Bioinspired Robust Mechanical Properties for Advanced Materials

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Nature is a substantial repository for various kinds of aquatic, terrestrial, and windborne plants and animals adapting extensively to sustain life on earth’s different environmental conditions. The organisms or structures of these natural creatures possess extraordinary properties or functionalities through well-organizing usually weak biopolymers and bioinorganic structures, which provide good examples to emulate their mechanical and structural characteristics in materials innovations and discoveries. This review collectively investigates the studies of the structures and mechanical properties of some representative fauna and flora with robust mechanical properties and the corresponding bioinspired materials with mimicking structures and mechanical properties. By learning from the natural structures with robust mechanical properties, bioinspired materials and composites with superior mechanical performance to the constituent materials have been designed and fabricated. Via this study, there is hope to draw some principles for designing innovative materials with extraordinary properties from existing common materials by learning from nature. It is expected that the understanding of the extraordinary natural mechanical properties and the robust bioinspired materials can provide some insights into the design of novel materials and composites.

1. Introduction

Evolution means life forms have found optimal ways to survive amid harsh environmental changes with their structural and functional specifications evolving significantly from primitive to extremely advanced forms. Scientists today look toward nature to develop superior quality materials, which surpass the properties of nature. Biomimicry and bioinspiration provide a key to developing advanced artificial materials having excellent performance and enhanced sustainment abilities as those natural targets.[1,2] The term “bioinspiration” conveys the idea of designing materials with better performance by learning from nature. Current researchers are intrigued by these adaptations and starting to develop man-made bioinspired nanomaterials with extraordinary properties based on natural species with interesting properties. The fabrication of bioinspired nanomaterials integrates the unique physical and chemical properties of well-evolved biological structures and functions with the well-applied artificial materials to create unique advanced materials that exceed their normal forms. The increasing demand for sophisticated materials for emerging applications has led to research to identify architecture and composition to create bioinspired materials and address global challenges.

Many biological structures have inspired structural innovations for antifogging, antifouling, hydrophobic surfaces, super wettability, extraordinary adhesion, wetting, optical behaviors,[3–9] and other fascinating architectures, including dense composites, porous materials, flexible membranes and films, and fibrous components. For example, some biological components have interesting surface properties such as gecko feet, spider silk, fish scales, pitcher plant (Nepenthes alata), nacre, and fly-eyes. Inspired by these elements in nature, researchers have endeavored to develop novel artificial bioinspired materials mimicking the surface geometry and/or chemistry for applications in self-cleaning coatings, super-adhesive fibers, optoelectronics, energy conversion and storage devices, and antivirus coatings.[10–15] For most engineered materials, mechanical properties are critical in terms of high strength, lightweight, and high stiffness acting as a driving force to investigate the remarkable material aspects of nature. Nature is a library of great fauna and flora, which has evolved with mechanically robust parts or organs to meet the functionality needs of strong, tough, and sharp qualities, and has inspired the researcher to design stronger and tougher materials.[16–19] Plausible progress in the investigations on both natural mechanically robust structures and bioinspired artificial advanced materials by learning from nature has been achieved in past decades. A review on recent progress in bioinspired mechanical properties and insights into the design
principles of bioinspired advanced materials with robust mechanical properties, therefore, are highly desired for materials research. In this review, we first summarize the biological structures with robust mechanical properties in the categories of natural toughness and strongness, natural sharpness and rigidity, and natural tolerance and resilience. Then, the bioinspired materials are summarized into the corresponding three categories. As an expansion on the applications of bioinspired advanced with robust mechanical properties, some typical examples of the mechanically bioinspired materials in emerging sustainable energy and environmental technologies are also briefly reviewed. Based on the detailed examples and discussion, a summary of this review together with an outlook on the challenges and perspectives are provided in the last section. Figure 1 presents the graphic summary of the structure of this work, where learning of the eminent mechanical properties of natural species is leading to the design of mechanical robust artificial materials and providing insights into bioinspiration and biomimicry.

2. Biological Structures with Robust Mechanical Properties

Biological structures have evolved in both fauna and flora in the processes of surviving predators, hunting for food, and succeeding in a competitive environment. All faunae have evolved with one or more artifacts, such as strong mechanical appendages of teeth, fangs, stings, armors, and crusts, which present surprisingly outstanding mechanical performances but share very common biocompositions, for example, proteins and biocarbonates. Similarly, plants have also developed diverse structures and strategies starting from seedpods to roots to sustain life against biotic and abiotic stressors with similar compositions consisting of celluloses, hemicelluloses, and lignin. Among versatile amazing properties of natural structures, super-stretchability, super-adhesion, extraordinary toughness and hardness, and fantastic resilience are a few of the ubiquitous characteristics that current research is investigating. The study of the relationships between the structure-mechanical properties of these specific parts and structures can provide general principles for designing advanced materials and composites.[20–23]

Collectively, biological structures can be either dense composites, porous materials, or flexible textures depending on the adaptations. Specific architectural alignments and compositions of these biological structures lead to overcoming the compromise of mutually exclusive inbuilt mechanical properties such as toughness and strength. Specifically, some natural structures provide not only light weight and low density but also high strength and high toughness through combining alternating soft and elastic proteins/biopolymers and hard but weak biocarbonates into a dense composite, which provide design principles for achieving light, strong, and tough artificial composites.

In this section, we intend to classify the natural robust mechanical properties into three representative categories, i.e., natural toughness and strongness, natural sharpness and rigidity, and natural tolerance and resilience. Typical examples

Figure 1. Graphic summary of designing mechanically robust artificial materials inspired by prominent mechanical properties from nature.
aligned with these categories are introduced and the mechanisms and the structural origins of the mechanical properties are summarized and discussed. It is expected that the understanding at the molecular level to the macroscale level of the extraordinary natural mechanical properties can provide some principles for the design of bioinspired materials.

2.1. Natural Toughness and Strength

Toughness and strength are mutually exclusive properties that incorporate elevated mechanical characteristics. Toughness is the resistance to fracture, which is generally measured by the energy needed to initiate a crack and can be classified as intrinsic and extrinsic toughening in the event of a fracture. The intrinsic toughness is more conspicuous in plasticity-associated materials as opposed to extrinsic toughness.[24] Specifically for brittle materials, we use fracture toughness to quantitatively describe the resistance of a material to brittle fracture when a crack is present or the ability of a material containing a crack to resist further fracture.[24]

In the field of materials, strength is the ability of a material to withstand an applied load without failure or plastic deformation. While we are very eager to get strong and tough materials, strength and toughness are mutually exclusive.[25] Usually, the lower the strength means the material is more ready for deformation and the stress is more easily to be distributed among defects and cracks for reaching higher fracture toughness.[25] However, strength and toughness indeed coexist in nature, which surely can provide us inspirations to overcome this challenge.

2.1.1. Tough and Strong Natural Nacre Structure

One representative biological structure for the coexistence of strength and toughness is nacre, which has attracted increasing research interest for its high strength, notable toughness, and beautiful light reflectance.[13,24–31] Nacre is the shell of the “mother of pearl” constructed from a layered brick-and-mortar structure and a similar structure has been identified within the shells of various types of shellfish and mollusks in either fresh water or salt water. Some very pioneer studies have revealed that the tensile strength and modulus of nacre along the lamellar direction are 80–130 MPa and 60–70 GPa, respectively.[18–22] Barthelet et al. studied the hardening mechanisms of layered structures including nacre and found the interfaces together with the tablet structure are important factors impacting crack propagation, strain, shear, and tension during mechanical impacts.[22]

Further studies demonstrated that the high level of toughness is the result of mineral bridge breaking between the calcium carbonate platelets and the shear resistance is caused by nano-asperities, viscoelastic glue provided by the organic layer, and tablet interlocking.[20–22]

Figure 2A presents the shell structure of a bivalve mollusk P. nobilis, where polygonal (or pseudo-hexagonal) aragonite (CaCO₃) platelets with a size of 0.5–1 μm in thick and 10–20 μm in wide are alternatively stacked with thin layers of biopolymer at a thickness of ≈5–30 nm (an interlamellar membrane) to form the nacre structure.[23] By using a high-resolution scanning/transmission electron microscope (S/TEM) combined with an in situ nanoindentor, Gim et al. observed the deformation and recovery behavior of the nacre of Pinna nobilis shell.[23] It reported that the interlocked nacreous aragonite responds to the strain as a continuous material and the organic boundaries restrict the crack propagation, which allows the storage of 80% mechanical energy. The viscoelastic nature of the nacre tablets leads to full recovery during the in situ TEM compression, which helps the tablets to bounce back to the previous position (Figure 2B). These properties of nacre are that although these have brittle minerals and weak polymers, synergistically these result in tough and hard biological materials.

In a very recent work reported by Liu et al., the interface strength and the fracture mechanism of the “brick-mortar” structure in an abalone shell (Figure 2C), a typical nacre structure, have been investigated by using microsized cantilever beam and bend samples.[26] The microsized samples with a length of ≈5 μm and thickness of ≈500 nm were cut by a focused ion beam (FIB) system and then tested under a nanoindentation tester. Compared with the interface strength of 41.5 MPa tested on bulk nacre, a high aragonite/biopolymer interfacial strength of ≈298 MPa was observed together with high toughness. As shown in Figure 2C, the high interfacial strength is resulted from the polymer chain bridging which connects two neighboring aragonite layers during the interface separation, and the high toughness is contributed by the crack deflection along the aragonite/biopolymer interfaces. The authors also analyzed the competition between the crack deflection to the interface and the penetration into the aragonite platelet as a function of interfacial angle and normalized energy release rate, by which the crack propagation is predictable if we can know the interfacial angle and interfacial toughness.

Based on the previous reports and the abovementioned examples, it can be concluded that the high strength and high toughness of the nacre structure are the results of a collective contribution from molecular scale boundaries to macro-scale brick and mortar structures that prevents crack deflection by harnessing toughness in the inbuilt architecture. On the one hand, the availability of mineral bridges offers the capability to prevent the lateral growth of cracks and the easy delamination along the aragonite interfaces, enabling the withstanding of high stress to achieve high strength. On the contrary, the relatively weak interfacial strength and the soft biopolymer, even though a value as high as 298 MPa has been reported by Liu et al., provide a perfect pathway for the dissipation and adsorption of mechanical energy and prevent the structure from catastrophic failure. Of note, the growth of nacre plates differs in bivalves and gastropods, while the formation of mineral bridges and existing polygonal nano tablets ultimately leads to similar toughening mechanisms, including crack deflection, fiber/tablet pull out, organic matrix bridging, dilatation band formation, nanograin rotation, and so on.[26] The unique microstructure makes the nacre outsmart other biological structures to exhibit not only high strength but also high energy dissipation/absorption capacity for high toughness.

2.1.2. Tough and Strong Natural Stomatopod Dactyl Club Structure

Besides the nacre structure, it has been found that the dactyl clubs of mantis shrimps (Odontodactylus scyllarus) have a high strain and impact tolerance property to withstand extreme forces.
Figure 2. Natural nacreous structures with exemplary toughness and strength. A) Photograph of bivalve mollusk *P. nobilis* and the microstructure response for compression showing the complete recovery of nacre tablets, and B) in situ transmission electron microscope (TEM) compression tests on the nacreous tablet showing the elastic recovery after a series of indentations due to the existence of organic inclusions in between the inorganic platelets. Reproduced under the terms of the Creative Commons CC BY license with permission.[23] Copyright 2019, The Authors. Published by Springer Nature. C) Micromechanical properties of the nacre structure cut from an abalone shell. Reproduced with permission.[26] Copyright 2020, Elsevier B.V.
Figure 3. Natural dactyl club structures with exemplary toughness and strength. A) Mantis shrimp \((Odontodactylus scyllarus)\) and dynamic finite element analysis (DFEA) on the stress distributions within the stomatopod dactyl club upon impact. Reproduced with permission\(^{[27]}\) Copyright 2012, American Association for the Advancement of Science. B) Depth-sensing nanoindentation analysis of the mechanical responses of the impact surface and the inner layers of a dactyl club under both dry and hydrated conditions. Reproduced with permission\(^{[28]}\) Copyright 2015, Springer Nature. C) Microstructure of the impact surface and the impact region of the dactyl club of the mantis shrimp. Reproduced with permission\(^{[31]}\) Copyright 2020, Springer Nature.
 (>700 N) at the point of strike.\cite{27,28,30–32} This type of shrimps, as shown in Figure 3A, has a set of hypermineralized hammer-like dactyl clubs, which provide high-velocity impacts to smash the heavily armored prey during feeding activities.\cite{27} The microstructural compositions and the microstructural alleviation of mechanical damage have been evaluated extensively. The dactyl club is the most electron-dense region of the stomatopod exoskeleton and can be divided into three distinct regions: the impact region, the periodic region, and the striated region.\cite{27} Different to the nacre, it has platelets with a high degree of crystallinity in hydroxyapatite and strengthened by an angular arrangement of chitin fibers as the matrix, the high level of toughness of the dactyl clubs is correlated with the fiber arrangement which introduced resistance to even micro cracks. In the study of Weaver et al., the dynamic finite element analysis (DFEA) revealed the stress distribution within the dactyl club upon mechanical impact. Even subjected to extremely high hydrostatic compressive stress, no catastrophic failure of the dactyl club was observed for its high ability to sustain high-level localized impact pressure.\cite{27} In this analysis, the toughening of the dactyl club is contributed by the gradient structures: 1) the outmost layer with a high level of mineralization is ultrahard to sustain maximum impact force; 2) a transit layer between the impact surface and the impact region provides modulus mismatch that acts as a crack deflector; 3) the periodic region under the impact region with pitch-graded helicoidal chitin fibers and modulus oscillation dissipates impact energy and shields catastrophic crack propagation.

