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Biological and bioinspired materials: Structure leading to functional and mechanical performance

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ABSTRACT

Nature has achieved materials with properties and mechanisms that go far beyond the current know-how of the engineering-materials industry. The remarkable efficiency of biological materials, such as their exceptional properties that rely on weak constituents, high performance per unit mass, and diverse functionalities in addition to mechanical properties, has been mostly attributed to their hierarchical structure. Key strategies for bioinspired materials include formulating the fundamental understanding of biological materials that act as inspiration, correlating this fundamental understanding to engineering needs/problems, and fabricating hierarchically structured materials with enhanced properties accordingly. The vast, existing literature on biological and bioinspired materials can be discussed in terms of functional and mechanical aspects. Through essential representative properties and materials, the development of bioinspired materials utilizes the design strategies from biological systems to innovatively augment material performance for various practical applications, such as marine, aerospace, medical, and civil engineering. Despite the current challenges, bioinspired materials have become an important part in promoting innovations and breakthroughs in the modern materials industry.

1. Introduction

Biological materials are ingeniously designed and optimized tools that are employed by nature for organisms to survive and thrive within challenging environments [1–4]. They represent the elegant strategies that fulfill a variety of not only mechanical but also functional needs [2,5,6], as they are generally simple in composition but efficient in performance [7–9]. This is distinct from most engineering materials that usually depend on complex chemicals or expensive manufacturing, and therefore often confront a tradeoff between properties (e.g., increasing weight to increase strength). Thus, biological materials have been an endless source of inspiration for developing novel materials and structures in recent decades. To actuate this inspiration, the first fundamental step requires revealing structure-property mechanisms and formulating systematic theories, which is known as Biological Materials Science [4,10–12]. This paves the road for the next exciting step of creating new advanced materials by providing essential insights with heretofore unexploited strategies from natural designs.

Along with this research rapidly developing to be at the frontier is the ever-expanding understanding and knowledge of biological materials themselves. By utilizing exquisite structures instead of chemical complexity, biological materials surpass their synthetic counterparts in many properties and functions. The key to efficiently secure these outstanding properties lies in their hierarchical structure [1–3,8,13]. For mechanical performance, this significantly amplifies the properties of the weak constituents, e.g., the shell nacre has high Young's modulus (70–80 GPa), high tensile strength (70–100 MPa) and high fracture toughness (4–10 MPa $m^{1/2}$) [14–16], although it is composed mostly of brittle minerals (at least 95% by volume [17]) that show a work of fracture that is about 3000 times less than that of the shell [16,18]. Meanwhile, hierarchical structures enable biological materials to achieve substantially higher performance per unit mass, e.g., the spider silk has a tensile strength of 1.1 GPa, which is comparable to that of high-strength steel (1.5 GPa) [19]; but considering the density (1.3 g/cm^3 versus 7.8 g/cm^3 [20,21]), the spider silk is more than four times stronger per unit mass. These have led to an increasing number of bioinspired high-performance structural materials, e.g., nacre-inspired strong and tough materials [22] and crustacean-inspired fracture-resistant composites [23].

In addition to their mechanical performance, biological structures also generate a diversity of interesting functions. For examples, lotus leaves have special surface topographies that allow self-cleaning [24],

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gecko feet employ a hierarchical structure that enables them to scale walls through dry adhesion [25], the beautiful colors of butterflies are realized by their microstructure interacting with light [26], and the fibrous structure of many plants leads them to self-deform with changes in humidity [27]. These intriguing functions obtained through the structures of relevant biological materials are reliable, durable, and nontoxic as additional advantages, and thus have been inspiring to functional materials for a variety of practical applications, e.g., high-performance bioinspired anticorrosion coatings [28], gecko-inspired high adhesion pads [29], nature-inspired reversible underwater adhesives [30], and bioinspired self-shaping composites [31].

In an aim to highlight the rapid, exciting development of this field in a way that is distinct from existing reviews (in which biological and bioinspired designs focusing on certain properties are usually discussed separately), this work addresses bioinspired materials from a vast number of fascinating biological materials in terms of functional and structural categories. Within each category, representative types of functions (superwettability, bioactivity, stimuli-responsiveness) and mechanical properties (light-weight and high-strength, light-weight and high-toughness) are detailed through paradigmatic biological and bioinspired materials. We illustrate the fundamentals for the specific property, then we discuss the insights in structure-property mechanisms from biological materials and their corresponding bioinspired materials that show exceptional functions/properties for relevant applications. We also provide our perspectives on the challenge and prospect of biological and bioinspired designs to further promote the development of advanced functional and structural materials.

2. Key strategies for bioinspired materials from the biological systems

Despite the constraints of weak constituents and mild synthesis conditions, biological materials show exceptional mechanical and functional properties that are coincidently important to many of the various engineering industries in human society. The core of this, simple in composition but efficient in performance, lies in their hierarchical structure, i.e., how the structural elements or building blocks are arranged and organized at multiple length scales. With additional advantages of high durability, reliability, and nontoxicity, these materials present resourceful inspirations for designing modern advanced materials. Indeed, looking into naturally refined biological materials for innovation to develop advanced materials has been an exciting area, especially in recent decades where a rapid research progress into bioinspired materials has taken place.

Generally, the development of bioinspired materials involves activities revealing their design principles from nature, which form the basis of novel structured materials that are fabricated to address certain engineering problems. This starts with discovering the unique, interesting phenomena of biological materials/systems and, through scientific analysis, the fundamental mechanisms underlying these phenomena are systemized/theorized, correlating to the needs of relevant engineering applications. The next step employs these natural principles to the design and fabrication of materials that show targeted mechanical and/or functional properties with enhanced performance (shown in Fig. 1). There are indeed challenges in this endeavor, such as the requirement of highly sophisticated fabrication instruments that are capable of producing these structures and the transfer of small-scale laboratory synthesis to mass production, but biological and bioinspired materials have provided numerous valuable insights in boosting innovation in a variety of areas including marine, aerospace, medical, transportation, and even housing, which are illustrated in this work. As an added benefit, this process allows us to reflect back on our understanding of biology and promote a more harmonious relationship between human society and the natural world.

3. Development of bioinspired materials

Here we discuss biological and bioinspired materials by focusing on two categories: functional and structural, both of which depend on their structures. Within each category, we will focus on particularly impactful and recent examples of bioinspired designs, and therefore provide a compact overview of the grand panorama of this field. Bioinspired functional materials will focus on designs that provide superwettability, bioactivity, and stimuli-responsiveness, while bioinspired structural materials will include designs that provide high strength with low weight and high toughness with low weight.

3.1. Bioinspired functional materials

3.1.1. Superwettability

Surfaces and structures with superwettability are found throughout nature, with examples including rose petals [32,33], lotus leaves [34,35], shark skin [36,37], exoskeletons of desert beetles [38,39] and cactus spines [40,41]. These structures employ superwettability to fulfill essential functions including providing an external barrier and directional liquid transport, to name a few. In terms of man-made materials, research on functional surfaces with specific wettability has made amazing progress over the last few decades, since these surfaces are also of important use in practical applications. From the fundamental viewpoint, superwettability of a surface can be achieved by the physical structure or the chemical constituents of the surface; nature generally adopts the former against the latter, due to the limited constituents available to nature materials. This provides a rich source of inspiration to develop bioinspired, textured structures with specific wettabilities.

One extensively studied type of superwettability is superhydrophobicity, which, in nature, utilizes surface structures to trap air pockets under liquid droplets so as to repel the droplets [42]. These surfaces are found in many plant leaves, where various nano/micro topographies are present. For example, the leaves of the Salvinia molesta plant behave as a superhydrophobic surface, even though the components on the leaf surface are hydrophilic (which is known as the “Salvinia effect”). This is due to the long-term air retention and effective stabilization of the air-water interface caused by the hierarchical structures on the leaf surface (Fig. 2a) [43]. Inspired by the Salvinia effect, Chen et al. [44] successfully fabricated a bioinspired superhydrophobic eggbeater-like structure inspired by the structure of the Salvinia molesta via a three dimensional (3D) printing technique (Fig. 2b). 3D printing technology can replicate the complex structures of natural materials at a finer scale, which provides a prerequisite for the successful preparation of bioinspired materials. The as-prepared eggbeater structure, made with a hydrophilic, photocurable material, shows remarkable superhydrophobic properties and water adhesion. This Salvinia-inspired structure was further applied to separate oil from water, with efficient removal of oil from the water in 6 s (Fig. 2b).

