Systematic numerical investigation of the role of hierarchy in heterogeneous bio-inspired materials

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Abstract:

It is well known that hierarchical structure is an important feature in biological materials to optimize various properties, including mechanical ones. It is however still unclear how these hierarchical architectures can improve material characteristics, for example strength. Also, the transposition of these structures from natural to artificial bioinspired materials remains to be perfected. In this paper, we introduce a numerical method to evaluate the strength of fibre-based heterogeneous biological materials and systematically investigate the role of hierarchy. Results show that hierarchy indeed plays an important role and that it is possible to “tune” the strength of bio-inspired materials in a wide range of values, in some cases improving the strength of non hierarchical structures considerably.

1. Introduction

It is known that many biological materials and organisms display fascinating physical and mechanical properties, which have up to now been hard to replicate in artificial materials and systems. One of these is the ability to combine exceptional strength and toughness, which occurs for example in nacre, bone and dentine (Espinosa et al., 2011; Pugno, 2006; Wegst and Ashby, 2004), or the smart adhesion which is found in spiders and geckos (Autumn et al., 2000; Autumn and Peattie, 2002; Foelix, 1996; Tian et al., 2006). An important feature underlying these properties is thought to be material structure and hierarchy (Fratzl and Weinkamer, 2007; Lakes, 1993). A prime example of this is spider silk, whose hierarchical structure ranges from nanostructure to macrostructure and consists of an amorphous network of chains and β-sheet crystals constituted by poly-(Gly-Ala) and poly-Ala domains (Ackbarow et al., 2007; Keten et al., 2010). Molecular dynamics and atomistic simulations have shown how the specific structure and bonding at molecular level affects macroscopic properties like strength and toughness (Bratzel and Buehler, 2012; Keten et al., 2010; Nova et al., 2010).

Aside from spider silk, a great number of biological materials are inherently structurally hierarchical. The hierarchical structure of tendon, taken from (Riley, 2005) is shown in Fig1a. Another example is the case of bone, where variability at the nanometer level lies 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. Various studies show the dependence of the mechanical properties of bone on all these parameters (Currey, 2002; Gibson et al., 1995; Launey et al., 2010; Rho et al., 1998; Weiner and Wagner, 1998). Other biological systems that have been studied to assess the role of hierarchy are tendons (Puxkandl et al., 2002), protein materials (Gao, 2006), Gecko adhesion (Yao and Gao, 2006), tissue

growth (Cranford and Buehler, 2011).

Given a hierarchical organization, various designs are possible, by altering the type and arrangement of the components at different levels. Hierarchy and functional grading frequently lead to variable mechanical properties at different length scales, i.e. overall mechanical properties are often quite different from those of constituents (Lakes, 1993; Pugno, 2006), and many natural materials can be considered an equivalent of artificial composite materials. For example, stiff biological materials are often composites with the smallest components mostly in the nanometer range (Gao and Ji, 2004). In the case of plants or insect cuticles, a polymeric matrix is reinforced by stiff polymer fibers, such as cellulose or keratin (Vincent, 1999), and in the case of bone or dentin even stiffer structures are obtained using a fibrous polymeric matrix reinforced by hard carbonated hydroxylapatite particles (Currey, 1999).

One possible hypothesis is therefore that the exceptional mechanical behavior of biological materials is due to two essential elements: hierarchy and material heterogeneity. To verify this conjecture, a number of theoretical models which include both these elements have been formulated, including molecular dynamics or atomistic simulations (Buehler et al., 2009; Currey, 1999, 2003; Gao and Ji, 2004; Pugno, 2006). A simplified numerical approach is the fiber bundle model (FBM) which has been extensively studied during the past years (Pradhan et al., 2010). 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. The Equal Load Sharing (ELS) formulation is most often adopted, whereby after each fiber break the stress is equally distributed on the intact fibers, neglecting stress concentrations in the vicinity of failed regions. Based on this model, we developed a hierarchical formulation of the FBM (“HFBM”) and

used it to calculate the space elevator cable strength including the role of defects (Pugno et al., 2008). With this model we also studied the strength and toughness of nanotube-based composites, starting from the properties and volume fractions of the fragile and ductile constituents (Bosia et al., 2010). In recent work, we addressed the issue of the synergy between hierarchy and material mixing to enhance the mechanical performance of composites, finding evidence that some hierarchical configurations lead to an improvement with respect to the non hierarchical case (Bosia et al., 2012) An important numerical study of damage evolution in hierarchical FBMs was also recently carried out by Mishnaevsky (Mishnaevsky, 2011).

However, despite the recent advances in this field, a systematic study addressing the role of *pure* hierarchy (independently of the specific material system it refers to), its interaction with material heterogeneity, and their effect on macroscopic mechanical properties is still missing. In this paper, we therefore wish to begin such a systematic study, and investigate the possibility of tuning and optimizing the strength of hierarchical fibre bundles composed of different fibre types as a function of hierarchy and distribution of different fibre types.

The paper is structured as follows: in Section 2, we introduce the numerical model used to calculate the strength of hierarchical fiber bundle architectures in composite materials and the evaluation procedure; in Section 3, we present results of calculations and their discussion; finally, conclusions and outlook are given.

2. Hierarchical Fibre Bundle Model

2.1 Model implementation

As mentioned above, a Hierarchical Fibre Bundle Model (HFBM) was adopted for simulations. The model used here is related to that proposed by Bosia *et al.* (Bosia et al., 2008) and Pugno *et al.* (Pugno et al., 2008). As with all FBMs, the individual fibres have randomly-assigned statistically-

distributed strengths, in our case according to the 2 parameter Weibull distribution (see Fig.2), which is described by the following equation (Weibull, 1939, 1951):

$$P(\sigma) = 1 - e^{-\left(\frac{\sigma}{\sigma_0}\right)^m} \quad (1)$$

The model is based on an Equal-Load-Sharing (ELS) FBM approach, replicated in a hierarchical scheme at various length scales ("levels") to predict from statistical considerations the mechanical behaviour of different hierarchical architectures. Also, in order to model heterogeneous fibrous media, the fibres of each bundle can assume different mechanical properties. The *k*-th fibre type is characterized by a Young's moulus E_k, length l_k, cross-sectional area A_k, and Weibull-distributed fracture strengths, the latter characterized by a scale parameter σ_{0k} and shape parameter m_k. The various types of fibres combine in forming bundles, with complex mechanical behaviour emerging from the mechanical properties and arrangement of the constituent fibres. The specimen's stress-strain behaviour is determined by imposing an increasing displacement and "rupturing" individual fibres in the bundle (i.e. setting their stiffness to zero) when their statistically assigned strength is exceeded. After each fracture event, the load is redistributed uniformly among the fibres in the same bundle as the fractured one (ELS). The bundle strength strength is obtained as the maximum stress value reached in the simulation before failure, i.e. when all parallel fibres of the bundle have failed. Since the fibre strengths are assigned randomly according to the Weibull distribution, results differ for each simulation, and average trends can be derived from repeated simulations.

