

# A Comprehensive Review on the Development of Natural and Biomimetic Bouligand Structure Materials

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## ABSTRACT

The Bouligand structure, also known as a helicoidal fibrous structure, appears extensively in natural biological tissues with excellent mechanical properties, such as the dactyl clubs of mantis shrimp, the exoskeletons of lobsters, and the scales of fish; because this highly unique layer-by-layer architecture is formed by stacking single-direction fibrous sheets in a spiral manner at specific angles, it can equip biological tissues with exceptional fracture resistance, anti-impact capabilities, and damage tolerance; if the relationship between this structure and its performance can be thoroughly understood, and this mechanism can then be applied to artificially manufactured systems, it would be of immense value for the development of new, high-performance composite materials; this article primarily lists classic examples of Bouligand structures in natural environments, carefully analyzes the internal principles that enable it to increase material toughness, which include crack deflection, crack twisting, and the bridging effect between fibers, and summarizes the main methods used in recent years to artificially biomimic and manufacture this structural material, such as 3D printing, material self-assembly, and assembly through shear forces, while also forecasting the roles this material could play in fields like aerospace, national defense, and medical health in the future.

## KEYWORDS

Bouligand structure; Biomimetic materials; Hierarchical architecture; Toughening mechanism; Composite materials

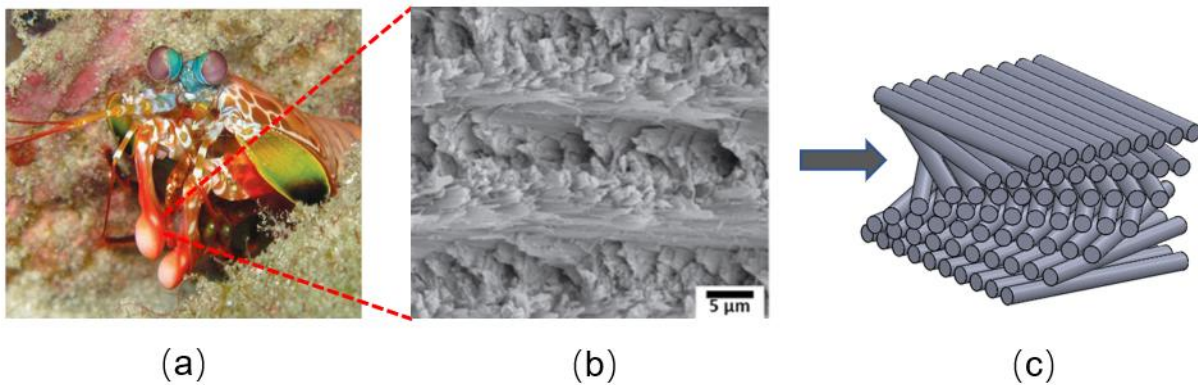
## 1. INTRODUCTION

After going through extremely long periods of species evolution, nature has nurtured countless structural materials with extremely high mechanical strength; although natural materials like bones, shells, and seashells are basically composed of ordinary microscopic basic substances such as relatively fragile minerals and softer organic matter, after undergoing highly ingenious and regular assembly across different levels and sizes, they can often present a combination of extremely high strength and excellent toughness that modern industrialized materials cannot match [1]; among the various natural structural patterns, the Bouligand structure has become a focal point deeply explored in materials science and biomimetics in recent years, precisely relying on its unique advantages in enhancing damage tolerance and resisting external impacts; this structure was first documented by a French biologist in the 1960s, and its most critical feature is taking microscopically aligned fibers, like chitin or collagen, forming them into a sheet first, and then continuously stacking these sheets vertically at a fixed rotation angle, thereby creating a three-dimensional shape looking like twisted plywood; since this architecture can be found everywhere in natural environments, especially growing most abundantly on biological parts that often have to withstand fast and forceful impacts, the purpose of this article is to comprehensively review the exploration status of this structure, explaining in detail its natural original appearance, mechanical conditions, artificial manufacturing methods, and where

it can be used in the future, thereby providing some inspiration for designing new biomimetic materials with better performance.

## 2. NATURALLY EXISTING BOULIGAND STRUCTURE SAMPLES

Many animals and plants rely on growing this structure in the most important positions of their bodies to meet the mechanical conditions needed to survive and catch prey, and these natural samples have brought very precious inspiration to researchers; point one is the dactyl club of the mantis shrimp, whose club section can be considered the most famous example in the natural environment, as the smashing-type mantis shrimp can swing its club at a fast speed of twenty-three meters per second and with several thousand newtons of force to smash seashells or crabs [2]; the impacting area and the intermediate connecting area of its club are mainly composed of densely packed chitin fibers containing minerals, and in this connecting area, the fibers are arranged in a perfect Bouligand stacking pattern, which can particularly effectively dissipate the forces striking from the outside and block those potentially fatal large cracks, thereby demonstrating extremely powerful damage tolerance, as shown in Figure 1; point two is the outer hard shells of crustaceans, such as the outer shells of lobsters, crabs, and insects, which are actually also built from chitin fibers using this structure, and this special arrangement not only protects their soft bodies from being injured by outside forces but also allows the fibers to slide against each other to coordinate with the body's various movements; because the stacking angles of the fibers in different parts of the body differ greatly, this also shows that living organisms have made flexible changes to adapt to the mechanical conditions in different places; point three includes the scales of fish like the arapaima or the ancient coelacanth, and even inside the bones of mammals, varying degrees of this similar spiral-like arrangement have also been found; it is exactly these special architectures that bring biological organisms a comprehensive protection that is lightweight, sturdy, and elastic [3].