Amini et al. further analyzed the micromechanical responses of the dactyl club under deformation by using a depth-sensing nanoindentation technique with both spherical and sharp contact tips (Figure 3B).\cite{28} The load–displacement responses under nanoindentation were recorded from both the impact surface and the inner layers. Under dry conditions, the impact surface presented a clear elastic–plastic transition with a gradual drop in yield stress moving towards the inner layers, where the yield stress was 3.5–5.0 GPa for the outer layers but it decreased to 1.6–2.1 GPa for the inner layers. Under hydrated conditions, the mechanical differences became more obvious, and a strain-hardening type behavior was observed in the indentation stress–strain response of the inner layers. Combining the mechanical response analysis with the microstructure characterizations, it is concluded that the impact surface has a quasi-plastic contact behavior resulted from the interfacial sliding and rotation of apatite nanostructure, resulting in significant energy dissipation by localized yielding, and the inner layers composed of chitin and amorphous mineral microchannels exhibited strain hardening, which leads to lower compressive strength but much higher yield strains to accommodate the damage energy as a result of microchannel densification.\cite{29,30}

Huang et al. carried out a detailed study on the nanostructure and the multiscale energy dissipation mechanism of the impact surface and the impact region.\cite{31} Figure 3C shows the microstructure of the impact surface and the impact region of the dactyl club of mantis shrimp, which is the most critical section to localize damage and avoid catastrophic failure from high-speed collisions.\cite{31} The impact surface is about 70 μm in thickness and consists of densely packed, bicontinuous, highly mineralized hydroxyapatite (HAP) nanoparticles embedded in an organic matrix. The impact region is a layer beneath the impact surface of about 500 μm and has a herringbone-like structure of nanocrystalline HAP mineralized chitin fibers. Compared to the nacre structure, the dactyl clubs are capable of a greater damage resistance with twice of energy absorption under similar strain impacts. This superior energy absorption ability is due to HAP nanoparticles integrated within the organic matrix. While the nanoparticles rotate and convert to accommodate the high strains, the interpenetrating organic network provides additional toughening and substantial damping by the formation of dislocations and amorphization.

Similarly, the mandible of crayfish (\textit{Cherax quadricarinatus}, freshwater crayfish) also possesses a specific outer layer comprising a crystalline, wear resistant, enamel-like apatite layer and softer amorphous minerals in the bulk.\cite{33} The composition gradient and the mechanical response behaviors are very close to those of dactyl clubs. The mechanical properties examined by nanoindentation showed that a gradient changes of hardness from 4.5–5 GPa of the outer crystalline apatite layer to about 1 GPa of the inner bulk of the mandible consisting of chitin and amorphous calcium carbonate. These layers possess an outstanding ability to dissipate the impact energy and resist the potential damage caused by predators.

Therefore, the toughness and strength of shrimp dactyl club is another extraordinary creation in nature and can be a principle to design impact-resistant materials based on growing oriented attached mesocrystalline structures around organic networks.\cite{32} These investigations provide us useful insights into designing synthetic multiscale/multicomponent materials and structures, which can be not only hard and strong on the contact surface but also tough and tolerant with the co-existence of soft phase and stiffness gradient along the transverse direction to bear high velocity and high load damage impact.

2.1.3. Tough and Strong Natural Bone Structure

In nature, the bone structure of either animals or humans is another excellent example of high strength and high fracture toughness and possesses unique microstructure and chemical compositions to prevent catastrophic fracture while sustaining superhigh strength. The cuttlebone within the cuttlefish, an inhabitant in the deep sea has been discovered recently as an ultra-lightweight cellular structure, which helps the cuttlefish maintain buoyancy in different seawater levels but can withstand water pressure of 20 atm. Figure 4A presents the photographs of cuttlefish and the mechanical responses of the cuttlebone under compression observed by a synchrotron micro-computed tomography (CT).\cite{14} The cuttlebone is consist of brittle aragonite with ≈93 vol% porosity. The relationship between the macroscopic response and the microstructure of cuttlebone was established to explain the reinforcement of the architecture and how it supports the pressure difference at various pressure levels using digital imaging of the structure–mechanical correlation. The observations indicated that the chambers and the septa are developed with a high stiffness along with subtle energy dissipation at failure. On compression, the aragonite structure showed three stages, including at first stepwise elastic deformation stage, a serrated stress plateau regime, and a densification stage, implying
an extremely high energy absorption of 0.6–1.5 MJ m$^{-3}$ at the density of only 180–260 kg m$^{-3}$. This work revealed that the cuttlebone is optimized to be lightweight, with high stiffness (8.4 MN m kg$^{-1}$), and high energy absorption (4.4 kJ kg$^{-1}$), simultaneously, so providing ideas on how to design low-density, stiff, and damage-tolerant cellular ceramics. The simulations, tailored in the chamber levels, suggest that the higher the waviness is, the lower the stress of failure (Figure 4B). A strength-controlled fracture and buckling mechanism was identified in this study and the waviness in the cuttlebone provided a high-level of energy absorption from its unique architecture, which will be beneficial for understanding mechanism of toughness.

The human bone is another representative example that possesses simultaneous strongness and toughness. The origin of the fracture toughness in the human cortical bone has been identified from a hard inorganic (mostly carbonated hydroxyapatite)—soft organic (mainly type I collagen) interconnected structure.[35–37] In a typical bone hierarchical structure, as shown in Figure 4D, at the macroscale, the bone consists of a compact and dense structure (cortical bone), which can be found at the surface of all bones, and a spongy or cancellous structure underneath the hard surfaces (trabecular bone). At the microscale, the trabecular bone structure is cancellous with struts of 100–300 nm in diameter, contains marrow, and contributes most to the functionalities of the bones.[35] The compact bone structure, which provides the greatest mechanical support to the body, comprises mineralized lamella osteons formed by collagen fibrils, linked by protein molecules or tropocollagen and Haversian canals. The structure resembles the composite and matrix structure akin to the nacre and dactyl club structures. Contributed to the hierarchical structure consisting of protein molecules, fibrils, and fibers (Figure 4E), the bone has an intrinsic toughening mechanism occurring at the nanoscale ascribed by the tropocollagen molecules and the collagen fibrils, which facilitate the molecular uncoiling and intermolecular sliding and avoid the propagation of the crack. An extrinsic toughening also occurs at the microscale resulted from the fibril arrays and ligaments, which deflect and bridge the cracks (Figure 4F).[24] Upon mechanical damage, the deformation of bones involves molecular levels of collagen fibrils and the bonding in tropocollagen resists catastrophic brittle failure.[36] The tensile strength and strain indicated that up to 40% of bone is sustained, due to the availability of H-bonds in collagen. The bone matrix absorbs energy thereby having an energy dissipation ability. While the crosslinking of collagen fibrils is less effective to sustain strength and toughness, the mineral bridges...
of hydroxyapatite formed in collagen fibrils play a major role in the continuous strongness and toughness of bones.

2.1.4. Tough and Strong Natural Wood Structure

Plants are one of the most ancient natural species and have been long existing in almost everywhere for hundreds of million years on the earth. Regardless of the environmental changes from era to era, they have thrived under every change. The diverse growth environments, of course, have adapted the plants with compatible structures and functionalities.\[38–42\] Equally important components in a plant with unprecedented high strength engineering properties are wood, seeds, and bark.\[42–44\] Other parts of the plants, such as wood stems, pinecones, and bamboo stems, also illustrate the existence of a stiffness gradient from the periphery to the center of the plant tissues, where the coexistence of pores and fibers enhances the sustainment and roughness of the plant hard tissues. Wood from the trees or large plants has been the most important structural material throughout human civilization. Wood is known to be one of the most abundant natural materials on earth, with its main compositions being lignin (20–35%), cellulose (30–50%), and hemicellulose (15–30%).\[42\]

Usually, lignin acts as a network to provide physical rigidity and strength via assembling all cellulose fibers together, and cellulose and hemicellulose are responsible for the microstructure and porosity of the transportation channels in plants. In mature plants, the cell walls saturated with lignin (sclerenchyma tissue) become hard and rigid to survive in the final stages of the life cycle. Besides the direct use of wood as timber for building constructions, the mechanical strength of wood has also been directly modified into artificial strong and tough materials. Based on the investigations on understanding the component-structure-functionality relationships, cutting-edge research on sustainable or biodegradable wood-derived structural or mechanical components is currently underway.\[45–47\]

Many soft or tough biostructures in plants have been studied by applying atomic force microscopy or nanoindentation characterizations, providing useful information on how plants build mechanical robust trunks and branches via these biostructures.\[47–49\] In a plant world, the trunks, stems, and some leaves are developed to maintain a gradient biological structure to reach mechanical strongness and toughness from biological lignin and cellulose fibers that usually have inferior mechanical performances.\[50–52\] Figure 5A presents the hierarchical structure of typical wood from the molecular scale (molecules of cellulose, hemicellulose, and lignin structures), nanoscale (molecular chains), microscale (elementary fibrils, microfibrils, fibrous bundles), then into bulk (wood bulk). With cellulose and lignin reinforcement, the natural wood exhibits exceptional tensile strength and toughness for building and construction applications.\[53\] A large fraction of pores exist in the natural wood, unfortunately, which results in a noticeable decrease in mechanical performance, such as hardness and modulus, even though they provide better damage energy absorption and deformation capability.

While the natural wood is yet inferior in mechanical properties, i.e., a strength of \(\approx 46\) MPa and work of fracture of \(\approx 0.39\) MJ m\(^{-3}\) for natural wood, densified wood with much enhanced strongness and toughness has been developed by partially removing the lignin and hemicellulose from the natural bulk wood via chemical treatment and then undergoing hot pressing.\[54,55\] As shown in the microstructures examined before and after densification (Figure 5A), the wood cells existing within natural wood were fully collapsed together with a reduction of the thickness of about 20% and a threefold increase in density. Surprisingly, as the mechanical properties presented in Figure 5B, the densified wood displayed a 12 times enhancement in tensile strength (\(\approx 548.8\) MPa) and tenfold improvement in the work of fracture (\(3.9 \pm 0.2\) MJ m\(^{-3}\)) compared with the natural wood. The toughening and strengthening mechanisms of the densified wood should be the high degree of aligned cellulose nanofibers and the increased interfacial contact as the result of wood cell walls packing and intertwining and the hydrogen-bond formation, breaking, and reformation at the molecular scale. The strong, tough, and lightweight densified woods make this type of material a promising potential in the applications in low-cost armor and ballistic energy absorption.

Inspired by the reinforcement mechanisms of natural wood, Wang et al. developed mechanically robust and hydro-stable cellulose-lignin composite straws from mixed cellulose microfibers, cellulose nanofibers, and lignin powders.\[56\] The mechanical properties of the straws strongly correlate with the content of lignin. As shown in Figure 5C, the tensile strength increased from \(\approx 40\) to \(\approx 70\) MPa with the additional amounts from 0% to 26% and then decreased dramatically to around 35 MPa with the further addition of lignin to 36%. Simulation and modeling were performed on how lignin and cellulose wood work together with the formation of hydrogen bonds to reach strongness and toughness (Figure 5D).\[56\] The hydrogen bonds formed more efficiently among the molecular chains with the presence of hemicellulose and lignin units than in the case without hemicellulose and lignin, indicating the critical role of hydrogen bonds among the cellulose chains and the improved efficiency contributed by the addition of lignin and hemicellulose.

