Crack Propagation and Toughening Mechanism of Staggered Structure of Hydroxyapatite Sheets

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Abstract. Based on the macro-microscopic analysis of the fracture path of cortical bone, it is shown that the crack path of the cortical bone deflected obviously during bending fracture, which indicates that the microstructure of the cortical bone affects the crack propagation of the bone. According to the microstructure of cortical bone, a staggered structure analysis model of hydroxyapatite (HA) sheets was established, and the effects of the structural parameters and volume content of hydroxyapatite sheets on the crack deflection coefficient and fracture toughness were compared and analyzed. The analysis results show that the structural parameters and volume fraction of hydroxyapatite sheets are positively correlated with the fracture toughness of bone. When the bone is fractured, the cracks deflected between the staggered hydroxyapatite layers to dissipate more energy, thereby improving the fracture toughness of the bone. The research results can provide useful guidance for the design and manufacture of high-performance bionic bone composites.

1. Introduction

Bone tissue is a natural composite material, after tens of millions of years selective evolution, it has excellent mechanical properties. It mainly consists of an organic phase with a volume fraction of 32~44% (90% of type I collagen and 10% of non-collagenous proteins, matrix) and an inorganic phase of 33~43% (hydroxyapatite crystals, HA crystal) [1]. Its stiffness is only one order of magnitude lower than that of HA crystals, its strength is 3~5 times that of pure proteins and minerals, and its fracture toughness is much higher than that of HA crystals [2].

At present, artificial high-content ceramic composites are far from the level of natural ceramics [3]. Numerous researches have attributed the excellent mechanical properties of bone biomaterials or bionic bone biomaterials to their unique structures at the nanometer scale, and have proposed many enhancement and toughening mechanisms. Gao et al. [4] proposed a tension shear chain model and analyzed its macroscopic properties through its microstructure. Jager et al. [5] proposed the stiffness analysis of staggered biocomposites and investigated the elastic modulus of bone and shell. Liu et al. [6] investigated an improved model based on Gao’s tension-shear chain model by considering the uneven distribution of stress between staggered layers. Based on previous research, Bar-On et al. [7] researched the general analytical expression of elastic modulus of staggered biocomposites. Duan et al.[8] investigated the mechanical characteristics of interlayer staggered structure, it is shown that the plastic deformation of the interlayer staggered zone mainly resulted from particle breakage and...
directional arrangement under high stress. Taylor [9] investigated the mechanical property of bone, quantifies the ease with which cracks propagate and defines a material's tolerance for pre-existing cracks and other stress concentrating features.

From the above research results, it can be known that the existing research mainly focuses on the staggered structure's influence on the stress distribution and the elastic modulus of material, while the research on the effect of the staggered structure on the fracture toughness of cortical bone is less. In this paper, a staggered structure analysis model was established, which composed of hydroxyapatite sheets and collagen based on microstructure characteristics and crack deflection of cortical bone when it breaks in a direction perpendicular to the axial direction. Quantitative comparative analysis was conducted to explain the bone fracture process, the cracks deflection dissipate energy between the hydroxyapatite layers was investigated, which illustrate the reasons of the greater fracture toughness of bone.

2. Material and Methods

2.1. Macro-micro analysis of cortical bone fracture path
Ten samples were selected along the axial direction of the cortical bone (the angle between the length direction of the sample and the axis is 0°), and a three-point bending test was used to apply external force to bend and fracture along the direction perpendicular to the axis. Then, the scanning electron microscope (TESCAN VEGA 3 LMH SEM) was used to observe and analyze the crack propagation path of the sample under external force. Macro and micro images of the fracture path and crack propagation process of the sample are shown in Figure 1. It can be seen from Figure 1 that during the bending fracture of the sample, the fracture path of the specimen undergoes obvious deflected, the fracture path is tortuous, the fracture path is longer, the fracture surface is rough, and the fracture process dissipates a large amount of fracture energy.

![Figure 1](image1.jpg)

Figure 1. Crack deflection when fractured perpendicular to the axis of the cortical bone. (A) Fracture specimen, (b) crack deflection of microcracks

2.2 Establishment of staggered structure analysis model
Cortical bone is mainly composed of meso-scale osteons and interstitial bone, and osteons are composed of more meso-scale concentric bone lamella. Each bone lamella is wound by mineralized collagen fibers, and its winding direction is at a certain angle with the axis of osteon [10]. The structure of the osteon and the arrangement of HA sheets in mineralized collagen fibers are shown in Figure 2. Figure 2a is a schematic diagram of a single osteon. The concentric layer (bone lamella) in osteon is composed of mineralized collagen fibers arranged in a certain direction, and the thickness of a single layer is about 3-7μm [11]. The mineralized collagen fibers are composed of collagen and
nano-scale HA sheets. The HA sheets with an average size of 50nm×25nm×3nm (Figure 2b) [12]. HA sheets are hard and brittle, and collagen is soft but highly deformable. It is assumed that the geometric parameters of the HA sheets are the same, and they are periodically and uniformly distributed in the matrix (Figure 2c).

According to the arrangement of the hydroxyapatite sheets in mineralized collagen matrix and the staggered structure of the mineralized collagen fibers shown in Figure 2 (c), a representative elementary volume (REV) model is selected as shown in Figure 3. It is assumed that the matrix and the HA sheets are isotropic materials, \( h \) is the thickness of the REV. The HA sheet is a uniform cuboid, and the length of the HA sheet is \( L_p \) and the thickness is \( h_p \). The REV model has a length of \( L \) and a height of \( h \). Based on this, the effect of the structural parameters and volume parameters of hydroxyapatite sheets on the crack inflection coefficient was analyzed. The geometric structure parameters in the REV can be expressed as:

\[
\rho = \frac{L_p}{h_p}, \quad \nu = \frac{2L_p h_p}{L \cdot h} \tag{1}
\]

Where \( \rho \) is the aspect ratio of the crystal plate; \( \nu \) is the volume ratio of hydroxyapatite sheets in RVE.

