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PAPER

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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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 millimetres. 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 nanometre 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.*

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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 imply 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 nanometres.[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 hydroxyapatite 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