Inspired by the successful approach to fabricating strong and tough wood via densification, some works have been carried out, for example, to fabricate highly elastic, ionically conductive, anisotropic, or elastic wood materials from natural wood.\[48–52,54–57\] Chen et al. reported the fabrication of an elastic wood that can sustain a compressive strain up to 70% but with negligible plastic deformation and a small energy loss coefficient, owing to the formation of a honeycomb-like structure with an interconnected cellulose fibril network with the partial removal of lignin and hemicellulose.\[57\] Similar to the fabrication of elastic wood, the bamboo stem has also been chemically treated to remove the lignin and hemicellulose from the lignocellulosic cell walls to obtain the well-aligned cellulose fibers.\[58\] After densification via hot-pressing with a 70% reduction in thickness, the densified bamboo reached a record high tensile strength of \(\approx 1\) GPa and a toughness of 9.74 mJ m\(^{-3}\), which is significantly greater than most structural materials including steel and alloys, and provides a promising candidate for future green building materials.

Zhu et al. proposed a scaling law of the mechanical properties of wood materials as a function of constituent cellulose fiber size and designed interesting cellulose-based nano-paper with both attractive strength and toughness.\[46\] The toughness and strength of the nano-paper can be increased simultaneously with the
Figure 5. Natural strong and tough wood structures. A) Formation of wood with unit cells from the molecular scale to microscale and then bulk. Reproduced with permission.[54] Copyright 2021, Wiley VCH. Fracture surface after tensile tests in natural and densified wood. Reproduced with permission.[55] Copyright 2018, Springer Nature. B) Corresponding tensile tests and comparison of strength and work of fracture. Reproduced with permission.[55] Copyright 2018, Springer Nature. C) Tensile strength variation with the change of lignin content. D) Simulations on how cellulose, lignin, hemicellulose, and hydrogen bond form during the densification of natural wood. Reproduced with permission.[56] Copyright 2021, Wiley VCH.
decrease of the diameter of cellulose fibers. An increase of 130- and 40-fold, respectively, in strength and toughness was achieved, when the fiber diameter decreased from 27 μm to 11 nm. The cellulose paper showed a remarkable tensile strength of 275.2 MPa and work of fracture of 0.13 MJ m⁻³, which confirms the scaling law of the mechanical properties of cellulose nano-paper that the smaller the fibers, the stronger and the tougher the fibril products.

The aforementioned examples demonstrate that the quintessential structure of lignin in plant cells has initiated ideas to fabricate materials with exceptional strength and toughness. Evaluating the contributions of the hemicellulose, lignin, and the bonding behaviors between the cellulose fibrils in constructing elastic and natural wood can surely provide hints into the design of advanced materials and composites with high both elasticity and toughness. Even though relatively few plants have inspired material development to date, it is expected that more intelligent materials and structures will be inspired by this largest group of creatures on earth.

The toughness and strength of organisms in nature, have evolved to survive in challenging environments. The intelligent configuration and organization of naturally weak and brittle materials with soft biopolymers in faunas has created hard and tough shells, armors, bones, and scales to survive from predators, prey on food, and minimize mechanical collisions. In plants, via a similar configuration philosophy to assemble the soft molecules and chains of lignin, cellulose, and hemicellulose into hard and strong trunks, stems, branches, tip of leaves, and bulk structures to sustain the plants in strong windy and snowy weathers. These natural structures presented superior strong and tough mechanical properties and provide us with lots of design principles to fabricate artificial strong and tough composites or materials from weak or brittle starting materials via proper bioinspiration and biomimicry.

2.2. Natural Sharpness and Rigidity

Sharpness and rigidity are another class of interesting properties that can be observed in natural species. Similar to the strongness and toughness reached via the intelligent assembly of weak and brittle components with soft and elastic molecules, the sharpness and rigidity are also the result of perfectly organized biostructures and biomaterials. The sharp and rigid parts of fauna and flora play critical functions in feeding and surviving. The higher the sharpens and rigidity, the lesser the force needed for penetration. Generally, sharpness is quantified by the force required to penetrate a substrate, and in terms of the appendages of organisms, the puncture ability, and the miniature of the body size. From the point of view of materials research, the studies on the natural sharpness and rigidity can help the design of robust and wear-resistant micromechanical components, needles and nails, and microchannels and tunnels.

Porcupines are well known for the rigid and easily abscised hairs or quills for self-defense (Figure 6A). The unique microstructure of the quills has a foam-like structure contained in a hard cortex cylindrical shell, which allows an easy release and penetration into the tissues of a predator when the porcupines are under threat (Figure 6B). Tee et al. studied the compression and buckling behaviors of the whole quill, the shell only samples without the form structure, and vice versa A three-stage deformation model was observed for the whole quill and the foam structure, which includes a linear early stage, followed by a plateau stage resulting in the buckling of the rigid shell and the support of the foam and stiffeners, and finally, the collapse and density of the quill (Figure 6C). This mechanical response of the porcupine quill coincided well with one previous study performed by Yang et al. The microstructure analysis of the quill demonstrated that the foam structure accommodates the wavy structure of the shell during the deformation, the cortex carries the majority of the mechanical load, and the robust connection between the foam and the cortex shell results in crushing properties. The lightweight and excellent collapsible energy absorption properties of the quills allow the porcupines to jab the sharks into the body of enemies quickly and deeply while maintaining the integrity of the quills. Therefore, the form structure in porcupine quills increases the buckling strength, the buckling strain, and the elastic strain energy absorption, which provides insights into the design of lightweight and buckle resistant materials.

Besides the unique buckling resistance and mechanical energy absorption performance, the characteristics of easy penetration and difficulty to remove porcupine quills have also been noted. Cho et al. studied the barb structure and examined the punctation performance of quills, which can inspire the design of needles (Figure 6D). The barbed quill is backward, allowing the low penetration force to penetrate tissue (0.33 ± 0.08 N) but resisting the quill inside the tissue when pulling out with a much-increased pull-out force of 0.44 ± 0.06 N (Figure 6E). It is also noted that a lower penetration force and a higher penetration ability were observed for the barbed quill than for the barbless quill. This interesting find can inspire us to design the needles or patches for the applications, where easy penetration and strong substrate adhesion are requested, such as local anesthesia, abscess drainage, vascular tunneling, etc.

Likewise, the scorpion has a sharp and rigid sting to protect it from and drive away predators (Figure 6F). The chemical composition along the sting has been characterized as main chitosan. The thick-walled cuticle of the sting consists of one exocuticle layer and one endocuticle layer. The exocuticle is homogeneous in terms of microstructure and an endocuticle is a multilayered structure composed of oriented chitosan fibers. The Young’s modulus of the exocuticle varies over ≈0.8–1.8 GPa along the axial direction, and is lower than 1 GPa for the endocuticle layer. The exocuticle has a stiffness higher than that of the endocuticle. This hollow structure in the scorpion sting allows the transport of venom and offers strong mechanical properties to penetrate the skins and tissues of prey or invaders and maintain the shape stability. In addition to experimental work, simulation insights (Figure 6F,G) have proposed that the stiffness and stress distribution of the scorpion sting are inversely correlated with the angle of penetration.

The insect stingers can provide us with more examples of natural sharpness and rigidity. The piercing stingers of the insects possess naturally elegant structures and robust mechanical properties. Through nanoindentation data computed into a 3D topographical image, a clear idea has been established about the structure and mechanical mechanism of the sharp stings of...
Figure 6. Natural sharp and rigid structures. A) Photograph of porcupine and porcupine quills. Reproduced with permission.\textsuperscript{[59]} Copyright 2012, Springer Nature. B) Cross-section of a quill and magnified image of internal architecture. Reproduced with permission.\textsuperscript{[60]} Copyright 2020, Elsevier Ltd. C) 3D structure and surface morphology change over compression of a quill. Reproduced with permission.\textsuperscript{[61]} Copyright 2021, Elsevier Ltd. D) Comparison of quill with a typical surgical needle and magnified areas showing the barbs of a quill. E) Representative curves of penetration and retraction for three types of quills, where the substrate is a muscle tissue. Reproduced with permission.\textsuperscript{[62]} Copyright 2012, National Academy of Sciences. F) Numerical simulation for scorpion stings. The optimum angle of insertion to reduce stress at the sting is calculated. G) Theoretical analysis of the optimum angle calculation to avoid higher stress on the tip. Reproduced with permission.\textsuperscript{[63]} Copyright 2016, Elsevier Ltd. H) Penetration and retraction forces of honeybee and wasp stings. I) Microstructure of stylets of wasp. J) Barb of the honeybee as seen in SEM micrographs. Reproduced with permission.\textsuperscript{[64]} Copyright 2015, The Company of Biologists. K) Photograph of a viper and fangs with respective SEM images.\textsuperscript{[67]} Copyright 2020, Elsevier Ltd. L) Comparison of fangs depending on diet and variation of sharpness index. Reproduced with permission.\textsuperscript{[68]} Copyright 2021, John Wiley & Sons, Inc.
insects. For example, a mosquito only needs ≈18 μN to pierce its proboscis into human skin, which is three orders of magnitude smaller than an artificial needle.[64] The style of honeybees and wasps have unique tactics for inserting venom into the host (Figure 6H–J).[65] The structure of a honeybee sting comprises one stylo and two lancets as well as a circular canal, while the paper wasp has a spiral morphology of the lancet to avoid hooking by the tissue fibers. It is proposed that the lancets provide extra reinforcement and support to the stings from mechanical failure. However, the structural difference of the honeybee sting makes it much more difficult to retract from a fibrous tissue (porcine skin) than the wasp, which verifies why honeybees always lose their stings after an attack. During penetration, the adjustable angle of insertion of the tip and a low puncturing force also plays a major role in avoiding mechanical failure.[65, 66]

Viper fangs can puncture their prey at a high striking rate and penetrate quickly into various skins. Most of the Viperidae genus possess fangs based on their habitat and food habits. Fangs are used to paralyze the prey which carries venom inside for fast numbing. Studies have assessed the sharpness index to understand the correlation between the fang angle and the puncture.[67, 68] The sharpness of fangs differs based on their eating habits. For example, a soil burrower has blunt fangs while prey-eating Viperidae reptiles have sharp fangs (Figure 6K).

Specifically, the easiness of penetration and puncture through the skin of prey was assessed by the rate of load approaching the target, the load or the stress initiating the crack during penetrating, and the ability to initiate crack propagation (Figure 6L). Puncture is efficient when attacked at a high velocity and it demonstrates that the time for one puncture takes only 50–90 ms, which is less than the blink of an eye. The viper fangs with a hollowed structure similar to the insect styloes are capable of withstanding stresses to avoid buckling at the point of striking, which is particularly critical in engaging with a higher striking velocity and large kinetic energy. Even though the study of the structure–mechanical properties relationships is yet rare, it is believed this typical class of sharp and rigid structures can provide us with ideas on the design of advanced materials and structures.

Based on the structures explored for sharpness and rigidity, it is clear that the incorporation of a rigid and dense outer layer with the inner cellular form or hollow structure can realize strong support for the sharp tip and simultaneous lightweight and energy absorption and dissipation for maintaining rigidity during penetration and punctation. The study of natural sharpness and rigidity is yet in an infant stage, but it will surely inspire the design of sharp and rigid structures for engineering applications.

2.3. Natural Tolerance and Resilience

2.3.1. Natural Damage-Tolerant Structures

According to engineering standards, tolerance is known as the accepted variation limits of a material that it is allowed to deviate its shape for proper function and is an indicator of the ability of a structure to sustain defects safely until it becomes unstable to maintain the integrity.[24] In this review, different to the toughness, which we defined as the property of a material to resist nanoscale or microscale cracks, the tolerance is a macroscale capability to sustain the overall structural stability and integrity. Resilience is the capability of a material to recover or self-heal from mechanical impact or damage, which plays an important role in the safe service, especially the long-term service, of the materials. In advanced materials, both the tolerance and resilience properties indicate the safety and endurability of materials in service.