Hierarchy is implemented as described by Pugno et al. (Pugno et al., 2008) and Bosia et al. (Bosia et al., 2008), schematically illustrated in Fig.3, i.e. the input mechanical behaviour of a level *i=h-1* "fibre" or subvolume is statistically inferred from the output deriving from hundreds of level

h simulations, that of a level *i=h*-2 subvolume from level *i=h*-1 simulations, and so on, down to the lowest hierarchical level *i=1*. Overall, the specimen is modelled as an ensemble of N_1 subvolumes arranged in a bundle. Each of these subvolumes is in turn constituted by N_2 subvolumes, arranged in a bundle as before. This scheme is applied for *h* "generations", up to a level *h* subvolume, which is constituted N_a type "*a*" fibres, N_b type "*b*" fibres, and so on.

Since at every single fibre failure the load is only redistributed among parallel fibres in the "local" bundle, fibre failures in different bundles "interact" only at the next hierarchical level. Thus, comparing hierarchical bundles with the same overall number of fibres and fibre-type percentages, but different hierarchical architectures, amounts to considering different stress redistribution schemes in the material. To simplify the problem, we initially consider only 2 fibre types (*a* and *b*, basically a "matrix" and a "reinforcement" as in composites) and symmetrical structures (each bundle is split into identical bundles at each hierarchical level). Thus, to define the overall bundle, we require the following parameters:

i. Fixed parameters:

N (integer): total number of fibres

E_a and E_b (real number): stiffnesses for fibres *a* and *b*

σ_a and σ_b (real number): Weibull scale parameters for fibres *a* and *b*

m_a and m_b (real number): Weibull shape parameters for fibres *a* and *b*

α (real number): Fraction of type *a* fibres, so that (1-α) is the fraction of type *b* fibres. $0 < \alpha < 1$

ii. Variable parameters:

h (integer)*:* number of hierarchical levels

n_1 , n_2 ...n_{h-1} (integers): number of parallel bundles at hierarchical levels *i=1,2...(h-1)*

n_a (integer): number of parallel bundles of type *a* fibres at the last hierarchical level *i=h*

n_b (integer): number of parallel bundles of type *b* fibres at the last hierarchical level $i=h$

Clearly, for $h>1$, any configuration with $n_i=1$ corresponds to a (h-1) architecture, e.g. a $h=3$ configuration with $n_1=1$, $n_2=2$, $n_a=5$, $n_b=10$ is identical to $h=2$, $n_1=2$, $n_a=5$, $n_b=10$. Therefore, for $h>1$, only configurations with $n_i>1$ are considered.

The number of free parameters depends on the number of hierarchical levels of the considered structures. Two equivalent parameters are the number of type *a* and *b* fibres, respectively, in each bundle at level *h*, i.e. $N_a = \frac{\alpha N}{n_1 n_2 ... n_{i-1} n_a}$ and $N_b = \frac{(1-\alpha)N}{n_1 n_2 ... n_{i-1} n_b}$. The only constraints on the variable parameters are thus that N_a and N_b must be integers. To be able to satisfy this constraint, N needs to be sufficiently large, however this increases computational time, so a compromise is required (typically $N=10^2 \div 10^4$). To avoid excessively time-consuming problems, we can consider initially $h<4$. Each simulation is repeated typically 10^2 times to obtain a mean strength for the considered structure.

To illustrate the procedure, let us consider an example of a 3-level hierarchical structure ($h=3$) with $N=3600$ and $\alpha=0.2$. The chosen mechanical parameters are E_a =1000 GPa and E_b =10 GPa for the Young's moduli, and σ_a =100 GPa σ_b=1 GPa, m_a =2 , m_b=3 for the Weibull scale and shape parameters, respectively. One possible structure with these parameters is shown in Fig.3a. Each "box" in the figure represents a fibre bundle. N_i indicates the number of fibres in each bundle at hierarchical level *i*. A fibre bundle at hierarchical level *i* is equivalent to a single fibre at hierarchical level *i*-1. The distinction between fibres *a* and *b* occurs only at the highest hierarchical level $i=h$. We observe here that a rule of mixtures (Gibson, 2007) for this non-hierarchical configuration yields a strength value of 18.44 GPa. This is because the rule of mixtures is strictly be valid only in the case of simultaneous failure of all fibres in the bundle, which does not occur because of the statistical distribution of the fibre strengths. Typically, in this study the non-hierarchical bundle reaches its

maximum stress (i.e. strength value) when about 50% of its constituent fibres (both *a* and *b*) have failed, leading to the cited strength value of about 9.1 GPa. Consequently, the rule of mixtures is only an upper bound for the mean strength, and does not provide a reliable estimation method in this case.

The strength of hierarchical structures with the same fixed parameters (specified above) is thus compared to the non-hierarchical (h=0) fibre bundle (Fig.3b), the "default configuration", where all fibres are in parallel (N=3600, N_a=720, N_b=2880). The latter configuration has a mean strength of 9.1 GPa. In order to evaluate the variation of bundle strength with hierarchy level and type, all possible configurations are systematically considered for h=1, 2 and 3. The number of possible configurations for the given parameters are 1260 for h=1, 4690 for h=2, and 8641 for h=3, respectively. These numbers are calculated numerically for by determining all n_1 (and n_2 if h=3), n_a and n_b values that give rise to integer values for N_a and N_b. Since single-fibre strengths are randomly assigned based on a Weibull distribution, there is some variability in results for each given configuration. Thus, simulations are repeated 10^2 times for each configuration and a mean value and standard deviation are calculated.

2.2 Hierarchical configuration analysis

Regarding the number of possible configurations for given N and α parameters, some general considerations can be made. Given an even number N of springs, if we denote by $\hat{h}$ the maximum number of levels allowed for N, this number is at most the total number of prime factors (distinct or not) of N. On the other hand, the total number $T(N)$ of different configurations must take into account the number of levels l =2...$\hat{h}$ and for each level l all related configurations. As explained above, at least two fibres must be present in each bundle at every level, or else the configuration

must be considered as belonging to the lower hierarchical level. $T(N)$ is significantly larger than N already for a limited value of $\hat{h}$. Indeed, if we consider the case where N is a power of 2 (namely $N=2^{\hat{h}}$) and $\alpha=0.5$, then we have $\hat{h}$ prime factors all equal to 2 and $\alpha N = (1-\alpha)N = 2^{\hat{h}-1}$. Hence, as N_a and N_b must be integers, we have a feasible configuration if the product $n_1 \cdot n_2 \cdot \ldots (n_l - 1) = 2^j$ for any integer $j = 1, \ldots, \hat{h} - 1$, where $l = 1, \ldots, j$ denotes the number of levels of this configuration. Notice that, as $n_1 \cdot n_2 \cdot \ldots (n_l - 1)$ are all integers, they are also a power of 2. Correspondingly, the total number of different combinations such that $n_1 \cdot n_2 \cdot \ldots (n_l - 1) = 2^j$ summed on all values of $l = 1, \ldots, j$ is exactly 2^j. On the other hand, given j, $N_a = \frac{2^{\hat{h}-j-1}}{n_a}$ and $N_b = \frac{2^{\hat{h}-j-1}}{n_b}$. Hence n_a and n_b can each have any value 2^x with $x = 0, 1, \ldots, \hat{h} - j - 1$. That is, for any given j the number of different combinations of n_a and n_b is given by $(\hat{h} - j)^2$. Overall, by summing upon all $j = 1, \ldots, h^* - 1$, we have that, for $N = 2^{\hat{h}}$, $T(N) = \sum_{j=1}^{\hat{h}-1} 2^j \left(\hat{h} - j\right)^2$. For instance, for $N = 4096 = 2^{13}$, we have $T(N) = \sum_{j=1}^{12} 2^j (13 - j)^2 = 48756$. When N is not a power of 2, the analysis becomes more tedious while the combinatorial explosion is even larger. Just to give a rough idea on the matter, if we suppose one wants to compute the number of different configurations with $\alpha=0.5$, $n_a = n_b = 1$ and number of levels $t = \hat{h}$: then, when the prime factors of N are all identical (as for $N=2^{\hat{h}}$), there is just one configuration. On the other hand, when the prime factors of N are all distinct, for any t-uple of these different factors assigned to t different levels, there are $t!$ different assignments of such factors to the levels (namely all possible permutations of a string with length t) correspondingly inducing $t!$ different combinations.

