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Toughness amplification in natural composites

Francois Barthelat*, Reza Rabiei

Department of Mechanical Engineering, McGill University, 817 Sherbrooke Street West, Montreal, Quebec, Canada H3A 2K6

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ABSTRACT

Natural structural materials such as bone and seashells are made of relatively weak building blocks, yet they exhibit remarkable combinations of stiffness, strength and toughness. This performance can be largely explained by their "staggered microstructure": stiff inclusions of high aspect ratio are laid parallel to each other with some overlap, and bonded by a softer matrix. While stiffness and strength are now well understood for staggered composites, the mechanisms involved in fracture are still largely unknown. This is a significant lack since the amplification of toughness with respect to their components is by far the most impressive feature in natural staggered composites such as nacre or bone. Here a model capturing the salient mechanisms involved in the cracking of a staggered structure is presented. We show that the pullout of inclusions and large process zones lead to tremendous toughness by far exceeding that of individual components. The model also suggests that a material like nacre cannot reach steady state cracking, with the implication that the toughness increases indefinitely with crack advance. These findings agree well with existing fracture data, and for the first time relate microstructural parameters with overall toughness. These insights will prove useful in the design of biomimetic materials, and provide clues on how bone fractures at the nano and microscales.

1. Introduction

Structural biological materials are increasingly attracting the attention of researchers and engineers for their remarkable performances (Wegst and Ashby, 2004; Barthelat, 2007; Ortiz and Boyce, 2008), and their structures and mechanics are therefore at the focus of intense research. Recent advances in experimental and modeling techniques have enabled significant progress in this area. For example in medicine, a better understanding of how bone deforms and fractures will improve the way drugs are targeted to treat diseases such as osteoporosis. In biomimetics, the structures and mechanics of hard biological materials such as bone and nacre are now inspiring novel bio-inspired materials with remarkable properties (Barthelat, 2007, 2010; Ortiz and Boyce, 2008; Munch et al., 2008).

Natural materials use a large variety of structures, mechanisms and ingenious designs to achieve high mechanical performance. Beyond this apparent diversity, examination at smaller length scales however reveals common structural patterns or "universal motives" (Buehler and Yung, 2009) found across biological materials. An excellent case of universal motives is the staggered structure, where stiff inclusions of high aspect ratio (long molecules, fibers or platelets) are embedded in a compliant and ductile organic matrix with some overlap, to form a staggered structure (Jackson et al., 1988; Jager and Fratzl, 2000; Kotha et al., 2001; Gao, 2006; Barthelat et al., 2007). A well known example of such structure is nacre from mollusk shells, where calcium carbonate tablets form a three dimensional staggered brick wall-like structure with softer protein and polysaccharide

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* Corresponding author. Tel.: +1 514 398 6318; fax: +1 514 398 7365.
E-mail address: francois.barthelat@mcgill.ca (F. Barthelat).

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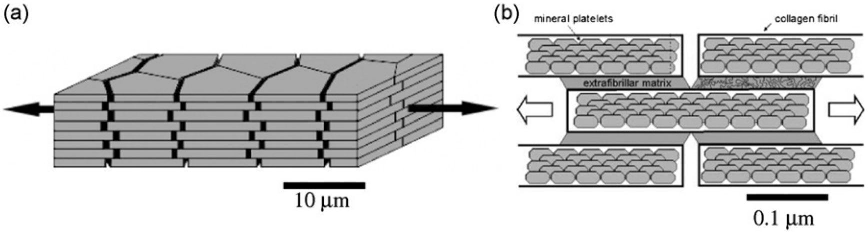

Fig. 1. Example of natural staggered structures: (a) nacre and (b) bone (Gupta et al., 2006).

layers at the interface (Jackson et al., 1988) (Fig. 1a). Under tensile stress the tablets can "slide" on one another, a key mechanism that generates significant deformation and energy dissipation. Another example is collagen fibrils, composed of staggered collagen molecules. In bone the fibrils are mineralized by nanometers size crystals, intercalated with the softer collagen molecules in a staggered fashion (Buehler, 2007) (Fig. 1b). In turn, those fibrils assemble into a staggered structure to form microscopic fibers, the building blocks of bone (Weiner and Wagner, 1998; Gupta et al., 2006). Gliding of the mineralized fibers on one another is an important deformation mechanism in bone (Gupta et al., 2006). Interestingly, bone also has a hierarchical structure where the staggered motif can be found over several length scales (Gao, 2006). In natural staggered composites the stiff inclusions are bonded by softer interfaces capable of maintaining cohesion over large separation distances. The materials found at these interfaces must have high resilience, and it is actually striking that these softer materials have similar structures and mechanics in nacre and bone (Smith et al., 1999; Fantner et al., 2005). Other identified examples of materials with staggered structures are tooth enamel and spider silk (Keten, 2010).

The question of how the staggered microstructure controls structural properties such as modulus, strength and toughness is of high relevance for biomedical and biomimetic applications. The modulus along the long axis of the inclusions can now be predicted with the models developed by Jager and Fratzl (2000) or by Kotha et al. (2001). Both of these models predict that high aspect ratio and high concentration of the inclusions lead to high modulus. To predict strength, shear-lag type models borrowed from composite theories can be used (Jackson et al., 1988). Toughness is more challenging to predict because there are multiple toughening mechanisms at work when a crack propagates in a staggered composite. For example, no less than ten toughening mechanisms were recently proposed for nacre (Mayer, 2005). The few models that captured specific toughening mechanisms in staggered structures include the fracture model by Okumura and de Gennes (2001), who examined the effect of the differential of stiffness between inclusions and interfaces on the toughness, and of the viscoelasticity of the interfaces. Gao (2006) has proposed a fracture model that incorporates bridging by the inclusions and the effect of the hierarchical structure on overall toughness. The possible implications of the small size of the inclusions on their tensile strength were first proposed by Currey (1977), and further elaborated by Gao et al. (2003), although this point remains controversial (Ballarini et al., 2005). Recent experimental work demonstrated the significant contribution of inelastic deformations on the toughness of nacre (Barthelat and Espinosa, 2007; Rabiei et al., 2010). In large regions around cracks the mineral tablets "slide" on one another (Fig. 1a), which dissipates a significant amount of energy that would otherwise be used for crack propagation (Barthelat and Espinosa, 2007). Fracture experiments on various types of nacres strongly suggest that this "process zone" is a prominent toughening mechanism (Rabiei et al., 2010). In bone, a similar sliding mechanism was recently demonstrated using synchrotron X-ray illumination (Gupta et al., 2006). Under tensile stress the mineralized collagen fibrils slide on one another, generating inelastic deformations (Fig. 1b). As opposed to nacre these mechanisms could not be observed directly in the vicinity of cracks, possibly because they involve smaller length scales that are masked by toughening mechanisms operating at larger scales (Ager et al., 2006; Peterlik et al., 2006). There is currently no comprehensive model to capture the mechanics of fracture in staggered composites and in particular, there is no model that properly captures the effect of the inelastic process zone. This is a significant lack, since for these materials it is the toughness which is by far the most spectacular property, and therefore the most attractive property to duplicate in biomimetic materials (Launey and Ritchie, 2009).

The aim of this paper is to provide a condensed approach to predicting relevant structural properties in staggered composites. Existing micromechanics models for modulus and strength are briefly reviewed first and their predictions are compared with available experimental data. Following the same philosophy, a model for the toughness of staggered composites incorporating the micromechanics of crack bridging and process zone is presented. Whenever possible each step of the model is validated with experiments. The results reveal new insights on how staggered structures fracture, and how microstructural parameters control the overall toughness.

2. Basic properties of staggered composites

2.1. Elastic modulus

The basis for modeling the elasticity of staggered structures is the two dimensional representative volume element (RVE) shown in Fig. 2 (Jager and Fratzl, 2000; Kotha et al., 2001). The stiff mineral inclusions (modulus E_m) have a length L