Mechanical properties of unidirectional nanocomposites with non-uniformly or randomly staggered platelet distribution

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\begin{abstract}
Unidirectional nanocomposite structures with parallel staggered platelet reinforcements are widely observed in natural biological materials. The present paper is aimed at an investigation of the stiffness, strength, failure strain and energy storage capacity of a unidirectional nanocomposite with non-uniformly or randomly staggered platelet distribution. Our study indicates that, besides the volume fraction, shape, and orientation of the platelets, their distribution also plays a significant role in the mechanical properties of a unidirectional nanocomposite, which can be quantitatively characterized in terms of four dimensionless parameters associated with platelet distribution. It is found that, compared with other distributions, stairwise and regular staggering of platelets produce overall the most balanced mechanical properties, which might be a key reason why these structures are most widely observed in nature.
\end{abstract}

1. Introduction

Natural biological materials such as bone, teeth, and nacre are nanocomposites made of protein and mineral (Currey, 1977, 1984; Jackson et al., 1988; Landis, 1995; Landis et al., 1996; Rho et al., 1998; Weiner and Wagner, 1998). The protein is compliant (Young's modulus 50–100 MPa) with relatively high ductility and high strength-to-modulus ratio (20–50%) (Jager and Fratzl, 2000; Ji and Gao, 2004a). The mineral, on the other hand, is stiff (Young's modulus 50–100 GPa) with relatively low fracture toughness ($\leq 1$ MPa m$^{1/2}$) and low strength-to-modulus ratio (0.1–0.6%) (Currey, 1980; Jackson et al., 1988; Ji and Gao, 2004a). Natural biological materials, however, are stiff and tough compared to their constituents. Their Young's modulus is $\sim 50$ GPa for shell and $\sim 20$ GPa for bone (Jackson et al., 1988; Jager and Fratzl, 2000; Ji and Gao, 2004a), comparable to that of mineral. The strength of natural biological materials is 100–300 MPa for shell and 100 MPa for bone (Jackson et al., 1988; Jager and Fratzl, 2000; Ji and Gao, 2004a; Menig et al., 2000, 2001), comparable to the strength of mineral. The fracture toughness of natural biological materials ranges from 2 to 7 MPa m$^{1/2}$ (Jackson et al., 1988; Ji and Gao, 2004a; Norman et al., 1995), which is several orders of magnitude higher than that of mineral (see, e.g. Ji and Gao (2004a) for a collection of mechanical properties). It is remarkable that natural biological materials can achieve mechanical properties that are far superior compared to their constituents.

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Previous studies have indicated that the staggered micro-/nanostructures in natural biological materials play a crucial role in their superior mechanical properties (Anup et al., 2007; Currey, 1977; 1984; Fratzl et al., 2004; Gao et al., 2003; Gao, 2006; Gupta et al., 2004, 2006; He and Swain, 2008; Ji and Gao, 2004a, b; Kamat et al., 2000; Laraia and Heuer, 1989; Liu et al., 2009, 2006; Nukala and Simunovic, 2005; Weiner and Wagner, 1998). Based on prior studies on the tensile behaviors of natural biological materials (Jackson et al., 1988; Jager and Fratzl, 2000; Kotha et al., 2001), Gao et al. (2003) and Ji and Gao (2004a) developed a tension–shear chain model to illustrate how natural biological materials achieve high elastic modulus and other mechanical properties. As shown by the macroscopic and TEM images of shell and bone in Fig. 1(a)–(d), natural biological materials adopt a generic nanostructure whose load transfer path follows a one-dimensional tension–shear chain shown in Fig. 1(e). The original tension–shear chain model of Gao et al. (2003) and Ji and Gao (2004a) is based on the following assumptions:

(i) The elastic modulus of protein is two to three orders of magnitude smaller than that of the mineral such that the protein cannot transfer any normal stress between the neighboring platelets; the load in the longitudinal direction \((z)\) can only be transferred via shear in protein.

(ii) The thickness \((h)\) of mineral is one or two orders of magnitude smaller than its length \(L\) such that the deformation in mineral is essentially one dimensional [i.e., depending only on \(z\), Fig. 1(e)].

(iii) The distribution of mineral repeats itself after two mineral platelets.

(iv) The neighboring platelets overlap half of their length along the longitudinal direction.

Furthermore, the gap between mineral platelets in the longitudinal direction \((z)\) is much smaller than the mineral length \(L\).

The tension–shear chain model with half overlapping length, hereafter referred to as regular staggering, can be used to interpret many underlying mechanisms in biocomposites. However, strictly speaking, there also exist other patterns of staggering in biocomposites, such as the stairwise staggering shown in Fig. 1(b) (Fratzl et al., 2004; Meyers et al., 2006, 2008; Rho et al., 1998). Moreover, non-uniform or even random platelet alignment can occur in biomimetic materials with “mortar–brick” micro-/nanostructure (Bonderer et al., 2008; Tang et al., 2003). One question then arises, why does nature choose regular and stairwise staggering? A possible answer is that these structures produce optimal mechanical properties compared to other distribution patterns. In comparison between regular and stairwise staggering, we note that the regular staggering has obvious weak cross-section planes as denoted by the dashed line in Fig. 1(e) such that the composite strength is only half of that of a continuous layering composite, while the stairwise staggered structure avoids the co-alignment of longitudinal gaps of platelets in concentrated weak planes and is therefore expected to have higher strength. The tension–shear chain model of Gao et al. (2003) and Ji and Gao (2004a) does not account for non-uniform platelet alignment and cannot address any question regarding the platelet distribution in biological and biomimetic nanocomposites.