Based on these observations, for the chosen value of N, the required calculation time for $h \geq 4$ becomes unacceptable, so that in further studies global optimization techniques have to be implemented to maximize strength or other required properties.

3. Results

3.1 Influence of hierarchy

First, we consider the case where only one type of fibre is present, i.e. α =1, to evaluate the influence of hierarchy only. The mean strength in the non hierarchical case here is 43.1 GPa). The allowable configurations are evaluated numerically for the chosen N and h parameters by determining all n_1 and n_a values that give rise to integer values for N_a. The numbering of the configurations is chosen so as to have increasing n_1 values as first criterion, and increasing n_a values as second. Figure 4 illustrates the calculations for the mean strength of the various hierarchical configurations for h=2. A quasi-periodic behaviour is found, related to the n_1 value (as highlighted in Fig.4b). Maximum strength is obtained when maximizing n_a values, for given n_1 values, and the overall maximum strength value is obtained when maximizing both n_1 and n_a (49.7 GPa). This corresponds to hierarchical architectures where bundles constituted of a minimal number of fibres are present, i.e. where minimum stress redistribution occurs in the material in the fracture process. In 9 cases the non-hierarchical mean strength is exceeded, although values are affected by some fluctuations, due to the statistical nature of the simulations.

3.2 *Influence of hierarchy and fibre mixing*

Next, to additionally evaluate the influence of material heterogeneity together with hierarchy, let us consider a "mixed" bundle with N=3600 and α =0.2. As explained above, the strength of various different hierarchical structures is evaluated for h=1, 2, 3. It is important to remember that some statistical variability remains in these data, as simulations are based on randomly assigned single-fibre strengths for fibre types *a* and *b*. Thus, while calculation results do not provide an absolute comparison between different configurations, results are strongly indicative of the relative strength

classification. Mean strength results for h=1 structures are shown in Fig.5. The x-axis represents the considered configurations (again ordered according to increasing n_a), whilst the corresponding strength is plotted on the y-axis in log scale. In this case, the parameters n_a and n_b are sufficient to define the structure, i.e. the different configurations are obtained by simply changing the number of bundles of fibres of type *a* and *b*, since only a single level of hierarchy is present. It is found that in this case hierarchy does not favour maximal strength, because the maximum value (8.71 GPa) is obtained for [n_a =1, n_b =1] (configuration number 1260 in Fig.5), which corresponds to the non-hierarchical case. However, many other "hierarchical" configurations generate similar strength values, and appear as local maxima in the plot in Fig.5. These favourable configurations are those where the number of parallel fibres is maximized, both for type *a* and type *b* fibres, i.e. when n_a and n_b are small and/or not too dissimilar in value. Conversely, the minima appear for large values of n_a or n_b, especially if one of the two exceeds a limiting value (e.g. n_a or n_b > 720). This is highlighted more clearly in Fig.6, where strength values are plotted vs. n_a and n_b in a 3-D plot.

From Figs. 5 and 6 it is apparent that the choice of hierarchical arrangement can lead to a variation in strength of more than an order of magnitude. These observations lead to the consideration that appropriate choice of fibre arrangements can provide the means to obtain tailor-made strength properties, starting from the same constituents. No strength improvement is obtained with respect to the non-hierarchical arrangement, proving that higher-level hierarchy is a key factor in this respect.

Results for h=2 are shown in Fig.7. Here, the number of available configurations is greater (a total of 4690), as a function of the additional parameter n_1. For each given n_1 value, the same quasi-periodic behaviour as in Figs. 4 and 5 is observed as a function of n_1, with maximum strength values obtained for simultaneous large values of n_a and n_b and a similar excursion between maxima

and minima. Thus, these results are consistent with those for α=1 (single fibre type) and h=2, and are in contrast with those for α=0.2 and h=1. This once again proves the importance of hierarchy in determining a qualitatively different behaviour. Here, for increasing n_1 values, the number of available (n_a, n_b) pairs decreases, so that the number of evaluated configurations also decreases and with it the "period" of the oscillations. The number of configurations with a mean strength greater than the non-hierarchical case is 28, with a maximum value of 10.1 GPa, so the introduction of fibre mixing improves the situation with respect to the single-fibre case.

The results obtained with the chosen fibre fraction α=0.2 are qualitatively confirmed for different α values. For example, the case α=0.5 is considered (Fig.8). Clearly, for this α value, the overall mean strength increases because of the larger fraction of "strong" fibres (type *a*), and the mean strength in the non-hierarchical case is 21.8 GPa. Again, a quasi-periodic behaviour is obtained in the configuration space, with the best configurations occurring for various n_1 values when maximizing n_a and n_b values, which corresponds to the cases of greatest of "local" stress redistributions. Some variability remains, due to the statistical nature of the simulations. A greater number of configurations (66) provide an improvement with respect to the non-hierarchical case, with a maximum mean strength value of 26.4 GPa (a 21% improvement). Results are compared in Tables 1 and 2 for h=2 and α=0.2 and α=0.5, respectively.

As shown in Fig.9, results are more complicated for h=3. There remains an oscillating quasi-periodic behaviour as a function of the hierarchical configuration, with local maxima and minima, as a function of the n_1, n_2, n_a and n_b values. As seen in Table 3, where the 10 most favourable configurations are reported, the maxima in mean strength can be found in configurations where at least one of these numbers is maximized, in particular the n_a value, relative to the "strong" type *a* fibres, so the analysis for h=2 is confirmed. Similar to lower-order hierarchies, maximal attained

strength values exceed the non hierarchical case (9.1 GPa) in 34 cases, i.e. a smaller number of cases with respect to h=2 and α = 0.5. This seems to indicate that hierarchical structure is all the more effective for greater high-strength fibre content percentage.

To better analyze these qualitative indications, a systematic study for higher hierarchical levels needs to be carried out. For h>3, it is not possible to analyze all configurations, due to computational time, but preliminary data (not reported) shows that the overall tendency highlighted at h=1, 2, 3 seems to continue, with a greater span between minimal and maximal strength values, and the more favourable configurations being close to those where the number of parallel fibres at level h is maximized.

4. Conclusions

We have presented a systematic procedure to evaluate the influence of heterogeneity and hierarchy in fibre bundle architectures using a Hierarchical Fibre Bundle Model. The first three hierarchical levels have been investigated and a considerable strength variability as a function of hierarchical configuration has been highlighted, with a mean strength improvement of up to 21% with respect to the non hierarchical case. The hierarchical configurations with better strength characteristics are those favouring the more spatially confined stress redistributions during fracture. The results in this paper are promising for the improvement of the performance of artificial bio-inspired architectures and the design of materials with tailor made properties. To derive more specific and quantitative conclusions, in future a wider investigation of the parameter space will be carried out. A greater number of hierarchical levels will be assessed, using global optimization techniques to avoid exceedingly cumbersome calculations and the emphasis will be particularly on metaheuristics based on local improvement techniques (Gendreau and Potvin, 2010). Also, other

properties will be investigated, including stiffness and energy dissipation, in order to address the problem of the simultaneous optimization of different material properties (e.g. strength and toughness), which is a commonplace feature in nature but remains to be effectively achieved in artificial materials.

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Fig4.: a) Simulations results for the mean strength of different fibre arrangements for h=2 and α=1; b) close-up on the first 250 configurations, highlighting the quasi-periodic dependency on the n_1 index. The dotted line indicates the mean strength value for the non-hierarchical case.

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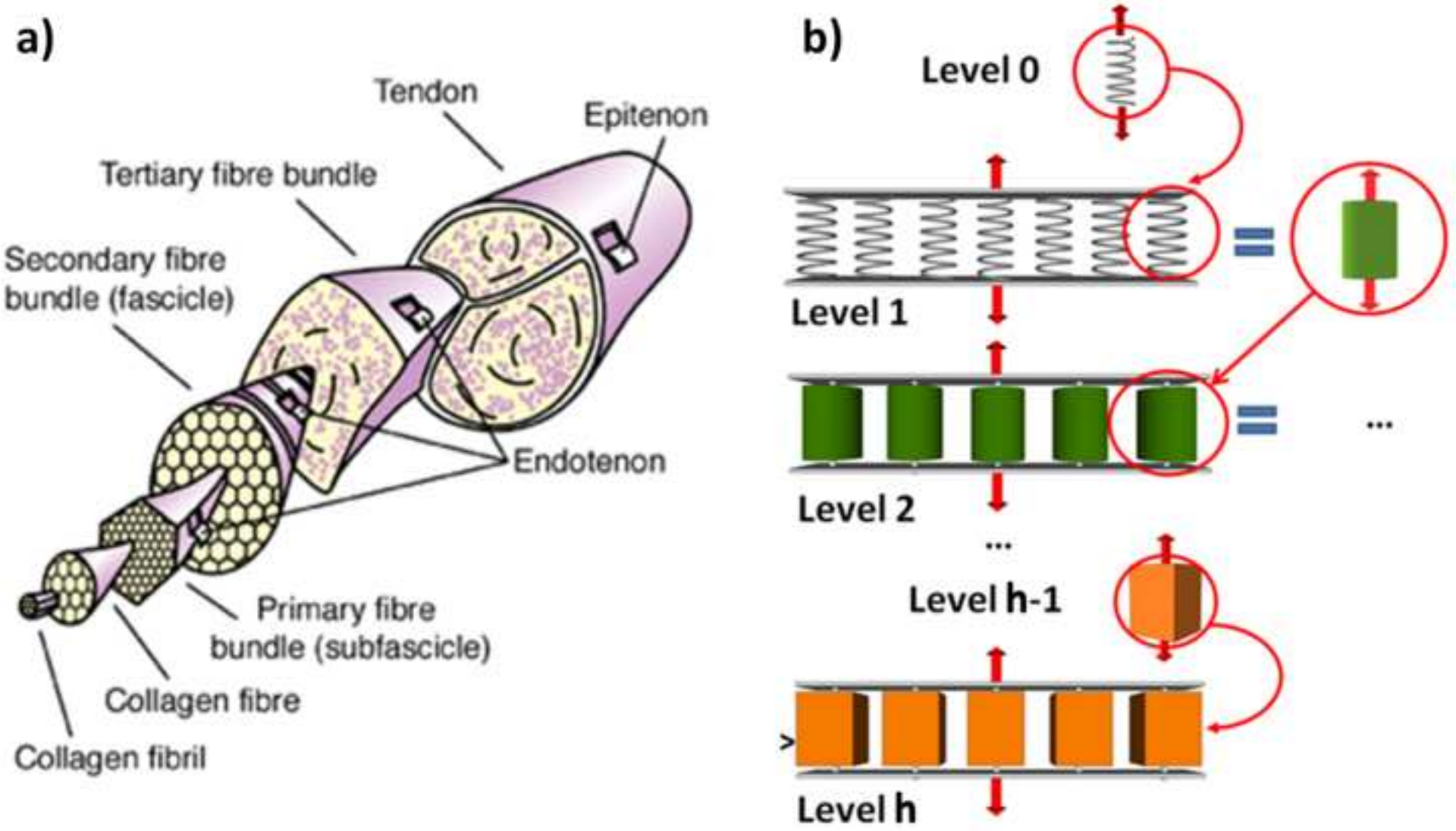

Figure 2

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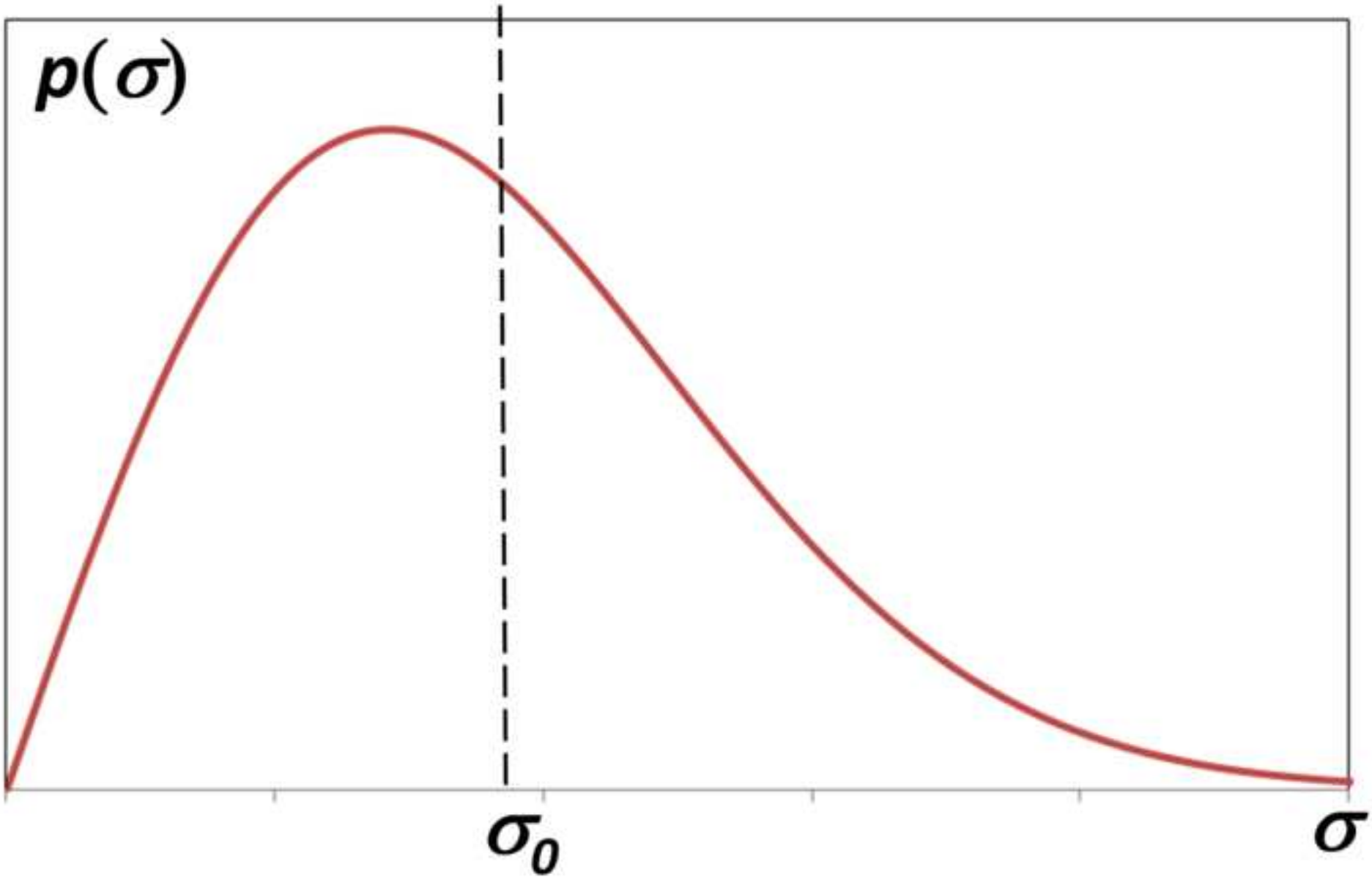

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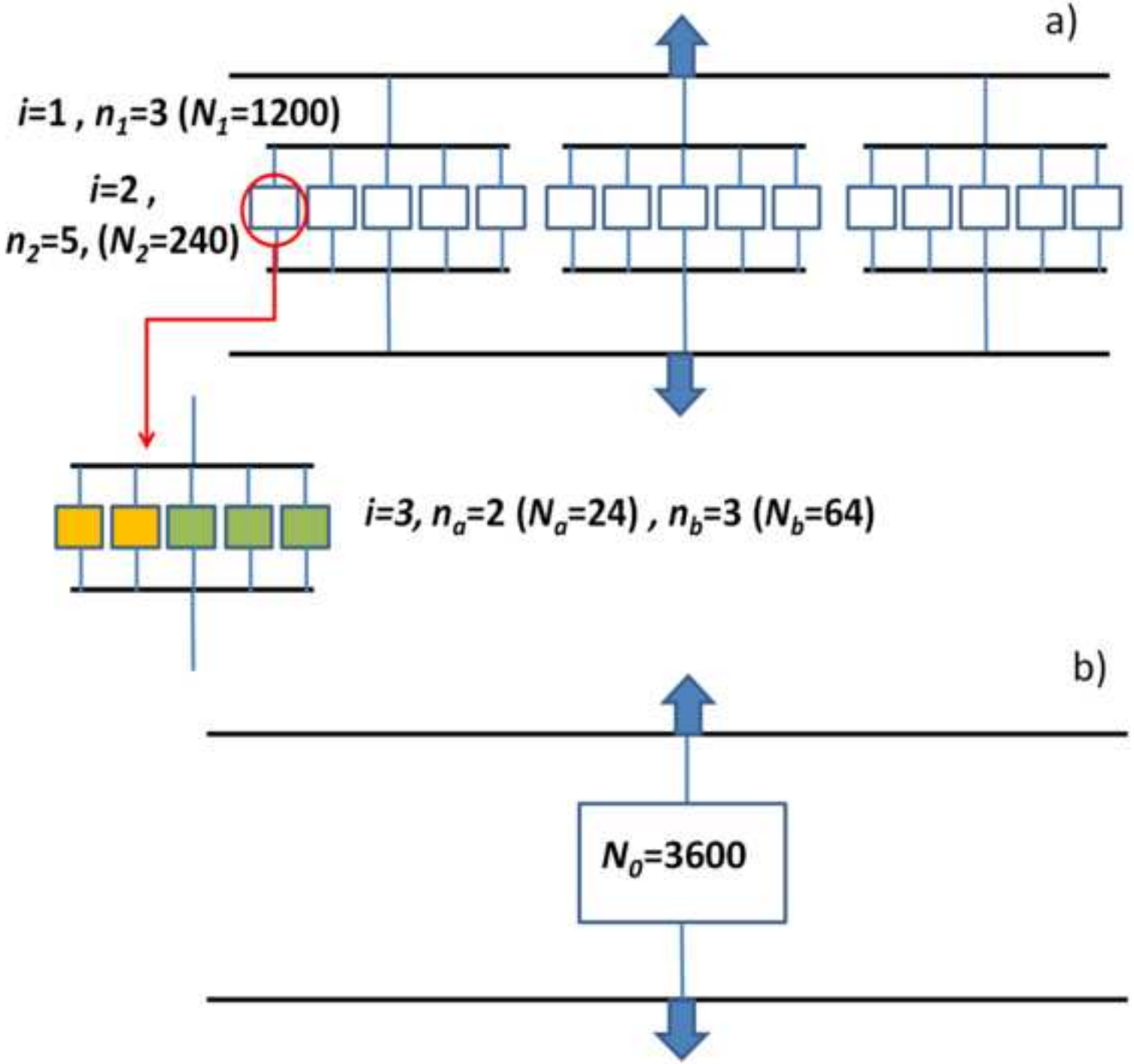

