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Research Paper

Multi-phonon scattering processes in one-dimensional anharmonic biological superlattices: Understanding the dissipation of mechanical waves in mineralized tissues

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ABSTRACT

The scattering of elastic waves in a one dimensional phononic (PnC) crystal composed of alternate collagen and hydroxy-apatite constituent layers is studied. These superlattices are metaphors for mineralized tissues present in bones and teeth. The collagen is treated as an open system elastic medium with water content which can vary depending on the level of stress applied. The open system nature of the collagen–water system leads to a non-linear stress–strain response. The finite difference time domain method is employed to investigate the propagation of non-linear mechanical waves through the superlattice. The spectral energy density method enables the calculation of the non-linear vibrational wave band structure. The non-linearity in the mechanical response of the collagen–water system enables a variety of multi-phonon scattering processes resulting in an increase in the number of channels for the dissipation of elastic waves and therefore for the dissipation of mechanical energy. These results provide an explanation for the relationship between bone fragility and decreased hydration.

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1. Introduction

Mineralized biological tissues, such as bone and tooth, are hierarchical composite structures composed of a stiff hydroxyapatite (HAP) mineral phase, a compliant proteinaceous collagen phase, and water. At the nanoscale, bone and teeth are constituted of a periodic assembly of alternating regions of collagen and HAP in a hydrated environment with a repeat unit cell size of 67 nm (Ten Cate 1980). This periodic composite structure, forming a one dimensional (1-D) superlattice, is believed to be responsible for the remarkable strength and toughness of these biological materials (Jager and Fratzl 2000; Currey 2002; Deymier-Black et al. 2010). At the micrometer scale, mineralized tissues exhibit a large network of interconnected porosity, tubules in dentin and canaliculi and lacunae in bone, which allow for the transfer of nutrients, waste, and water throughout the tissue (Boyde and Lester 1967; Dillaman et al. 1991). This porosity allows bone and teeth to remain in equilibrium with water, maintaining hydration of the tissues. Water molecules exhibit a variety of different interactions with the HAP and collagen including the formation of water-bridges within the collagen helix, filling channels within the HAP, and surface hydration of the collagen and HAP phases (Bertinetti et al. 2007; De Simone et al. 2008; Ravikumar and Hwang 2008). Three-point bend and notch testing indicate that hydration has a significant impact on the mechanical properties of mineralized tissues resulting in increased elastic moduli as well as decreased toughness and loss of plastic behavior (Kahler et al. 2003; Kruzic et al. 2003; Nyman et al. 2006). Hydration increases the non-linear behavior of collagen as well as its elastic modulus while increasing its toughness (Grant et al. 2008; Gevorkian et al. 2013). It is theorized that these changes in the collagen mechanical behavior due to water content are the major cause for the changes in overall tissue mechanics. Although these tests provide important information about how hydration affects material properties, they provide no information about the dynamic properties of these tissues.

The number one cause of fracture in both bones and teeth is trauma or impact. To avoid fracture, the energy from sudden impacts must be dissipated in order to limit the formation of stress concentrators that can lead to tissue failure and fracture. It is therefore essential to understand how the composite structure of bone and teeth reacts to these dynamic loads which are so often responsible for fracture. In this study, it is theorized that the non-linear behavior of collagen in equilibrium with water provides a means of filling vibrational band gaps that arise from the periodicity of the HAP/collagen structure assembly. Under high load (high deformation) conditions, the non-linearity of the mechanical response of the collagen–water system may open up multiphonon scattering channels leading to a filling of the band gaps in the vibrational band structure of the HAP/collagen superlattice. We model the collagen/water system within the context of the thermodynamics of stressed open systems. This model leads to non-linear stress–strain relationships of the collagen. The non-linear model of the open collagen/water systems is incorporated into a dynamic model of the HAP/collagen superlattice. The propagation of elastic waves through that superlattice is investigated using the finite difference time domain (FDTD) method in conjunction with the spectral energy density (SED) method. Vibrational (phonon) wave band structures of the HAP/collagen/water system show that at high amplitudes, vibrational waves can interact with each other through multiphonon scattering channels that can fill the band gaps inherent in the band structure of elastic superlattices. This band-gap filling facilitates the propagation of vibrations in a larger range of frequencies, providing an effective mechanism for dissipation of mechanical energy, thus limiting the risk of failure.

This paper is organized as follows. In Section 2, we introduce the model of the open collagen/water system. This system is modeled within the context of the thermodynamics of stressed solid solutions. This model results in the formulation of the non-linear stress–strain response of the collagen/water solid, i.e. a strain dependent elastic modulus. Section 2 also presents the model of the HAP/collagen periodic structure in the form of a 1-D superlattice as well as the methods of FDTD and SED that are employed to investigate the dynamic response of the superlattice. In particular we focus on the calculation of the vibrational/phonon band structure of the superlattice as a function of the energy (amplitude) of the propagating elastic waves. In Section 3, we report the results of the calculations and provide an analysis of the effect of the non-linear elastic modulus. In particular, it is shown that high amplitude (energy) waves can interact with each other through multiple phonon scattering processes. These interactions lead to the opening of new channels for the dissipation of the elastic energy over a wider range of frequencies compared to the case of low energy waves.

Finally, conclusions are drawn in Section 4as to the relationship between the observed behavior and bone fragility due to decreased hydration.

2. Models and methods

2.1. *Thermodynamics of stressed solid solution*

To address the problem of the mechanical behavior of bone material in the presence of water, we develop the chemo-mechanical equations of states of materials that can adsorb fluids under stress based on the work of (Larche 1985; Larche and Cahn 1985). The total internal energy of the material is obtained as an integral of an internal Helmholtz energy density f:

$$E = \int_{V'} f' \; dV' \tag{1}$$

where the energy density is given by

$$f' = f'(T, \varepsilon, ..., c_I^{'}, ...) \tag{2}$$

the prime indicates that all densities are relative to the reference state for measuring strain. T, ε, and c_I' are the temperature, strain and molar density of chemical constituent "I", respectively. We consider K variable the chemical species in the chosen materials. The differential form of Eq. (2) takes the form:

$$df'(T, \varepsilon_{ij}, c_I^{'}, ...) = s'(T, \varepsilon_{ij}, c_I^{'}, ...)\; ds' + \sigma_{ij}(T, \varepsilon_{ij}, c_I^{'}, ...)\; d\sigma_{ij} + \sum M_{I,K}(T, \varepsilon_{ij}, c_I^{'}, ...)\; dc_I^{'} \tag{3}$$