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Rapid Assembly Processes of Ordered Inorganic/organic Nanocomposites

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1. Introduction

The material of seashell nacre is of great interest to material scientists due to its superior mechanical properties and unique bio-compatibility. Natural seashell nacre consists of about 95% of aragonite (a mineral form of CaCO3) and a few percent of biological macromolecules; yet its work of fracture is about three orders of magnitude higher than its mineral constituency. The superior strength and toughness of seashell nacre are attributed to the robust nanostructure in which the protein collagen layers (10–50 nm thick) and aragonite tablets (200–900 nm thick) form an ordered brick-and-mortar structure. The macromolecules act as strong adhesives while the aragonite tablets as the rigid building blocks (Smith et al., 1999). The structure of seashell nacre has inspired material scientists to design and develop various advanced biomimetic materials in the last two decades.

A macroscopic analogue of nacre structure led to the development of laminated silicon nitride ceramic composites which exhibit superior toughness to monolithic ceramics (Clegg et al., 1990; Wang et al., 2002), however, the laminated structure in micrometer scale may not have the same performance as the one with nano-scale laminated structure. In recent years, there is an increasing interest in the development of biomimetic organic/inorganic nanocomposites. These new materials hold promise for high performance orthopaedic materials, chemical sensors and photonic functional materials.

The formation of seashell nacre is a slow process in nature. Seashell absorbs mineral elements and organic matters in seawater to form inorganic nano-platelets and macromolecules, which deposit alternately to form compact brick-and-mortar structure of nacre. There are several approaches to mimic this process. One approach is called layer-by-layer assembly, by which ultrathin films of multilayered structure were prepared by sequential adsorption of a cationic polyelectrolyte and individual platelets of the negatively charged silicate mineral (Kleinfeld & Ferguson, 1994). Kleinfeld and Ferguson reported the incorporation of these clay sheets into thin films by the electrostatic layer-by-layer assembly (LBL). The structural investigation of film thickness and interlayer spacing of clay particles was accomplished by Kotov et al (Kotov et al., 1997) through a series of surface-sensitive techniques. The determination of the mechanism of the layer-by-layer alternating self-assembly of the clay multilayers was reported (Kotov et al., 1997; Kleinfeld & Ferguson,
First, given the difference between testing methods, the reduced Young's modulus above cannot be directly compared with Young's modulus (~48.5 GPa) of nacre in the recent three-point bend test.

Second, in addition to ordered layered structure, interfacial compatibility of the organic and inorganic components is a key factor. From this aspect, only certain types of polymer are effective in dramatically enhancing mechanical properties of such composite films. Thus, whether AAER is most suitable or not is still unknown.

Third, the thicknesses of organic layers and inorganic layers in our nano-laminated films are much thinner than those in nacre. In natural nacres, the biopolymer layers are usually 10–50 nm thick, providing necessary space for tight folding of polymer chains and certain degree of cross-linking of polymer. In comparison, in our laminated structure, polymer is confined within the interlayer space of smaller than 2 nm. Thus, the degree of cross-linking of AAER with its percentage in total organic content is probably low, consistent with the result that no distinction in FTIR spectra and XRD patterns were observed between the as-deposited HMMT film and the heat-treated HMMT film. Meanwhile, aragonite layers in nacre are 200–900 nm thick, hundreds of times thicker than the clay layers in our film. This may well explain why natural nacre adopts the micro/nano composite structure but not the nano/nano composites structure. Research on the preparation of micro/nano laminated organic–inorganic composites is being conducted by our group.

Fourth, properties of clay platelets are fairly different with aragonite. Clay platelets are extremely compliant, while aragonite is much more rigid. Additionally, CaCO₃ blocks have nano asperities that are about 30–100 nm in diameter, 10 nm in amplitude, providing additional friction when one block is sliding on the other.

4.3. Summary
The special assembly method—hydrothermal-electrophoretic assembly was successfully developed to prepare AAER/MMT nanocomposites that mimic nacre, both in structure and composition. The thickness of the nanocomposites film is controllable and can reach to more than 20 μm.

In this process, AAER plays four important roles as: intercalation agent in the hydrothermal process, binder around intercalated or non-intercalated platelets, stabilizing agent for MMT suspension, and improving the electric conductivity of MMT by AAER-intercalated.

Reduced Young's modulus was improved from 2.9±0.4 GPa for NMMT film to 5.0±1.0 GPa for HMMT film even at a low polymer content contained in the composite. The brick-and-mortar nacre-like structure is mainly attributed to the improved mechanical properties by incorporating extra energy-absorbing mechanisms during elastic deformation.

5. Conclusions
This chapter has summarized three processes that can produce laminated biomimetic nanocomposites. The high-speed centrifugal process can produce nanocomposites up to a thickness of 200 μm within minutes. The thick films produced have similar organic content and mechanical properties compared to that of lamella bones. The electrophoretic
deposition of monomers and intercalated montmorillonite clay followed by ultraviolet initiated polymerization can produce dense laminated nano-composite films up to tens of µm. The composite film exhibits four-fold improvement in Young’s modulus and hardness over monolithic polycrylamide polymers. Electrophoretic deposition combining intercalated montmorillonite nano-plates and polyelectrolyte such as acrylic anodic electrophoretic resin (AAER) can produce nanocomposites with organic content of 5 wt% to 15 wt%. The composites obtained have good uniformity and significant improvement in Young’s modulus and strength over monolithic montmorillonite films. These methods hold promise to fabricate laminated biomimetic materials at increased deposition rate. With the development of synthetic hydroxyapatite nanoplates (Le et al, 2009), these methods will enable the fabrication of a new generation of biomimetic nanocomposites for bone substitutes. This is becoming an area of great interest to clinicians as well as materials scientists.

6. References


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Nature’s evolution has led to the introduction of highly efficient biological mechanisms. Imitating these mechanisms offers an enormous potential for the improvement of our day to day life. Ideally, by bio-inspiration we can get a better view of nature’s capability while studying its models and adapting it for our benefit. This book takes us into the interesting world of biomimetics and describes various arenas where the technology is applied. The 25 chapters covered in this book disclose recent advances and new ideas in promoting the mechanism and applications of biomimetics.

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