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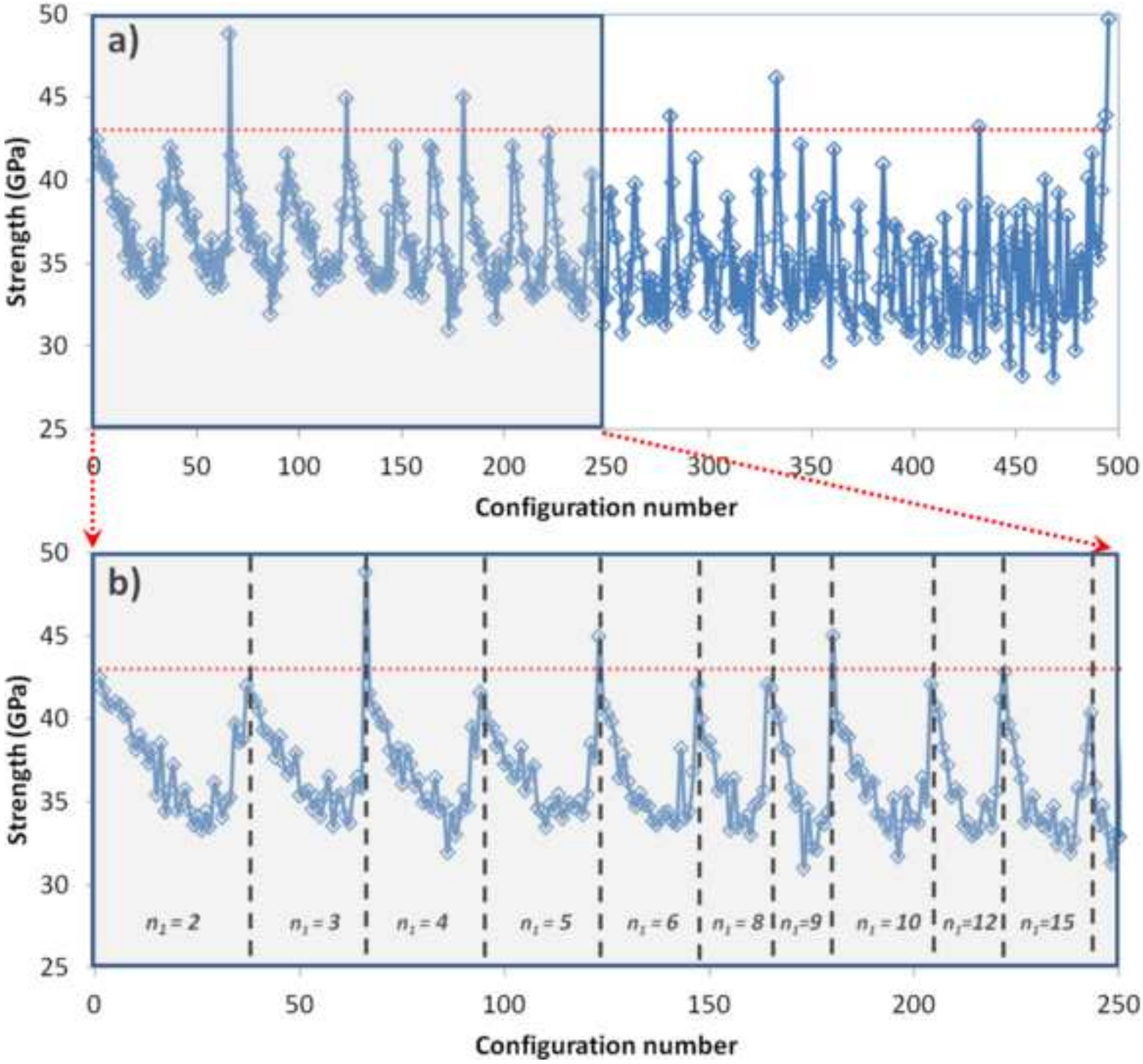

Figure 5

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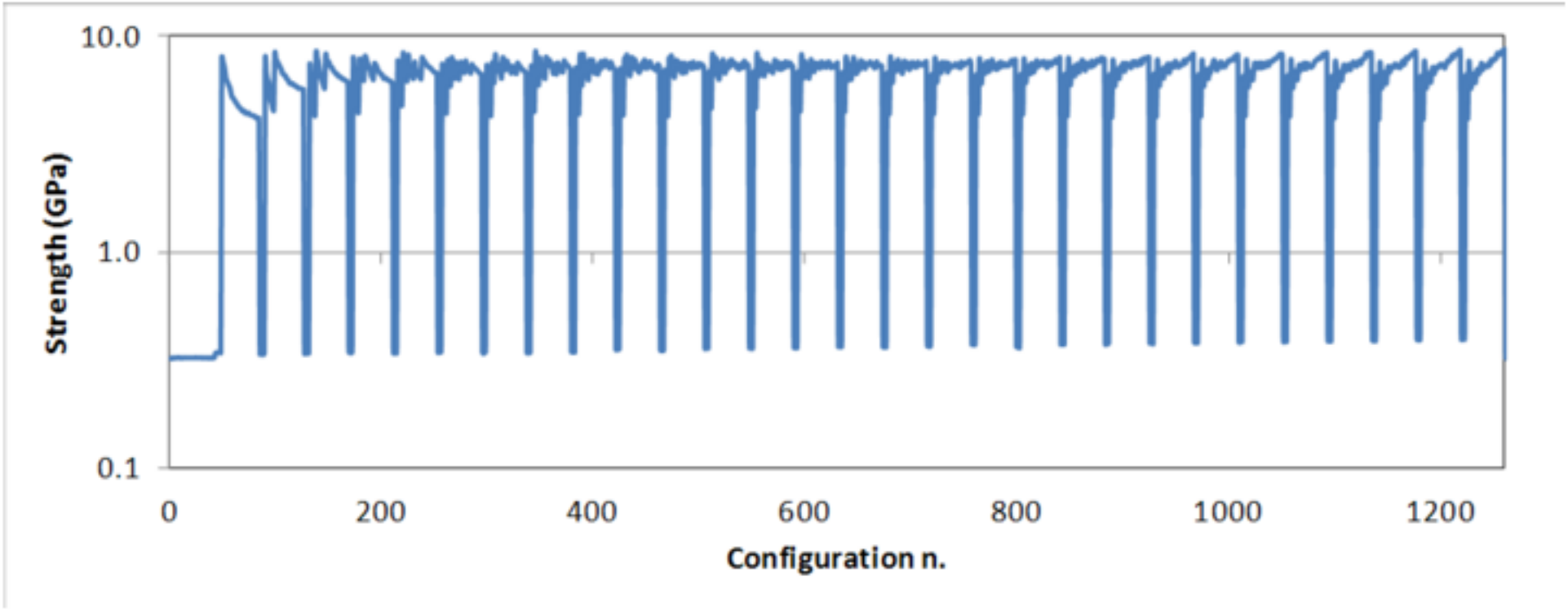

