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Influence of geometric and material parameters on the behavior of nacreous composites under quasi-static loading

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A B S T R A C T

Biological structures, such as nacre from mollusk shells exhibit a tremendous balance of strength and toughness compared to traditional engineering materials. The mollusk composite shell is mainly comprised of a brittle mineral but exhibits superior fracture toughness. Learning from the microstructural design of nacreous composites could lead to the development of more effective protective composite systems. In this study, a nacreous composite model is constructed and validated by experimental results under uniaxial tensile loading. Parametric studies are conducted to investigate the influences of several geometric and material parameters on the energy dissipation and damage distribution in the composite. Different cohesive laws governing the mechanical behavior of the bonding matrix, variations in tablet geometries and waviness of the tablet interface are considered. It was found that the orientations of the cohesive bonds to the applied load and the cohesive law used to describe the behavior of the matrix strongly influenced the hardening behavior and damage distribution in the composite, as opposed to the interfacial geometry of the tablets, which has been the focus of many studies on nacre.

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1. Introduction

Extreme loads that may arise from blasts, impacts from storm debris, ballistics, barge impacts on bridge piers, and so on continue to claim lives and cause severe damage to infrastructure world-wide [\[1,2\].](#page--1-0) Consequently, the demand for lightweight highperformance protective structures has increased significantly. In general, lightweight composite structures offer significant advantages, owing to their light weight, high energy absorption and high strength, whereas traditional engineering materials typically sacrifice one for the other $[3]$. This makes composite structures suitable under a range of protective applications against extreme loadings, such as blast, ballistics, fire, and so on $[4-10]$. Biological structures utilize composite architectures to construct effective defense mechanisms against predators. For instance, it is believed that the mollusk shell [\(Fig. 1a](#page-1-0)) has optimized a two-layer armor system over millions of years of evolution to protect its soft tissues from extreme loads. They are typically subjected to these loads over their lifetime due to potential predator attacks, tidal rock impacts and immense hydrostatic ocean pressures. The armor system of the mollusk is believed to be the most efficient because the bulk of its

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structure is mainly comprised of a brittle mineral, but it exhibits superior fracture toughness and crack arresting capabilities [\[11–](#page--1-0) [13\].](#page--1-0) The relationship between the structure of the mollusk shell and the mechanisms by which it achieves superior fracture toughness is very important to the field of protective structural engineering, because it can potentially lead to the development of a superior lightweight composite structure for protective applications, or improve the structural resilience of existing composite systems.

Specifically, red abalone seashells [\(Fig. 1](#page-1-0)a) are built from an armored structure that consists of a stiff brittle outer calcium carbonate layer and a tough nacreous layer at its inner surface. The tough nacreous layer is found in other bivalve, gastropod and even cephalopod species, such as sea snails, squid and octopus. It is well known that the nacreous layer is composed of 95% vol. aragonite, a brittle mineral, but exhibits remarkable toughness [\[15–19\].](#page--1-0) As can be observed in [Fig. 1](#page-1-0), nacre shows a hierarchical structure over several length scales: the outer shell at the macro scale ([Fig. 1](#page-1-0)a); the staggered microstructure [\(Fig. 1](#page-1-0)b) and the platelet arrangement in one of the layers ([Fig. 1](#page-1-0)c). Many researchers have observed the similarity between nacre's microstructure and masonry [\[20–24\].](#page--1-0) More specifically, aragonite platelets (randomly shaped polygons) are staggered and stacked over many layers (approximately 0.5 μ m thick), which are bonded by a soft organic matrix [\(Fig. 1b](#page-1-0)) and c). Although the organic matrix (roughly 30 nm thick) has been

Fig. 1. a) Brick and mortar structure of an abalone shell; b) Elevation view of nacre's brick and mortar microstructure; c) Voronoi-shaped polygonal platelets found in each nacreous layer; and d) Staggered platelet configuration in nacre [\[14\].](#page--1-0)

typically treated as an inter-layer adhesive in recent investigations (e.g. $[25-27]$), recent work has also demonstrated the importance of the matrix as inter-granular cohesive bonds between nacre's polygonal tablets (Fig. 1c), which can significantly influence the stiffness of the composite [\[28\]](#page--1-0). Furthermore, other features that have been identified as potential contributors to nacre's remarkable toughness are: nano-asperities on the tablet surfaces, which facilitate interlocking between the conforming tablets [\[29–32\];](#page--1-0) mineral bridges that reinforce the tablets at the interface [\[33–](#page--1-0) [36\]](#page--1-0); and waviness on the surfaces of the tablets that impede tablet sliding, thereby inducing a hardening effect that distributes dam-age effectively throughout the composite [\[28,37\].](#page--1-0) These features indicate that the strength and stiffness of nacre may be influenced by the tablet aspect ratio [\[3,38\]](#page--1-0).

Other investigations have attempted to establish the structurefunction relationship of nacre's microstructural and nanoscale features (volume fractions, tablet aspect ratio, overlap length, unfolded protein molecules of the biopolymer matrix, etc.). By extending Volkersen's shear lag lap joint model utilized by Wei et al. [\[39\],](#page--1-0) which demonstrated the importance of the overlap length between adjacent nacreous layers, Ghazlan et al. [\[20\]](#page--1-0) showed that the waviness of the tablet surfaces facilitates a more uniform shear stress distribution across the interface prior to yielding. Qi et al. [\[40\]](#page--1-0) reported that the unfolding of protein molecules is a key toughening mechanism that occurs in nacre's biopolymer matrix, which is not well understood. They studied the effects of pretension, which are present in the membrane and solid states of these molecules. Their results obtained from a molecular model showed that pretension has a significant effect on triggering the unfolding of molecules at lower stresses, which is believed to be a key toughening mechanism found in staggered composites similar to nacre. Barthelat et al. [\[28\]](#page--1-0) reconstructed the nacreous tablet structure through optical imaging of a red abalone specimen and subjected it to uniaxial tension. They verified that the intergranular cohesive bonds have a profound influence on the strength and stiffness of nacre. Tran et al. [\[14\]](#page--1-0) constructed a voronoi model with inter-granular cohesive elements inserted between nacre-like tablets to mimic nacre's tablet structure. They also found that the cohesive bonds directly influence the overall strength and stiffness of the composite. Therefore, the voronoi model, in which its geometry is generated in a controlled manner, is able to mimic the tablet arrangement found in nacre quite well. Flores-Johnson et al. [\[27\]](#page--1-0) investigated the performance of a nacre-like composite plate under blast loading, which consisted of several monolithic layers bonded together by adhesives. They found that the nacre-like composite had a profound effect on reducing the back face velocity of the panel compared with a monolithic plate of equal mass. Other investigations have also found that nacre has tremendous abilities to arrest crack propagation, which is believed to be attributed to intrinsic and extrinsic toughening mechanisms that operate in front of and behind a crack tip, respectively [\[41\]](#page--1-0).

This paper introduces a novel design approach for mimicking the key features of nacre's hierarchical structure, namely the voronoi-shaped tablets, multilayer architecture, intergranular cohesive and interlayer adhesive bonds. A staggered voronoi representative volume element (RVE) is constructed to capture the randomness found in the nacreous tablet structure and the model is validated using experimental results by [\[28\]](#page--1-0) under quasi-static in-plane tensile loading. Parametric studies are subsequently conducted to study the influence of the tablet geometries, adhesive and cohesive properties on the strength, stiffness and toughness of the composite. The nacre-mimetic model was developed into a computer aided design (CAD) specification format such that it can be fabricated with rapid prototyping techniques such as 3D printing for a wide range of applications with complex geometries, including knee and elbow pads, gloves, helmets, crash mats, and so on. Preliminary 3D printed prototypes are presented in [\[14\].](#page--1-0)

2. Representative model of nacre's microstructure

2.1. Generating the nacre mimicking geometry

Several researchers have commented on the similarity between nacre's tablet arrangement and a voronoi diagram (Fig. 1d) [\[28,42,43\].](#page--1-0) Barthelat et al. [\[28\]](#page--1-0) generated a representative volume element that mimics nacre's tablet structure by reconstructing the tablet contours obtained from optical imaging of a red abalone specimen. This process makes it challenging to develop nacremimetic composite systems, because there is limited control over the geometry and arrangement of the tablets in each layer for practical applications. Although the tablet geometry in nacre appears to be randomly distributed, there is some structure to it, and several researchers are able to determine an average tablet size (around 8 μ m) and an average overlap length between adjacent layers, which is approximately one-third of the tablet area [\[39,44,45\].](#page--1-0) There are limited investigations into the influence of the cohesive bonds between the tablets, as well as the tablet shapes and sizes on the load sharing capability of nacre. To this end, a novel

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