag{15}$$

Equations 14–15 describe the nominal strength dependency not only on the volume fraction of inclusions (geometric effect, by D), but also on the nature of such inclusions (material effect, by Φ). Note that, in the homogeneous case ($\Phi = 1$), Eq. 15 provides $D^* = 0$ and consequently $\sigma_0 = \sigma_N$ (Fig. 3). On the other hand, if the

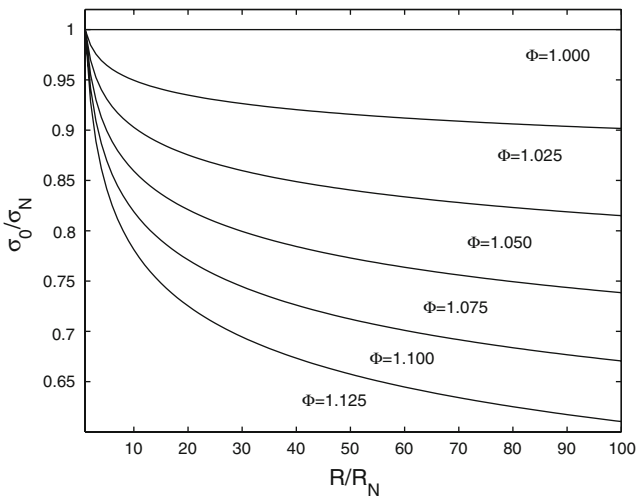

Fig. 3 Scaling law of the nominal strength σ_0 for different values of Φ (n = 9, n_h = 8)

specimen is made only of inclusions ($\Phi = n/n_h$), it is not difficult to demonstrate that $D^* = 2 - D$ and the same scaling predicted by Eq. 6 is recovered (Fig. 3).

The present model, by modelling different matrix properties at each level, can hence be considered as an extension of the model presented in Sect. 3 (Pugno 2006, Pugno and Carpinteri 2008), which is based on purely geometrical considerations. For a further generalization see the Appendix.

Under the hypothesis mentioned above, the same scaling occurs clearly for stiffness:

$$E_0 = \left(\frac{R_N}{R}\right)^{D^*} E_N, \tag{16}$$

where E_0 and E_N are the Young's moduli of the bar and of the inclusions at level N, respectively, and D^* is provided by Eq. 15.

5 Conclusions

In this work, the strength and hardness of hierarchical materials are investigated. The study has focused on the behaviour of biomaterials subjected to uniaxial loading. Despite these hierarchical structures show a self-similarity character only over a finite range of scales, it is possible to model their scaling properties at each level by means of a fractal approach. Recursive relationships are presented, based on a renormalization group transformation.

Appendix

In Sect. 3 a multiplicative process model has been proposed for the prediction of strength in hierarchical biomaterials: stresses in the hard phases are modelled at each level according to different matrix properties (Eq. 1) by means of a renormalization group transformation. As already stated, this model has been proven to provide a natural extension of the fractal model presented in Sect. 2.

It is interesting to note that, if the multiplicative process involves all the cells and not only the stiffer ones (Eq. 10), for infinite levels of hierarchy (i.e., for $N \to \infty$) the stress distribution shows a multifractal behaviour: in other words, the ligament is characterized by the union of fractal sets, each with its own fractal dimension (Feder 1988). Furthermore, even more interesting for our purposes, the force F concentrates on a fractal set of dimension D_1, whose expression has been derived following the procedure presented in (Turcotte 1997):

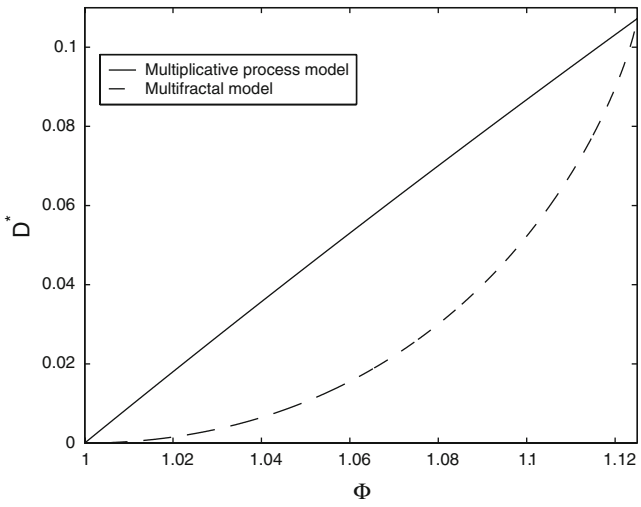

Fig. 4 Fractal exponent D^* versus Φ ($n = 9$ and $n_h = 8$): multiplicative process model (*solid line*) and multifractal model (*dashed line*). If $\Phi = 9/8 = 1.125$ the classical Sierpinski carpet is recovered and $D^* = 2 - (\log 8/\log 3)$ (Eqs. 15, A2)

$$D_1 = -\frac{2}{\log n} \times \left[\left(\frac{n - n_h\Phi}{n}\right)\log\left(\frac{n - n_h\Phi}{n(n - n_h)}\right) + \left(\frac{n_h\Phi}{n}\right)\log\left(\frac{\Phi}{n}\right)\right] \quad \Phi \in (1, n/n_h). \tag{A1}$$

This effect of concentration is called *curdling*.

Hence, according to (Carpinteri 1994b) the following scaling law for strength is obtained:

$$\sigma_0 = R^{-D^*}\sigma^*, \tag{A2}$$

where $D^* = 2 - D_1$ and σ^* is the so-called fractal strength, with the anomalous dimensions $[\mathrm{F}][\mathrm{L}]^{-D^*}$.

Note that, for the extreme cases ($\Phi = 1$, $\Phi = n/n_h$), the values of the fractal exponent D^* provided by the multifractal and the multiplicative models coincide ($D^* = 0$ and $D^* = 2 - D$). On the other hand, a different behaviour is observed for the intermediate cases, as shown in Fig. 4 for $n = 9$ and $n_h = 8$.

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