In nature, many animals have armor-featuring scales or skins consisting of small pieces of scaly units to accommodate mechanical impact or damage and prevent the tenuous inner organs from wear and tear. Fish is one of the greatest numbers of aquatic dwellers and has typical armor-like scales covered on the whole body to provide mechanical protection, environmental camouflage, and underwater maneuverability.[25, 69–78] In general, fish possesses four types of scales, namely cosmoid (lungfishes and some fossil fishes), placoid (sharks and rays), ganoid (bichirs, bowfin, paddlefishes,gars, surgeons), and elasmoid scales (can be further divided into cycloid and ctenoid, for most bony fishes).[69] Among them, the elasmoid scales and the ganoid have been well investigated for their excellent hardness and mechanical tolerance found in most common fish and some ancient fish, such as arapaima gar (Figure 7A) (Table 1).[69–71]

Gil-Duran et al. studied the composition, microstructure, and mechanical properties of the cycloid scales of the Megalops Atlanticus, an Atlantic tarpon.[71] The scale is composed of collagen and hydroxyapatite within three well-defined layers from the bottom to the top, and the collagen fibers are arranged in piles with preferred orientation along the longitudinal direction of the fish. In terms of mechanical properties, this fish scale had a Young’s modulus of ≈195–300 MPa, a modulus of toughness of 3.6–4.4 MPa, and tensile strength of 22.6–24.3 MPa. The Young’s modulus of the scales from the head was significantly greater than that of the middle and tail regions, as a result of the orientation variation of the collagen fibers arranged in the piles within the scale.

The effect of the microstructure and orientation of arrangement of the collagen fibers on the biomechanical properties of the coelacanth fish and the common carp scales were recently investigated in terms of fracture toughness, penetration, and shear strain (Figure 7B).[73] In the carp scale, a typical elasmoid scale with an oval shape, the exterior surface consists of an ultrathin discontinuous mineral layer on top of the mineralized woven collagen fibrils and the two underlying collagenous components.[73] The Young’s modulus values ranged from 521.5 ± 125 MPa to 463.5 ± 92.3 MPa for the whole carp scale, which is slightly larger than the 290 ± 0.08 MPa measured as an averaged value for scales from the whole-body length.[73] Particularly, the high mechanical performances resulted from the structural reinforcement of collagen fibers in the carp scale have been detailed investigated, as shown in Figure 7C.[70] At the first stage under tensile loading, cracking and peeling off occurred in the highly mineralized external layer, where the lamellar collagen fibrils with orientations close to the loading axis subjected to tensile strains were reoriented toward the loading direction by interfibrillar shearing, delamination, and relaxation (Figure 7Ba–c). Further increased the loading, the lamellae failed by delamination and rotation and were unable to recover from the tensile and compressive plastic deformation (Figure 7Bd). In general, the collagen fibrils of the carp scale can adapt to applied load by lamellar rotation, fibrillar deformation and sliding, and interlayer separation and thus has the capability to dissipate stored elastic energy more effectively and with better
Figure 7. Mechanical tolerance in biological structures. A) Photographs of a common carp and an alligator gar, and B) fracture models of carp scales observed by scanning electron microscope (SEM), where stretching, delamination, twisting, and buckling during crack formation are indicated. Reproduced with permission.[70] Copyright 2018, Wiley-VCH. C) 3D rendering of bouligand scale structure and the deformation mechanism of carp scale. Reproduced with permission.[73] Copyright 2020, Elsevier Inc. D) Damage tolerance tested surfaces after penetration by a shark tooth. Reproduced with permission.[70] Copyright 2018, Wiley-VCH. E) Fracture resistance of alligator gar scales in wet/dry states and different directions. Reproduced with permission.[74] Copyright 2013, Elsevier Ltd. F) Photograph of a Knobby starfish. G) SEM images on fractured adjacent planes. Inset shows a dislocation. H) Fracture evolution process showing damage bands and densified regions. Reproduced with permission.[80] Copyright 2022, American Association for the Advancement of Science. I) Photograph of a Chiton and optical and SEM images of its teeth array known as radula. Reproduced with permission.[81] Copyright 2014, Wiley-VCH. SEM images of mineralized particles entangled within chitin nanofibers. J) Atomic force microscopy (AFM) phase contrast map on an early-stage teeth development. Reproduced with permission.[82] Copyright 2022, Wiley-VCH.
toughness (Figure 7Be,f). Even though the deformation mechanism is a bit different between the coelacanth scale and the carp scale aroused by the less threading fibrils in the carp scale, the coelacanth scale has the collagen fibrils in the form of bundles embedded into a matrix comprising fibers perpendicular to the layered, double-twisted, Bouligand structure, which provide rigidity to the scales and resistance to deformation.\(^7^3\) As shown in Figure 7D, the coelacanth scale is not sensitive to the precracks during deformation, owing to the existence of collagen fiber bundles. Contributed by the reinforcement effect of the collagen fibrils, damage energy can be dissipated by the bridging, delamination, and relaxation of the fiber bundles, endowing the scale with outstanding damage tolerance.

Bruet et al. studied the structure–mechanical properties of an individual ganoid scale of Polypterus senegalus—one of the earliest fish which retains many features of the dermal armour of ancient palaeoniscoids.\(^7^2\) The scale has a four-layer structure consisting of ganoine, dentine, isopedine, and a bone basal plate from the outer to the inner surface. This organic–inorganic nanocomposite layers deliver appealing mechanical properties and toughness. The residual impressions imaged by the atomic force microscopy (AFM) on these four layers revealed the plastic nature of indentation. The Young’s modulus decreased from 62 to 17 GPa and the hardness decreased from 4.5 to 0.2 nm from the outer ganoine layer, composed of highly mineralized non-collagenous structure to the inner basal osseous layer. This unique gradient layered structure simultaneously maintains the advantageous mechanical properties of hardness and stiffness, of the outer ganoine and the facile energy dissipation by the underlying layers and avoids the disadvantage of radial cracking. Figure 7E shows the crack resistance of ganoine scales of alligator gar under wet and dry conditions. Anisotropy of the collagen fibril orientation, the crack propagation differs.\(^7^4\)\(^-\)\(^7^8\) Therefore, the fish scale provides outstanding damage tolerance via a unique arrangement of collagen fibrils and bundles. It should also be noted that the fish scale works as a whole to provide tolerance to withstand external mechanical damage and impact via the synergic response of the overlapped tough scales to dissipate damage energy.\(^7^9\) The unique arrangement of the scaly units has inspired the design of armors.

Besides fish scales, a very recent study identified the knobby starfish shell to have an enhanced damage tolerance.\(^8^0\) A single crystalline microlattice consisting of calcite with excellent mechanical tolerance is identified in a knobby starfish, Protoreaster nodosus (Figure 7F). The skeleton of the starfish has a diamond periodic structure, which exhibits composite gradients and atomic-level dislocations to convey remarkable energy absorption capability for avoiding structural failure under mechanical damage (Figure 7G,H). The remarkable damage tolerance of the starfish is resulted from the atomic-level dislocations that occurred undergoing local compression, which provides us with another interesting case on building damage-tolerant structures and materials.

Chiton’s radula is another interesting example of damage or wear and tear resistance. This type of animal eats algae by rasping or scraping off the hard rock surfaces using the radulae, which must endure mechanical damage and resist wear and tear during rasping. The radulae are composed of mineralized mesocrystallines and chitin-binding organic matrix, where the minerals are mainly magnetite and iron phosphate (Figure 7I). A typical core–shell architecture was identified in this ultra-hard structure. The hard shell is made of magnetite crystals embedded in nanorods parallel to the long axis, where the nanorods of the chitin organic matrix direct the formation of mineral crystals. The soft core is organic-rich hydrated iron phosphate. Owing to this architecture and chemical composition, high shock and damage resistance are ensured. The hard outer shell has one of the highest reported average moduli thus far, reaching 129.6 ± 6.2 GPa and an average hardness of 10.3 ± 1.5 GPa. The investigation of the biologically controlled phase development of the hard magnetite-containing shell of the chiton teeth revealed that a phase transition of the ultrafine nanoparticles from ferrihydrite crystals to magnetite is facilitated by an organic matrix of chitin fibrils (Figure 7J), which leads to the growth of highly aligned single-crystalline nanorods and contributes to the strength and damage tolerance at mature stages.\(^8^2\)

### 2.3.2. Natural Resilient Structures

Figure 8 presents some typical examples of resilience and adhesion in biological structures.\(^8^4\)\(^-\)\(^9^1\) As mentioned earlier, resilience can be defined as a material’s capacity to reverse deformation or to recover from damage. Resilience associated with hydration has been identified in some plants, such as pinecones. The hydration status is the passive stimulation of such as sclereids and sclerenchyma fibers.\(^8^4\) Figure 8A shows the brown coloration occurring due to dead cells. In the pinecones, the fiber orientation changes with the microfibrillar angles (MFA), which differ from layer to layer. Biologically, pinecones open in dry conditions and close during wet weather to secure
Figure 8. Resilience and adhesion of biological structures to navigate environmental changes. A) Photograph of a pinecone. In dry and hydrated states, the cells and fiber movements depend on the microfibrillar angle (MFA). B) SEM micrographs indicate the upper cellulose fibrils and lower region consist of lignified sclereids. C) Schematic illustrations of a possible mechanism for reversible bending of pinecone scales. Reproduced with permission. Copyright 2021, Elsevier Ltd. D) Nanoscale architecture inside the fibrils and cells of pinecones. Reproduced with permission. Copyright 2021, Springer Nature. E) Photograph of lotus petiole and schematic illustration of the internal architectural structure of a lotus stem. Reproduced with permission. Copyright 2017, Royal Society of Chemistry. F) SEM images of lotus fiber in the stem, cross-section showing the spiral structures in fibers, arrangement of vessels and tracheid, and the helical structure in individual fibers. Reproduced with permission. Copyright 2014, Wiley-VCH. G) Physical properties of lotus fibers. Reproduced with permission. Copyright 2020, Taylor & Francis, Inc. H) Photograph of mussels. Illustrations on the upper right show protein adhesion using the foot, and protein secretion in the microenvironment. Images in the lower right show adhesion and tensile strength distribution in an event of detachment. Reproduced with permission. Copyright 2017, The Company of Biologists. I) Characterization of longitudinal ducts. J) Schematic of plaque formation. Reproduced with permission. Copyright 2021, American Association for the Advancement of Science. K) Photograph of a gecko foot and SEM image shows the setae on the foot pad. Images below show protein structures and the near-edge X-Ray absorption fine structure (NEXAFS) analysis for examining the molecular structure for adhesion explained at the molecular level. Reproduced with permission. Copyright 2022, American Ceramic Society. L) Numerical simulation to calculate the pull-out force of keratin from a dry, hydrophobic surface. M) Modeling of dry gecko keratin for adhesion. Reproduced with permission. Copyright 2022, The Royal Society of Chemistry.
Adhesion is another important mechanical property, which describes the tendency of dissimilar particles or surfaces to cling onto another surface or particles. The adhesion force is usually from intermolecular interactions at the interfaces. Some recent discoveries of some super-adhesion phenomenon, however, indicate that the mechanical interlink of nanofibers could also be one important mechanism. Strong adhesion of bivalves or mussels on the surfaces of rocks and piers has attracted attention to understanding the adhesion mechanism and the development of engineering materials, especially for working under wet conditions. Figure 8H shows the pedestal of a mussel used to attach to a hard surface. The microenvironment of the foot has been examined. Proteins, 3,4-dihydroxyphenylalanine (DOPA), and ligand forming chemicals, also known as metal storage particles (MSP) form adhesive byssus fibers reinforced by protein-metal coordination. The adhesion of mussels depends solely on external excretion facilitated by the foot pad. The plaque secretion, illustrated in Figure 8J, indicates the storage of two distinct metal ions: iron and vanadium. The selective extraction of these ions mixing with adhesive proteins via a microfluidic-like process within interconnected microchannels to create porous and adhesive plaque filaments to stick onto hard surfaces. The superstrong adhesion of the mussels has inspired the design of next-generation metallopolymers and adhesives for engineering applications.

Similarly, gecko adhesion also has been noted for plenty of years and the secrets of climbing on the vertical smooth glass surfaces via van der Walls (vdw) interactions also has inspired the design of super-adhesive materials and interfaces. Further investigation shows that the molecules of keratins within the gecko setae arrays have similar importance to the nanofibrillar geometry of the gecko pads. Figure 8K presents the microstructure of the gecko foot and the micro setae terminated by spatula shafts on the footpad that provide the superstrong adhesive. A molecular structure analysis of the keratin molecules within the spatula tissue was studied by using a NEXAFS technology. It is identified that the keratin proteins have a well-aligned β-strand structure and the fibrils within the setae can be aligned either downward or parallel to the direction of shafts, which can change the status of adhesion. When the protein fibrils are not twisted, strong adhesion occurs, and pull-off happens where the twisting occurs. This adaptability of fibers thus modulates the adhesion for the gecko setae. The molecular simulation further provides insights into the gecko pull-off at a molecular level with the change of the humidity of the environment. It is interesting that larger pull-off forces are needed to detach the wet gecko keratin than the dry state, owing to the interplay between capillary bridges and a mediator effect of water for enhancing the pull-off forces. These studies confirmed that the adhesion and detachment of gecko foot depend on the molecular composition and the angular orientation of keratin fibers, which provide molecular understanding of the super-adhesion of gecko foot structure for designing engineering structures for adhesion and pull-off.