The purpose of this paper is to investigate the effects of non-uniform or random staggering (alignment) of platelets on the mechanical properties of biocomposites. Since there are many possible staggered structure patterns, an interesting question is why regular staggering and stairwise staggering are most frequently observed in nature. From a fabrication point of view, it would seem that random staggering does not require high precision control and has a clear advantage from the entropic point of view. Why is it not selected by nature? The present study is motivated by these questions and by

![Fig. 1. The tension–shear chain model of biological nanocomposites. (a) Macroscopic image of bone and (b) TEM image of its generic nanostructure. (c) Macroscopic image of shell and (d) TEM image of its microstructure. (e) Schematic illustration of the tension–shear chain model proposed by Gao et al. (2003) and Ji and Gao (2004a).](image-url)
the general interest to understand the basic design principles of biological systems. In particular, we will extend the tension–shear chain model to account for non-uniform or random staggering of platelets in a unidirectional composite structure. The paper is outlined as follows. In Section 2, the assumption (iv) above is relaxed to extend the tension–shear chain model to the case of an arbitrary overlap length \((1 - \xi)L\), as shown in Fig. 2, where \(0 \leq \xi < 1\) represents platelet distribution, and \(\xi = \frac{1}{2}\) degenerates to regular staggering considered by Gao et al. (2003) and Ji and Gao (2004a). The elastic modulus and strength are obtained analytically in terms of \(\xi\), as well as other properties of the platelet and matrix. In Section 3, we further relax the assumption (iii) above to extend the tension–shear chain model to \(n > 2\), where \(n\) is the number of platelets within each period, as shown in Fig. 3. In stairwise staggering, each platelet is shifted up by a distance \(L/n\) with respect to the platelet on its left. The solutions obtained then pave the way for the study of general non-uniform platelet distribution in Section 4, and random platelet distribution in Section 5. The implications of the present study on natural biological materials, including the optimal platelet distribution to achieve high stiffness, strength, and resilience (i.e., the elastic energy storage capacity) of composite materials, are discussed in Section 6.

2. Tension–shear chain model for regular staggering with offset

The tension–shear chain model is extended to a regularly staggered structure with an arbitrary overlap length between neighboring platelets in this section. As shown in Fig. 2, all platelets are assumed to be parallel to the \(z\) direction, and are shifted to every other neighbor by a distance \(\xi L\), which gives the overlap length \((1 - \xi)L\), where \(L\) is the platelet length,
0 ≤ \xi < 1; \xi = \frac{1}{2} degenerates to the case considered by Gao et al. (2003) and Ji and Gao (2004a). The present configuration will be referred to as regular staggering with offset.

Fig. 2(b) shows a unit cell of the model. The thickness of the matrix layer is related to the platelet thickness h and volume fraction \phi by \((1−\phi)/\phi\)h. The unit cell is subjected to a displacement \(A\), as shown in Fig. 2(c). The shear stresses in the matrix, denoted by \(\tau^t\) and \(\tau^s\), respectively, for the upper and lower parts of the matrix [Fig. 2(c)], are assumed to be constant. The force equilibrium requires

\[(1−\xi)\tau^t−\xi\tau^s = 0.\]  

(1)

The normal stress in the (first) platelet is obtained from the force equilibrium as

\[
\sigma_p = \begin{cases} 
\frac{2}{h} (\tau^s z, & 0 ≤ z ≤ \xi L, \\
\frac{2}{h} \tau^t (L−z), & \xi L < z ≤ L.
\end{cases}
\]  

(2)

The strain energy in the matrix is

\[
2 \frac{(1−\phi)h}{\phi} \left(\xi L \left(\xi\xi^t\right)^2 + (1−\xi)L \left((1−\xi)\xi\xi^t\right)^2\right) \frac{2}{G_m}
\]

where \(G_m\) is the shear modulus of the matrix. The complementary energy of the unit cell is

\[
\Pi_c = 2h \int_0^L \frac{\sigma_p^2}{2E_p} dz + 2 \frac{(1−\phi)h}{\phi} \left(\xi L \left(\xi\xi^t\right)^2 + (1−\xi)L \left((1−\xi)\xi\xi^t\right)^2\right) \frac{2}{G_m} − FA
\]

where \(F\) is the axial force on the platelet given by

\[
F = 2\tau^t \xi L.
\]  

(4)

Minimization of the complementary energy by imposing \(d\Pi_c/d\xi^t = 0\) gives the shear stress \(\xi\) as

\[
\tau^t = \frac{A}{4 \xi L^2 + (1−\phi)h} \frac{1−\phi}{\phi(1−\xi)G_m}.
\]  

(5)

The average stress \(\sigma\) in the composite is

\[
\sigma = \frac{F}{2h + 2(1−\phi)h/\phi} = \frac{\phi F}{2h}
\]

which can be rewritten via Eqs. (4) and (5) as

\[
\sigma = \frac{A}{3\phi E_p + \phi^2(1−\phi)\rho^2 G_m}.
\]  

(6)

where \(\rho = L/h\) is the platelet aspect ratio. Since \(A/L\) represents the average strain \(\varepsilon\), Young’s modulus is obtained from the ratio of average stress to average strain,

\[
E = \frac{1}{3\phi E_p + \phi^2(1−\phi)\rho^2 G_m} = \frac{1}{\phi E_p} \frac{1}{\frac{1}{3} + \frac{1}{\phi^2(1−\phi)\rho^2}}.
\]  

(7)

where

\[
\alpha = \frac{\phi \rho^2 G_m}{3(1−\phi)E_p}
\]  

(8)

is a parameter combining the effects of platelet volume fraction \(\phi\) and aspect ratio \(\rho\), as well as the matrix and platelet elastic moduli \(E_m\) and \(E_p\). Young’s modulus, normalized by \(\phi E_p\), is a universal function of \(\alpha\) and \(\xi\), which represents platelet distribution. For \(\xi = \frac{1}{2}\), Eq. (7) degenerates to Gao et al. (2003) and Ji and Gao (2004a) except for the factor \(\frac{1}{\phi}\). This is because Eq. (7) is derived based on the principle of minimum complementary energy, which is known to give rise to a lower bound for Young’s modulus. Eq. (7) gives an improved estimate of Young’s modulus compared to the finite element calculations by Ji and Gao (2004a). This improvement can be partly understood by considering the limit of \(\phi\) approaching 1, which corresponds to a material with vanishing organic matrix, i.e. with only platelets left. In this limit, the platelets are not continuous. The gaps between the platelets tend to soften the material similar to internal cracks, which would reduce the effective Young’s modulus below \(E_p\). We should point out that our analysis is intended only for cases when the
tension–shear chain model is approximately valid, which requires that the aspect ratio fall below a critical value (Chen et al., 2009). For very large aspect ratios, the applied load will be carried by the stiff platelets alone and that the shear stress in the soft matrix vanishes except near the ends of the mineral platelets. This stress field obviously violates the basic assumption of the tension–shear chain model that the shear stress in the soft matrix remains uniform.

Fig. 4 shows Young’s modulus $E$, normalized by the platelet volume fraction and modulus $\phi E_p$, versus $\xi$ for a large range of $\alpha$ from 0.021 to 83. Due to the symmetry with respect to $\xi = \frac{1}{2}$, only the curves for $0 \leq \xi \leq \frac{1}{2}$ are plotted. Young’s modulus achieves a maximum value of

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

at $\xi = \frac{1}{2}$, which corresponds to the case of regularly staggered platelets (without offset) as studied by Gao et al. (2003) and Ji and Gao (2004a). The limit $\xi = 0$ corresponds to 100% overlap of platelets, which leads to weak planes, and therefore vanishing Young’s modulus. Moreover, it is worth noting that the moduli associated with regularly staggering of long platelets are insensitive to the non-dimensional overlap length $\xi$ when it is close to $\frac{1}{2}$.

The composite strength is defined as the average stress in the composite at which the maximum shear stress in the matrix reaches the matrix shear strength $\tau_m^{\text{critical}}$, or the maximum normal stress in the platelet reaches the platelet strength $s_p^{\text{critical}}$,

$$\max\{\tau_L, \tau_U\} = \tau_m^{\text{critical}}$$

or

$$2\tau_L \frac{\xi}{\rho} = s_p^{\text{critical}}$$

These give the composite strength as

$$\sigma_{\text{critical}} = \begin{cases} \phi \sigma_p^{\text{critical}} \frac{\rho}{2 \rho'_{\text{critical}}}, & \rho \leq \rho'_{\text{critical}}, \\ \phi \sigma_p^{\text{critical}} \frac{1}{2}, & \rho > \rho'_{\text{critical}}, \end{cases}$$

where

$$\rho'_{\text{critical}} = \frac{1}{1 - 1 - 2 \xi} \frac{\sigma_p^{\text{critical}} \tau_m^{\text{critical}}}{s_p^{\text{critical}}}$$

is the critical aspect ratio that separates matrix failure ($\rho \leq \rho'_{\text{critical}}$) from platelet failure ($\rho > \rho'_{\text{critical}}$). The composite strength, normalized by $\phi \sigma_p^{\text{critical}}$, is a universal function of the platelet aspect ratio $\rho$ and $\rho'_{\text{critical}}$, and the latter depends on the platelet-to-matrix strength ratio and $\zeta$ (representing platelet distribution).

Fig. 5 shows the composite strength $\sigma_{\text{critical}}$ normalized by the platelet volume fraction and strength $\phi \sigma_p^{\text{critical}}$, versus $\xi$ for the platelet-to-matrix strength ratio $\sigma_p^{\text{critical}} / \tau_m^{\text{critical}} = 10$ and platelet aspect ratio $\rho$ ranging from 8 to 500. The solid line represents platelet failure, and is given by a universal relation $\sigma_{\text{critical}} = \phi \sigma_p^{\text{critical}} / 2$. The dotted lines represent matrix failure, and they depend on the platelet aspect ratio $\rho$. The regular staggering ($\zeta = \frac{1}{2}$) generally gives the maximum composite
strength, which is \( \sigma_{\text{critical}} = \phi \sigma_p / 2 \) if the aspect ratio \( \rho \) exceeds \( \sigma_{\text{critical}} / \tau_{\text{critical}}^m \). The limit \( \xi = 0 \) gives vanishing strength due to the co-alignment of platelet gaps in weak planes. However, if \( \rho > \sigma_{\text{critical}} / \tau_{\text{critical}}^m \) then all \( \xi \) could achieve the maximum composite strength \( \sigma_{\text{critical}} = \phi \sigma_p / 2 \). Similar to the moduli, the strengths of the regularly staggered composites with long platelets (aspect ratio \( \rho > \sigma_{\text{critical}} / \tau_{\text{critical}}^m \)) are insensitive to the non-dimensional overlap length \( \xi \) when it is close to \( \frac{1}{2} \), which implies some robustness of the biocomposites.

3. The tension–shear chain model with stairwise staggering

In this section, the tension–shear chain model is extended to the case of a periodic distribution of platelets with stairwise staggering as observed in bone [see Fig. 1(b)]. Fig. 3(a) shows the unit cell of a periodic distribution with \( n \) platelets in each period. Each platelet is shifted by \( L/n \) with respect to the adjacent platelets, and therefore gives an overlap length of \((n-1)/L\). The model degenerates to that of Gao et al. (2003) and Ji and Gao (2004a) when \( n = 2 \).

Fig. 3(b) shows the unit cell of the model consisting of \( n \) platelets and matrix. It is subjected to a displacement \( \Delta \), as shown in Fig. 3(c). The shear stresses in the matrix, denoted by \( \tau^t \) and \( \tau^u \), satisfy the force equilibrium

\[
\frac{n-1}{n} \frac{\tau^t}{h} + \frac{1}{n} \frac{\tau^u}{h} = 0. \tag{13}
\]

The normal stress obtained from force equilibrium is given separately over three regions in each platelet:

1. the bottom region \([0, (L/n)]\) of length \( L/n \), of which the top \((z=L/n)\) corresponds to the gap of the platelets next to the left side wall of the unit cell;
2. the middle region \((L/n, (n-1)/n)/L)\) of length \((n-2)/n)L\), of which the top \((z=(n-1)/n)L)\) corresponds to the gap of the platelets next to the right side wall of the unit cell; and
3. the top region \([(n-1)/n), L]\) of length \(L/n)\):

\[
\sigma_p = \begin{cases} 
(t^t + \tau^u) z / h, & 0 \leq z \leq L / n, \\
(t^t + \tau^u) L / nh, & L / n < z < (n-1) / n, \\
(t^t + \tau^u) (L-z) / h, & (n-1) / n \leq z < L.
\end{cases} \tag{14}
\]

