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Research Paper

# Protein viscosity, mineral fraction and staggered architecture cooperatively enable the fastest stress wave decay in load-bearing biological materials

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ABSTRACT

One of the key functions of load-bearing biological materials, such as bone, dentin and sea shell, is to protect their inside fragile organs by effectively damping dynamic impact. How those materials achieve this remarkable function remains largely unknown. Using systematic finite element analyses, we study the stress wave propagation and attenuation in cortical bone at the nanoscale as a model material to examine the effects of protein viscosity, mineral fraction and staggered architecture on the elastic wave decay. It is found that the staggered arrangement, protein viscosity and mineral fraction work cooperatively to effectively attenuate the stress wave. For a typical mineral volume fraction and protein viscosity, an optimal staggered nanostructure with specific feature sizes and layouts is able to give rise to the fastest stress wave decay, and the optimal aspect ratio and thickness of mineral platelets are in excellent agreement with experimental measurements. In contrary, as the mineral volume fraction or the protein viscosity goes much higher, the structural arrangement is seen having trivial effect on the stress wave decay, suggesting that the damping properties of the composites go into the structure-insensitive regime from the structure-sensitive regime. These findings not only significantly add to our understanding of the structure-function relationship of load-bearing biological materials, and but also provide useful guidelines for the design of bio-inspired materials with superior resistance to impact loading.

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## 1. Introduction

Bone, tooth, antler and a variety of sea shells are known as load-bearing biological materials, composed of mineral and biopolymer and achieving extraordinary mechanical properties that allow them to support the body weights of animals, resist external forces and impacts, and protect the interior soft organs. Among these load-bearing biological materials, bone is a typical representative which has one of the most delicate hierarchical architectures. Fig. 1 schematically shows the hierarchical architecture of cortical bone, with up to seven levels of structural hierarchy overcrossing the length scales from several nanometers to hundreds of micrometers (Rho et al., 1998; Weiner and Wagner, 1998). With the hierarchical structure, bone acquires superb stiffness, strength and toughness, both of its constituent materials have significant drawbacks, protein being very soft while mineral (mainly hydroxyapatite crystals) being very brittle. To unveil the underlying mechanistic origin, a significant number of experimental, theoretical as well as computational studies have been performed to investigate its constituents, structural organization and structure-based stiffening, strengthening and toughening mechanisms (Currey, 1984, 2002, 2003; Delmas et al., 1984; Dorozhkin and Epple, 2002; Espinosa et al., 2009; Ethier and Simmons, 2007; Fratzl and Weinkamer, 2007; Lipson and Katz, 1984; Meyers et al., 2008; Nyman et al., 2006; Orgel et al., 2006; Rho et al., 1998).

At the nanoscale of bone, it was found that platelet-shape mineral crystals are embedded in a protein matrix in a staggered pattern (Jäger and Fratzl, 2000). The staggered arrangement of reinforcements is also widely seen in other types of load-bearing biological materials at the nanoscale irrespective of the mineral volume fraction and the aspect ratio of the mineral crystals (Mayer, 2005; Meyers et al., 2008; Ten Cate, 2003), and seems to be a generic structural feature of load-bearing biological materials. The staggered nanostructure has been rationalized in the perspective of mechanics and materials science by means of theoretical modeling, computational simulations, and relevant experiments on bio/biomimetic-materials, separately. A number of theoretical models including the tension-shear chain model and some other similar ones, combined with some finite element simulations, were built up to reveal the working mechanism of the staggered nanostructure. It was shown that the staggered structure, in which mineral platelets mainly bear tensile force and protein mainly transfers tensile force between neighboring mineral platelets via shear deformation, can synergize the advantages of both mineral and protein while avoiding their drawbacks, and thus provide good mechanical properties in the terms of stiffness, strength and toughness (Jäger and Fratzl, 2000; Ji and Gao, 2004a; Lei et al., 2013, 2012; Zhang et al., 2010). Interestingly, Guo and Gao (2006) speculated that the staggered arrangement should be a result of optimization and evolution in nature, and tried to prove it by finding the optimal distribution of a hard and brittle inorganic phase in a soft and ductile organic phase through a topological optimization procedure based on genetic algorithm. Their results showed that the staggered arrangement is the stable solution for simultaneously optimizing the stiffness and toughness of the composites. In addition, there have been many successful biomimetic artificial composites with staggered nano-/micro-structure fabricated in the laboratory through the state-of-the-art fabrication technologies such as the layer-by-layer assembly (Tang et al., 2003), ice-templated method (Munch et al., 2008), and 3D printing (Dimas et al., 2013; Espinosa et al., 2011). These biomimetic materials all exhibited mechanical properties much surpassing their conventional counterparts, which further validated the superiority of the staggered arrangement.

The building block of the inorganic component in bone is Calcium Phosphate (Hydroxyapatite) which is a naturally occurring mineral form of calcium apatite. The mineral crystals are nucleated and deposited within the collagen holes and overlap zones, as well as on the surface of collagen fibrils, through a complicate process of bio-mineralization (Landis et al., 1993, 1996a, 1996b). It is worth noting that the thickness of mineral crystals at the bottom level of load-bearing biological materials generally ranges from several nanometers to tens of nanometers. Why does nature construct materials from the nanoscale up? In the perspective of mechanics and materials, it has been argued that 1) nanoscale crystals usually have much higher stiffness and strength

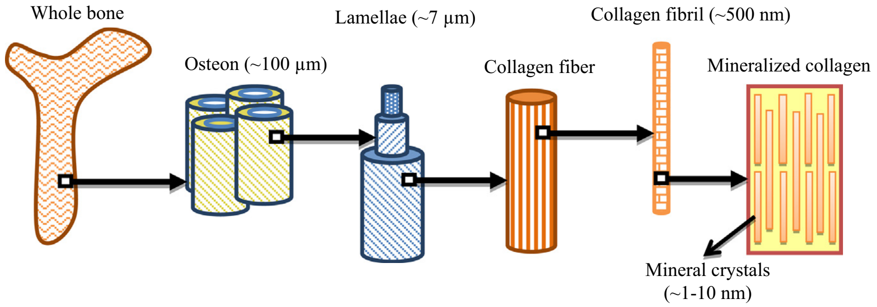

**Fig. 1 – Schematic of bone hierarchical structure. At the highest hierarchical level or the largest length scale, cortical bone is composed of osteones (∼100 μm) which are the main part of the bone structure. Lamellae (∼3–7 μm) form the next structural hierarchy of the bone, composed of collagen fiber arrays (∼1–7 μm). At the next level, mineralized collagen fibrils (∼300 nm to 1 μm) consist of collagen molecules (∼300 nm) and embedded hydroxyapatite nanocrystals (∼1–10 nm). At the smallest level, mineralized collagen structure is known as the nanostructure of the bone (Hulmes, 2002; Jäger and Fratzl, 2000; Meyers et al., 2008; Puxkandl et al., 2002; Rho et al., 1998).**