With similar importance to the nano- and/or microstructure for superstrong adhesion, the chemical compositions and the molecules at the contact surface are also critical to forming a strong and tough joint interface and to reaching simultaneous strong adhesion and easy pull-off onto the surfaces. The progress achieved in the molecular level understanding of the superstrong
adhesion provides insights into designing bioinspired adhesive, mechanical robust tapes, smart robotics, etc.

3. Bioinspired Mechanically Robust Advanced Materials

The above-classified examples give us ideas that there are remarkable mechanical properties in nature, which are reached accordingly with structural adaptations from some weak and soft constitutional biomaterials, such as proteins, carbonates, hydroxyapatite, cellulose, lignin, etc. The extraordinary mechanical properties of the natural structures surely provide us with many inspirations for designing advanced materials from inferior raw materials. To mimic the structures and obtain the exceptional properties in man-made materials, it is imperative to understand the microstructure and the structure–property relationships of natural structures. To date, a considerable number of studies on bioinspired mechanically robust materials have been reported by mimicking the structures of flora or fauna from molecular-scale structures to overall macrostructures. In this section, we discuss the interesting mechanical aspects of advanced materials learned from natural structures which are conspicuous in achieving strongness, toughness, rigidity, sharpness, tolerance, and resilience.

Some properties are mutually incompatible in the artificial materials world. For example, it is difficult to simultaneously achieve high strength and high toughness. The strength of hardness represents the resistance of materials to non-recoverable deformation, while toughness means the resistance of materials to fracture and is measured as the energy needed to cause a fracture. The elaboration of resistance to plastic deformations and the associated mechanisms with a trade-off of toughening is a tactic seen in high-strength materials, and the ductile materials with high toughness usually have lower strength. The natural materials with hierarchical architectures, such as seashells and cortical bonds, however, exhibit far more successful toughness while maintaining high strength, relying on both intrinsic and extrinsic toughening mechanisms, which provide approaches to achieving high strength and toughening artificial materials.

The natural successful examples give us inspirations that, if we can realize the mimicking structures at different length scales as the natural structures, we then can fabricate artificial advanced materials with the targeting mechanical performance. To date, plausive bioinspired materials have been innovated by learning the best of nature.

3.1. Bioinspired Tough and Strong Advanced Materials

3.1.1. Nacre Structure Inspired Strong and Tough Advanced Materials

As we mentioned, there are difficulties in designing advanced artificial materials, but it is fortunate that the species in nature have demonstrated both strength and toughness via the rational assembly of soft biopolymers and brittle and weak calcium carbonates. The brick-and-mortar structure with alternative stacking of soft proteins and calcium carbonate in the nacre structure is the key to achieving high fracture toughness with a facture work of 700–1800 J m⁻². High strength has been observed in biological nacres and seashells often being plastic in their inbuilt nature, where the intrinsic deformation was found to be prominent in the nanoscale while the extrinsic deformation occurred at the micrometer scale. Inspired by the strong and tough nacre structure, bioinspired materials composed of hard nano mica layers and soft polyimide (PI) have been fabricated to reach superior mechanical stability for potential aerospace material in low Earth orbit (LEO). In this design, the nano mica works as the brick to significantly enhance the mechanical properties, such as the nanocomposite possessing surface hardness of 0.37 GPa, tensile strength of 125 MPa, and Young’s modulus of 2.2 GPa, together with the enhanced temperature and UV resistance.

Amini et al. recently reported nacre-inspired tough glass with superior mechanical properties, which comprises SiO₂ and modified poly(methyl methacrylate) (PMMA) with similar refractive indexes (Figure 9A). This bioinspired design endows the glass with failure in a gradual fracture mode rather than a catastrophic mode, by which the fracture toughness and work of fracture were enhanced by ≈55% and 30%, respectively. Specifically, the fracture toughness (KIC) of the bioinspired glass was twice the estimated KIC contributed by the SiO₂ tablet alignment. During fracture, the delamination and deformation follow the extrinsic toughening mechanism, where the polymer binds the tablets and bridges them during fracture until complete debonding or formation of microscopic cavities. Figure 9A demonstrates the fracture surfaces of the bioinspired glass and the natural nacre structures, which show very similar fracture modes and crack propagation and deflection paths. It is interesting that the alignment of the SiO₂ tablets can be altered depending on centrifugation force during the fabrication process, which can provide the altered strength and toughness in the bioinspired glass composite to suit different work conditions.

Direct fabrication of artificial nacre structures has also led to the development of a synthetic nacre capable of exhibiting similar structures with high hardness, high strength, and high toughness as natural nacres (Figure 9B). In this process, a mesoscale “assembly-and-mineralization” approach was proposed to mimic both the chemical composition and hierarchical structure of the natural nacre. The nacre-inspired artificial materials are fabricated using calcium carbonate (brushite) micro platelets as the inorganic building blocks and sodium alginate (SA) as the biopolymer matrix for its biocompatibility, flexibility of the polymeric chain, and availability of carboxyl and hydroxyl chains for interfacial interactions. This bioinspired structure showed better mechanical properties than the natural structure where the impact strength was ≈7.1 KJ m⁻², fivefold higher than that of the natural nacre (≈1.4 KJ m⁻² for Cristaria plicata), which means that the artificial nacre is tougher than the natural nacre. The flexural strength of the artificial nacre structure was ≈267 MPa, while ≈172 MPa was the ultimate for the natural nacres. The synthetic nacre structure could be more plastic, the stiffness recorded as ≈18.6 GPa for the nacre-mimicking bulk material, whereas it was ≈48.9 GPa for a natural nacre. The fracture toughness, KIC, of the synthetic nacre was ≈1.9 MPa m²/2, which is very close to the toughness of the well-evolved natural nacre structure, such as ≈2.4 MPa m²/2 for C. plicata.
Figure 9. Nacre or brick-and-motor structure bioinspired materials with toughness and strength. A) Nacre-inspired tough glass comprises SiO₂ and modified Poly(methyl methacrylate) (PMMA) with various orientations of layered structures fabricated via centrifugation at different speeds, and the fracture surface of the bioinspired glass compared with the natural nacre tablets. The plots show the strength and fracture toughness of the glass fabricated at different centrifugation forces. Reproduced with permission. Copyright 2021, AAAS. B) Schematic illustrations of bottom-up fabrication of artificial nacre, and SEM observation of the fracture of the nacre structures. Reproduced with permission. Copyright 2017, Springer Nature. C) MXene/reduced graphene oxide (rGO) nanosheets brick-and-motor structure (MrGO) with 1-aminopyrene (AP)-disuccinimidyl suberate (AD) interlayer molecules for ultrahigh toughness, high tensile strengths, and high electrical conductivity. Molecular dynamic simulations on toughening mechanisms of the MrGO and MrGO-AD sheets. Reproduced with permission under terms of CC BY 4.0. Copyright 2020, The Authors. Published by Springer Nature. D) Dense MXene thin films by using a sequential bridging process (SBM) with the addition of hydrogen and covalent bonding agents with enhanced tensile strength, toughness, and conductivity. Reproduced with permission. Copyright 2021, AAAS.
Membrane technology has been inspired by the brick and motor structure as well. Figure 9C shows the use of MXene/reduced graphene oxide (rGO) nanosheets as a foundation for fabricating super tough and foldable membranes.[109] Based on low porosity and high ordering stacking of MXene and rGO nanosheets into a brick and motor structure and crosslinked by a conjugated molecule (1-aminopyrene-disuccinimidyl suberate, AD), ultrahigh toughness, high tensile strengths, and high electrical conductivity were reached. The addition of AD is critical in reaching high mechanical properties and sustaining structural integrity under deformation, which crosslinks the surface groups in-between the MXene/rGO sheets and enhances the rigidity via π-π bridging. Contrary to MXene/rGO film without AD, the MXene/rGO-AD shows better responsibility under deformation with sliding between MXene layers and crack deflection and bridging while the strength is well secured.

Recently, Wan et al. reported the facilitation of dense MXene thin films by using a sequential bridging process (SBM) with the addition of hydrogen and covalent bonding agents, such as sodium carboxymethyl cellulose (CMC) and boron.[101] Figure 9D presents the fabricated SBM thin film, which exhibited much fewer and thinner voids than the MXene only film, because of the insertion of CMC molecule chains as filler and binder between the MXene platelets can bridge the adjacent platelets more closely. With the introduction of both hydrogen and covalent agents to reach densification and strengthening, dense SBM films with reduced voids and decrease porosity exhibited superior mechanical properties. The SBM films had a tensile strength of 583 ± 16 MPa, a Young’s modulus of 27.8 ± 2.8 GPa, and toughness of 15.9 ± 1.0 MJ m−3, which are ~7-, 5-, and 12-fold increase, respectively, compared to the MXene-only films.

Therefore, the formation of a brick-and-mortar structure can form strong and tough composites starting with the soft materials, which usually have low hardness and low deformation resistance, and the hard inorganic materials with a brittleness or low toughness nature. This important learning provides a principle on how to build strong and tough advanced materials from alternative weak and soft materials and with the modification of interfacial interactions for improving the interlayer synergistic response towards mechanical impact and deformation.

3.1.2. Wood Structure Inspired Strong and Tough Advanced Materials

As we have discussed, in a plant world, the trunks, stems, and some leaves of plants are developed to maintain gradient biological structures to reach mechanical robustness from biological lignin and cellulose fibers that usually have inferior mechanical performances. The natural plant structure can surely provide us with many ideas to design gradient mechanical structures to meet the requirements of both strength and toughness. Inspired by natural wood, Yu et al. fabricated bioinspired polymeric woods by low-temperature curing of the preformed polymeric matrices synthesized via ice-template-induced self-assembly and followed freeze-drying.[102] As shown in Figure 10A, via accurate control of the freezing rate, the artificial polymeric woods made from phenolic resin (artificial CPF wood), melamine resin (artificial CMF wood), and the melamine resin-graphene oxide composite (PF/GO composite wood) presented similar structures with that of natural balsa wood were successfully obtained. Contributed by the anisotropic honeycomb-like microstructures, the bioinspired polymeric woods manifested high compressive strengths (35–45 MPa) and elastic moduli (653–700 MPa) along the longitudinal direction. Furthermore, this type of bioinspired wood also exhibited outstanding acid corrosion resistance without the decrease in mechanical properties and much improved thermal insulation and fire retardancy compared to the natural wood. This work provides a new method to fabricate wood-inspired structural materials with much improved chemical and physical properties by selecting proper candidate raw materials.

The gradient feature of the natural plants described earlier has also inspired the design of bioinspired materials with gradient microstructure and thus extraordinary mechanical performances or responsive behaviors. By adapting the gradient structure and mechanical stiffness of the natural wood, Frey et al. designed smart materials by implementing bioinspired structural, chemical, and mechanical gradients into the delignified wood (Figure 10B).[103] Through tuning the fiber orientation and mimicking the adaptive fiber alignment in trees, the stiffness of the delignified wood reached 35 GPa and the strength was up to 270 MPa, owing to the close contact between the mechanically interlocked fibers and the hydrogen bonding for excellent stress transfer. In another example, Sato et al. proposed a bamboo-inspired optimal design of functionally graded, stiffening hollow cylinders through rational distribution of the fiber reinforcement phases.[104] Quantitatively matching the vascular bundle distribution of wild bamboos, an optimal design principle for fiber-reinforced cylindrical composites to realize high-stiffness and lightweight properties was proposed. Recently, by emulating bamboo’s mechanical efficiency, a tube wall material with tube gradient structures was fabricated by using a freezing casting process, in which the highly aligned, honeycomb-like porosity was generated by the formation of an ice scaffold template. The bamboo-inspired porous material possessed a similar structure to a real bamboo, in which a stiff outer shell supported by a porous, elastic inner layer formed. This unique structure delayed the onset of deformation and failure upon mechanical loadings and exhibited excellent mechanical efficiency.[105]

Besides the directly structural applications of the flora-inspired materials, the unique structure and mechanical properties of plants have also provided inspiration for the design of functional materials for energy devices or electronic devices. Attracted by the excellent mechanical performance of bamboo, Sun et al. designed a bamboo-inspired nanostructure consisting of graphitic carbon nanofiber via electrospinning for the application in flexible, foldable, and twistable energy storage devices.[106] Owing to the excellent mechanical stability under continuous dynamic operations of bending (90°) and twisting (180°) of the bioinspired nanofibers, the flexible all-solid-state supercapacitor with the free-standing bamboo-inspired electrode exhibited 100% capacitance retention after 10 000 charge/discharge cycles. Inspired by the gradient mechanical structures of the biological systems, highly mechanically sensitive materials have been developed for high-performance electronic skin (e-skin)
Via simultaneous controlling of the tactile stress transfer to an active sensing area and the electrical current through the gradient structures, a flexible e-skin sensor based on a stiffness gradient and conductivity gradient multilayer allowed efficient transfer and localization of applied stress to the sensing area, exhibited ultrahigh piezoresistive sensitivity.

3.1.3. Bone Structure Inspired Strong and Tough Advanced Materials

Contributing to the hierarchical structure consisting of protein molecules, fibrils, and fibers, the bone has an intrinsic toughening occurring at the nanoscale ascribed by the tropocollagen.
molecules and collagen fibrils for intermolecular sliding and avoiding the propagation of crack, and an extrinsic toughening to deflect and bridge the cracks. Inspired by the Haversian bone structure, Flavia et al. developed a bone-inspired structure into fiber-reinforced composites to achieve enhanced fracture toughness and balanced stiffness and strength (Figure 11).\textsuperscript{[108,109]} In this design, carbon fibers, glass fibers, and an epoxy matrix were applied to mimic the osteons, cement sheaths, lamellar, and interstitial tissues of the osteonal secondary bone structure. In comparison with the laminate composites and other structural materials, the bone-inspired design presented much enhanced mechanical properties in terms of tensile strength, Young’s Modulus, fracture toughness, and stiffness, with the fracture toughness improving by 86% and 26%, respectively, compared to a previous design and some classic laminated composites.

The unique mechanical properties of bone structures have inspired the development of biomaterials based on polymers and polypeptides. Recently, Xu et al. summarized the bone-inspired materials with typical inorganic–organic hybrid structures for the regeneration of hard tissues, such as collagen and non-collagenous proteins.\textsuperscript{[110]} For example, polyacrylic acid (PAA) with a high molecular weight of 450 kDa was used to mimic the collagen structure and the low molecular weight PAA was applied to mimic the osteocalcin, a protein within the collagen fibril. The PAA-crosslinked collagen-inspired structure, after intrafibrillar mineralization, provided better
biomechanical properties.\cite{111} In another case, peptide-amphiphile molecules were introduced to mimic the natural extracellular matrix via cross-link, self-assembly into a nanofibrous structure, or electropinning.\cite{112-114}

Bone structures have also inspired the development of additive manufacturing of advanced mechanical structures that are light in weight. Audibert et al., for example, extracted the shape, size, and orientation of porosities from the bone structure of either avian species or terrestrial mammalians and applied these for topological optimization and bioinspired structural designs.\cite{115} The bone-inspired steel structures with around 65% density were fabricated by a 3D metal printing method. The terrestrial mammalian inspired beam presented a flexural stiffness and a failure force increase of 17% and 7.5%, respectively, compared with the beam with topological optimization, attributed to the homogenization of the stress field and the reduced pore size. This work demonstrates that bioinspiration is relevant/applicable not only for materials innovations but also for additive manufacturing and paves a new way in designing lightweight but strong and tough mechanical structures.

### 3.2. Bioinspired Sharp and Rigid Advanced Materials

Natural sharp structures such as porcupine quills and stingers of the honeybee have evolved with backward oriented barbs to ensure easiness of penetration. However, the difficulty of retraction after attack without buckling and fracture failure is noteworthy, whereas some biological species, such as mosquito proboscis, have forward barbs only. This is to ensure easy retraction other than easy penetration. Artificial needles and nails with synchronized high sharpness and rigidity for either easy penetration or easy retraction are highly desired in practical applications. Figure 12A shows a honeybee-stinger-inspired microneedle fabricated by 3D printing with barbs for easy skin insertion and difficult removal.\cite{116} Compared with barbless parent needles which needed a penetrating force of 50.8 mN, the bioinspired needles can penetrate a substrate with a lesser force of 41.7 mN. For a displacement of 1.5 mm, the penetration force and work needed for the bioinspired needles with barbs were 124.9 mN and 48 μJ, which are 1.6 and 1.25 times, respectively, lesser than the barbless parent needles (200.7 mN and 60 μJ). The energy distribution of the barbs at penetration was studied via a finite element analysis (FEA), which shows that the lesser the stress distribution at the tip, the easier for penetration (Figure 12B). The barbs of the bioinspired microneedle with additional stress distribution at their tip besides at the needle tip help separate the direct contact between the microneedle and the tissue and thus reduce the friction force during insertion. At retraction, extremely high stress was distributed at the tip areas of the barbs, which reaches the rupture limit of the skin tissue to be interlocked with the barbs from pulled back. The difficult retraction characteristic is requested in the microneedle patches, which need to sustain within the skin tissue for the full release of vaccine or medicine. These bioinspired microneedles with the features of easy penetration and difficult retraction have promising applications in tissue adhesives, transdermal drug delivery, biopotential monitoring, and so on.

As we discussed in the previous section, the porcupine quill has a sharp tip and buckling resistant body, which provide simultaneous low-force penetration for easy insertion and high rigidity to prevent from breaking. The internal architecture of the quill that provides high buckling and fracture resistance has inspired the construction of tough structures (Figure 12C). Inspired by the internal architecture of the porcupine quill, 3D printing manufacturing the bioinspired structures with higher rigidity has been realized, which provides a new idea on advanced additive manufacturing and mechanically rigid advanced materials (Figure 12C).\cite{117} The porcupine quill has been investigated for its sharpness and also the interesting barb structure covered on the quill tip. Currently, it is yet no relevant study on combining the characteristics of lightweight and rigidity together with the sharpness to design light, rigid, and sharp nails and needles, which should be an interesting research topic.

Very recently, a novel microneedle patch with the capability to shrink the wound and remodel the local tissue was designed by learning from the oriented disc-shape lamprey teeth.\cite{118} Figure 12D presents bioinspired microneedle arrays mimicking the lamprey buccal cavity occupied with sharp teeth to achieve the dual functions of penetration and directional traction for rapid wound repair. The bioinspired oriented antibacterial sericin microneedles (OASM) were fabricated from a negative poly(dimethysiloxane) (PDMS) mold. The lengths of these microneedles were \( \approx \)1.5–2.5 mm. The failure force of OASM was around 1.2 N per needle, which is sufficient to penetrate the skin. Compared with regular sericin needles, a higher dragging force was examined for the OASM, which restricts the wound size and accelerates wound healing.

With the increasing focus on public health and people’s well-being, advanced microneedles that combine multifunction of easy penetration, improved skin adhesion, painless and rapid healing insertion, sufficient drug containing and delivering, and high rigidity and toughness for safe operation, more and more reports on bioinspired microneedles appeared by learning from the sharp insect stingers, viper fangs, mosquito proboscis, etc.\cite{119,120} It is clear that the natural sharpness and rigidity provide us with plenty of opportunities for engineering innovations and clinical translation.

### 3.3. Bioinspired Materials with High Tolerance, Resilience, and Adhesion

#### 3.3.1. Bioinspired Resilience

Mechanical tolerance and resilience are associated with the capability of mechanical structures and materials for sustaining the integrity under impact and damage, which are especially important for the service under extreme and harsh environments. Nature has many good examples of tolerance and resilience to provide new ideas for designing advanced materials with high tolerance and resilience.

Resilience has been proven in lotus fibers for the twisting structures with a helicoidal arrangement, which allows elastic recovery after mechanical unloading. The lightweight, flexibility, and resilience add explicit qualities to lotus fibers as a good example to inspire the innovation of artificial materials. Guan et al. fabricated a spiral hydrogel bacterial cellulose (BC) fiber inspired by the structure of lotus fibers (Figure 13A).\cite{121} As shown in the schematic...
Figure 12. Bioinspired advanced materials with excellent sharpness and rigidity. A) SEM images of parent needle, honeybee-stinger bioinspired needle, and barbs on the bioinspired needle. Sequential images of penetration-retraction of bioinspired needle and barbless needle. B) Stress distribution simulated in barbless and bioinspired needle. Calculated depths and performance in penetration and retraction. Reproduced with permission. Copyright 2018, American Chemical Society. C) SEM images indicate the before and after compression of the porcupine quill. Reproduced with permission. Copyrights 2021, Elsevier Ltd. Additive manufacturing of porcupine quill mimicking internal structure. Reproduced with permission. Copyright 2021, Springer. D) Schematics of a lamprey buccal teeth bioinspired microneedle patch. Poly(dimethyl siloxane) (PDMS) mold was used for the preparation of orientated short and long needles. Lengths of antibacterial loaded needle 2.5 mm. Image of mouse skin tested with trypan blue staining after using the micro-needle patch. Reproduced with permission. Copyright 2022, American Chemical Society.
drawing. A 3D bacterial cellulose (BC) hydrogel with nanofiber networks was first fabricated, and then long strips were cut from the hydrogel under an applied tangential force to obtain ultra-strong and ultra-tough BC hydrogel fibers with a bionic spiral structure.

The toughness of the bioinspired BC hydrogel fibers was ≈116.3 MJ m⁻³, which is ninefold higher than the original BC hydrogel fibers. The reason for the increase is the twisting along with enhanced linkage. These lotus fiber bioinspired hydrogel
fibers could be a promising hydrogel fiber for surgical sutures and other biomedical applications based on their high strength, high stretchability, high energy dissipation, high hydrophilicity, porous structure, and excellent biocompatibility. In another work, the bioinspired green composite bundle of twisted lotus fiber coated with poly(vinyl alcohol) (PVA) was fabricated (Figure 13B). In this design, the twisting angle of the lotus fiber bundle and the PVA content were optimized, and a loading of 22.4% PVA and a twisting angle of 20° reached the highest mechanical properties. The optimized bioinspired fibers GCF-II with a diameter of 80 μm reached a tensile strength of 622.5 ± 60.3 MPa, while the tensile strength for the pure PVA was only 76.1 ± 4.2 MPa. Further cross-link and heat-treatment at 60 °C for 2 h improved the interfacial strength between the PVA and the lotus fiber, while the Youngs’ modulus improved to 22.4 ± 2.9 GPa. Considering the low density of lotus fiber, the specific tensile strength of the bioinspired fibers reached 629 kN m⁻¹ kg⁻¹, which is higher than other natural fibers, including cotton, jute, hemp, sisal, and ramie fibers. Even though the resilience of the lotus-fiber-inspired hydrogel or green fibers has not been specifically characterized, it is confirmed that the twisting structure played a key role in the significantly enhanced mechanical properties of the same materials but without the spiral feature.

Bioactuation toward temperature, moisture, and solar light of natural plants is also one typical response for resilience. By learning from the self-regulatory capability of sunflower plants for the day–night rhythm to the solar illumination conditions, a bioinspired soft phototrophic adaptive soft tubular actuator with rapid prompt photo-response toward the unconcentrated light source was fabricated by using MXene-reinforced liquid crystal elastomer (MXene-LCE). As shown in Figure 13C, the phototropic sunflowers biomimicking adaptive photovoltaic system exhibited the maximum light irradiation density, enhanced photovoltaic efficiency of the solar cells can be achieved. It is expected that the bioinspired actuators with external stimulus responsibilities have promising application potential in developing self-regulated solar cells, intelligent robotics, environmental sensors, responsive energy and environmental devices, etc.

The resilience of natural structures and organs is usually reflected in the way of self-healing of the wonders or fractures. Inspired by the wound healing capability of biological systems, pH-sensitive self-healing hydrogel has been fabricated. Figure 13D demonstrates the bonding and physical state deformation of the acylhydrazone-based polymer hydrogels with pre-installed urease and urea. After a fracture, with the treatment of acidic urea buffer liquid, the dynamic acylhydrazone bonds within the urease-containing hydrogel can be activated to heal the fracture surfaces. After healing, a slow generation of ammonia occurs to decrease the local pH value and restabilize the self-healing hydrogel. From this case, it is indicated that hydrogels can process the self-healing ability with the addition of a chemical stimulus.