Figure 6

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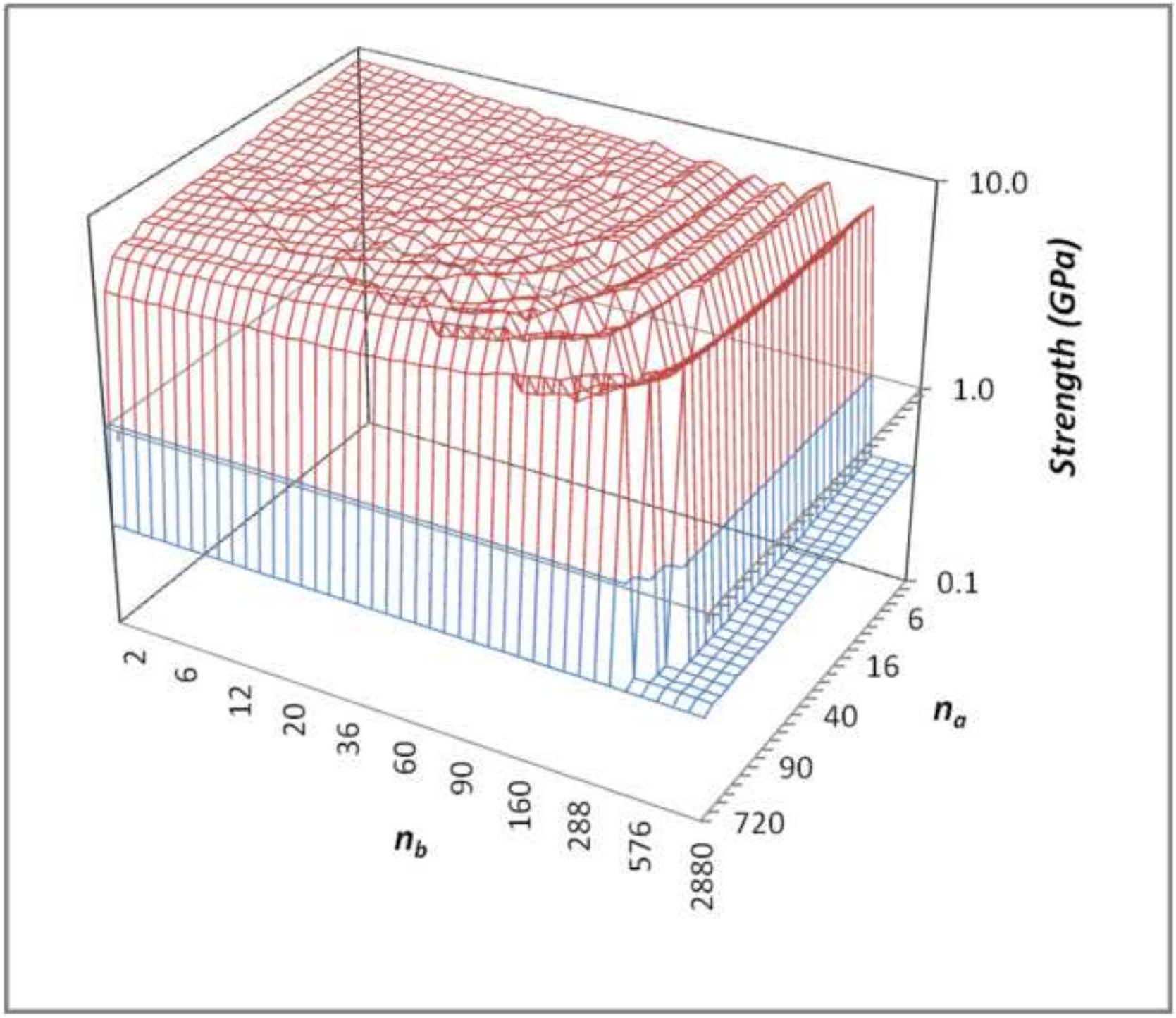

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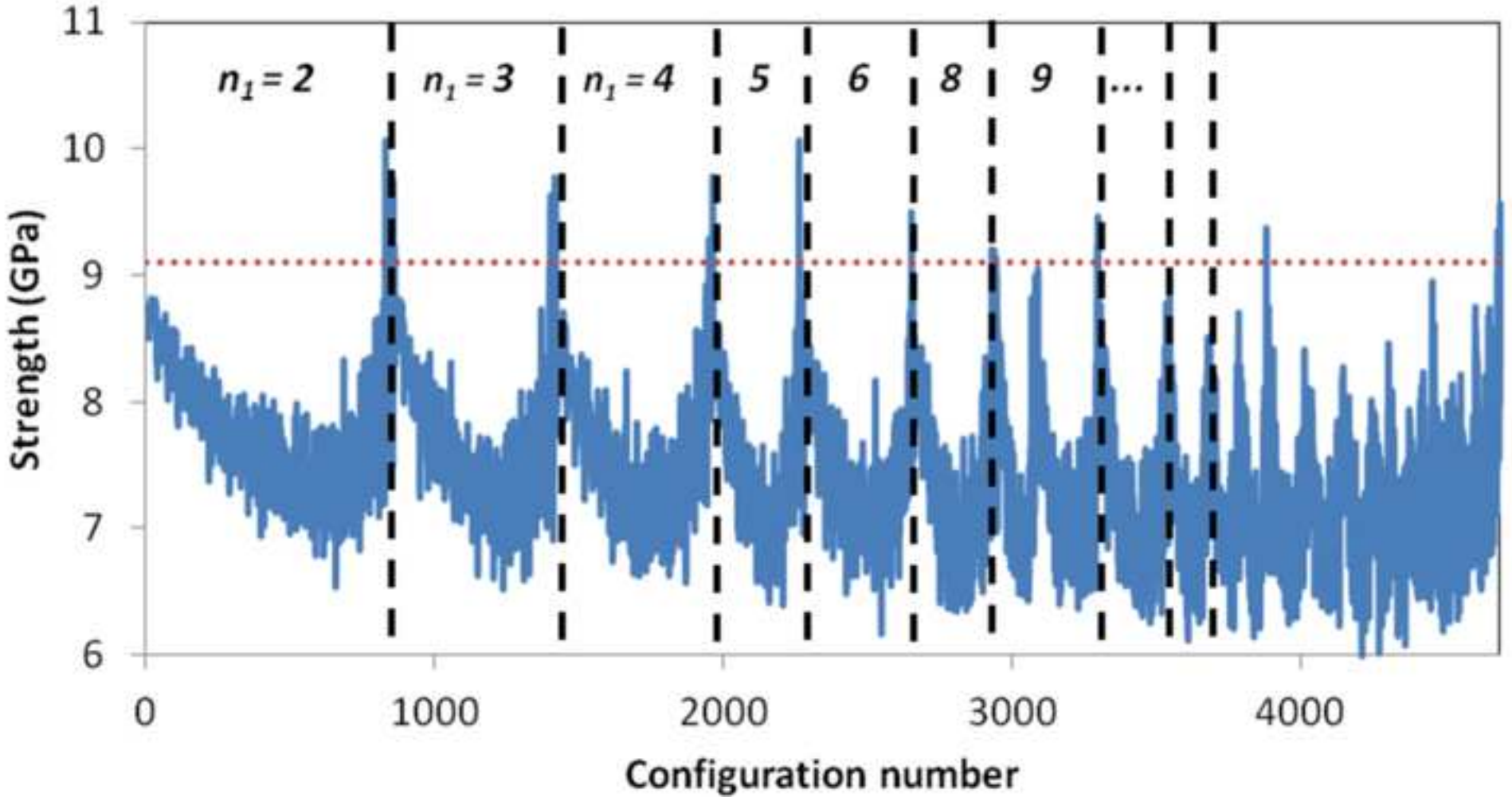

