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Bioinspired 2D Nanomaterials for Sustainable Applications

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The increasing demand for constructing ecological civilization and promoting socially sustainable development has encouraged scientists to develop bioinspired materials with required properties and functions. By bringing science and nature together, plenty of novel materials with extraordinary properties can be created by learning the best from natural species. In combination with the exceptional features of 2D nanomaterials, bioinspired 2D nanomaterials and technologies have delivered significant achievements. Here, the progress over the past decade in bioinspired 2D photonic structures, energy nanomaterials, and superwetting materials, is summarized, together with the challenges and opportunities in developing bioinspired materials for sustainable energy and environmental technologies.

1. Introduction

Millions of years of evolution has produced unique structures and specialized functions in all creatures on the earth to cope with the environment. Fascinating interfacial phenomena observed in various natural species, such as vivid iridescent colors, unique light-response behaviors, superwetting properties, etc.,[1–4] offer excellent opportunities to develop artificial nanomaterials with similar functionalities, through manipulating the interface structures and chemistries based on inspirations from nature. To date, many bioinspired structures or interfaces have been constructed with some unique properties or functions, which cannot be observed in their constituent materials. For example, the exceptional mechanical, surface and optical properties observed in bone,[5,6] nacre,[7,8] gecko feet,[9] spider silk,[10] fish scales, butterfly wings,[11,12] etc., have been attracting increasing research efforts. Recent advances include nacre inspired damage tolerant materials;[13,14] fish scale and armadillo shell inspired artificial armors;[15,16] lotus leaf and cicada wing inspired self-cleaning surfaces;[17–19] insect compound eye inspired antifogging coatings;[20,21] gecko feet and mussels inspired superadhesive materials;[22,23] desert beetle and cactus inspired water harvesting;[24,25] opal, butterfly wings, and chameleon inspired photonic materials;[26–29] insect-trapping plant (e.g., nepenthes and sarracenia) inspired ultrafast water transport;[30] fish-gill and cactus-needle inspired oil–water separation,[31] etc. There is no doubt that learning from nature opens new doors for innovation in materials and sustainable technologies. Owing to the great diversity and complexity of nature, the well-evolved natural structures corresponding to specific functionalities may vary from 1D nanofibers or nanoneedles to 2D nanosheets or nanoplates or even 3D multiscale-ordered structures.[32] A good understanding of the structure–property relationships is essential for the design and fabrication of bioinspired nanomaterials.

2D nanomaterials have achieved some significant triumphs in various applications, benefitting from their unique structural characteristics and relevant chemical and physical properties.[33–35] In fact, 2D nanostructures also widely exist in nature and generate some amazing functionalities, which offers us great opportunities to further expand the design and fabrication of 2D nanostructured materials, devices, and technologies.[36] By integrating 2D nanomaterials with the bioinspired strategies, innovative materials and technologies have been proposed and realized. In this research news, recent progress that mainly achieved over the past decade in bioinspired materials and technologies based on 2D nanomaterials for targeted sustainable energy and environmental technologies, such as energy conversion and storages, environmental remediation, etc., is reviewed and discussed. As shown in **Figure 1**, three topical subjects, including bioinspired 2D photonic structures, bioinspired 2D energy nanomaterials, and bioinspired 2D superwetting materials, along with the challenges and opportunities will be the focus of this article, and give an overall perspective to this emerging and promising research area.

2. Bioinspired 2D Photonic Materials

To survive in the wild world, natural species have evolved various unique structures with fascinating optical functionalities, such as glitzy structural colors for attracting prey or mates,[37] tunable camouflage colors for escaping from predators,[38] antireflection function of compound eyes for weak light vision,[36] and so on. These structures, as known as photonic crystal structures, have inspired the design of novel photonic micro/nanostructures and some smart optical devices.

Among the various natural photonic structures, one class consists of periodically stacked 2D multilayers, also known as Bragg Stacks, have been found in many natural organisms, such as plants, insects, and marine benthos.[39] Multiple pairs of 2D layers with different refractive indexes in Bragg Stacks can generate iridescent structural colors through constructive

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Figure 1. a) Schematic images of neon tera fish,[59] seashell,[64] and *C. rajah* beetle.[49] (*In clockwise sequence, the same hereinafter*) b) Schematic images of Asian *Arowana* fish,[81] fly compound eyes,[20] and fish gill.[82] c) Schematic images of plant leaf,[77] *Electrophorus electricus*,[68] and honeycomb.[67] d) Photonic applications of incidence angle,[64] incidence wavelength,[49] and iridescence by magnetic actuation.[59] e) Wettable applications of spilled oil collection,[82] superhydrophobic surface,[81] anti-fogging coating and contact angle.[20] f) Energy applications of voltage-driven cell,[72] photocatalysis,[29] solar cell,[66] optoelectronics,[34] and rechargeable battery.[35] a) Neon tera fish: Reproduced with permission.[59] Copyright 2019, American Chemical Society. Seashell: Reproduced with permission.[64] Copyright 2019, Wiley-VCH. *C. rajah* beetle: Reproduced with permission.[49] Copyright 2015, Royal Society of Chemistry. b) *Arowana* fish: Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/).[81] Copyright 2015, The Authors; published by Springer Nature. Fly compound eye: Reproduced with permission.[20] Copyright 2014, Wiley-VCH. Fish gill: Reproduced with permission.[82] Copyright 2017, American Chemical Society. c) Plant leaf: Reproduced with permission.[77] Copyright 2014, Nature Publishing Group. *Electrophorus electricus* Reproduced with permission.[68] Copyright 2017, Nature Publishing Group. Honeycomb: Reproduced with permission.[67] Copyright 2019, Wiley-VCH. d) Incidence angle: Reproduced with permission.[64] Copyright 2019, Wiley-VCH. Incidence wavelength: Reproduced with permission.[49] Copyright 2015, Royal Society of Chemistry. Magnetic actuation: Reproduced with permission.[59] Copyright 2019, American Chemical Society. e) Spilled oil collection: Reproduced with permission.[82] Copyright 2017, American Chemical Society. Superhydrophobic surface: Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/).[81] Copyright 2015, The Authors; published by Springer Nature. Anti-fogging coating and contact angle: Reproduced with permission.[20] Copyright 2014, Wiley-VCH. f) Voltage-driven cell: Reproduced with permission.[72] Copyright 2017, Wiley-VCH. Photocatalysis: Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/).[29] Copyright 2016, The Authors, published by Springer Nature. Solar cell: Reproduced under the terms of the CC-BY 4.0 Creative Commons Attribution 4.0 International License (https://creativecommons.org/licenses/by/4.0/).[66] Copyright 2017, The Authors, published by Springer Nature. Optoelectronics: Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/).[34] Copyright 2018, The Authors, published by Springer Nature. Rechargeable battery: Reproduced with permission.[35] Copyright 2017, American Chemical Society.

interference, when the optical thickness (nd) of a certain pair of AB layers meets the conditions of Equation (1)[40]

$$2\left(n_{\mathrm{A}}d_{\mathrm{A}}\cos\theta_{\mathrm{A}}+n_{\mathrm{B}}d_{\mathrm{B}}\cos\theta_{\mathrm{B}}\right)=m\lambda \tag{1}$$

where n is the refractive index, d is the physical thickness of each layer, θ is the angle of propagation in the corresponding layer, and λ is the wavelength of light giving the highest reflectivity, and m is an integer. Because of this interesting optical property, organisms with Bragg Stacks on their surfaces present a wide range of brilliant colors, creating a vivid world. Inspired by nature 2D photonic structures, many delicately designed 2D photonic materials with fluctuating structural colors have been developed with applications across broad areas.[41–45] Compared with pigment color, structural color can offer ultrahigh saturation, brightness, and vivid

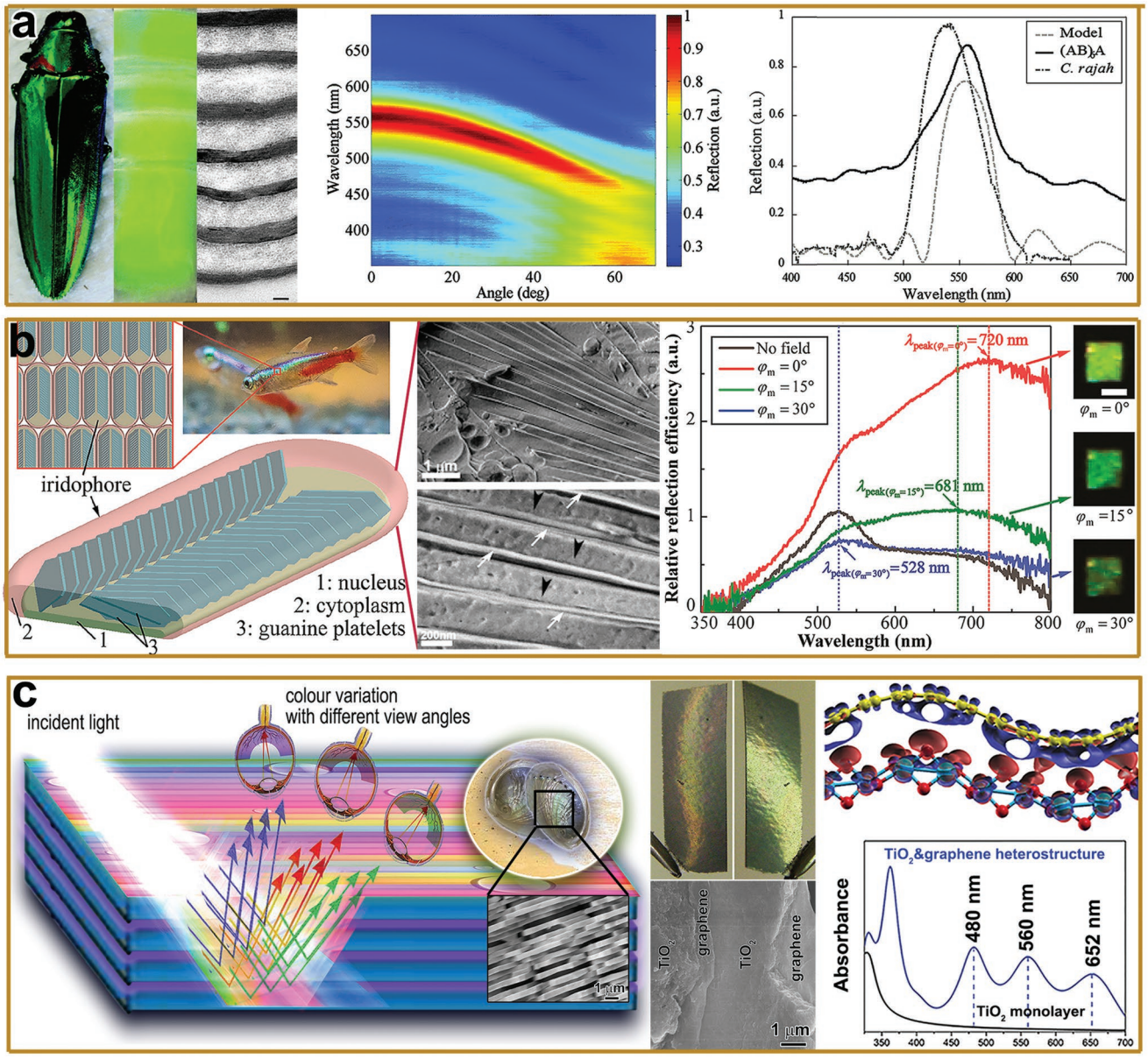

Figure 2. Bioinspired 2D photonic nanomaterials. a) The optical photographs and SEM image show the *C. rajah* beetle; the assembled artificial Bragg stacks with green color and the corresponding microstructure; the color plots show the reflection-incidence angle spectrum for the artificial stacks; the right figure shows the reflection spectra of *C. rajah* elytron and the fabricated stack. Reproduced with permission.[49] Copyright 2015, Royal Society of Chemistry. b) Schematic illustration of neon tetra and its photonic structures; the iridescence demonstration indicates color variation from bright yellow to dark green with different magnetic alignment angles φ_{m} = 0–30°. Reproduced with permission.[59] Copyright 2019, American Chemical Society. The SEM images show the microstructures of the guanine crystals (with arrows) and cytoplasm layers (black arrows). Reproduced with permission.[58] Copyright 2015, Wiley-VCH. c) Schematic illustration shows the angle-varying structural change of seashell structures; the inset shows the microstructure of natural seashell; the optical and SEM images show the seashell-inspired nanostructure made from graphene and 2D-TiO_2 nanosheets; the right part shows the interlayer electronic coupling and calculated light absorption spectrum of bioinspired heterostructure. Reproduced with permission.[64] Copyright 2019, Wiley-VCH.

iridescence, or the change of color with angle of incidence of the light.

One well-known case is the brilliant iridescent colors found in the elytra of some beetles. For example, buprestid beetles have beautiful green iridescence on their elytra (wing cases), which originates from multiple stacks of periodically ordered chitin-protein pairs.[46] The multilayers of chitin–protein pairs in beetles consist of a few thin layers of chitin in a proteinaceous matrix with different refractive indexes. When the optical thickness of the chitin layers genetically grow to approach one-quarter the wavelength of visible light (Equation (1)), one or more colors will be generated by constructive interference. Inspired by the attractive structural colors of beetle elytra, nanoparticles which change color with environment have been realized through a zinc oxide (ZnO) flower-like structure using quasi-ordered scattering.[47] Similarly, an optical diode design was inspired by *Plusiotis batesi* beetles, which have a unique helix structure in their elytra for a conventional selective reflection.[48] Tzeng et al. fabricated a synthetic analogue with bioinspired iridescent colors by mimicking the color appearance of a *Chrysochroa rajah* beetle.[49] As shown in **Figure 2**a, the artificial Bragg Stack of optically reflective pairs with alternating low and high refractive indices were made from colloidal silica/cellulose nanocrystals and polyelectrolyte/clay thin layers assembled by a layer-by-layer deposition method, and the bioinspired thin film showed similar iridescence as the beetle elytra.

By learning from the Bragg Stacks and the iridescent color in biological species, some other artificial materials with well controlled thickness and refractive indices have been fabricated to realize structural colors, such as the spin-coated mesoporous titanium dioxide/silicon dioxide (TiO_2/SiO_2) multilayer stacks,[50,51] layer-by-layer assembled TiO_2/SiO_2 nanoparticles,[52] thermally evaporated thin films of calcium fluoride (CaF_2) and zinc sulfide (ZnS),[53] polymers,[54] and so on, for the applications in sensors, smart windows, and soft robotic devices.[55] Besides the direct iridescent colors generated by reflection and interference, enhanced fluorescence emission properties have also been inspired by a *Hoplia coerulea* beetle, whose scales contain fluorescent molecules ingrained in 2D multilayers of thin flat chitin layers separated by air/chitin mixing layers.[56]

2D photonic structures based on guanine crystals have been observed in some hydrobios.[57] As shown in Figure 2b, tunable multilayer photonic structures in neon tetra fish can generate iridescent colors from blue to indigo with the change of light environment by tuning the tilt angle between guanine crystals and cytoplasm layers of the 2D guanine–cytoplasm pairs.[58] Based on this understanding, a dynamic iridescence actuated by magnetic nanopillars has been fabricated, which allows the structure to tilt the substrate when the angle of the applied magnetic field is changed and this can lead to programmable iridescent display under ambient light.[59] This type of guanine-based photonic crystals has also been identified in different types of fishes, such as Japanese *Koi* fish and *Sardina pilchardus* fish. These fish are covered by a large area of guanine photonic crystal stacks with numerous reflecting units. These stacks generate a broadband, wavelength-independent reflector over the whole visible light spectrum when the incident angle approaches Brewster's angle.[60] In contrast to the above broadband reflectance, *sapphirinid* copepods generate wavelength-independent reflectance in response to the luminous environment by controlling the thickness of the cytoplasm layers of the guanine–cytoplasm photonic crystal pairs.[61] Such incredibly tunable 2D photonic systems in *sapphirinid* copepods are rarely observed even in nature, and they have not yet been successfully mimicked in artificial systems. The major challenges in fabricating such a class of bioinspired photonic structures are finding suitable 2D nanostructures and materials to replace the guanine crystals and cytoplasm layers and realizing the precisely controllable layer-by-layer assembly of the 2D crystals and layers. Although some pioneering studies have been performed on the guanine-based bioinspired materials, for example, through the self-assembly of peptides or DNA to realize unique photonic performance,[62,63] there is still a long way to go in developing this type of bioinspired material.

Very recently, Sun et al. fabricated a sea-shell inspired 2D photonic nanostructure by using a facile vacuum-filtering technique to assemble graphene and atomically thin 2D TiO_2 nanosheets in a layer-by-layer manner to mimic the iridescent, well-arranged, layered brick-and-mortar microstructures of natural sea-shells (Figure 2c).[64] The bioinspired photonic structure with 16 alternating thin graphene layers and thick 2D TiO_2 layers exhibited significant and beautiful green-red strip-like colors under both dispersion-dominated and reflection-dominated conditions, resulting from its well-designed thickness and well-aligned 2D interfaces. When this type of unique bioinspired nanostructure is employed in optoelectronic devices, significantly enhanced photocurrent and much improved responsivity and sensitivity have been achieved, attributed to the significantly enhanced interlayer charge transfer formed between the 2D–2D heterostructure and the homostructure of the bioinspired photoelectrode. Kolle and co-workers have fabricated a concave 2D photonic structure consisting of 2.5 mm thick concave gold layer and 20 nm thick carbon film with a conformal multilayer of thin titanium dioxide/aluminum oxide (TiO_2/Al_2O_3) films to simulate the color mixing effect resulting from deformed multilayer stacks in the wings of *Papilio blumei* butterfly. This proved that bioinspired nanostructures could display the same optical features as the natural butterfly wing scale.[43] Furthermore, introducing stimuli-responsive materials into photonic structures can achieve color shifting with the change of environments. For example, Wang et al. fabricated bioinspired hybrid photonic crystals consisting of alternant thin films of TiO_2 and organic polymer layers, which have reversible color shifting capacity over the full visible spectrum in response to different water–vapor concentrations as a result of swelling of the polymer layers.[65]

Therefore, based on the understanding on the structure–optical property relationships found in natural species, plenty of 2D bioinspired photonic structures have been developed and fabricated. These bioinspired 2D nanostructures presented mimicking extraordinary colors exhibited in natural species and are very promising in applications in optical displays, sensors, anticounterfeit technologies, solar energy harvesting and conversion devices, etc.

3. Bioinspired 2D Energy Materials

Development of high-efficiency energy conversion and storage devices has attracted great research interest, due to the urgent

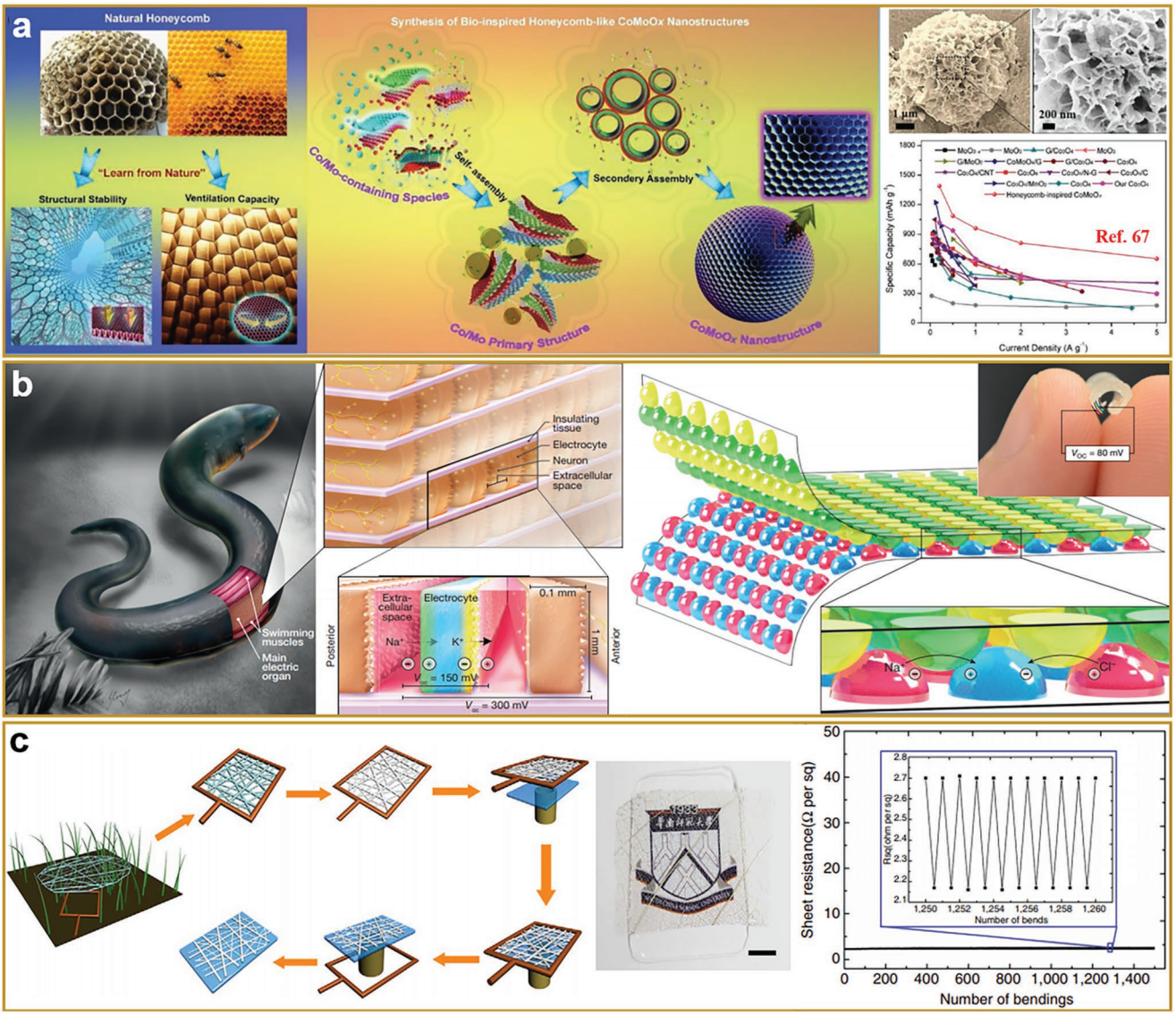

Figure 3. