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1 Introduction

Nature has always been a source of inspiration for the development of novel materials, devices, and architecture in human society. Load-bearing biological materials in nature, such as bone, teeth, antler, and shell, are composites of biopolymers and minerals [1–6]. While biopolymers are soft and tough, minerals have the opposite and complementary properties of being stiff and brittle. Through well-ordered and efficiently organized structures of constituent materials, biocomposites have achieved remarkable mechanical properties with stiffness and strength comparable to those of mineral, and toughness several orders of magnitude larger than that of mineral [6–8]. The structure-property relationships of load-bearing biological materials such as bone and shell are of great interest to mechanicians, materials scientists, biologists, even clinicians, and intense current efforts are being made to develop bio-inspired synthetic composites.

Recent years have witnessed many studies aimed at developing synthetic composites that mimic the microstructures of biological materials. Tang et al. [9] synthesized nanostructured artificial nacre by sequential deposition, sometimes referred to as layer-by-layer assembly, of polyelectolytes and montmorillonite clay. The resulting "brick-and-mortar" composites exhibited tensile stiffness (\sim 10 GPa) and strength (\sim 100 MPa) comparable to those of natural nacre and bone. Using a similar technique of layer-by-layer assembly, Podsiadlo et al. [10] fabricated thin films with montmorillonite clay platelets aligned in polyvinyl alcohol,

Elastic Bounds of Bioinspired Nanocomposites

Biological materials in nature serve as a valuable source of inspiration for developing novel synthetic materials with extraordinary properties or functions. Much effort to date has been directed toward fabricating and understanding bio-inspired nanocomposites with internal architectures mimicking those of nacre and collagen fibril. Here we establish simple and explicit analytical solutions for both upper and lower bounds of the elastic properties of biocomposites in terms of various physical and geometrical parameters including volume fraction and moduli of constituents, and aspect ratio and alignment pattern of stiff reinforcements. Numerical analyses based on the finite element method are performed to validate the derived elastic bounds. [DOI: 10.1115/1.4023976]

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treated them with glutaraldehyde to enhance the inorganic/organic interface, and made strong (strength $\sim 400 \text{ MPa}$) and stiff (modulus ~106 GPa) nanocomposites. Bonderer et al. [11] chose Al_2O_3 and chitosan as components and developed nacre-like platelet reinforced polymer films with high strength (\sim 315 MPa), stiffness $(\sim 10 \text{ GPa})$, and ductility (rupture strain $\sim 21\%$). Munch et al. [12] adopted an ice-templating technique to fabricate ceramic-based nanocomposites with multiscale structural features mimicking nacre and excellent properties, including a toughness 300 times higher than that of their ceramic component. The fact that bioinspired nanocomposites made in the laboratory can often attain mechanical properties comparable to or surpassing their natural counterparts is mainly attributed to broader choices of constituent materials for synthetic systems rather than better structural design. In contrast, biological materials rely on relatively weak inorganic compounds available in nature for biomineralization to achieve amazing mechanical properties in all aspects, which is still far beyond the state-of-the-art of man-made materials. In other words, there is ample room for further improvement of the existing bioinspired composites through optimal design of micro- and nanostructures. In addition, many novel materials such as carbon nanotube and graphene have great potential for applications as key constituents in bioinspired structural or functional composites [13-15].

Previous studies have shown that the staggered nanocomposite structure in biological materials plays a crucial role in determining their mechanical properties [8,16–27]. Figure 1(a) shows a generic nanostructure of shell in which reinforcements are aligned in a "brick-and-mortar" pattern, while Figs. 1(b) and 1(c) exhibit the stairwise staggering patterns of collagen fibrils in bone and tendon. Early studies were primarily focused on mechanical

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Fig. 1 Typical staggered nanostructures observed in loadbearing biological materials. (a) The staggered alignment of mineral platelets in shell, also referred to as the "brick-andmortar" structure; (b) the stairwise alignment of hydroxyapatite nanocrystals in mineralized collagen fibrils of bone; (c) the "stairwise staggering" alignment of tripocollagen molecules in collagen fibrils of tendon, with a periodic unit cell comprising four tripocollagen molecules. Note that the staggered alignment of mineral crystals in bone is a result of mineralization of collagen fibrils.

behaviors of the "brick-and-mortar" structure [6-8,16,26-28]. Gao and his coworkers [8,16] developed a tension-shear chain model and illustrated how the "brick-and-mortar" structure can lead to high stiffness and toughness at the same time. Zhang et al. [29] proposed a two dimensional framework to determine elastic properties of an arbitrarily staggered structure (including "brickand-mortar" and "stairwise staggering" as special cases) based on the principle of minimum complementary energy, in which the effects of reinforcement distribution and pattern are represented by four dimensionless factors. Lei et al. [30] extended the two dimensional framework of Zhang et al. [29] to three dimensions and predicted the variations of several mechanical properties with respect to the aspect ratio of mineral reinforcements. While these recent studies have revealed new insights on the mechanical properties of biocomposites, a serious limitation is that the principle of minimum complementary energy can only be used to derive a lower bound of elastic properties. The present paper is devoted to establishing both upper and lower bounds for the elastic properties of nanocomposites with staggered nanostructure inspired from biocomposites such as shell, bone, and collagen fiber.

The paper is outlined as follows. In Sec. 2, we first derive an upper bound based on the principle of minimum potential energy, and then a lower bound based on the principle of minimum complementary energy. The derived bounds are compared to results from finite element analysis in Sec. 3 and main conclusions are summarized in Sec. 4.

2 Derivation of Elastic Bounds

There exist several micromechanics models to capture effective elastic properties of composites. One prominent example is the Mori–Tanaka method, which has been shown unable to provide an accurate assessment for the effective Young's modulus associated with the staggered nanostructure of biocomposites [8,29]. The failure of conventional micromechanics models could be attributed to their intrinsic limitations in properly accounting for the effects of microstructure. In the following, we will apply principles of minimum potential and complementary energies to derive the upper and lower elastic bounds based on kinematic relationships between different components in the staggered nanostructure of biological materials.

2.1 Upper Bound. Figure 2(a) shows a bio-inspired staggered nanostructure whose unit cell consists of *n* platelets aligned in a staggered manner in the matrix, each column of the platelets shifting upward by a distance equal to 1/n the length of the platelet relative to its left adjacent column (Fig. 2(b)). Under an applied elongation Δ , a kinematically admissible strain field is assumed as follows (Fig. 2(c)): The hard platelets are assumed to experience a uniform strain ε_* , and the shear strains in the soft matrix are assumed to have two characteristic values,

$$\begin{cases} \gamma^{L} \\ \gamma^{U} \end{cases} = \frac{\Delta - L\varepsilon_{*}}{h_{m}} \begin{cases} \frac{n-1}{n} \\ \frac{1}{n} \end{cases}$$
(1)

where Δ is the total elongation of the unit cell, *L* is the length of platelets, and $h_m = h(1/\phi - 1)$ is the thickness of the soft layer, *h* being the thickness and ϕ the volume fraction of platelets. It can be shown that this strain field is roughly compatible and satisfies the displacement boundary conditions.

For a unit cell consisting of *n* layers of platelets (seen in Fig. 2), the strain energy stored in the platelets is $nE_p\varepsilon_*^2hL/2$, and that in the matrix is



Fig. 2 Generalized stairwise staggering alignments mimicking collagen fibrils. (*a*) Schematic nanostructure of biocomposites; (*b*) the unit cell (solid box) with staggering number *n*, and (*c*) the assumed kinematically admissible strain field under a longitudinal elongation Δ .

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Fig. 3 (a) The assumed static equilibrium stress field in the stairwise staggered nanostructure under a longitudinal elongation Δ , (b) the distribution of shear traction along a hard platelet, and (c) the distribution of normal stress in the platelet

$$n \left[\frac{G_m}{2} \left(\gamma^L \right)^2 \cdot \frac{L}{n} \cdot h_m + \frac{G_m}{2} \left(\gamma^U \right)^2 \cdot \frac{(n-1)L}{n} \cdot h_m \right]$$
$$= \frac{G_m L (\Delta - L\varepsilon_*)^2}{2h_m} \cdot \frac{n-1}{n}$$

Thus the potential energy in the unit cell is

$$\Pi = n \left[\frac{E_p h L \varepsilon_*^2}{2} + \frac{G_m L (\Delta - L \varepsilon_*)^2}{2h_m} \cdot \frac{n-1}{n^2} \right]$$
(2)

Minimization of the potential energy with $d\Pi/d\varepsilon_* = 0$ gives

$$\varepsilon_* = \frac{(n-1)G_m L\Delta}{(n-1)G_m L^2 + n^2 E_p h_m h} \tag{3}$$

Substituting Eq. (3) into Eq. (2) yields an effective Young's modulus,

$$E = \frac{d^2 \left[\frac{\Pi}{Ln(h+h_m)} \right]}{d\Delta^2} \cdot L^2 = \left[\frac{1}{\phi E_p} + \frac{n^2(1-\phi)}{(n-1)\phi^2 \rho^2 G_m} \right]^{-1}$$
(4)

where $\rho = L/h$ is the aspect ratio of platelets.