In another case, also inspired by the self-healing property of biological systems, Fu et al. developed a self-healing structural color hydrogels by incorporating glucose oxidase (GOX) and catalase (CAT) filled glutaraldehyde cross-linked bovine serum albumin (BSA) hydrogel into a methacrylate gelatin (GelMA) inverse opal scaffold (Figure 13E). In this composite, the opal photonic nanostructure provides structural stability to the hybrid hydrogel, while the protein hydrogel filler aids the self-healing capability via the reversible covalent attachment between the glutaraldehyde and the lysine residues of proteins. Mechanical properties were measured on both the intact composites and the self-healed composites from cut pieces. It shows that the self-healed composites exhibited almost similar stress–strain deformation characteristics to the intact composites. This unique hybrid hydrogel structure exhibited excellent elasticity, high recovering ability, and attractive reversibility. The self-healing properties of the bioinspired materials will provide promising potential in many engineering applications such as constructions, buildings, and extremely environmental devices with much prolonged lifetime and enhanced structural stability and safety.

3.3.2. Bioinspired Damage Tolerance

Fish scales have mechanical properties that protect the inner soft organs and prevent hunters via their gradient structures and unique stacking sequence, which have inspired the design of ancient armors and modern robust coatings. By learning from the tunable responses towards maneuverability, camouflage, and mechanical damage by changing the orientation of fish scales, Sun et al. developed fish-scale inspired multifunctional ZnO coatings, which can realize different mechanical, wetting, and optical responses on one material but with different orientations of the scale-like nanostructures (Figure 14A). It is interesting that, even though all the coatings were made from ZnO, the coatings with flatfish scale-like nanostructures at a (002) preferred orientation were much softer (0.7 GPa for bioinspired coatings vs 4.9 GPa for referential ZnO nanomaterials) but more damage tolerant (energy dissipation of 163.86 pJ), while the bioinspired coatings with highly titled nanostructures at a (101) preferred orientation presented high hardness (3.55 GPa) but less damage tolerant (energy dissipation of 22.5 pJ). From this case, we can have a clear view that damage tolerance can be achieved from the arrangement of brittle subunits inspired from the fish scales.

Inspired by the function of the fish scale in protecting the inner soft organs, Funk et al. fabricated a synthetic fish skin for the protection of soft materials (Figure 14B). The bioinspired fish scale materials were made from a low-modulus elastic mesh or “dermis” layer that holds relatively rigid plastic scales to mimic the leptoid-like scales in that of Mullus surmuletus or the striped red mullet. The mechanical tolerance of the bioinspired materials was examined during in-plane deformation, flexure, and indentation. Upon mechanical damage, the overlapping of scales transferred the applied load from a single scale to the adjacent scales, which disperses the localized applied force over a larger area for a much less stress concentration and avoids the localized damage to the underlying soft materials. The results demonstrated that the synthetic fish scale outperformed the biological fish scale under deformation for providing mechanical protection to the soft materials underneath the bioinspired coatings.

In a similar way, as inspired by the flexible armor of elasmoid fish, bioinspired armor consisting of stiff plates and a soft matrix was designed to provide protection against penetration events
Indentation and three-point bending tests were performed on the bioinspired armor materials. The indentation tests demonstrated that low inclination angles and high-volume friction of the stiff plates offered better penetration resistance and higher penetration stiffness, and the bending tests exhibited that the high inclination angle and the high-volume friction of the stiff plates compromised the flexibility. Therefore, via careful selection of the microstructural parameters of the bioinspired armor, optimized designs for reaching grand protection against penetration while preserving flexibility can be realized.

The teleost fish scales consisting of a hard bony outer layer and a soft inner cross-ply of collagen fibrils have been reported to be capable of dispersing external loadings but providing superior flexibility. By learning from this, a bioinspired structure composed of hard and stiff SiC ceramic as the outer layer and soft aluminum as the inner layer was constructed by Liu et al. (Figure 14D). Through this bioinspired design, the area density of the bioinspired armor was reduced by 12.5% while the same ballistic performance maintained the same at an impact velocity of 878 m s\(^{-1}\) compared with pure SiC scales. The energy dissipation mechanism during the ballistic tests was ascribed to the brittle failure of the brittle SiC scales and the plastic deformation of the inner Al layer, and the Kevlar fabrics connected to the scales contributed to maintaining the structural integrity and the redistribution of the impact energy.

Therefore, based on the excellent examples of fish-scale-inspired structures and coatings, it is concluded that the fish scale has inspired the design of the artificial materials to achieve lightweight, flexibility, and superior damage tolerance. By learning from the fish scales, the enhanced strength and the damage tolerance can be reached. Especially, the surface and bulk properties of the fish-scale bioinspired materials can be tuned by adjusting the organization and orientation of the constituting nanostructures, which provides an extra option in advanced materials design. Further investigations on the selection of more versatile materials to suit different work conditions and the optimization of stacking models as well as the discovery of micro-scale structure-property relationships are needed for this promising class of bioinspired materials.

### 3.3.3. Bioinspired Superstrong Adhesion

Adhesion has been widely investigated in engineering applications for joining two components together via surface interaction, and nature has also provided many good examples of how to achieve superstrong adhesion. As we have discussed, mussels can attach to smooth rocks super-strongly via the metal ion coordinated proteins formed on the footpad. The secreted adhesive proteins have been examined are rich in catecholic amino acid 3,4-dihydroxyphenylalanine (DOPA) and positively charged lysine. This mechanism provides a clear-cut scenario for generating superstrong adhesives for working under both wet and dry conditions. Figure 15A illustrates three pressure-sensitive adhesives (PSAs) inspired by mussel with different
catechols and cationic amines configurations, namely, adjacent catechols and cationic amines (PSA-Lys-DA), catechols and cationic amines distributed randomly (PSA-DAc-ABA), and cationic amines without catechol (PSA-Lys-PEA). The adhesion properties of the bioinspired adhesives were evaluated by a surface force apparatus. It shows that the addition of catechols enhanced energy dissipation and adhesion of PSA. Due to hydrophobic interactions between molecules, the adhesion forces in water were much larger than those in an ethanol/water solution. This study provides insights into the influence of molecular structure and configuration of mussel-inspired adhesives on the adhesion under different conditions.

Different to the superstrong adhesion provided by strong molecular interaction in mussels, gecko feet display strong adhesion towards smooth surfaces via vdw interactions. In previous studies, it has been reported that the adhesion and friction forces originating from the vdw forces between the submicron-sized spatulae and the substrate and the fast switch ability are from the natural digital hyperextension (DH) of gecko toes. The understanding of the adhesion and deadhesion of the gecko foot has been well explored for bioinspired strong adhesion and fast adhesion-detachment switch ability. By learning the gecko feet, a shape memory polymer (SMP)-based switchable dry adhesive (SSA) was developed. The highest adhesion reached 332.8 kPa but it is easy for detachment at 3.73 kPa. Due to the use of SMPs, the gecko feet-inspired adhesive can be easily recovered into its initial shape. Figure 15B presents the structure of the gecko foot and the bioinspired dry adhesive. Via the bioinspired design, a switchable glass transfer system was designed by using thermostet amorphous epoxy-based SMP.

Another gecko feet-inspired reversible dry adhesive made from polydimethylsiloxane (PDMS) micropillar arrays and MoO$_{3-x}$ quantum dots (MoO$_{3-x}$ QDs) was recently reported (Figure 15C). The temperature-induced reversibility is resulted from the thermal response of MoO$_{3-x}$ QDs to the external near-infrared laser irradiation and then the thermal expansion occurs of the PDMS micropillar structure. At a normal state, the PDMS micropillars provide strong adhesion to the gecko foot. For detachment, photo-induced thermal expansion makes the adhesive patch curl up to decrease the contact with the surface and thus weak adhesion force. In another instance, a gecko foot-inspired slanted shape memory microcone array (SSMMA) with tunable adhesion forces was developed (Figure 15D). The adhesion/deadhesion reversibility of this bioinspired adhesive is achieved by altering the orientation and the contact areas of the microarrays as the responses to the changes in temperature and pressure. Under applied pressure, the SSMMA presented exclusive adhesion properties as a result of increased contact area between the collapsed microcones. Being reheated, the SSMMA changed back to the initial point contact state with weak adhesion. Interestingly, the adhesion force of the SSMMA can also be tuned by changing the spacing between the microcones.

The reversible adhesion carries both resilience and adhesion adapted from nature to create responsive smart materials. The superstrong adhesive can be resulted from both chemical bonding and vdw interaction and can provide strong adhesion in...
either wet or dry conditions. The insights into the superstrong adhesion and the rapid switchable adhesion/deadhesion will accelerate the development of smart materials with promising application potentials in construction, materials joining, transport and transfer of smooth structures, soft robotics, etc.

4. Bioinspired Mechanically Robust Materials in Sustainable Applications

In addition to the focus on the mechanical properties of flora and fauna bioinspired materials for engineering applications, the natural robust structures have also inspired the development of many functional materials in emerging sustainable applications by taking advantage of both the robust mechanical properties and the functional properties.[134,135] It is expected that the application of bioinspired materials with strong mechanical properties in sustainable energy and environmental technologies will not only expand the range of application of bioinspired materials, but also provide new opportunities for enhancing the performance of energy and environmental devices.

Osmotic pressure and capillary force involved in transpiration are very typical mechanical phenomena that play critical roles in natural plants. By using the capillary force mechanism, Park et al. developed a leaf-inspired energy-harvesting foam (LIEHF) for effective solar evaporation-based energy harvesting.[136] In this design, polydimethylsiloxane (PDMS) with macro- and microporous structures together with conductive carbon materials were applied to mimic the cellulose fiber structures in the leaves. The LIEHF presented improved solar evaporation efficiency for streaming potential/current to generate hydro voltaic electricity. It was recorded that a size of 15 × 15 × 10 mm$^3$ of LIEHF generated an open-circuit voltage, a short-circuit current, and a power density of 254 mV, 42 μA, and 1 μW cm$^{-2}$, respectively, under 1 Sun irradiation. Similarly, applying the same principle of capillary action in transpiration, nanogenerators were developed by using natural wood.[137,138] Benefiting from the solar evaporation and the capillary pressure of the microporous natural wood structure, streaming potential/current was generated for the pressure gradient formed inside the microchannels of hydrolyzing cellulose, hemicellulose, and lignin.

2D/2D-layered structures by learning from the natural structures, such as nacres and bonds, have demonstrated robust mechanical properties. Besides the achieved strongness and toughness, ultrafast electrolyte or mass transport has been identified within the 2D-based structures.[139,140] For example, Mei et al. observed the ultrafast nutrients transport via the layered membrane of bamboo to support the Guinness Record of the fastest growth rate (≈40 mm h$^{-1}$).[141] This study revealed that the multilevel distribution of interlayer spaces of the 2D stacked bamboo membrane structure provides both nanofluidic ultrafast transport for water molecules and ions within closely stacked layers and facile bulk water wetting and permeation through the loosely stacked 2D layers. The bamboo-membrane bioinspired artificial membrane with multilevel interlayer space distributions was fabricated from graphene oxide, carbon nanotubes, and 2D CO$_2$O$_4$ nanosheets. As expected, the bioinspired membrane exhibited excellent wettability and permeability as well as ultrafast ion transport properties. When used as the anode for Li-ion batteries (LIBs), also contributed by the excellent mechanical properties to sustain the integrity, the bioinspired membrane electrode presented excellent initial and cycling capacity. Particularly, the ultrafast ion transport of the membrane allowed high-rate charging at a rate up to 2 A g$^{-1}$ and the close pack at both nano- and microscale endorsed the electrode a superhigh volumetric capacity above 1400 mAh cm$^{-3}$, which outperforms many reported promising electrodes for LIBs.

The bioinspired materials have also been applied widely in sustainable environmental technologies, such as the fish-gill-inspired one-step water-oil collection and separations.[142] In plants, the gradients of osmotic pressure, transpiration, and guttation in natural leaves have inspired the high-efficiency water purification and production, which is also a good example of applying the mechanical aspect of the natural structure for sustainable environmental applications. A plant-leaf-inspired sunlight-driven water purifier was designed, which consisted of poly(N-isopropylacrylamide) (PNIPAm) chains coated melamine skeleton with a PNIPAm-modified graphene (PG) coated membrane at the outside.[143] Being immersed in polluted water, the water purifier absorbed a large amount of clean water, then under solar irradiation, the PG membrane adsorbed sunlight and converted it into heat to produce water via transpiration and guttation at a rate of 4.2 kg m$^{-2}$ h$^{-1}$ and anionic rejection of >99% under 1 Sun. The leaf structure can also inspire the development of oil-water separation membranes. A taro leaf-skin-inspired oil purification nanonet was fabricated for oil-water emulsion separation.[144] The micro/nano hierarchical leaf surface led to a bioinspired membrane with good channel connections, and multiscale roughness, together with the features of robust underwater oleophilic, low water adhesion, and high-water intrusion pressure for separating water-in-oil emulsions under ultralow driving pressure. This technology is very promising in developing next-generation oil spill cleanup and recovery.