![Figure 2. Structure schematic diagram of osteon and HA sheets in cortical bone. (a) Osteon model, (b) mineralised collagen fibril, (c) periodic distribution of HA sheets in organic collagen matrix](image)

![Figure 3. Selection of representative elementary volume (REV) in staggered structure](image)

The staggered structure of the hydroxyapatite crystals makes the fractures not break through the layers of the hydroxyapatite in the hard phase, but constantly deflects in the direction of the collagen layer in the soft phase during the fracture process. And lead to multiple micro-cracks, dissipate a lot of fracture energy, thereby improving the fracture toughness of bone. Since the HA sheets with a high strength and stiffness accounts for a large proportion in the material, this is conducive to improving the strength and stiffness of the bone. In order to further study the effect of the structural parameters and volume content of HA sheets on the fracture characteristics of bone, the effect of crack deflection on fracture toughness will be explained from the perspective of fracture energy.
From the definition of work, the work done by force \( F \) is equal to the force \( F \) times the distance moved in the direction of the force. That it is \( W = F \times S \).

Assume that the interfacial resistance to collagen cracking is \( \tau \), crack propagation distance during the crack penetration is \( H \), as shown in Figure 3, then the distance \( H \) can be described as [13]

\[
H = h + \frac{hL_p^v}{2h_p}
\]

Thus, the fracture energy required for the complete fracture of a REV is

\[
W_1 = \tau H = \tau \left( h + \frac{hL_p^v}{2h_p} \right)
\]

When the crack is fractured perpendicular to the surface of the specimen, if no crack deflection occurs, the required fracture energy for complete fracture is

\[
W_1 = \tau \times h
\]

From Eq. (1)-(3), it can be known that the dissipated fracture energy with a crack deflection is greater than the fracture energy without a crack deflection \( \Delta W \).

\[
\Delta W = \tau H = \tau \frac{hL_p^v}{2h_p}
\]

Based on the above formulas, it can be concluded that the larger the thickness of the composite material containing the staggered structure, the more the volume fraction of the HA sheets, the greater the interface resistance when the collagen is cracked, and the larger fracture energy required. Furthermore, the larger the aspect ratio of a single HA sheet, the greater the fracture energy required.

From the above analysis results, it can be seen that the fracture energy is related to the length of the crack, that is, it is directly related to the number of crack deflection. The more the number of crack deflection, the longer the crack, and the more fracture energy required during fracture. Assuming the crack deflection coefficient is \( \lambda \), the expression can be written as:

\[
\lambda = \frac{H}{h} = 1 + \frac{L_p^v}{2h_p} = 1 + \rho \frac{v}{2}
\]

It can be known from the Eq. (5) that the crack deflection coefficient \( \lambda \) is only related to the volume fraction \( v \) of the hydroxyapatite sheets and the aspect ratio \( \rho \) of a single hydroxyapatite sheet.

3. Results and discussion

According to the analysis of the staggered structure, the more the number of crack deflection, the longer the crack propagation path, and the more the fracture energy required during fracture. Based on the relationships between the crack deflection coefficient and the structural parameters and volume content of HA sheets, a quantitative analysis was performed in order to more intuitively reflect the effect of HA sheets on the fracture toughness of cortical bone.

In the staggered structure, when the volume content of HA is constant, the effect of the aspect ratio \( \rho \) of the HA sheets on the crack deflection coefficient is shown in Figure 4. From the analysis results (Figure 4), it can be seen that when the volume content of the HA sheets is constant, the crack deflection coefficient is positively related to the aspect ratio of the HA sheet. And with the increase of HA volume content, the crack deflection coefficient also increased significantly. Figure 5 shows the relationship between the crack deflection coefficient and the volume content of HA sheets in a staggered structure. It can be known from the analysis results that when the aspect ratio of the HA sheets is constant, the crack deflection coefficient is positively related to the volume content of the HA sheet.
The analysis results show that the volume content of HA sheets and their structural parameters affect the fracture toughness of cortical bone. Within a certain range, the larger the aspect ratio, the volume content of HA sheets and the crack deflection coefficient, the larger the required fracture energy, the greater the fracture toughness of the cortical bone, so that the fracture toughness of the cortical bone is also greater.

4. Conclusion
According to the fracture path of the cortical bone during the bending fracture of the sample, the macro-microscopic structure observation and analysis were performed. A staggered structure analysis model of HA sheets was established, and the effects of the structural parameters and volume content of the HA sheets on the crack deflection coefficient and fracture toughness were analyzed. The following conclusions are obtained:

(1) When the cortical bone is bent and fractured, the crack is obviously deflected, which is related to the staggered structure of the hydroxyapatite sheets in the cortical bone.

(2) The thickness of the staggered structure and the structural parameters of HA sheet affect the fracture process of cortical bone. The greater the thickness of staggered structure and the volume fraction of the HA sheet, the greater fracture energy are required.

(3) When the volume content of HA sheets is constant, the crack deflection coefficient is positively related to the aspect ratio of the HA sheets, and the crack deflection coefficient also increases significantly as the volume content of hydroxyapatite sheets increases.

(4) In a certain range, the larger the aspect ratio and volume content of hydroxyapatite sheets, the greater the fracture toughness of cortical bone, which can provide useful guidance for the design and preparation of bionic bone high-performance composite materials.
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