The strain energies in the platelets and matrix within each unit cell are

\[
h \int_0^L \frac{\sigma_p^2}{2E_p} \, dz \quad \text{and} \quad n \left( 1 - \phi \right) h \frac{L (t^t)^2}{2G_m} + \frac{(n-1)L (t^u)^2}{2G_m}.
\]
respectively. The complementary energy in the unit cell is

\[
\Pi_C = n \left[ \int_0^L \frac{\sigma_2^2}{2E_p} \, dz + \frac{(1 - \phi)hL(\tau^*)^2}{n\phi} + \frac{(n-1)(1 - \phi)hL(\tau^*)^2}{2C_m} \right] - (n-1)F_A
\]

\[
= \left[ \frac{(3n-4)L^3}{6(n-1)^2hE_p} + \frac{n(1 - \phi)hL}{2(1 - \phi)n\phi C_m} \right] (\tau^*)^2 - L\tau^* F_A.
\]

where

\[
F = (\tau^* + \tau^*)L = \frac{\tau^* L}{n-1}
\]

is the axial force on each platelet. Minimization of the complementary energy with \(d\Pi_C/d\tau^* = 0\) gives the shear stress as

\[
\tau^* = \frac{A}{\frac{3(n-4)}{n^2(1-\phi)\phi^2 E_p} + \frac{n^2(1-\phi)}{(n-1)\phi^2 \phi^2 C_m}}
\]

The average stress in the composite is

\[
\sigma = \frac{(n-1)F}{nh + (1 - \phi)h/\phi}
\]

which can be rewritten via Eqs. (16) and (17) as

\[
\sigma = \frac{1}{\frac{3(n-4)}{n^2(1-\phi)\phi^2 E_p} + \frac{n^2(1-\phi)}{(n-1)\phi^2 \phi^2 C_m}}
\]

where \(\rho = L/h\) is the platelet aspect ratio. Its ratio to the average strain \(A/L\) gives Young's modulus of the composite as

\[
E = \frac{1}{\frac{3(n-4)}{n^2(1-\phi)\phi^2 E_p} + \frac{n^2(1-\phi)}{(n-1)\phi^2 \phi^2 C_m}} = \frac{\phi E_p}{\frac{n(3n-4)}{3(n-1)\phi^2 E_p} + \frac{n^2}{3(n-1)\phi^2 C_m}}
\]

where \(\alpha\), as defined in Eq. (8), serves as a unique parameter combining the effects of platelet volume fraction \(\phi\) and aspect ratio \(\rho\), and matrix and platelet elastic moduli \(C_m\) and \(E_p\). Young's modulus, normalized by \(\phi E_p\), is a universal function of \(\alpha\) and the number of platelets \(n\) within each period. For \(n=2\), it degenerates to Gao et al. (2003) and Ji and Gao (2004a) (except for a factor \(\frac{4}{3}\) associated with the term \(1/\phi E_p\), resulting from the minimization of complementary energy). For the limit of platelet volume fraction \(\phi \rightarrow 1\) or platelet aspect ratio \(\rho \rightarrow \infty\), Young’s modulus approaches \((3(n-1)^2/n(3n-4))\phi E_p\), which increases from \(\frac{4}{3}\phi E_p\) for \(n=2\) to \(\phi E_p\) for \(n=\infty\), where \(\phi E_p\) is the Voigt (upper) limit.

Fig. 6 shows Young’s modulus \(E\), normalized by the platelet volume fraction and modulus \(\phi E_p\), versus \(n\) for a large range of \(\alpha\) from 0.021 to 83, which corresponds to the platelet aspect ratio ranging from 8 to 500 for a fixed platelet volume fraction \(\phi=50\%\) and platelet-to-matrix modulus ratio \(E_p/C_m=1000\). For relatively small \(\alpha\leq0.021, 0.13,\) and 0.83 (i.e., platelet

![Fig. 6. Plots of Young’s modulus over period n for different values of \(\alpha = \phi p^2 G_m/3(1-\phi)E_p\) for a stairwise staggered nanocomposite.](image-url)
aspect ratio $\rho = 8, 20, \text{ and } 50), \text{ Young's modulus reaches the maximum}
\[ E = \frac{1}{4 \phi E_p + 4(1 - \phi) \rho^2 \mu^2 G_m} \]
at $n=2$, which corresponds to the case considered by Gao et al. (2003) and Ji and Gao (2004a). However, for large $\alpha = 3.3 \text{ and } 83 \text{ (i.e., platelet aspect ratio } \rho = 100 \text{ and } 500), \text{ Young's modulus reaches the maximum away from } n=2. \text{ (For a finite } \rho, \text{ the limit } n \to \infty \text{ corresponds to weak planes, and therefore vanishing Young's modulus.)}

It can be shown analytically that, for $\alpha \leq 1$, Young's modulus of the composite always achieves the maximum at $n=2$. For $\alpha > 1$, the maximum Young's modulus becomes

\[ E = \frac{\phi E_p}{(\sqrt{1 + 8z} + 1)^3(3\sqrt{1 + 8z} - 1)} \]
at $n = (1 + \sqrt{1 + 8z})/2$. Combining these results, the maximum Young's modulus can be expressed as

\[ E = 3\frac{\phi E_p}{\sqrt{\alpha}} \]

which increases with $\alpha$, and approaches $\phi E_p$ as $\alpha \to \infty$, where $\phi E_p$ is the Voigt (upper) limit.

The composite reaches its strength at critical shear stress in the matrix $\tau^m = \tau^m_{\text{critical}}$ or at critical normal stress in the platelet $\sigma_p = \sigma_p^{\text{critical}}$. Thus the composite strength can be written as

\[ \sigma_{\text{critical}} = \begin{cases} \phi \sigma_p^{\text{critical}} \frac{n-1}{n} \rho_{\text{critical}}^{\text{critical}}, & \rho \leq \rho_{\text{critical}}^{\text{critical}}, \\ \phi \sigma_p^{\text{critical}} \frac{n-1}{n}, & \rho > \rho_{\text{critical}}^{\text{critical}}. \end{cases} \]

where

\[ \rho_{\text{critical}}^{\text{critical}} = (n-1) \frac{\sigma^{\text{critical}}}{\tau^{\text{critical}}} \]

is the critical aspect ratio that separates matrix failure ($\rho \leq \rho_{\text{critical}}^{\text{critical}}$) from platelet failure ($\rho > \rho_{\text{critical}}^{\text{critical}}$). The composite strength, normalized by $\phi \sigma_p^{\text{critical}}$, is a universal function of $n$, platelet aspect ratio $\rho$ and $\rho_{\text{critical}}^{\text{critical}}$.