Surfaces with superwettability are capable of providing a number of practical properties, e.g., altering hydrodynamic drag, antireflection, and self-cleaning [41,45–49]. For example, shark skin has numerous micron-sized riblets that contribute to their efficient swimming motion (Fig. 2c). These riblets, which are oriented along the longitudinal axis of shark's body, can effectively reduce the resistance of water flow over the skin surface. Zhang et al. [49] designed bioinspired polydimethylsiloxane (PDMS) films with superhydrophobicity that mimicked the structure of shark skin by a replication technique and subsequent chemical modification. The fabricated PDMS films show larger water contact angles than the non-grooved structures (Fig. 2d and e). Therefore, these PDMS films provide superhydrophobicity as well as self-cleaning (Fig. 2f). As these PDMS films trap air, their interaction with the surrounding fluid is a gas-liquid interface as opposed to a solid-liquid interface. Consequently, the fluid can freely flow over the surface, which causes effective slippage and reduces flow resistance.

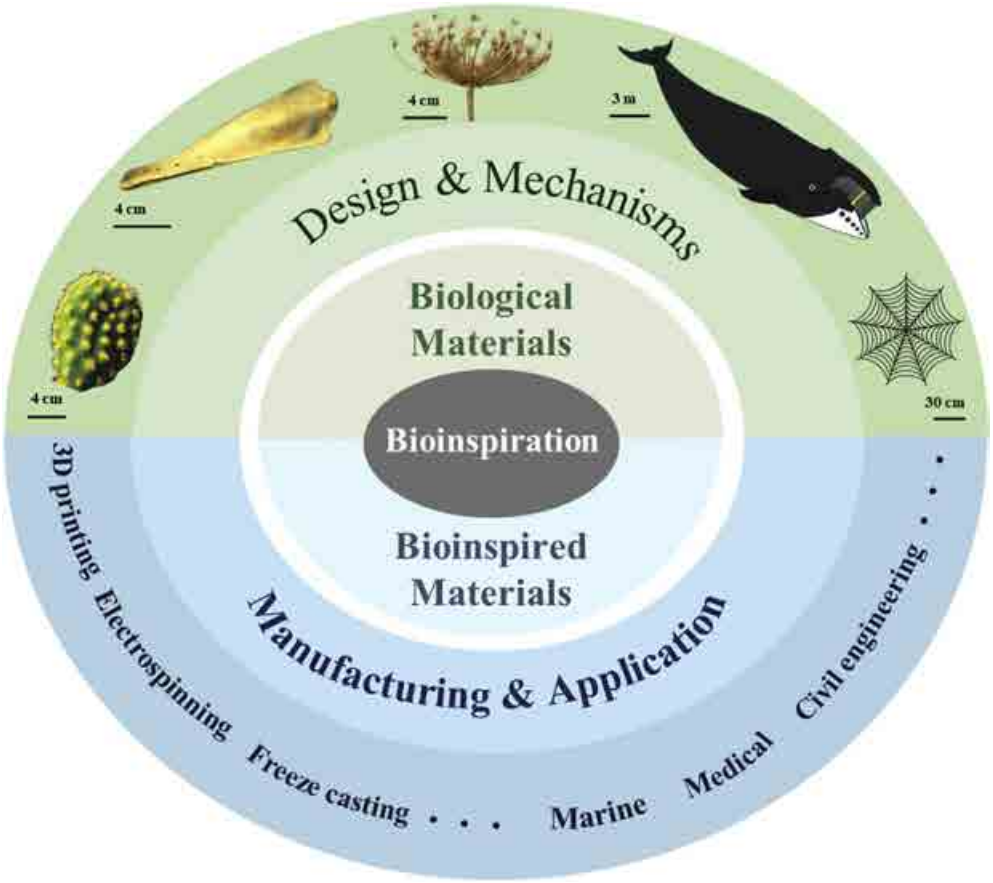

Fig. 1. The development of bioinspired materials from natural prototypes.

Therefore, these bioinspired PDMS films with superhydrophobic surfaces have an excellent drag reduction effect (up to a maximum of 21.7%), which is even greater than shark skin (where the maximum is 5.4%) (Fig. 2g).

Another function of superwettabile surfaces is directional liquid transport, which is exhibited in nature in cactus spines. Given their arid environment, these cacti spines collect water through a hierarchical surface structure. Fig. 2h shows that the conical spines of cacti consist of three parts with different structural features, the oriented barbs, gradient grooves, and belt-structured trichomes, which are staggered from the tip to the base, respectively [45]. Subtle integration of the three parts creates a gradient in the Laplace pressure and a gradient in the surface-free energy, therefore enabling the droplets to move towards a desired direction for collection so as to nourish the cactus. Specifically, the droplets on the surface of cactus spine are usually driven to the side with a larger radius (that is, the base of the spine) due to the gradient of the Laplace pressure; at the same time, the microgrooves on the spine are sparser at the base (less rough) than at the tip (more rough), and this gradient roughness produces a gradient of surface-free energy, which generates a driving force to drive the water droplets toward the base of the spine. Inspired by the water-collection principle of cacti, Jiang et al. [48] designed a bioinspired water collector with dual gradients via gradient electrochemical corrosion and gradient chemical modification (Fig. 2i). The artificial cactus spine collects water drops at a relatively high speed of about 2.08 μL s^{-1} and transports it quickly with a velocity of around 20.05 μm s^{-1} (Fig. 2j).

3.1.2. Bioactivity

Biological organisms consist of materials that are inherently biocompatible and bioactive, thus effectively fulfilling the functions of life. This provides important guidance to the design and development of advanced biomaterials for biomedical applications. In essence, the bioactive function arises from the special structure and composition of biological materials, which interact with cells/tissues. Based on this, there has been a significant amount of research effort towards fabricating novel bioinspired structures that show superior biocompatibility and bioactivity. Typical techniques include 3D printing, chemical modification, self-assembly, electrospinning, and templating, to name a few. Each fabrication technique has its own unique advantages, which can be employed to obtain different morphologies. For example, bioactive scaffolds for bone tissue engineering have been prepared by combining 3D printing and a biomimetic mineralization process (Fig. 3a) [50]. The biomimetic mineralization involves in-situ formation of hydroxyapatite (HAp) by the reaction of calcium ions and phosphate ions in simulated body fluid on the surface of the scaffolds. This allows the HAp to be more uniformly distributed in the scaffold than materials that are directly mixed with HAp particles. In addition, this structure has a similar chemical structure and similar material properties to natural bone minerals. Therefore, it has a higher biological activity and better promotes cells proliferation and growth [51–54]. After biomimetic mineralization these scaffolds have better mechanical properties to meet the needs of bone tissue repair, while scaffolds without biomimetic mineralization tend to be soft and are prone to collapse.