Besides the above-listed examples, there are too many excellent reports that appeared recently on the sustainable application of bioinspired mechanically robust mechanical properties, as a few summarized in Figure 16.[145–151] These examples demonstrate how bioinspiration can expand our knowledge in designing advanced functional materials for applications in sustainable energy and environmental technologies by learning from the unique wettability, transport, and transpiration features of the natural structures, while employing their excellent mechanical performance or unique mechanical phenomena proactively or passively. Even though the raw materials are still the conventionally available common materials, the fabricated bioinspired materials exhibit much improved structural or functional properties through the manipulation of surface, interface, or channel structures or the modification via proper functional groups, and thus deliver significantly enhanced performances of the sustainable energy or environmental devices. It is sure that bioinspiration not only offers us many new opportunities in fabricating novel structures or advanced materials, but also paves a new way in designing novel configurations of devices by learning the specific functionality of biological fauna and flora.
5. Summary and Outlook

The natural structures have achieved robust mechanical properties via the multilevel organization of weak building blocks, which provide plenty of inspiration in designing artificial materials and mechanical structures. Especially, some natural structures, such as nacre structures, dactyl club structures, and bone structures, are amazing examples to reach extraordinary strength and toughness by interlinking the weak and brittle bioinorganic components with the soft and elastic biopolymers into integrated bulk structures. Some insects and animals have evolved with sharp and rigid organs, such as stings, teeth, quills, etc., which provide easy penetration into tissues of prey and predators at very low forces. And some species have been equipped with tolerant and resilient armors and skins, which are also realized by the ingenious organization of common and weak biopolymers, biochitins, biocarbonate components into hierarchically ordered structures, etc. to be capable of absorbing significant mechanical energy and recovering from damage and deformations. From these representative examples, we can conclude that the precise architecture of building blocks of soft polymers and brittle inorganic materials can achieve lightweight materials and composites with high hardness, high strength, high toughness, and promising self-healing properties. In other words, if we can have good learning from these fantastic natural examples, we can enable the design of lightweight and mechanically strong, and resilient structural materials in an energy and material saving way from the existing weak and brittle materials. Therefore,

Figure 16. Bioinspired advanced materials with robust mechanical properties in sustainable applications. i) Reinforcement: Mechanically robust lattices inspired by deep-sea sponge skeletons with high buckling resistance. Reproduced with permission.[145] Copyright 2021, Springer Nature. Dragonfly wing patagium-inspired electromagnetic interference (EMI) shielding materials made from MXene framework with exceptional thermal stability, fast NIR-responsive healing performance, and excellent EMI shielding capability. Reproduced with permission.[146] Copyright 2021, Elsevier. Cellulose straws developed via taking advantage of bonding between three major constituents; better reinforcement is executed. Reproduced with permission.[56] Copyright 2021, Wiley-VCH. ii) Environment: Multifunctional reconfigurable topography/ferrofluid-containing liquid-infused porous surfaces (FLIPS) to remove algal biofilms. Reproduced with permission.[147] Copyright 2018, Springer Nature. A strong, biodegradable, and recyclable bioplastic made from natural wood by extracting and generating in situ lignin. Reproduced with permission.[148] Copyright 2021, Springer Nature. iii) Water purification: Taro leaf surface structure bioinspired nanonet with nanofiber membrane for oil purification. Reproduced with permission.[149] Copyright 2019, Royal Society of Chemistry. Nasal cavity lining inspired membrane surface for oil–water separation. Reproduced with permission.[148] Copyright 2022, Wiley-VCH. iv) Energy: leaf-inspired energy-harvesting foam for effective solar evaporation-based energy harvesting. Reproduced with permission.[146] Copyright 2021, American Chemical Society. Natural wood inspired electricity harvesting from water evaporation. Reproduced with permission.[137] Copyright 2020, American Chemical Society. Bamboo-membrane-inspired ultrafast ion transportation for superior energy storage. Reproduced with permission.[141] Copyright 2021, Wiley-VCH. v) Water harvesting: Janus surface structure of red cloverleaf with wettability and water harvesting. Reproduced with permission.[150] Copyright 2022, American Chemical Society. Bioinspired hierarchical hydrophilic/hydrophobic/bumpy Janus membrane for highly efficient fog collection. Reproduced with permission.[151] Copyright 2021, Elsevier.
biomimicry and bioinspiration would be a promising way to bridge the gap between our demands on the extraordinary properties and the limiting available materials. By employing this concept, even though plenty of grand challenges exist in bioinspiration and biomimicry in terms of material synthesis and structural design, bioinspired materials possess superior mechanical, physical, and chemical properties to the constituent materials have been confirmed as a highly feasible approach to material innovations.

While materials innovation via bioinspiration by learning the best from nature is a promising approach toward exceptional outstanding, many challenges exist in this field. 1) Natural structures and systems are too complex to be well understood. Regardless of how diverse nature is, the discovery of even defined natural structures and functionalities needs the combined expertise from different disciplines of biology, chemistry, physics, materials, and engineering, and requests close collaborations among the researchers with the distinct backgrounds but well-coordinated research skills. Given that bioinspiration is a multidisciplinary study, the research in this field will result in significant knowledge advances. 2) Exploring the underlying principles and establishing the structure–property relationships of biological structures still need to be addressed in a comprehensive way. Particularly, the understanding on the structures from nano- to micro- and macro-scale is the fundamental to bioinspiration, which needs the employment of advanced analysis facilities and equipment, such as advanced microscopes, advanced spectrometers, molecular level structural analysis, etc. 3) The difficulty in examining the microstructure at the fresh state of specimens is another challenge in analysis. Some natural structures exhibit totally different properties in the fresh state to those in the preserved or treated state, while only the latter is suitable for structural and property characterizations. Fortunately, the technological advances, such as the development of cryo-electron microscopy, allow the characterization of the structures of enzymes and proteins. 4) The specific structural or functional advantages of the natural species are generally a result of synergistic combinations of the multiscale ordering of structures, which significantly brings challenges and difficulties in structural mimicking or inspiring, particularly in scalable preparation at a low cost. The manufacturing capabilities of manipulating atomic-level structures, nanoclusters, micro-aggregations, and nanostructures in various dimensionalities have been sufficient for the fabrication of bioinspired materials at low-level ordering. The integration of multiscale ordered and intercrossed structures, however, is yet hindered by both synthetic methods and facilities. 5) The environmental durability and stability of the biological structures are usually not satisfactory in some harsh/extreme environments or in long-term service. These issues could be partly solved with the choice of mechanically or chemically robust artificial materials, but the environmental sensitivity associated with the structural delicacy of the bioinspired materials should be specifically considered during materials design. 6) Structure specificity will be another challenge for the use of bioinspired materials in various scenarios. Modifications and improvements on the bioinspired structures will be needed in the applications in practical scenarios. Nevertheless, bioinspiration provides solutions to design mechanically robust artificial materials from weak starting materials and even create more sophisticated and superior structures and materials. Bioinspired materials offer a pathway to meet the increasing demand for smart and robust materials brought by the rapid growth of the artificial intelligence world but the limited known elements and materials. Using advanced processing techniques, such as additive manufacturing, more fine and delicate bioinspired structures will be developed as smart and intelligent materials for emerging functional and engineering applications.

Acknowledgements

This work was supported by the Australian Research Council through an ARC Discovery Project (DP200103568) and two ARC Future Fellowship projects (FT180100387 and FT160100281). B.W. is supported by a Faculty of Science Scholarship of Queensland University of Technology (QUT).

Open access publishing facilitated by Queensland University of Technology, as part of the Wiley - Queensland University of Technology agreement via the Council of Australian University Librarians.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

bioinspired materials, mechanical properties, sharpness and rigidity, tolerance and resilience, toughness and strength

Received: December 26, 2021
Revised: April 23, 2022
Published online: June 1, 2022

[1] L. Jiang, L. Feng, in Bioinspired Intelligent Nanostructured Interfacial Materials, World Scientific, Singapore 2010.
[2] G. F. Swiegers, Bioinspiration and Biomimicry in Chemistry: Reverse-Engineering Nature, Wiley, Hoboken, NJ 2012.
[3] K. Autumn, Y. A. Liang, S. T. Hsieh, W. Zesch, W. P. Chan, T. W. Kenny, R. Fearing, R. J. Full, Nature 2000, 405, 681.
[4] F. Vollrath, D. P. Knight, Nature 2001, 410, 541.
[5] H. Chen, P. Zhang, L. Zhang, H. Liu, Y. Jiang, D. Zhang, Z. Han, L. Jiang, Nature 2016, 532, 85.
[6] Z. Sun, T. Liao, W. Li, Y. Dou, K. Liu, L. Jiang, S.-W. Kim, J. Ho Kim, S. Xue Dou, NPG Asia Mater. 2015, 7, e232.
[7] Z. Tang, N. A. Kotov, S. Magonov, B. Ozturk, Nat. Mater. 2003, 2, 413.
[8] Z. Sun, T. Liao, L. Sheng, J. H. Kim, S. X. Dou, J. Bell, Mater. Today Chem. 2016, 1–2, 84.
[9] Z. Sun, T. Liao, K. Liu, L. Jiang, H. Kim, S. X. Dou, Small 2014, 10, 3001.
[10] A. Lafuma, D. Quéré, Nat. Mater. 2003, 2, 457.
[11] A. K. Geim, S. V. Dubonos, I. V. Grigorieva, K. S. Novoselov, A. A. Zhukov, S. Y. Shapoval, Nat. Mater. 2003, 2, 461.
[12] Y. Zhang, J. Mei, C. Yan, T. Liao, J. Bell, Z. Sun, Adv. Mater. 2020, 32, 1902060.
[13] Z. Sun, T. Liao, W. Li, Y. Qiao, K. Ostrrikov, Adv. Funct. Mater. 2019, 29, 1901460.
[14] Z. Sun, K. Ostrrikov, Sustain. Mater. Technol. 2020, 25, e00203.
[15] J. Mei, T. Liao, H. Peng, Z. Sun, Small Methods 2022, 6, 2101076.
[141] J. Mei, X. Peng, Q. Zhang, X. Zhang, T. Liao, V. Mitic, Z. Sun, *Adv. Funct. Mater.* 2021, 31, 2100299.

[142] Y. Dou, D. Tian, Z. Sun, Q. Liu, N. Zhang, J. H. Kim, L. Jiang, S. X. Dou, *ACS Nano*, 2017, 11, 2477.

[143] H. Geng, Q. Xu, M. Wu, H. Ma, P. Zhang, T. Gao, L. Qu, T. Ma, C. Li, *Nat. Commun.* 2019, 10, 1512.

[144] J. Zhang, J. Ge, Y. Si, F. Zhang, J. Yu, L. Liu, B. Ding, *Nanoscale Horiz.* 2019, 4, 1174.

[145] M. C. Fernandes, J. Aizenberg, J. C. Weaver, K. Bertoldi, *Nat. Mater.* 2021, 20, 237.

[146] J. Xu, T. Liu, Y. Zhang, Y. Zhang, K. Wu, C. Lei, Q. Fu, J. Fu, *Matter* 2021, 4, 2474.

[147] W. Wang, J. V. I. Timonen, A. Carlson, D.-M. Drotlef, C. T. Zhang, S. Kolle, A. Grinthal, T.-S. Wong, B. Hatton, S. H. Kang, S. Kennedy, J. Chi, R. T. Blough, M. Sitti, L. Mahadevan, J. Aizenberg, *Nature* 2018, 559, 77.

[148] Q. Xia, C. Chen, Y. Yao, J. Li, S. He, Y. Zhou, T. Li, X. Pan, Y. Yao, L. Hu, *Nat. Sustain.* 2021, 4, 627.

[149] B. Wang, H. Dai, C. Zhang, Z. Dong, K. Li, L. Jiang, *Adv. Mater. Interfaces* 2022, 9, 2102311.

[150] C. Mohd, K. Majid, S. Lone, *ACS Appl. Mater. Interfaces* 2022, 14, 4690.

[151] Y. Su, L. Chen, Y. Jiao, J. Zhang, C. Li, Y. Zhang, Y. Zhang, *ACS Appl. Mater. Interfaces* 2021, 13, 26542.

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