Fig. 7 shows the composite strength $\sigma_{\text{critical}}$ normalized by the platelet volume fraction and strength $\phi \sigma_p^{\text{critical}}$ versus $n$ for the platelet-to-matrix strength ratio $\sigma_p^{\text{critical}}/\tau^{\text{critical}} = 10$ and platelet aspect ratio $\rho$ ranging from 8 to 500. The solid curve represents platelet failure, and is given by a universal relation $\sigma_{\text{critical}} = ((n-1)/n)\phi \sigma_p^{\text{critical}}$. The dotted curves represent matrix failure, and they depend on the platelet aspect ratio $\rho$. For each platelet aspect ratio $\rho \leq \sigma_p^{\text{critical}}/\tau^{\text{critical}}$, the dotted curve is always lower than the solid curve such that the maximum composite strength is reached at $n=2$, and is given by $\frac{1}{8} \phi \rho \tau^{\text{critical}}$. For each platelet aspect ratio $\rho > \sigma_p^{\text{critical}}/\tau^{\text{critical}}$, the intercept of solid and dotted lines gives the

![Fig. 7. Plots of strength over number (n) of platelets in one period for different platelet aspect ratios for a stairwise staggered nanocomposite.](image-url)
maximum composite strength
\[ \frac{\phi \sigma_{\text{critical}}^m}{\rho \sigma_{\text{critical}}^p} \]
which is reached at \( n = 1 + \rho (\sigma_{\text{critical}}^m / \sigma_{\text{critical}}^p) \). The maximum composite strength approaches \( \phi \sigma_{\text{critical}}^p \) as \( \rho \to \infty \).

4. The tension–shear chain model with arbitrary staggering

The tension–shear chain model is extended to the case of arbitrary distribution of parallel platelets in this section. Fig. 8(a) shows a unit cell consisting of the matrix and \( n \) platelets that are arbitrarily distributed, and the end of \( i \)th platelet is denoted by \( z_L(0 \leq z_L < 1, i = 1, 2, \ldots, n) \) in the global coordinate \( z \). This arbitrary distribution repeats itself outside the unit cell. A local coordinate \( \tilde{z} = z - z_L \) is introduced in Fig. 8(b) such that the ends of \( (i-1) \)th, \( i \)th, and \( (i+1) \)th platelets are \( z_L \), 0, and \( z_{L_{\text{right}}} \), respectively, where \( \tilde{z}_{L_{\text{left}}} = \tilde{z}_L - \tilde{z}_i \) if \( \tilde{z}_L - \tilde{z}_i \geq 0 \), \( \tilde{z}_{L_{\text{left}}} = \tilde{z}_L - \tilde{z}_i + 1 \) if \( \tilde{z}_L - \tilde{z}_i < 0 \); \( \tilde{z}_{L_{\text{right}}} = \tilde{z}_{L_{\text{left}}} + 1 \) if \( \tilde{z}_{L_{\text{left}}} + 1 \geq 0 \), \( \tilde{z}_{L_{\text{right}}} = \tilde{z}_{L_{\text{left}}} - \tilde{z}_i + 1 \) if \( \tilde{z}_{L_{\text{left}}} + 1 < 0 \); and the 0th and \((n+1)\)th platelets are the same as the \( n \)th and 1st platelets, respectively. Let \( t_{i_{\text{left}}} \) and \( t_{i_{\text{right}}} \) denote the shear stress between \( i \)th and \((i-1)\)th platelets [Fig. 8(c)], \( t_{i_{\text{left}}} \) and \( t_{i_{\text{right}}} \) the shear stress between \( i \)th and \((i+1)\)th platelets, and \( A \) the displacement on the unit cell.

The force equilibrium of the \( i \)th platelet gives
\[ t_{i_{\text{left}}} t_{i_{\text{right}}} - t_{i_{\text{left}}} (1 - t_{i_{\text{left}}}) + t_{i_{\text{right}}} - t_{i_{\text{right}}} (1 - t_{i_{\text{right}}}) = 0. \]
(23)

An equilibrium stress field, which satisfies the above equation and will be used to calculate the complementary energy, can be written as
\[
\begin{bmatrix}
   t_{i_{\text{left}}} \\
   t_{i_{\text{right}}} \\
   t_{i_{\text{right}}} \\
   t_{i_{\text{right}}} \\
\end{bmatrix}
= \tau \cdot \begin{bmatrix}
   1 - \tilde{z}_{L_{\text{left}}} \\
   1 - \tilde{z}_{L_{\text{right}}} \\
   \tilde{z}_{L_{\text{left}}} \\
   \tilde{z}_{L_{\text{right}}} \\
\end{bmatrix},
\]
(24)
where \( \tau \) is to be determined. This stress field degenerates to Eqs. (1) and (13) for the platelet distributions in Sections 2 and 3, respectively. The normal stress in the \( i \)th platelet is obtained from the force equilibrium as
\[ \sigma_i = \frac{\tau}{h} \left[ (2 - \tilde{z}_{L_{\text{left}}} - \tilde{z}_{L_{\text{right}}}) \tilde{z}_i - \tilde{z}_{L_{\text{left}}} \langle \tilde{z} \rangle - \tilde{z}_{L_{\text{right}}} \langle \tilde{z} \rangle \right], \]
(25)
where the function \( \langle \cdot \rangle \) is defined by
\[ \langle x \rangle = \begin{cases} 
0, & x < 0, \\
1, & x \geq 0.
\end{cases} \]

The strain energy in all platelets in each unit cell is
\[ \sum_{i=1}^{n} \frac{h}{2E_p} \int_0^L \frac{\sigma_i^2(\tilde{z})}{2E_p} d\tilde{z} = n \frac{L^3}{2E_p} \tau f_p, \]

\[ \text{Fig. 8. Arbitrarily staggered nanocomposite structure. (a) Schematic of arbitrary staggering and (b) the unit cell (solid box) in a global coordinate system. Dashed box describes the local environment of platelet } i, \text{ with local coordinates defined in (b) and deformed configuration under a longitudinal displacement } A \text{ in (c).} \]
where

\[ f_p = \frac{1}{3n} \sum_{i=1}^{n} \left\{ \left( \frac{\zeta_i^{\text{left}}}{\zeta_i^{\text{right}}} \right)^2 + \left( \frac{\zeta_i^{\text{right}}}{\zeta_i^{\text{left}}} \right)^2 - \left( \frac{\zeta_i^{\text{left}}}{\zeta_i^{\text{right}}} \right) \right\} \tag{26} \]

is a dimensionless factor depending only on the distribution of platelets. The strain energy in the matrix is

\[ n\left((1-\phi)hL/2\phi G_m\right)\tau_2 f_m, \]

where

\[ f_m = \frac{1}{2n} \sum_{i=1}^{n} \left[ \zeta_i^{\text{left}} + \zeta_i^{\text{right}} \right] \tag{27} \]

is another dimensionless factor depending only on the distribution of platelets. The total force on the composite is \( n f_m \tau_2 \), which can be derived from Eq. (25). The complementary energy in the unit cell is

\[ \Pi_c = n\left[ \frac{L^3}{3\phi E_p} f_p + \frac{(1-\phi)hL f_m}{2\phi G_m} \right] \tau_2^2 - n f_m \tau_2 A, \tag{28} \]

where the last term represents the external work. Minimization of the complementary energy with \( d\Pi_c/d\tau_2 = 0 \) gives

\[ \tau_2 = \frac{L^2 f_p}{\phi E_p f_m} + \frac{(1-\phi)h}{\phi G_m} \tag{29} \]

The average stress in the composite is

\[ \sigma = \frac{1}{\phi E_p f_m} + \frac{1}{\phi G_m} \frac{A}{1-\phi}, \tag{30} \]

where \( \rho = L/h \) is the platelet aspect ratio. Its ratio to the average strain \( A/L \) gives Young’s modulus of the composite as

\[ E = \frac{\phi E_p}{\phi E_p f_m} + \frac{\phi G_m}{\phi G_m f_m} \frac{A}{1-\phi}, \tag{31} \]

where \( z \) is defined in Eq. (8) and

\[ \beta_1 = \frac{f_p}{f_m}, \tag{32} \]

\[ \beta_2 = \frac{1}{f_m}, \tag{33} \]

are two dimensionless factors representing the effect of the non-uniform distribution of platelets on Young’s modulus of the composite. It is interesting to note that, although there are so many distribution patterns for short platelet reinforced composites, all the information can be condensed into only two non-dimensional factors, \( \beta_1 \) and \( \beta_2 \), in determining the composite stiffness. Therefore, we can refer to \( \beta_1 \) and \( \beta_2 \) as the stiffness distribution factors, which can be computed by Eqs. (26), (27), (32) and (33) for any distribution \( \zeta_1, \zeta_2, \ldots, \zeta_n \). For the distribution in Section 2, \( \beta_1 = 4 \) and \( \beta_2 = 1/(\zeta(1-\zeta)) \). For the distribution in Section 3, \( \beta_1 = n(3n-4)/(3n-1)^2 \) and \( \beta_2 = n^2/(n-1) \). Table 1 lists the stiffness distribution factors for some typical distribution patterns.

<table>
<thead>
<tr>
<th>Table 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>Summary of stiffness and strength distribution factors for unidirectional composites with regular staggering, regular staggering with offset, stairwise staggering, random staggering and continuous layering.</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Distribution</th>
<th>Stiffness distribution factors</th>
<th>Strength distribution factors</th>
<th>Normalized critical aspect ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \beta_1 )</td>
<td>( \beta_2 )</td>
<td>( \beta_4 (\rho &gt; \rho_{\text{critical}}) )</td>
<td>( \beta_4 (\rho &lt; \rho_{\text{critical}}) )</td>
</tr>
<tr>
<td>Regular staggering</td>
<td>( \frac{4}{3n} )</td>
<td>( \frac{4}{3n-1} )</td>
<td>2</td>
</tr>
<tr>
<td>Regular staggering with offset</td>
<td>( \frac{4}{3n} )</td>
<td>( \frac{4}{3n-1} )</td>
<td>2</td>
</tr>
<tr>
<td>Stairwise staggering</td>
<td>( \frac{n(3n-4)}{3n-1} )</td>
<td>( \frac{n^2}{(n-1)^2} )</td>
<td>( n/(n-1) )</td>
</tr>
<tr>
<td>Random staggering</td>
<td>( \frac{2}{3} )</td>
<td>6</td>
<td>3</td>
</tr>
<tr>
<td>Continuous layering</td>
<td>1</td>
<td>–</td>
<td>1</td>
</tr>
</tbody>
</table>
The composite reaches its strength when the maximum shear stress in the matrix reaches the matrix shear strength \( \tau_{\text{critical}} \), or the maximum normal stress in the platelet reaches the platelet strength \( \sigma_{p\text{critical}} \). Combined with Eqs. (24) and (25), these give the composite strength as

\[
\sigma_{\text{critical}} = \begin{cases} 
\phi \tau_{\text{critical}} \frac{1}{\beta_4} \rho, & \rho \leq \rho_{\text{critical}} \\
\phi \sigma_{p\text{critical}} \frac{1}{\beta_3}, & \rho > \rho_{\text{critical}}
\end{cases}
\]

where

\[
\rho_{\text{critical}} = \frac{\beta_4 \sigma_{p\text{critical}}}{\beta_3 \tau_{\text{critical}}}
\]

is the critical aspect ratio that separates matrix failure (\( \rho \leq \rho_{\text{critical}} \)) from platelet failure (\( \rho > \rho_{\text{critical}} \)), and \( \beta_3 \) and \( \beta_4 \) are two non-dimensional factors representing the effect of non-uniform distribution of platelets on the strength of the composite, and are given by

\[
\beta_3 = \frac{1}{f_m} \max \left\{ \left( 2 - z_i \right) \min \{ z_i, 1 - z_i \} \left( 1 - \max \{ z_i, 1 - z_i \} \right), \ i = 1, 2, \ldots, n \right\},
\]

\[
\beta_4 = \frac{1}{f_m} \max \left\{ \frac{1}{2} z_i, 1 - \frac{1}{2} z_i, \ i = 1, 2, \ldots, n \right\}.
\]

Similarly, all distribution information can be condensed into another two non-dimensional factors, \( \beta_3 \) and \( \beta_4 \), in determining the composite strength, and we name them the strength distribution factors. For the distribution in Section 2, \( \beta_3 = 2 \) and \( \beta_4 = 2/(1 - |1 - 2z_i|) \). For the distribution in Section 3, \( \beta_3 = n/(n-1) \) and \( \beta_4 = n \). The strength distribution factors for some typical distribution patterns are listed in Table 1.