Along with single-material 3D printed structures for applications in skin, bones, muscles, and so on [55–57], the central nervous system is challenging to mimic due to the complexity of its structure and function [58,59]. Spinal cord tissue contains different types of cells, and the arrangement of these cells has a highly controlled spatial distribution. This spatial distribution is the key to mimic the spinal cord architecture, which plays an important role in controlling the differentiation of cells. Recently, McAlpine et al. [60] fabricated a heterogeneous bioinspired

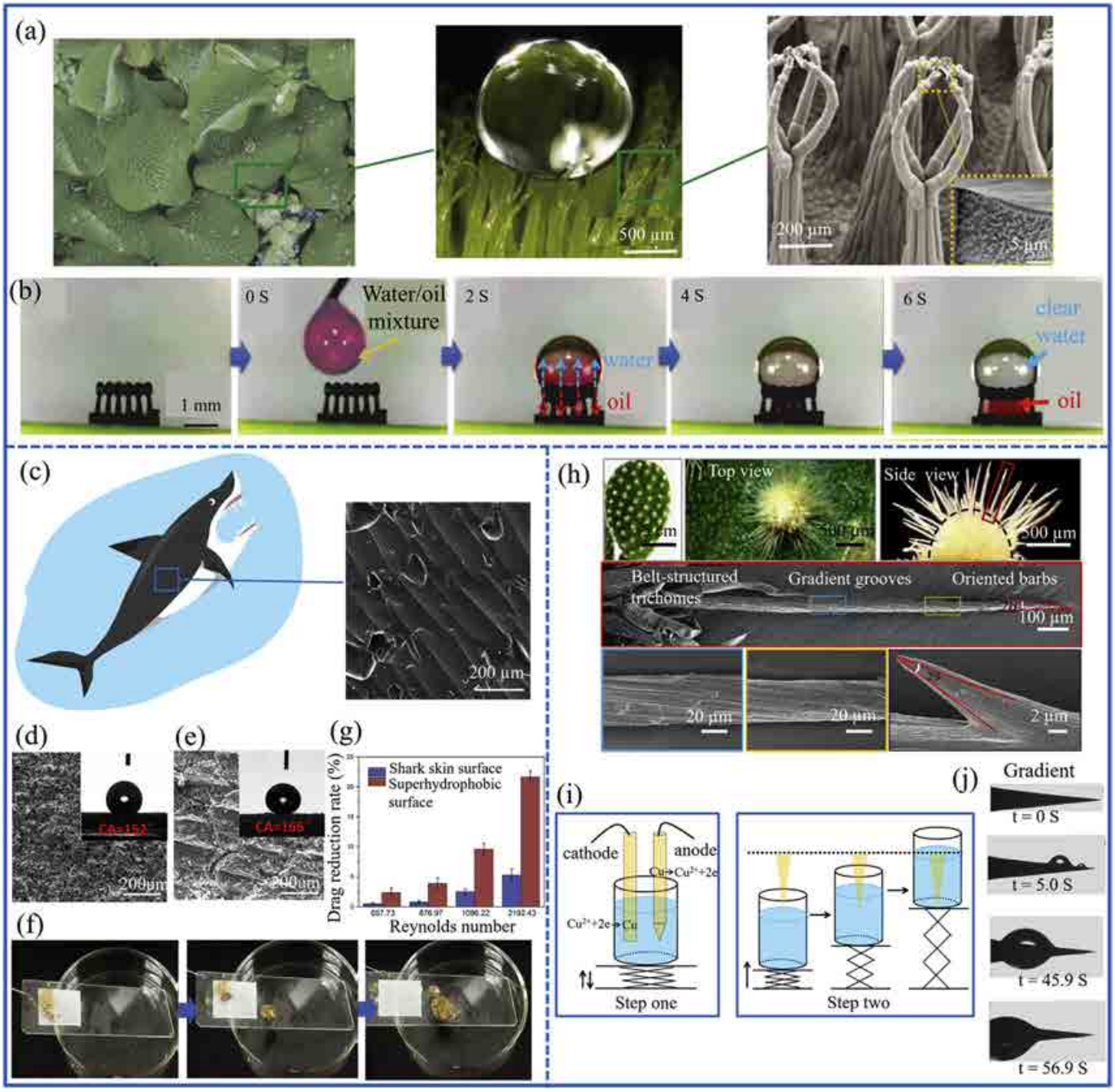

Fig. 2. Bioinspired functional materials with superwettability. (a) Optical and SEM images showing the morphology of Salvinia molesta leaves at different magnifications. (b) Oil/water mixture separation by the printed eggbeater-like structures [44]. (c) A schematic diagram of a shark and SEM image of shark skin. SEM images and water contact angle photographs of PDMS films with (e) and without (d) a bioinspired, grooved structure. (f) The self-cleaning effect of biomimetic PDMS films. (g) The drag reduction rate of shark skin surfaces and superhydrophobic surfaces at different Reynolds numbers [49]. (h) Appearance and surface structures of the cactus. (i) A diagram of the fabrication of bioinspired water collectors through gradient electrochemical corrosion and gradient chemical modification. (j) Microscopic observations of typical water-collection processes on artificial cactus spines with dual gradients [45,48].

spinal cord by multi-material 3D bioprinting in which clusters of spinal neuronal progenitor cells (sNPCs) and oligodendrocyte progenitor cells (OPCs) can be accurately positioned in different parts of the scaffold during assembly (Fig. 3b–d). The precise positioning of cell types allows the bioinspired spinal cord to model the native tissues as closely as possible, which enhances the bioactivity of the scaffold. These bioinspired spinal cords successfully showed differentiation of functionally mature neurons during in vitro culture, which allows for further differentiation of sNPCs and OPCs into neurons projecting axons and oligodendrocytes that myelinate the axons, respectively (Fig. 3e). This bioinspired process provides a novel approach to prepare scaffolds with better bioactivity for mimicking the central nervous systems in vitro and thus provides a novel technique for treating neurological diseases.

In addition to printing, electrospun nanofibrous scaffolds show features that simulate the structure of the extracellular matrix (ECM) and thus provide an avenue for tissue repair and regeneration [61–63]. However, electrospinng generally fabricates two-dimensional (2D) fibrous membranes, which must be expanded into the three-dimensional (3D) geometry of organs while also mimicking the surface morphology of collagen fibrils in the ECM. To address this, Mi et al. [64] prepared

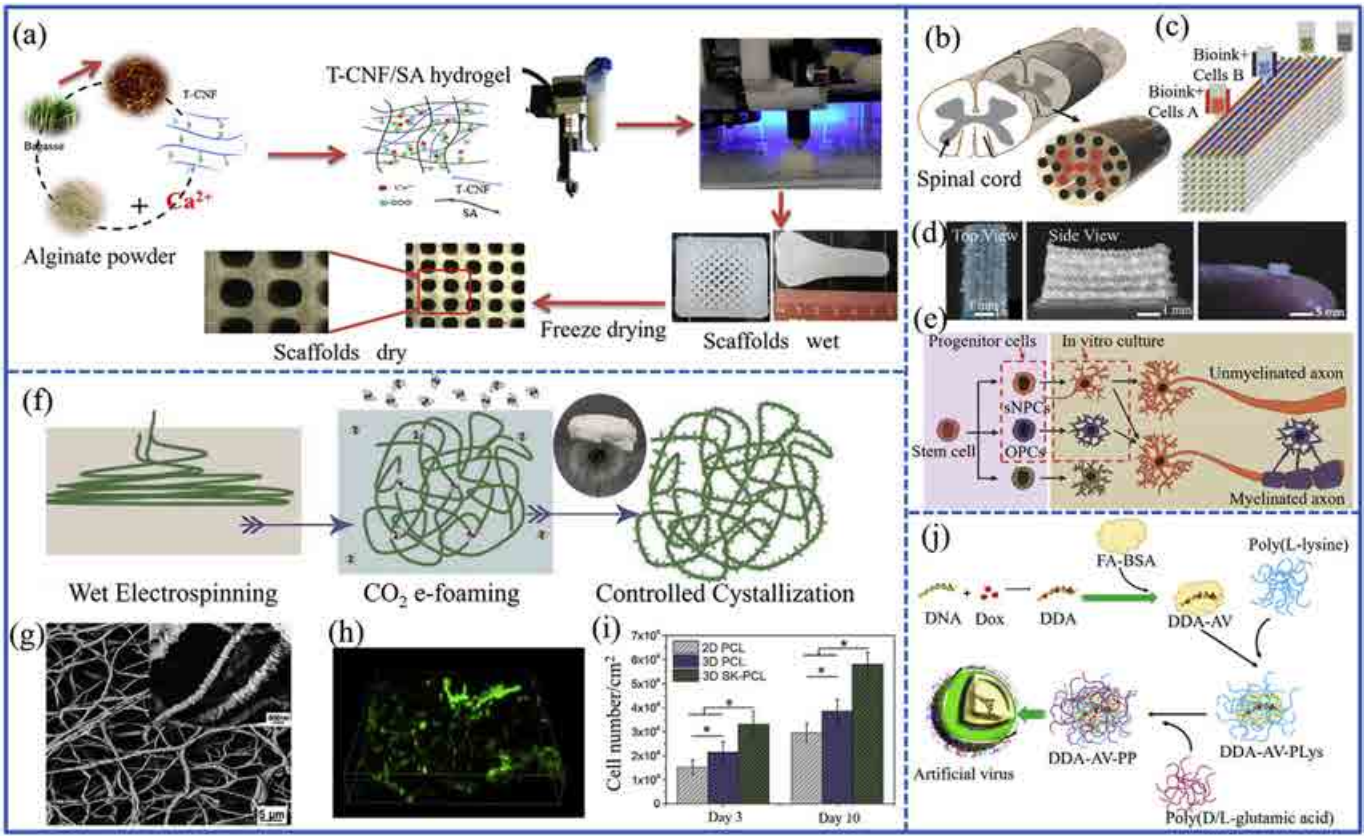

Fig. 3. Bioinspired functional materials with bioactivity. (a) A fabrication process for 3D printing scaffolds from TEMPO-oxidized cellulose nanofibrils/sodium alginate hydrogels [50]. (b) A schematic of the spinal cord and a design for a 3D bioprinted multichannel scaffold that models the spinal cord. (c) A schematic overview of the 3D bioprinting process. (d) The as-prepared scaffolds. (e) A schematic of the induced pluripotent stem cell reprogramming and differentiation into sNPCs or OPCs [60]. (f) A schematic illustration of the bioinspired nanofiber scaffold preparation process. (g) The morphology of 3D SK-PCL nanofibers at different magnifications. (h) Fluorescent images of live/dead assay results for human fibroblasts cultured on an as-prepared scaffold. (i) Cell proliferations results from an MTS assay of human fibroblast cells cultured on different materials for 3 days and 10 days [64]. (j) A schematic representation of the formation of artificial viruses [67].

biomimetic scaffolds, specifically a 3D nanofibrous foam (3D-PCL), with high biological activity by means of wet electrospinning, carbon dioxide (CO_2) foaming, and controlled crystallization (Fig. 3f). Compared with a 2D structure, the 3D-PCL is more conducive to cell infiltration and growth through the scaffolds. This nanofibrous foam was treated in a PCL/pentyl acetate solution to introduce a shish-kebab nanostructure onto the nanofiber surface (3D-SK-PCL) by controlled crystallization (Fig. 3g). Such nanostructures on the nanofiber surface are favorable for cell adhesion and migration [65,66], and can further improve the bioactive function of the scaffold. As can be seen in Fig. 3h, the HEF1 human fibroblast cells cultured on the 3D-SK-PCL scaffolds spread and elongate well, exhibiting a healthy state, and the cell population in the 3D bioinspired scaffold is significantly higher than that of other scaffolds (Fig. 3i). This is attributed to the structural features of the 3D-SK-PCL, which mimic the real 3D geometry required by living cells and the nanotopography of native collagen fibrils, thus stimulating cell attachment, migration and growth.

Another notable bioinspired design that provides enhanced bioactivity involves mimicking the sophisticated structures of viruses to develop high-efficiency targeted drug delivery carriers. Viruses consist of nucleic acid molecules surrounded by a protective coat of protein. Some substances on the coat surface, such as glycoproteins, can specifically bind to the receptors on the surface of host cells to achieve cell invasion. Inspired by this, the in vivo bioactivity of artificial materials can be improved by mimicking this viral nanostructure with a host-recognizable shell, which enables the ability to target the material to biologically relevant cells/tissues. Pramod et al. [67] established an artificial virus system with a core-shell structure for targeting tumor tissues with a simple and predictable self-assembly technology (Fig. 3j). In the artificial virus, the core is composed of adduct of doxorubicin and DNA. Doxorubicin is a specific anti-cancer drug, which is also toxic to healthy cells; therefore, it can be included in the artificial virus to avoid unnecessary toxicity. The outer layer is folate tethered albumin. Folate receptors are frequently overexpressed on cancer cells but exhibit limited expression on normal cells [68,69], which allows the artificial viruses to recognize and adhere to the tumor tissue. The outermost layer of the artificial virus is a polypeptide complex composed of poly(L-lysine) and poly(G/L glutamic acid), which dissolves in acidic pH [70]. Therefore, as the artificial virus reaches the tumor tissue, the complex dissolves and the drug releases, thus accomplishing targeted drug delivery.

3.1.3. Stimuli-responsiveness

Ingeniously designed by nature, a number of natural materials exhibit effective sensing and actuating functions based on passive structures that involve nonliving tissues. In biological systems, an external stimulus interacts with and alters the structure in different degrees, therefore producing a complete cycle of sensing and responsive behavior. This usually involves an energy change depending on the exact procedure, e.g., thermal energy into strain energy for sensing light and color changing (via changed periodic structure). Such a process eliminates the complexities of dealing with the living cellular activity or complicated chemical constituents, but still obtains precise control and enables a wide variety of sensing and responding behaviors, such as transforming signals of force, light, heat, humidity, etc. into electrical, mechanical or other forms of responses [26,71–76]. These provide

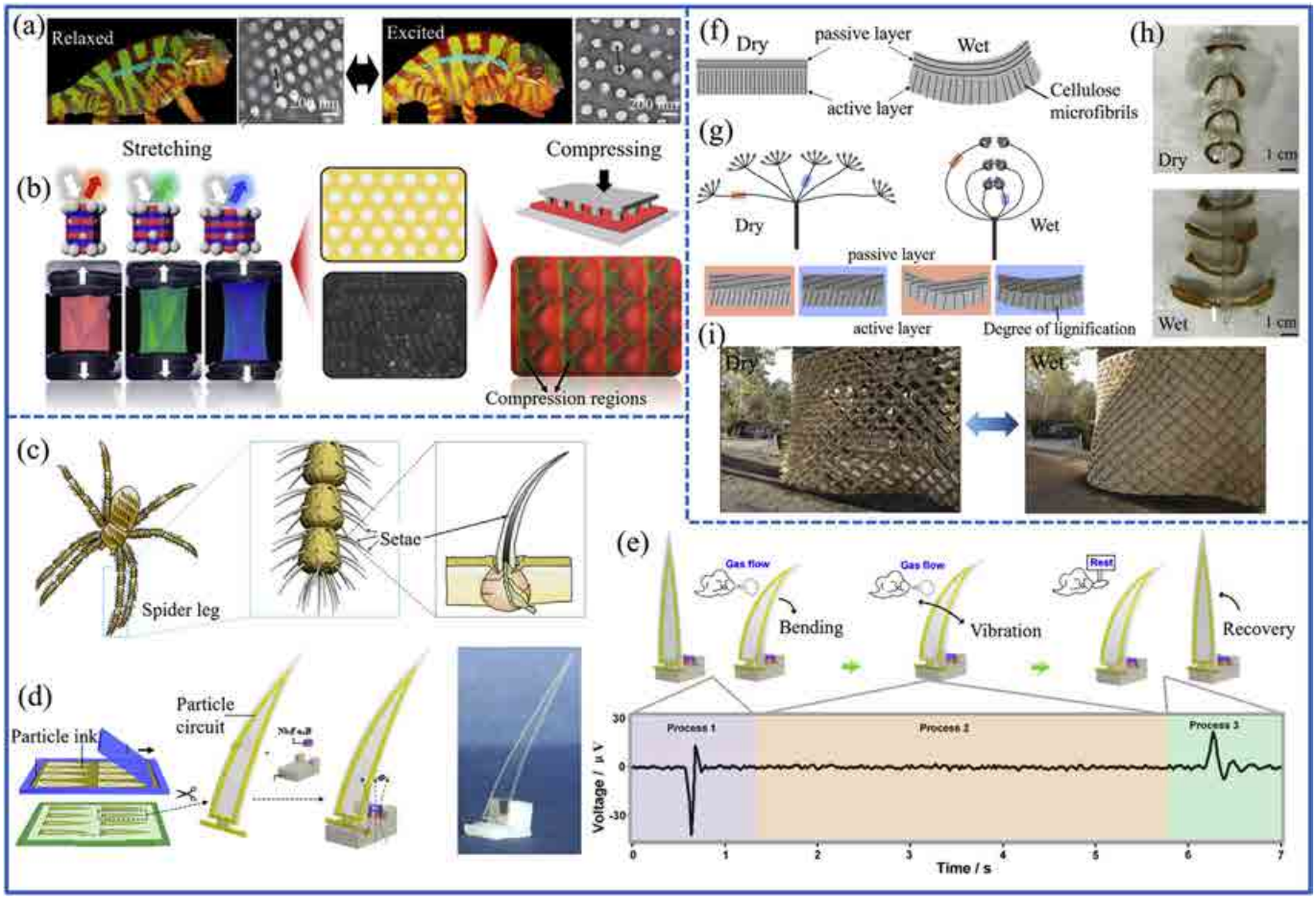

Fig. 4. Bioinspired functional materials with stimuli-responsiveness. (a) The relaxed and excited chameleon showing green color (left) and yellow color (right), respectively. Periodic changes in regular arrays of guanine nanocrystals are shown by transmission electron microscope images [77]. (b) The color change of artificial film during stretching and compression [74]. (c) Illustrations of fine hairs on the exoskeleton of spiders. One hair acts as a wind sensor to sense nearby wind changes. (d) Schematic illustrations of the preparation process of bioinspired wind sensors and optical photographs of an as-prepared sensor. (e) The electrical signals as output from the as-prepared wind sensor [75]. (f) Typical hygroscopic plants with a bilayer structure. (g) Hygroscopic deformation of the D. *carota* umbel. (h) Changes in the shape of synthetic pinecone scales with a similar bilayer structure under dry and wet conditions [31,84]. (i) The exterior wall of a building that mimics the humidity sensing of pinecones [83].

inspiration especially suitable for developing bioinspired, engineered, smart structures and devices.

Among the many intriguing natural sensors, the chameleons are notable for their ability to rapidly adjust their colors between a concealed (camouflaged) state and a highly visible (excited) state [77] (Fig. 4a) when fighting or courting. Studies reveal that this color change is achieved primarily by actively tuning the lattice of guanine nanocrystals within the iridophore cells. By mimicking the photonic structure of the iridophore cells of chameleons, mechanochromic elastomers containing a non-close-packed array of silica particles were designed [74]. In this bioinspired sensor, particles of rigid silica nanocrystals are embedded in a matrix of elastomer to form non-close-packed crystals. As the sensor undergoes a rapid volume change, the lattice parameters change while maintaining the lattice structure, which changes the refraction of light, resulting in dramatic color changes throughout the visible range. The sensor exhibits a color shift of red to blue under stretching, and a color change of red to green under compressing (Fig. 4b). Impressively, just like their biological inspiration, this color change is reversible. These mechanochromic sensors have promising applications in various fields such as large-scale wallpaper, signboard displays and optical recording.

Another interesting sensing and responding system is spider hair, which acts as a wind sensor that allows it to sense nearby airflow changes caused by predators or prey (Fig. 4c) [78]. Inspired by animal hair sensors, Su et al. [75] reported self-powered wind sensors based on flexible magnetoelectric material systems (Fig. 4d). The bioinspired sensor is composed of electrical and magnetic components. The electrical component includes silver nanoparticles on a thin polyethylene terephthalate (PET) film, created by screen printing, which mimics the triangle shape of spider's fine hairs. Thus, this part is flexible (for sensing wind) and conductive (for signal output). The magnetic component is NdFeB, which is combined with the electrical component through a 3D printing-assisted approach. This magnetic component constantly provides magnetic flux through the electrical component. The sensor will thus produce a unique electrical output (i.e., voltage) as the wind blows past it (Fig. 4e). When the air flow blows, the sensor bends rapidly and generates a negative voltage (-45.2 μV). Subsequently, the sensor bounces slightly due to inertia, which leads to a small reverse peak. Then as the wind blows continuously, the magnetic flux keeps consistent and thus no clear peaks in voltage appear (Fig. 4e). After removing the air flow, the PET film returns to its original state, and a positive voltage (20.3 μV) occurs due to the change of the relative position of the electrical and magnetic components. This wind-to-electrical signal relationship can be further quantified, which makes such sensors promising for applications that require the air flows measurements in harsh environments.

In addition to the animal kingdom, many immobile plants also show fascinating sensing and responding behaviors, such as hygroscopic,

photoinduced and force-driven features, which are of particular interest for practical applications [31,79–83]. One well-known example is the hygroscopic deformation of the pinecone scales [31] and the carrot umbel [84]. Upon dehydration, the corresponding structural components open for seed dispersal (through bending deformations of the constituent parts), and reversibly close for seed protection upon hydration (Fig. 4f). This hygroscopic deformation has been attributed to the orientation of cellulose microfibrils, the lignin distribution, and the tissue composition, which is usually analyzed through a bilayer structure (lines in Fig. 4g represent the cellulose microfibrils and the intensity of grey background lignification). The swelling properties differ between the two layers, e.g., the lower, active layer swells and elongates longitudinally as water enters into the matrix while the upper, passive layer does not, therefore resulting in an overall bending deformation. Based on this, artificial pinecone scales were fabricated through carefully aligning the reinforcements within the matrix to create the bilayer structure. This allows the artificial pinecone to exhibit similar humidity-induced deformation (Fig. 4h). Inspired by this, Reichert et al. manufactured bioinspired building skins that can sense humidity changes in the environment and automatically adjust the "skin" movement to open and close for the building (Fig. 4i) [83]. This provides a new approach to alleviate today's demand for high-tech electronic or mechanical systems, as this design allows the reversible movement without utilizing motors.

Through unraveling the mechanisms of representative biological materials featuring superwettability, bioactivity and stimuli-responsiveness, control of the hierarchical structure is the key to obtaining the desired functions. After formulating the design principles and then leveraging advanced fabrication technologies, such as high-resolution and/or multi-material 3D printing, chemical modification, replication, eletrospinning, self-assembly, and magnetic-assisted composite formation, a variety of bioinspired materials with enhanced, target functions can be developed for a diversity of applications through modulating their structures.

3.2. Bioinspired structural materials

3.2.1. Lightweight and high-strength materials

Many biological materials show superior mechanical properties by being light-weight and strong, in spite of their limited, weak chemical constituents (i.e., polymers and minerals). The key of obtaining this high strength combined with low weight without a complex system of available chemical constituents lies in the diverse, hierarchical structures of biological materials [85]. This provides invaluable inspiration for developing advanced composite materials for various modern industries, such as aviation, aerospace, marine, and land transportation.

Cellular structural materials, such as honeycomb [86], cancellous bone [2], cuttlebone [87–89], are typical lightweight materials with favorable strength. Taking cuttlebone as an example, it has a high porosity (93%) and excellent mechanical properties, which are balanced via a hierarchical structure from the nano- to the micro-scale. As shown in Fig. 5a, in the cuttlebone, aragonite nanofibers with an organic phase (about 4.5 wt%) form micrometer-thick lamellae, which are separated and supported by numerous, evenly distributed, micrometer-thick pillars, resulting in a highly complex porous structure that is able to sustain the pressure associated with their deep sea environment [88]. Burghard et al. fabricated hierarchical porous scaffolds using vanadia (V_2O_5) nanofibers to mimic this intricate architecture by ice-templating (Fig. 5b). The porosity of these bioinspired scaffolds is 99.8%, which is even higher than that of the cuttlebone. The compressive stress-strain curves in Fig. 5c show that, compared the scaffolds with the same ultra-high porosity but random microstructures (V_2O_5-0), the cuttlebone-like scaffolds (V_2O_5-1) have superior mechanical properties in terms of their compressive strength, which is about twice that of the V_2O_5-0. This is because of the regularly arranged rectangular pores in the V_2O_5-1, which can distribute the applied stress more effectively than the randomly assembled pores in the V_2O_5-0. In addition, increasing the vanadia nanofiber concentration of the cell wall leads to significantly enhanced mechanical properties, e.g., the Young's moduli of the fabricated cuttlebone-like scaffolds with different nanofiber concentrations are ten to fifty-five times higher than that of the V_2O_5-0 (1.73 ± 0.71 kPa). This is due to the rectangular pores minimizing the lateral motion of the lamellae, thereby increasing the Young's modulus.

The stems of many plants also show structural characteristics that make them strong enough to withstand the environmental stressors from their habitats (e.g., wind, rain, ocean currents) while also minimizing weight [90–92]. One type of natural strong materials is thalia dealbata; it is a perennial plant that has an impressive height/diameter ratio (~200–350), which requires the stem to be strong enough to survive within frequent wild winds. Bai et al. revealed that its porous stems exhibited a structure of oriented lamellar layers along the growth direction with interconnected bridges (Fig. 5d) [92]. This hierarchical structure is successfully mimicked in graphene aerogels by employing bidirectional freezing techniques (Fig. 5e). Moreover, as a general method, the bidirectional freezing achieves multiscale architectural control in a scalable manner, which can be extended to many other material systems. The as-prepared graphene aerogel can support more than 6000 times of its self-weight with around 50% strain. Moreover, it can fully recover without obvious permanent deformation after unloading (Fig. 5f), as about 85% of the original compressive strength is retained after 1000 compressive cycles at 50% strain (Fig. 5g and h). These results show that exceptional strength and low weight can be simultaneously achieved from the structural design of the thalia dealbata.

Natural wood is low-cost and sustainable, and a variety of studies have been devoted to enhancing the absolute mechanical strength of wood and bioinspired wood for advanced engineering applications [93–96]. In natural wood, lignin represents one amorphous matrix material that glues cellulose fibrils and plays a major role in determining the overall strength [97]. Inspired by the wood structure, Yu et al. [98] showed a strategy for large-scale fabrication of polymeric woods with similar amorphous polyphenol matrix materials (phenol-formaldehyde resin and melamine-formaldehyde resin) by a self-assembly and thermocuring process of traditional resins (Fig. 5i). Compared with natural woods, the polymeric woods show comparable compressive properties (a compressive yield strength up to 45 MPa, Fig. 5j and k). Moreover, the axial compressive performance of bioinspired woods is better than that of other wood-like materials, such as ceramic-based foam materials (Fig. 5k). The density of the polymeric woods shows a wider range than that of other engineering materials, which indicates a good tunability in weight and strength through control of the microstructure and the fabrication parameters.

Another well-known light weight and strong biological material is the spider silk. It has recently become clear that the excellent mechanical properties are mainly attributed to the structure, which involves highly ordered and dense hydrogen-bonded β-sheet crystals within a semi-amorphous protein matrix (Fig. 5l). Inspired by the hierarchical nanostructure of spider silk, Chen et al. developed a bioinspired composite film, which was fabricated by introducing graphene quantum dots (GOD, mimicking the β-sheet crystals) into polyvinyl alcohol (PVA, as the protein matrix) (Fig. 5m) [99]. Investigating the mechanical performance of the bioinspired GOD-reinforced PVA composite films reveals that controlling the content of GOD can greatly improve the mechanical properties. Specifically, when the content of GOD reaches 5.0 wt %, the yield strength and elastic modulus of the PVA are enhanced by about 66% (152.5 MPa) and 88% (up to 4.35 GPa), respectively when compared to 0.0 wt % (Fig. 5n).

3.2.2. Lightweight and tough materials

Distinct from traditional engineering materials, where achieving the two mutually exclusive properties of strength and toughness presents a

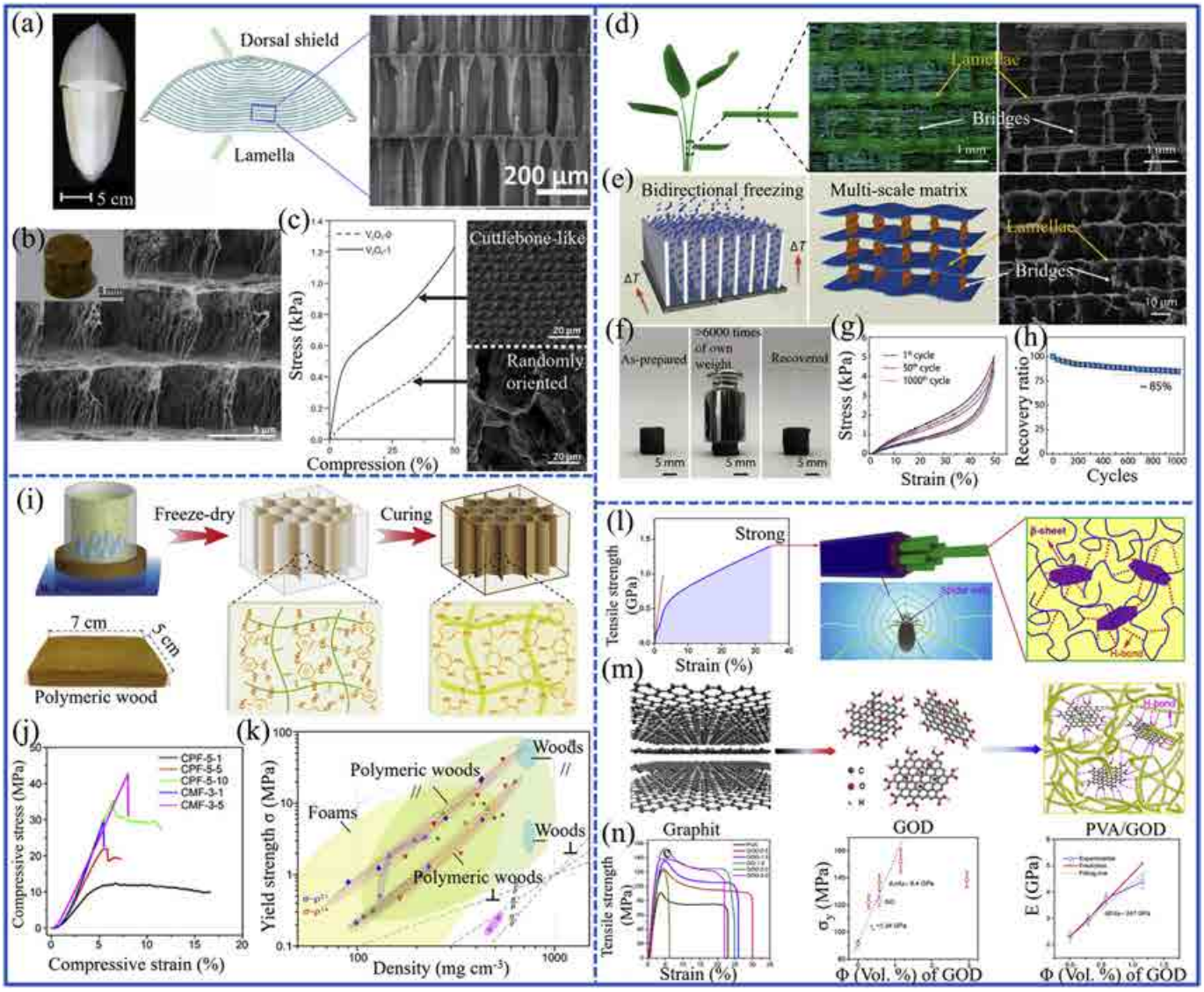

Fig. 5. Bioinspired structural materials that are lightweight and strong. (a) An image of the entire cuttlebone, a schematic representation of its transverse cross-section and an SEM image of its framework. (b) An optical image and SEM image of a bioinspired scaffold based on cuttlebone. (c) Stress-strain curves (left) of V_2O_5 nanofiber scaffolds with different structures (right) [87,89]. (d) An optical image and SEM images of a thalia dealbata stem. (e) A schematic diagram of the preparation process of bioinspired graphene aerogels and an SEM image of an as-prepared graphene aerogel. (f) Optical images showing an aerogel compressed and recovered with no obvious permanent deformation. (g) Representative stress-strain curves and (h) the strength recovery ratio of an aerogel compressed and recovered after 1000 cycles [92]. (i) Fabrication route of bioinspired polymeric woods. (j) Axial compressive stress-strain curves of typical polymeric woods. (k) An Ashby chart plotting the compressive yield strength versus density for polymeric woods and other engineered materials [98]. (l) A schematic representation of the tensile behavior and hierarchical microstructure of spider silk. (m) The fabrication process of GOD-reinforced PVA composite films. (n) Mechanical performance of bioinspired polymer composite films [99].

significant challenge, most biological materials are exquisitely designed to exhibit high-toughness while maintaining sufficient strength. Typical light-weight and tough materials in nature encompass the highly mineralized (e.g., seashell nacre and teeth), intermediately mineralized (e.g., bone and fish scales), and non-mineralized (e.g., human skin and wood). The relevant structure and mechanical properties of these materials have been extensively characterized, with an aim to distill the essential principles for developing new advanced composites with high toughness, favorable strength, and light weight. Fundamentally, toughness is the amount of energy that the material can absorb/dissipate through deformation while sustaining load without failure, which has been largely attributed to the soft, polymeric constituents. The salient structural toughening mechanisms in representative highly, intermediately and non-mineralized biological and bioinspired materials are discussed here.

Highly mineralized biological materials, such as mollusk shells and teeth, have attracted extensive attention for their exceptional strength and toughness [16,100,101], considering that the dominant brittle constituent (> 95 wt%) of calcium carbonate is the same material as classroom chalk [18]. It is known that the mineral components are responsible for providing the overall stiffness and hardness; while the organic constituents (which exhibit significant deformability), despite a meager content, play a crucial role in enhancing the toughness. They organize into weak interfaces with intricate architectures between the mineral building blocks, therefore directing the propagation of cracks into more tortuous configurations and dissipating more energy to improve the overall toughness [7]. In enamel, mineral rods (about 4–6 μm in diameter, composed of 50–70 nm hydroxyapatite crystalline rods) and proteins constitute the bulk material and the weak interface, respectively. The cracks are guided to propagate along the weak interface,

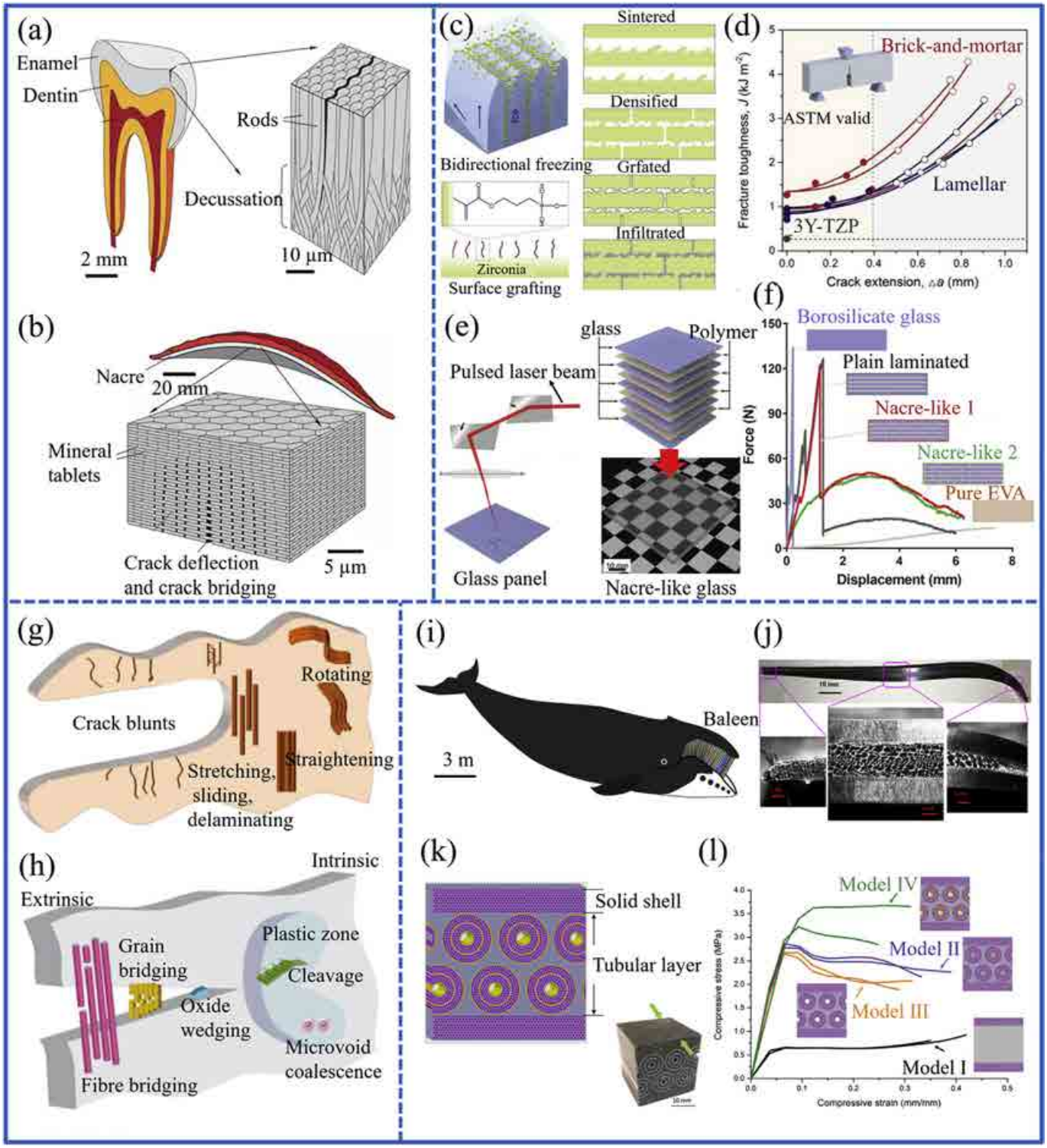

Fig. 6. Bioinspired structural materials that are lightweight and tough. (a) An overview of human teeth and the microstructure of enamel. (b) An overview of nacre and its toughening mechanisms [103]. (c) Schematic illustrations showing the formation process of nacre-mimetic composites. (d) *J*-integral fracture toughness with crack extension for lamellar and brick-and-mortar composites as compared to monolithic 3Y-TZP ceramic [107]. (e) Fabrication protocol for nacre-like glass panels. (f) Puncture force-displacement curves for pure borosilicate glass and pure EVA panels, plain-laminated panels, and nacre-like panels [111]. (g) Schematic illustrations of toughening mechanisms in skin. (h) Intrinsic and extrinsic toughening mechanisms of human bone [7]. (i) A schematic of the bowhead whale; the blue arrow indicates the direction from which the baleen hangs. (j) Transverse sections of the baleen plate. (k) A structural model of the baleen plate; the inset shows the printed model where green arrows indicate the loading direction for (l) structural models I, II, III, and IV with their corresponding compressive behavior [131].

circumventing the region where the mineral rods are located, thus avoiding a catastrophic failure (Fig. 6a) [102,103]. Similarly, in nacre the proteins and polysaccharides hold the microscopic tablets of calcium carbonate together to form “strong-weak” microscopic features. The organic phase is extremely important in the processes of controlling the crack propagation and energy dissipation (Fig. 6b) [4,104–106]. Inspired by this, Ritchie et al. [107] fabricated bioinspired dental materials by freeze-casting of zirconia polycrystals doped with yttria (3Y-

TZP) in suspension and further densification with a methacrylate resin, which resulted in composites with nacre-like lamellar and brick-and-mortar architectures (Fig. 6c). The *J*-integral fracture toughness curve in Fig. 6d shows rising R-curve behaviors for both the lamellar and brick-and-mortar composites, indicating stable crack propagation and increased crack-growth resistance that are different from the instantaneous cracking of 3Y-TZP ceramics. Moreover, the critical *J*-integral toughnesses of the two composites are about 1.2 and 1.7 kJ m^{-2}, respectively, which are about four times and six times higher than 3Y-TZP ceramics (Fig. 6d).

In addition to the crack deflection and crack bridging, the sliding mechanism in nacre can also dissipate a large amount of mechanical energy, which makes nacre deformable and tough [108–110]. Inspired by this, Barthelat et al. prepared bioinspired toughened glass by laser engraving and lamination fabrication methods [111]. In the process of preparation, the contours of the tablet are carved on borosilicate glass sheets by laser beam, then the engraved glass sheets are stacked with ethylene-vinyl acetate (EVA) as a mortar, and then laminated to obtain a transparent bioinspired glass (Fig. 6e). Puncture tests show that, compared to ordinary glass and laminated glass, the nacre-like glass produces a more ductile response with large deformations and high puncture energy (area under the force-displacement curve). This is because the tablets in nacre-like glass can slide on one another over large volumes and absorb a large amount of mechanical energy (Fig. 6f).

Intermediate and non-mineralized biological materials are remarkably tough, which is realized through their polymeric constituents and structural organization, both of which contribute to large deformations while carrying load. Biological collagenous materials encompass non-mineralized tissues such as skin, arteries, and eye corneas and modestly mineralized including bone and fish scales. Their mechanical behaviors ranging from the molecular to macroscales have been widely studied [112–115]. The toughening mechanisms of a non-mineralized collagen, skin, are illustrated in Fig. 6g [7,112,116,117]. The collagen fibers in the skin tissue are curved and arranged in multiple orientations. When being stretched the fibers rotate to align with the tensile direction and straighten. During this process, the collagen produces multiple toughening mechanisms including fibril straightening, reorienting, stretching at the molecular scale, and delaminating at a larger microscale, all of which absorb significant energy and ultimately improve the toughness and tear resistance of the skin.

For intermediately mineralized biological materials, human bone is a remarkable material that shows superior toughness and strength. It is composed of nanoscale hydroxyapatite crystals and collagen molecules, which then bundle into fibers and further organize into microscale lamellae, mesoscale osteons and the mature cortical bone. The hydroxyapatite constituent of bone plays a dominant role in strength [118,119], elasticity [120–123], and creep properties [124,125], while the collagen component accounts for the deformability and energy absorption. The toughening mechanisms in bone are usually classified into either extrinsic and intrinsic [126–128], depending on if the mechanisms operate behind or ahead of the crack tip, respectively (Fig. 6h) [2,129]. Intrinsic toughening acts at the nano-scale to inhibit damage and takes the form of the stretching and sliding of collagen fibrils to form plastic zones around crack-like defects. Extrinsic toughening mechanisms operate at the micrometer-scale and principally act on the crack wake to shield the local stresses/strains. They generally toughen the entire material via processes such as crack deflection, constrained microcracking and crack bridging, which effectively resist crack propagation. Both types of mechanisms result in a higher crack-driving force required to propagate cracks and thus increase the overall toughness of materials.

In addition to collagenous materials, biological keratinous materials represent some of the toughest biological materials. As one notable representative, baleen (Fig. 6i) is a keratin-based structure in the mouth of baleen whales, providing a life-long filter-feeding function (Fig. 6j). This indicates that baleen must be light enough for ease-of-use and mechanically sustain a variety of forces from water and prey. This is enabled by a hierarchical structure, in which key features include macroscale sandwich-tubules, microscale tubular lamellae, and a nanoscale filament-matrix with mineral structures [130]. Baleen shows significant fracture toughness, with the toughness *J*-integral reaching about 18 kJ m^{-2}, which makes baleen among the toughest biological keratinous materials. Much of this is due to the structural toughening mechanisms of crack redirection, fiber bridging and whitening. Based this, Wang et al. prepared a series of baleen-like models through multi-material 3D printing (Fig. 6k and l) [131], and revealed that the baleen-model has the best overall properties when compared to similar structures that do not include all of the baleen's toughening mechanisms. It is further demonstrated that aside from viscoelasticity, the structure plays an important role in impacting the strain-rate behavior of these materials.

Many biological materials represent exceptional structural materials that are lightweight, tough and sufficiently strong. These same properties have always been a core pursuit of almost all modern engineering fields such as aerospace, navigation transportation, automobiles. Two effective strategies from biological materials are the employment of cellular designs and hierarchical structures, which both endow increased resistance to deformation and failure while also introducing diverse toughening mechanisms. Fabrication techniques such as freeze-casting, self-assembly, thermocuring, 3D printing, and laser engraving are being further developed for the next step of scaling-up and mass production of advanced structural materials.

4. Perspectives and summary

Nature is the ultimate designer, having been refining materials and structures over millions of years. It presents a cornucopia of biological materials with intriguing properties, which provide resourceful inspiration for developing various novel materials. This has generated an impressive increase both in our fundamental understanding of biological materials and in the creation of bioinspired materials with diverse properties. Along with the rapid and tremendous growth of this field that has resulted in many fruitful outcomes, there are the challenges that need to be addressed for the ultimate promising future of biological and bioinspired materials.

With respect to biological materials science, since hierarchical structures range from the atomic/molecular to the macroscales, accurate characterization at each of these length scale to gain a thorough and in-depth understanding requires highly advanced and sophisticated techniques. Recent development in nanotechnology and computational simulation can supplement this to a certain degree, but some important observations at very small scales, e.g., the molecular toughening mechanisms of protein fibrils and chains, are still hard to be experimentally realized. To compound this, these techniques need to account for the delicate nature of the polymers that make up biological materials, which can be damaged by electron beams, desiccation, and vacuum conditions. At the same time, deducing relevant mechanics theories that connect and integrate the mechanisms taking effect at different length scales is a tough task, as current theories mostly work for homogeneous, single-scale structures. Moreover, at a general level, there have been a large amount of studies characterizing different biological materials; however, the search of conserved/unifying principles underlying these diverse natural phenomena is more important for practical applications and will allow for implementation in a broad variety of engineering applications.

With respect to creating bioinspired materials, it has to be admitted that the inherent efficiency of biological materials is difficult to duplicate, as the hierarchical structure involves fabricating delicate architectural details that are beyond the capability of most current nanotechnologies. In addition, most of these fabricated high-performance materials bioinspired from nature are synthesized within the laboratory

with limited size and quantity, and the properties are evaluated individually without uniform standards. Besides, the fabrication processes that are capable of guaranteeing structural features at multiple length scales are usually time-consuming and high-cost, which poses a great challenge for mass production at the industrial level.

Together with challenges are the opportunities and potential for the bright prospect of bioinspired materials. With the fast development of this research, the above challenges and many others will be addressed, which will contribute to translating the fundamental understanding of biological materials into practical engineering applications via bioinspired materials. Future materials to fulfil the ever-increasing demands from diverse industries would need to possess both high mechanical properties and multifunctionalities, while also being environmental-friendly. These are coincidently the inherent advantages of many biological materials. Thus bioinspired materials from biological systems show great potential in dominating the next generation of advanced materials design. In the current work, we formulate key strategies from biological to bioinspired materials, and discuss a variety of materials in terms of functional and structural categories. Through focusing on limited but essential properties with representative biological and bioinspired materials, we hope to have delivered a brief overview of the grand panorama of this field.

CRediT authorship contribution statement

Yayun Wang: Investigation, Data curation, Writing - original draft. **Steven E. Naleway:** Resources, Writing - review & editing. **Bin Wang:** Conceptualization, Supervision, Writing - review & editing.

Declaration of competing interest

None.

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