**Figure 1.** Schematic diagram of the basic morphology of Bouligand structure: (a) Mantis shrimp dactyl club, (b) Microstructure, (c) Geometric configuration

## 3. MECHANICAL PRINCIPLES ALLOWING THIS STRUCTURE TO MAKE MATERIALS TOUGHER

The reason it has such formidable fracture resistance is not actually because the microscopic substances comprising it are so special, but because when a cracking fissure spreads within this complex spiral three-dimensional space, the path it takes is irregular, and the principles that can make the material tougher mainly include the following aspects; one aspect is the deflection and twisting of the crack, where when those cracks encounter the boundaries between different layers inside this structure, because the direction of the fiber arrangement is constantly changing, it is very difficult for the crack to continue breaking along a straight line, and it is more willing to turn towards the direction of the weaker boundary surface of the fiber arrangement, being forcefully squeezed out of the area

experiencing the greatest stress; and as this crack spreads forward in circles between different layers, the foremost part of the crack is forced to twist along with it, thereby creating a highly complex and twisted cracking surface, and in this way, because the cracking route is greatly lengthened, creating a new fracture surface requires consuming far more energy [4]; another aspect is the bridging effect between fibers, which means that during the process where this structure is about to break apart, the fibers sandwiched in the middle layers are usually not all cut off at once, but rather build a bridge-like connection across the two sides of the cracking fissure, and these bridging fibers can help share a portion of the external pressure, generating a force that resists the widening of the crack, thereby very effectively reducing the overly concentrated stress at the very tip of the crack and delaying the process of the crack continuing to widen [5]; yet another aspect is the redistribution of stress and the consumption of energy, as this spiraling three-dimensional architecture brings the material a characteristic of performing similarly regardless of which planar direction the force comes from, which allows the material to steadily withstand pulling or shearing forces from any direction; at the same time, the sliding of the sandwiched fiber networks against each other, the breaking and reconnecting of internal chemical bonds, and the deformation occurring in the soft basic substances all provide a set of methods for vastly consuming the external impact energy.

#### **4. VARIOUS METHODS FOR ARTIFICIALLY MIMICKING AND MANUFACTURING THIS STRUCTURAL MATERIAL**

Having been inspired by these ingenious architectures in nature, researchers have figured out several ways to build this biomimetic material, which can generally be summarized into the following types; the first point is utilizing 3D printing technology, which is a highly effective method especially when building fiber-reinforced composite material structures at visibly larger sizes and millimeter scales [6]; as long as the printing route, the extrusion speed, and the flow state of those tiny fibers in the ink are well controlled, one can very accurately grasp the orientation of the fibers in the plane and the rotation angle between different layers; taking the use of electric or magnetic fields to help control the orientation of the short fibers inside the printing ink as an example, researchers have already used this to create that kind of biomimetic mantis shrimp club material that is particularly good at withstanding impacts; the second point relies on microscopic assembly and shearing methods, which means borrowing external shearing forces to guide those elongated microscopic fibrous substances, like carbon nanotubes or cellulose nanocrystals, making them line up, and combined with the technique of stacking several layers together, one can create a Bouligand structure that is regular at the nanoscale; the relatively common practices here include stretching while shearing, smearing while shearing like brushing paint, and heating while pressing tightly; the team led by Academician Shuhong Yu from the University of Science and Technology of China proposed a construction concept for a moderately ordered structure, where they pulled a net-like nanofiber gel film along one axis, guiding the fibers to align in a semi-regular state and lock together, and then stacked them in circles, ultimately creating a new biomimetic material that is particularly outstanding in both overall mechanical performance and stability; the third point is the method of letting the material assemble itself, where for some elongated nano-basic substances, as long as their concentration in water reaches a specific point, they will naturally generate a chiral liquid crystal state, and this liquid crystal structure, once the water dries, can turn into a perfectly flawless nanoscale spiral three-dimensional architecture; this is actually a preparation routine that builds slowly from the very bottom up and saves a lot of energy, making it perfectly suitable for creating strengthened thin-film materials with special optical functions.

#### **5. FINAL SUMMARY AND PREDICTIONS FOR FUTURE USES**

Just as comprehensively discussed previously, precisely because the Bouligand structure has excellent characteristics of increasing toughness, resisting impacts, and withstanding damage, it has

drawn the ultimate blueprint for designing composite materials that have both high performance and multiple functions; after thoroughly dismantling the original samples in nature and very accurately turning the mechanical principles into models, scientists can now rely on methods like 3D printing, moderately ordered assembly, and letting liquid crystals self-assemble to build artificial biomimetic materials carrying this structure across different size levels; looking into the future, these artificially created structural materials will have very broad areas of application; for example, in the aerospace and lightweight transportation industries, lightweight composite material shells that are particularly capable of withstanding impacts from space rocks or tiny particles can be developed; and in the national defense and military sectors, new high-performance bulletproof vests, safety helmets, and protective armor can be manufactured; moving to the medical and healthcare field, because human bones and cartilage also feature a similar layered spiral three-dimensional architecture, these artificial biomimetic structural scaffolds are very promising for use as tissue repair materials capable of consuming impact energy, which can satisfy the mechanical and biological growth requirements when replacing joints and repairing bone defects; however, currently, there are still quite a few difficulties encountered when researching this biomimetic material, which include how to perfectly piece together fibers as small as nanometers inside relatively large bulk materials, how to make the areas where different layers touch stronger, and how to mass-produce them in factories on a large scale; as long as these tough challenges can be resolved, it will greatly push fiber-reinforced composite materials into a new era where they become tougher and the degree of damage can be effectively controlled.

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