For an arbitrary distribution of platelets, Young’s modulus of the composite, normalized by \( \phi E_p \), is a universal function of \( z \) and the stiffness distribution factors \( \beta_1 \) and \( \beta_2 \). The strength of the composite, normalized by the platelet volume fraction and strength \( \phi \sigma_{p\text{critical}} \), is a universal function of platelet aspect ratio \( \rho \), platelet-to-matrix strength ratio, and the strength distribution factors \( \beta_3 \) and \( \beta_4 \).

The introduction of the stiffness and strength distribution factors makes it easier to compare the corresponding mechanical properties between the different distribution patterns, and obviously, the smaller stiffness/strength distribution factors imply higher stiffness/strength. Here, as an example, considering the herringbone-type distribution of \( n \) platelets in each period shown in Fig. 9, the factor \( \beta_3 = (3n^3 - 2n^2 - 8n + 8)/3(n-1)^2 \) is larger than (or equal to) its counterpart in Section 3, and \( \beta_3 = n^2/(n-1) \) remains the same. Therefore the distribution of platelets in Fig. 9 gives a smaller Young’s modulus of the composite. The factor \( \beta_3 = 2 \) is larger than (or equal to) its counterpart in Section 3, and \( \beta_4 = n \) remains the same. This leads to a smaller strength of the composite.

**Fig. 9.** A unidirectional nanocomposite structure with the herringbone-type platelet distribution \( n=6 \). Solid box denotes its unit cell.
5. The tension–shear chain model with random staggering

Random platelet alignment usually emerges in biomimetic materials with “mortar–brick” micro-/nanostructure (Bonderer et al., 2008; Tang et al., 2003). For random distributions of platelets, $\zeta_i$ representing the end of $i$th platelet (i=1,2, ..., n) is a random variable between 0 and 1. The average values of factors $f_p$ and $f_m$ in Eqs. (26) and (27) are:

$$f_p = \frac{1}{3} \int_0^1 \int_0^1 \left\{ \left( \zeta_i^{left} + (\zeta_i^{right})^2 \right) \left( 1 - \max(\zeta_i^{left}, \zeta_i^{right}) \right) \right\} d\zeta_i^{left} d\zeta_i^{right} = \frac{7}{180}. \quad (38)$$

$$f_m = \frac{1}{2} \int_0^1 \int_0^1 \left[ \zeta_i^{left}(1-\zeta_i^{right}) + \zeta_i^{right}(1-\zeta_i^{left}) \right] d\zeta_i^{left} d\zeta_i^{right} = \frac{1}{6}. \quad (39)$$

These give $\beta_1 = \frac{5}{3}$ and $\beta_2 = 6$ such that Young’s modulus of the composite with random distribution of platelets is

$$E = \frac{1}{\frac{7}{12} \phi E_p + \frac{5\phi}{\rho^2 G_m}} = \frac{5\phi}{10 + 7\phi}. \quad (40)$$

where $\chi$ is given in Eq. (8). For very large platelet aspect ratio, Young’s modulus is $E = \frac{5\phi}{10 + 7\phi} E_p$.

For random distributions of platelets with $\zeta_i$ (i=1,2, ..., n) between 0 and 1, $\max(\zeta_i^{left}, \zeta_i^{right})$, $1-\zeta_i^{left}$, $1-\zeta_i^{right}$, $i=1,2, ..., n$ may reach 1 such that Eq. (37) gives $\beta_1 = 6$ since $f_m = \frac{5\phi}{10 + 7\phi} \max(2-\zeta_i^{left} - \zeta_i^{right}, \min(z_i^{left}, \zeta_i^{right})), 1(\zeta_i^{left} + \zeta_i^{right})$ (1-$\max(\zeta_i^{left}, \zeta_i^{right})$), $i=1,2, ..., n$ may reach $\frac{1}{2}$ at $\zeta_i^{left} = \zeta_i^{right} = \frac{1}{2}$ such that Eq. (36) gives $\beta_2 = 3$. The strength of the composite with random distribution of platelets is

$$\sigma_{\text{critical}}^p = \begin{cases} \phi \sigma_{\text{critical}}^p & \text{if } \rho \leq \rho_{\text{critical}}^p, \\ \phi \sigma_{\text{critical}}^p \frac{1}{3} & \text{if } \rho > \rho_{\text{critical}}^p, \end{cases} \quad (41)$$

where

$$\rho_{\text{critical}}^p = \frac{2\sigma_{\text{critical}}^p}{E_p G_m}. \quad (42)$$

For very large platelet aspect ratio, the strength is $\sigma_{\text{critical}}^p = \frac{3}{5} \phi \sigma_{\text{critical}}^p$.

For random distribution of platelets, Young’s modulus and strength of the composite may reach $\frac{5}{3}$ and $\frac{5}{3}$ of those of the continuous layering composite.