Figure 8

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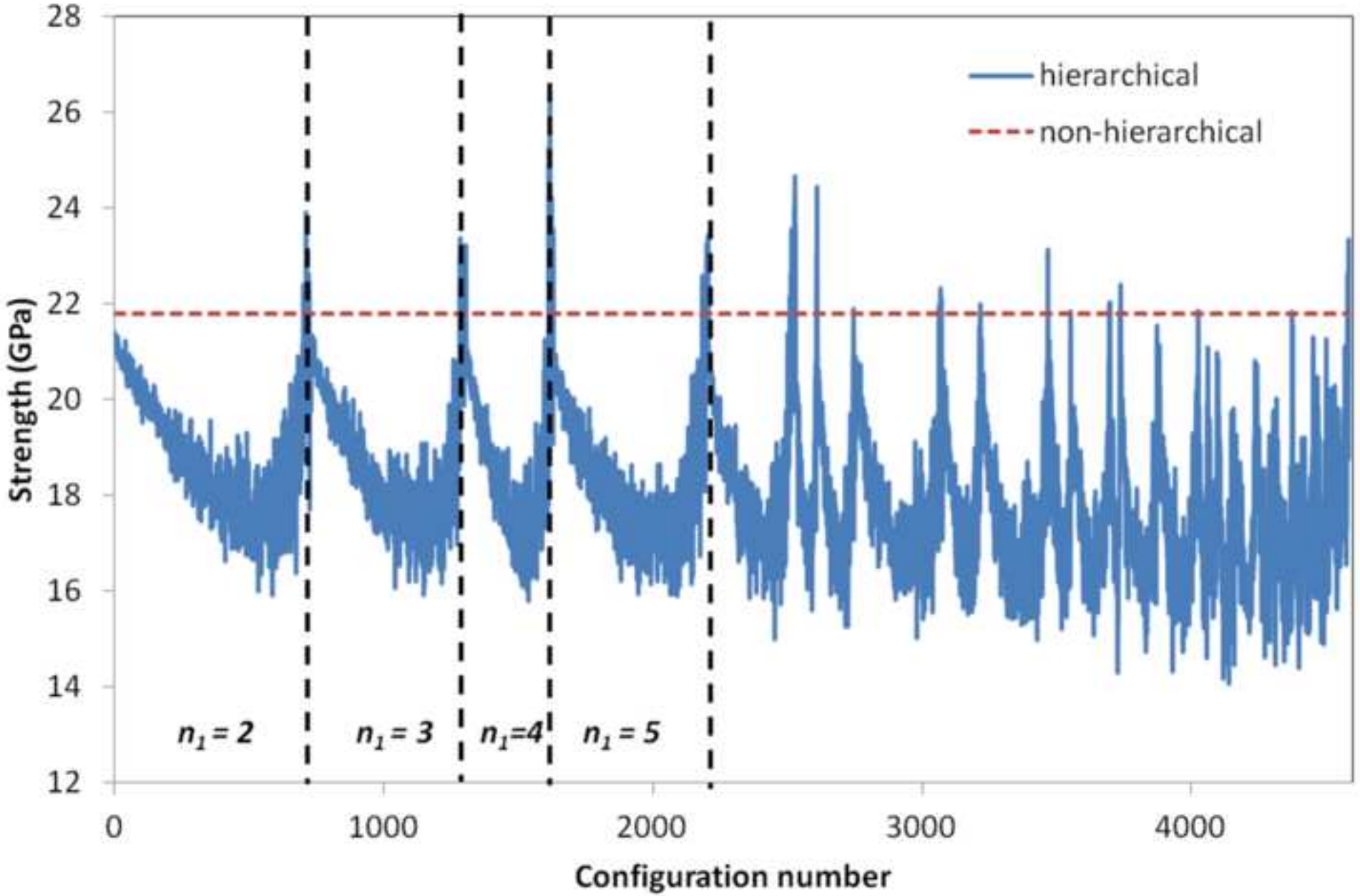

Figure 9

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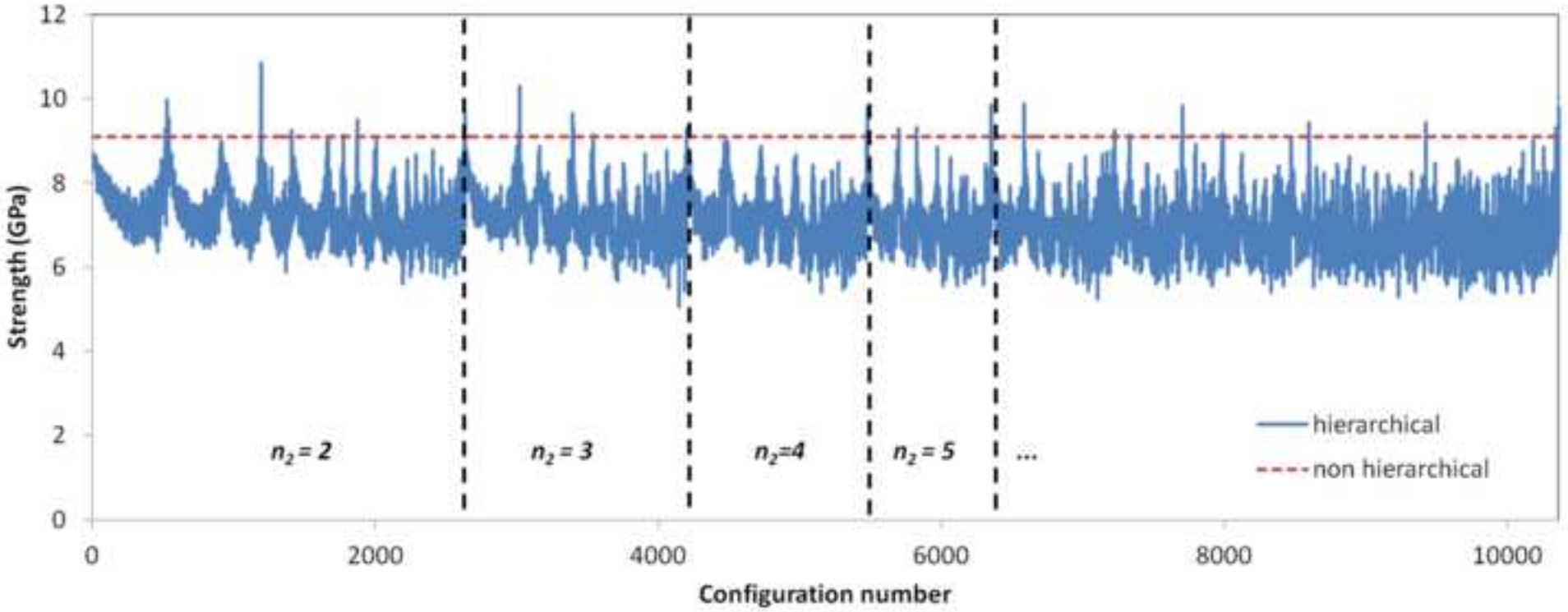

Tables

Table 1

n_1	n_a	n_b	Strength (GPa)
5	144	144	10.07
2	360	720	10.07
2	360	20	9.81
3	240	12	9.78
2	360	10	9.76
4	180	2	9.75
3	240	192	9.64
3	240	6	9.63
3	240	64	9.57
720	1	1	9.57

Table 2

n_1	n_a	n_b	Mean Strength (GPa)
4	450	9	26.4
4	450	2	25.2
6	300	100	24.6
8	225	75	24.4
4	450	45	24.2
6	300	60	24.0
2	900	12	23.9
6	300	300	23.6
6	300	3	23.6
4	450	450	23.5

Table 3

Configuration n.	n_1	n_2	n_a	n_b	Mean Strength (GPa)
1192	2	4	90	4	10.8
3020	3	2	120	480	10.3
527	2	2	180	36	10.0
6582	6	2	60	6	9.9
6354	5	144	1	1	9.9
7704	8	90	1	4	9.8
5480	4	180	1	2	9.8
10359	360	2	1	2	9.8
3393	3	4	60	24	9.7
2634	2	360	1	1	9.6