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Research Paper

On the mechanical behavior of bio-inspired materials with non-self-similar hierarchy

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ABSTRACT

Biological materials exhibiting non-self-similar hierarchical structures possess desirable mechanical properties. Motivated by their penetration resistance and fracture toughness, the mechanical performance of model materials with non-self-similar hierarchical structures was explored and the distinct advantages were identified. A numerical model was developed, based on microscopic observation of enamel prisms. Computational simulations showed that the systems with non-self-similar hierarchy displayed lateral expansion when subjected to longitudinal tensile loading, which reflected negative Poisson's ratio and potential for greater volume strain energies when compared with conventional materials with positive Poisson's ratio. Employing the non-self-similar hierarchical design, the capability of resilience can be improved. Additionally, the non-self-similar hierarchical structure exhibited larger toughness, resulting from the large pull-out work of the reinforcements. The findings of this study not only elucidate the deformation mechanisms of biological materials with non-self-similar hierarchical structure, but also provide a new path for bio-inspired materials design.

1. Introduction

Hierarchical structure is an important feature of many biological materials (Bechtle et al., 2010; Sen and Buehler, 2011). For example, bone has seven levels of hierarchy (Rho et al., 1998; Weiner and Wagner, 1998). The mineral crystals and protein are the primary building blocks and combine to form the reinforcements of the composite structure in the next hierarchical level. For this system, the composites in the n-th level is identical to that in the $(n+1)$-th level, which is self-similar arrangement (Jager and Fratzl, 2000; Gupta et al., 2006; Ji and Gao, 2004). This structural characteristic is also observed in mineralized tendons where collagen molecules are parallel to each other to constitute the collagen fibrils, which further form fascicles at the micron scale (Arsenault et al., 1991; Puxkandl et al., 2002; Gupta et al., 2004).

The unique microstructure of these biological materials is the secret to their mechanical properties. For instance, the fracture toughness of bone reportedly reaches up to 25 MPa m$^{0.5}$ (Koester et al., 2008), which is about 83 times that of its major constituent, hydroxyapatite (Bajaj et al., 2008). The high toughness of bones is attributed to the hierarchical

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structure, which gives rise to several potent toughening mechanisms (Vashishth et al., 2003; Nalla et al., 2004, 2005). At large length scales, crack bridging by uncracked ligaments in the crack wake are the main toughening mechanism, which reduce the stress intensity in the vicinity of crack tip, leading to the rising crack growth resistance curve (Nalla et al., 2005). At small length scales, collagen fibers acted as ligament bridges to contribute to the enhanced toughness of bone (Nalla et al., 2004). The study by Gao et al. (2005) revealed that the hierarchical design of gecko's feet consisting of setae, further containing many spatulae, enable switching between attachment and detachment easily. The flaw tolerant adhesion of a single fiber resulting from its small size can be achieved in macroscopic material systems, such as the gecko's foot, through hierarchical design (Yao and Gao, 2006). Here, the hierarchical design leads to robust adhesion and explains why geckos can climb the walls and ceilings.

Inspired from the fact that many biological materials adopt hierarchical design, Yao and Gao (2007) proposed a self-similar hierarchical model to develop a good understanding of effects of hierarchy on the mechanical properties of material systems. It was discovered that by utilizing hierarchical design the high strength and short range force (for example van der Waals interaction) can be transformed into low strength but long range force, increasing the work of adhesion for biological adhesive systems. For biological hard tissues, hierarchical design can amplify the fracture energy and make the materials become insensitive to flaws (Yao and Gao, 2007; Gao, 2006). Li et al. (2012a) investigated delamination of materials with hierarchical wavy interfaces and concluded this specially designed interface relieved stress concentration at the crack tip, leading to homogeneous stress distribution in the vicinity of crack tip and thereby promoting fracture resistance of laminated materials. In addition to improving fracture toughness, hierarchical geometry can be used to tailor stiffness and strength. For instance, the study by Li et al. (2012b) showed that both the tensile and shear modulus of suture joints increased with increase in hierarchical levels. Motivated by the structure of natural suture joints, Zhang et al. (2012a) proposed an interfacial strengthening strategy utilizing characteristics of interface roughness, and further increased the interface strength through hierarchical design. The advantages of structural hierarchy have inspired material designs with other unique properties as well. For example, Zhang et al. (2012b) designed multifunctional hierarchical wrinkled surfaces capable of switching between superhydrophobicity and superhydrophilicity under applied strains.

The previous studies have provided important insights into the power of utilizing structural hierarchy to tailor the mechanical properties of materials. Nevertheless, they were based on self-similar hierarchical structures. Non-self-similar hierarchical structures are also widely observed in natural biological materials. The difference in orientations between the reinforcements in the lower and higher length scales is a typical non-self-similar hierarchical structure for hard tissues. For instances, the nanocrystallites in a single mineral platelet, which serves as reinforcement in biocomposites of nacre, exhibit different crystal orientations (Li and Huang, 2009). Enamel is a biocomposite consisting of prisms bounded by a protein-rich region, while at the nanoscale the single prism is also a nanocomposite composed of protein matrix and mineral crystals acting as the reinforcements (White et al., 2001). However, the mineral crystals in enamel prism display a non-uniform arrangement. In the prism head region, the axes of mineral crystals were parallel to the prism axis (i.e., exhibiting self-similar structure), while in the tail region the mineral crystals become gradually inclined to the prism axis (i.e., a non-self-similar structure) (Spears, 1997; An et al., 2012); enamel transitions from a self- to a non-self-similar structure. Non-self-similar hierarchical structures can also be found in scales of alligator gars (Yang et al., 2013), *Arapaima gigas* scales (Lin et al., 2011), and turtle carapaces (Achrai and Wagner, 2013). These observations suggest that non-self-similar hierarchical structures are important, which calls for the development of a greater understanding.

The primary goal of this study was to investigate the mechanical advantages of the materials with the non-self-similar hierarchical structure, as well as to develop a good understanding of the deformation mechanism of this type of materials. The stress transfer mechanism of biological composites with non-self-similar hierarchical structure was explored via numerical simulations, and the corresponding mechanical properties were identified. Additionally, the pull-out behavior of the materials was studied, and the underlying mechanisms were elucidated.

2. Methods

2.1. Microscopic observation

To illustrate the non-self-similar hierarchical characteristic of biological materials, the microstructure of human tooth enamel was investigated using a scanning electron microscope (Helios NanoLab 600i, FEI, USA). Human third molars were collected from a participating dental hospital in Shanghai, China, with approved protocols issued by the International Review Board in Shanghai University. The molars were sectioned axially (EC400, Shengyang Kejing Instrument Co., Ltd. China) and the sectioned slices were polished successively utilizing abrasive papers with mesh of #600–#5000.

The microstructure of enamel reflects the typical feature of the non-self-similar hierarchical structure. The overall cross section of enamel prisms is shown in Fig. 1(a). Enamel prisms have irregular shapes with well defined boundaries. The hierarchical structure of enamel is evident from the oblique view of a prism, as shown in Fig. 1(b). Inside the enamel prism numerous HAP crystals were observed with non-uniform orientations. The HAP crystals are inclined with respect to the prism axis, indicating a typical non-self-similar hierarchy.

2.2. Numerical modeling

The present study was focused on characterizing the deformation mechanisms of the general materials with non-self-similar hierarchical structure, rather than a specific biomaterial. Without loss of generality, a two-scale model biological