6. Discussions and conclusions

Table 1 lists the stiffness and strength distribution factors for several typical distribution patterns. For completeness, the distribution factors for continuous layering structure are also included by directly comparing its stiffness and strength with Eqs. (31) and (34). From these equations, it is noted that increasing the aspect ratio of platelets $\rho$ can improve both the stiffness and strength of the staggered composites. When the aspect ratio $\rho$ (or $\chi$) is relatively large, according to Eq. (31), the stiffness distribution factor $\beta_1$ plays a more dominant role than $\beta_2$. Similarly, when $\rho > \rho_{\text{critical}}$, the strength will depend on the distribution factor $\beta_2$ based on Eq. (34), and will be independent of $\beta_2$. It is found from Table 1 that among various staggered distributions, the stairwise staggering with relatively large $n$ has the smallest $\beta_1$ and $\beta_2$, and therefore has higher stiffness and strength when the aspect ratio $\rho$ is large. To obtain a direct comparison, we select a group of typical material parameters $\phi=50\%$, $E_p/G_m=1000$, $\sigma_{\text{critical}}^p/\tau_{\text{critical}}^m=10$, and draw the normalized stiffness and strength as functions of the aspect ratio $\rho$ in Figs. 10(a) and (b) for different distribution patterns. The stiffness and strength of continuous layering composites are used to normalize the corresponding quantities.

It is noted from Fig. 10(a) that for large aspect ratio $\rho$, the stairwise staggering with $n=5,10$ has higher stiffness than regular staggering and random staggering distributions. The prediction by the Mori–Tanaka method (Mori and Tanaka, 1973; Weng, 1990; Zhao and Weng, 1990) is also plotted in Fig. 10(a) and it obviously underestimates the stiffness. It should be pointed out that the Mori–Tanaka method, as a classical homogenization-based mesomechanical approach, can only account for the influence of the volume fraction, shape and orientation of inclusions, but cannot take the distribution of reinforcements into account. In contrast, our study indicates that the distribution has significant effect on the stiffness.

Fig. 10(b) shows that the regular staggering has the highest strength at small aspect ratios $\rho$ (e.g., $\rho \leq 15$), but stairwise staggering with larger period $n$ (e.g., $n=5,10$) would achieve higher strength provided that the aspect ratio is large enough.
Every distribution tends to reach its strength limit \( \sigma_{\text{critical}} = \beta_3 \) when the aspect ratio \( \rho \) is beyond the corresponding critical value \( \rho_{\text{critical}} \). Higher strength limit is realized by stairwise staggering with larger period \( n \), which has smaller \( \beta_3 \) as indicated in Table 1. It is worth noting that the strength of random staggering is significantly lower than that of controlled distribution such as regular staggering and stairwise staggering.

From Fig. 10(a, b) and Table 1, it seems that the continuous layering composite is the best distribution with respect to stiffness and strength, but why does nature not adopt this microstructure? An answer to this question will require consideration of failure strain \( \varepsilon_{\text{critical}} = \frac{\sigma_{\text{critical}}}{E} \) and energy storage capacity \( w_{\text{critical}} = \frac{\sigma_{\text{critical}}^2}{2E} \), which also depend on the distribution via the four distribution factors \( \beta_1, \beta_2, \beta_3, \) and \( \beta_4 \). Fig. 10(c) shows the failure strain as a function of the aspect ratio \( \rho \) for regular staggering, stairwise staggering \( (n=5,10) \), random staggering, and continuous layering distributions. The failure strain is normalized by the failure strain of continuous layering structure, equal to platelet failure strain \( \varepsilon_{\text{critical}} = \frac{\sigma_{\text{critical}}}{E} \). It is found that, different from the stiffness and strength, the failure strain \( \varepsilon_{\text{critical}} \) of staggering distribution decreases as the aspect ratio \( \rho \) increases, and the continuous layering distribution has the lowest failure strain. Hence, there should exist an optimal aspect ratio \( \rho \) that gives best the overall best mechanical properties. In this sense, the energy storage capacity may serve as an index in evaluating the overall mechanical properties, since it is essentially the product of the strength and the failure strain. Fig. 10(d) shows the energy storage capacity as a function of the aspect ratio \( \rho \), normalized by that of continuous layering distribution \( w_{\text{critical}} = \frac{\sigma_{\text{critical}}^2}{2E} \). Fig. 10(d) suggests that each energy storage capacity curve for staggering distribution reaches its peak at a critical aspect ratio. The peak values for regular staggering and stairwise staggering \( (n=5,10) \) are several times and even one order larger than that of the continuous layering case. The peak value of the energy storage capacity for random staggering is less than that of regular staggering and stairwise staggering \( (n=5,10) \) but almost two times of that for the continuous layering composites.
In summary, we have developed analytical models on the stiffness, strength, failure strain, and energy storage capacity of unidirectional platelet-reinforced composites with arbitrary distribution. The following conclusions can be drawn:

1. It is found that stairwise staggering (including regular staggering with \( n=2 \)) could achieve overall excellent performance, namely high stiffness and strength comparable to those of the reinforcing platelets as well as large failure strain and energy storage capacity comparable to those of the soft matrix. This may be a key reason why regular and stairwise staggering are most widely observed in natural biological materials, and may be regarded as the optimal design resulting from millions of years of evolution. The performance of random staggering is obviously lower than stairwise staggering, which indicates that precisely controlled microstructure is an efficient way to further improve performance of biomimetic and other man-made composites.

2. Besides the volume fraction, shape, and orientation of the reinforcements, the distribution of platelets also plays a significant role in the mechanical properties of unidirectional composites. Four distribution factors, the stiffness distribution factors \( \beta_1, \beta_2 \) and the strength distribution factors \( \beta_3, \beta_4 \), are identified to completely characterize the influence of the reinforcement distribution, and they can be easily computed. It should be emphasized that classical homogenization-based mesomechanical approaches, such as the Mori–Tanaka method, cannot take this distribution effect into account.

3. The analytical models and distribution factors developed in the present paper allow one to compare different designs of unidirectional composites and may guide the development of high performance biomimetic and other man-made composites.

Acknowledgments

BL acknowledges the support from National Natural Science Foundation of China (Grant nos. 10702034, 10732050, 90816006) and National Basic Research Program of China (973 Program 2007CB936803). YH acknowledges the support from ONR Composites for Marine Structures Program (Grant N00014-01-1-0205, Program Manager Dr. Y.D.S. Rajapakse) and the NSF (Grant #0800417). The work of HG was partially supported by the A*Star Visiting Investigator Program “Size Effects in Small Scale Materials” hosted at the Institute of High Performance Computing in Singapore.

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