According to the principle of minimum potential energy, Eq. (4) should be an upper bound for the effective stiffness. Interestingly, for the regular "brick-and-mortar" structure n = 2, Eq. (4) becomes $E = \left[(1/\phi E_p) + (4(1-\phi)/\phi^2 \rho^2 G_m) \right]^{-1}$, which is identical to the corresponding stiffness formula derived by Gao et al. [8,16] using a different approach.

2.2 Lower Bound. The principle of minimum complementary energy can be used to derive a lower bound for the elastic stiffness of biocomposites. In this case, a static equilibrium stress field is assumed (Fig. 3),

$$\left\{ \begin{array}{c} \tau^{L} \\ \tau^{U} \end{array} \right\} = \tau_{*} \left\{ \begin{array}{c} \frac{n-1}{n} \\ \frac{1}{n} \end{array} \right\}$$
 (5)

where τ_* is to be determined. It can be shown that with the above shear stress filed, the total force on every hard platelet is in equilibrium in the axial direction. The normal stress in a hard platelet can then be obtained from force equilibrium as

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$$\sigma(z) = \begin{cases} \frac{1}{h} \left(\tau^L + \tau^U \right) \cdot z, & z \leq \frac{L}{n} \\ \frac{1}{h} \left(\tau^L + \tau^U \right) \cdot \frac{L}{n}, & \frac{L}{n} < z < \frac{(n-1)L}{n} \\ \frac{1}{h} \left(\tau^L + \tau^U \right) \cdot (L-z), & z \geq \frac{(n-1)L}{n} \end{cases}$$
(6)



Fig. 4 The finite element mesh in a representative unit cell for (a) the regular "brick-and-mortar" structure with n = 2 and (b) a "stairwise staggering" structure with n = 5

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Fig. 5 Variations of elastic bounds for Young's modulus as a function of the aspect ratio of platelets for (a) the "brick-and-mortar" structure with staggering number n = 2 and (b) the "stairwise staggering" structure with n = 5. In both (a) and (b), a moderate volume fraction $\phi = 50\%$ of mineral is adopted. The dash-dotted line is the upper bound while the solid line is the lower bound. The filled square data points are from finite element analysis. The predictions from the Voigt and Mori–Tanaka models are shown as the dashed and dotted lines, respectively.

The strain energy stored in hard platelets is thus $n \int_0^L (\sigma^2(z)/2E_p)hdz = ((3n-4)L^3/6n^2hE_p)\tau_*^2$, and that in the matrix is $(n-1)h_mL\tau_*^2/2nG_m$. The total axial elongation force on the unit cell is $((n-1)/n)L\tau_*$, which can be derived from Eq. (6), and the complementary energy in the unit cell is therefore

$$\Pi_C = \frac{(3n-4)L^3}{6n^2hE_p}\tau_*^2 + \frac{(n-1)h_mL\tau_*^2}{2nG_m} - \frac{n-1}{n}L\tau_*\Delta$$
(7)

Minimization of the complementary energy with $d\Pi_C/d\tau_* = 0$ yields

$$\tau_* = \frac{\Delta}{\frac{(3n-4)L^2}{3n(n-1)hE_p} + \frac{h_m}{G_m}}$$
(8)

The average stress in the composite is

$$\sigma = \frac{1}{\frac{n(3n-4)}{3(n-1)^2} \frac{1}{\phi E_p} + \frac{n^2}{n-1} \frac{1-\phi}{\rho^2 \phi^2 G_m}} \frac{\Delta}{L}$$
(9)

Dividing σ by average strain Δ/L gives the Young's modulus of the composite as

$$E = \left[\frac{n(3n-4)}{3(n-1)^2}\frac{1}{\phi E_p} + \frac{n^2}{n-1}\frac{1-\phi}{\rho^2\phi^2 G_m}\right]^{-1}$$
(10)

Compared to the upper bound given in Eq. (4), the lower bound only has a different coefficient for the first term in the bracket. For the regular "brick-and-mortar" structure n = 2, Eq. (10) becomes $E = \left[\frac{4}{3}(1/\phi E_p) + (4(1-\phi)/\rho^2\phi^2 G_m)\right]^{-1}$, which has been derived previously by Zhang et al. [29].

