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New classes of tough composite materials—Lessons from natural rigid biological systems

G. Mayer

Department of Materials Science and Engineering, Box 352120, University of Washington, Seattle, WA 98195-2120, United States

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Abstract

The structures and properties of a new class of composite materials, containing a predominantly high volume fraction ceramic or glass phase, combined with minor organic (adhesive) phases, have been studied. These composites have unusual combinations of mechanical properties, such as stiffness, strength, and toughness. They are based on the architecture of a rigid natural material, the nacre structure, such as those found in the shells of the abalone *Haliotis rufescens*, and those of other mollusk shells. The mechanisms underlying these properties have also been studied. Analogs (utilizing high-performance engineering materials), that mimic many of the mechanisms underlying those superior combinations of properties, have been built. The results of the foregoing investigations are discussed. It was found that the toughness of segmented composite beams which have high volume fractions of ceramic (89 v/o) exceeded those of continuous layered beams, as well as the monolithic ceramic (alumina) on which they are based.

Keywords: Biomimicry; Tough composite materials; Toughening mechanisms

1. Introduction

A scheme for new classes of composite materials, containing a predominant (95% or higher volume fraction) ceramic or glass phase, combined with minor protein phases, which have unique characteristics, has been investigated. This study has been based on prior investigations of rigid structures found in nature, mainly from the studies of nacre, which is found in a number of mollusk shell structures. The seminal paper that inspired this work was Ref. [1], by Jackson, et al. One of the significant results of that research is shown in Fig. 1, where the mechanical properties of the nacre structure (found in *Pinctada*) were compared with those of synthetic composites that had high volume fractions of ceramic filler. The key findings were that the architecture of the nacre structure provided not only superior stiffness, but strength and toughness (not shown) as well. The material at v/o of 1.0 is aragonite (100% $CaCO_3$). The multiple data points and spread for nacre, at v/o of 0.95 reflect different directions of test, and wet as well as dry conditions.

We have reviewed a number of the salient characteristics of nacreous materials, which persuade us that these can be of high technological importance. These are:

1. Attractive combinations of stiffness, together with strength (e.g., see below).
2. High levels of toughness, or energy-absorbing capability, with unusual resilience, even after loading that results in large amounts of fracture.
3. Development of unusual combinations of energy-absorbing mechanisms that bring about this toughness.

During the past several years, our group at the University of Washington has studied natural systems, including the nacreous structure found in the shell of red abalone *Haliotis rufescens* as well as those of many other shells (the actual structure of nacre is shown in Fig. 2b., and a schematic diagram is indicated in Fig. 3).

The schematic diagram shows that each tablet or platelet (which is hexagonal when viewed from the top) is surrounded on all sides by a thin organic layer. That layer consists of several components, one of which has been shown to exhibit unusual extensibility and resilience. The component which is responsible for those properties has been identified at the

E-mail address: gmayer@u.washington.edu.

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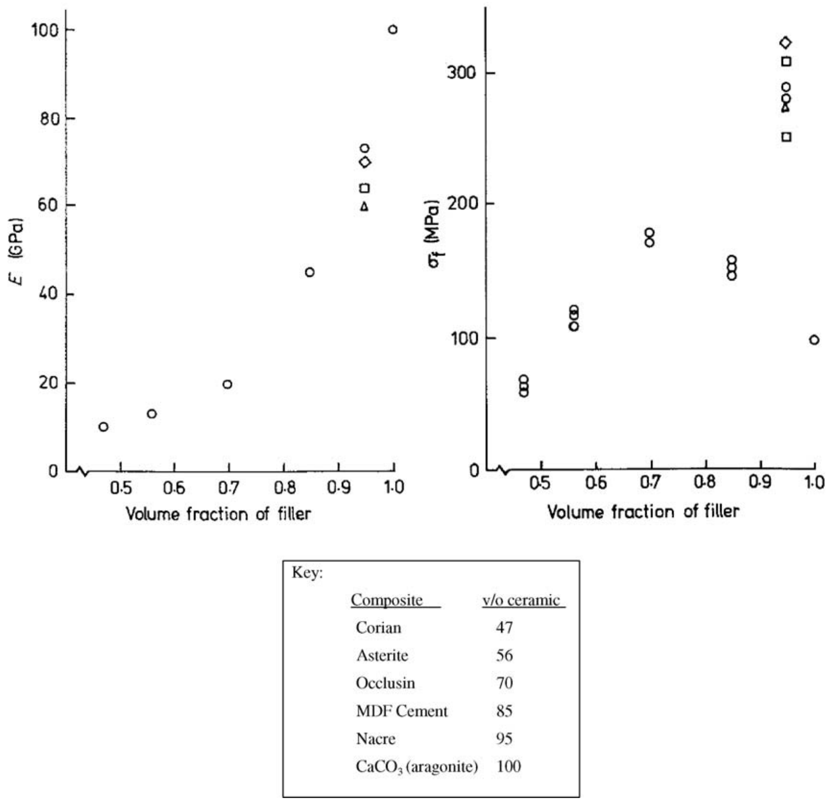