3 Comparison With Finite Element Analysis

Finite element analysis (FEA) of the bio-inspired staggered structure was carried out to verify the elastic bounds derived in the previous section. Figure 4(*a*) shows an FEA model of a unit cell of the "brick-and-mortar" structure (n = 2) under axial displacement loading. Periodic boundary conditions are applied in all directions. The FEA model of a unit cell for a stairwise staggering structure with n = 5 is shown in Fig. 4(*b*). Here, the unit cell is defined as the smallest structure unit for which periodic boundary conditions hold. In all FEA calculations, the modulus ratio between hard platelets and soft matrix is taken to be $E_p/G_m = 1000$, and the Poisson ratio is taken to be $\nu_p = \nu_m = 0.3$ for both hard and soft



Fig. 6 Variation of elastic bounds for Young's modulus as a function of the aspect ratio of platelets for (*a*) the "brick-and-mortar" structure with staggering number n = 2 and (*b*) the "stairwise staggering" structure with n = 5. A large volume fraction $\phi = 90\%$ of mineral is adopted. The dash-dotted line is the upper bound while the solid line is the lower bound. The filled square data points are from finite element analysis. The predictions from the Voigt and Mori–Tanaka models are shown as the dashed and dotted lines, respectively.

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phases. Note that the longitudinal gap between adjacent platelets is at least one order of magnitude less than the platelet length so that its influence is negligible.

Figures 5(a) and 5(b) plot the variations of normalized stiffness bounds together with the FEA results with respect to the platelet aspect ratio ρ for two different staggering patterns n=2, 5,respectively. The volume fraction of hard platelets is taken to be $\phi = 50\%$, similar to that of bone. It can be seen that the FEA results are sandwiched between the derived elastic bounds except for some slight deviations for aspect ratios less than 100. The deviations at small platelet aspect ratios can be attributed to some simplified assumptions in our theoretical framework, such as the assumption of pure shear in the soft matrix. Comparison of Figs. 5(a) and 5(b) shows that the elastic bounds become tighter as the stagger number n increases. This can be easily understood from comparing Eqs. (4) and (10); the latter indeed becomes closer and closer to the former as the stagger number n becomes larger and larger, since $n(3n-4)/3(n-1)^2 \rightarrow 1$ as $n \rightarrow \infty$. The predictions by the classical Voigt and Mori-Tanaka models, representative of typical micromechanics methods, are also included in Fig. 5. The Voigt model can be expressed as $E = \phi E_p + (1 - \phi) E_m \approx \phi E_p$ with the Young's modulus of the soft matrix E_m omitted. The Mori–Tanaka model [31] is one of the most popular Eshelby-type methods for estimating the effective elastic properties of composites. For simplicity, here the rectangular shape of the platelets is represented by an ellipsoid so that the formulae of effective moduli derived by Zhao and Weng [32] can be used directly. Figure 5 suggests that, while our upper bound gradually approaches the Voigt limit as the aspect ratio increases, the prediction from the Mori-Tanaka model substantially underestimates the FEA results. This limitation of conventional micromechanics methods, including the Mori-Tanaka model, stems from their homogenization schemes that are not able to account for the influence of specific micro/nanostructures.

Figures 6(a) and 6(b) plot the variations of normalized stiffness bounds together with FEA results with respect to the platelet aspect ratio ρ for two different staggering patterns n=2, 5, respectively, and a large volume fraction of hard platelets, $\phi = 90\%$, similar to that of shell. Overall, the trends shown in Fig. 6 for $\phi = 90\%$ are almost the same as those in Fig. 5 for $\phi = 50\%$. The elastic bounds are seen to provide reasonably tight envelops for the FEA results, especially when the platelet aspect ratio is larger than 100. In contrast, the predictions from the Mori-Tanaka model are substantially lower than the FEA results.

4 Conclusions

It is well known that the well-ordered nano/microstructures of biological materials play key roles in their mechanical properties. Conventional homogenization-based micromechanical models fail to provide a reliable estimate for the Young's modulus of biocomposites, as they primarily account for the influence of volume fraction, shape, and orientation of inclusions but not the specific nano/microstructures of materials. In contrast, our derived elastic bounds here are based on models developed specifically for shell-/ bonelike biological materials. Comparison with finite element analysis (FEA) suggests that the derived elastic bounds provide a tight envelop for the Young's modulus of biological or bioinspired composites. While our models are simplistic in several aspects, the elastic bounds given in Eqs. (4) and (10) provide simple and powerful analytical formulae for the stiffness of biological and bio-inspired composites. In addition, the elastic bounds can also be similarly derived for more complicated alignments of platelet (two-dimensional problem) or fiber (three-dimensional problem) reinforcements.

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