Fig. 1. Stiffness and fracture strength of several ceramic/polymer composites with high ceramic content [1]. The spread of data for nacre represent directional variance, and wet and dry conditions. (With permission, Kluwer Academic Publishers Jl. of Materials Science.).

University of California-Santa Barbara as *Lustrin A* [3]. It is noted that, the (nominally) hexagonal platelets are not of uniform size, and are not always hexagonal, but the composite structure is always space-filling.We have, additionally, built structures of synthetic materials based on our interpretations of the energy-absorbing mechanisms of the nacre structure. In our best estimate, those mechanisms include:

1. Creation of new surface area by delamination and/or fracture (multiple microcracking is included here).
2. Crack diversion.
3. Pull-out of the ceramic phase from the minor organic component.
4. Asperities on the platelet surfaces may also be involved in the pull-out process as well as may provide resistance against pull-out of the platelets [4].
5. Hole formation at the ends of the displaced ceramic phase elements, which may be related to stress-whitening, at larger deformations [5].
6. High level of anchoring of the organic (adhesive) phase(s).
7. Ligament, or filament formation in the organic phase(s), which are viscoelastic, as well as highly resilient.
8. Crack bridging by ligaments of the organic phase(s).
9. Strain-hardening of the organic phase(s), accompanied by prior chain extension, breaking of cross-links, etc.

The relative contributions of at least one of the foregoing mechanisms (the role of asperities) has been brought into question [6], but the major importance of the minor proteinaceous organic phase(s) in toughening is well established.

Early on, there was concern about trying to mimic architectures at the micron level. This is a formidable task in considering the assembly of a bulk structure. Then, we became aware of the concept underlying LAST® (*L*ight *A*pplique *S*egmented *T*ile) armor, which had been developed by the Foster-Miller Corporation [7] more than a decade ago, for defense applications. An array of these tiles is shown in Fig. 4, below. The hexagonal tiles of the ceramic phase (Coors alumina, or a Lanxide ceramic material) are surrounded on all sides by thin rubber layers, which act as traps for the shock waves that are generated by striking projectiles. The entire array is mounted with Velcro® fasteners onto the steel plates of fighting vehicles. The actual performance of the LAST tiles has been verified, and the efficacy of the array of tiles for ballistic protection has been confirmed.

Thus, damage to the structural array is often localized to a single tile, depending on the size of the projectile. These similarities between the nacre structure and the LAST array encouraged the idea that the concept of the nacre architecture, and its mechanical attributes, was valid on more than just the micron scale, and we proceeded to design and build a macroscopic model system. It should be noted that the LAST armor does not simulate nacre from the structural standpoint precisely, since the minor phases in nacre are adhesive, and are not so in the LAST family of materials. And, there are other differences, also, such as the uniformity of tiles, and the use of Velcro®. However, for the future, a scaling factor may have to be considered, and the structure optimized. Finally, since the hexagonal structure of the plates in nacre (and in LAST tile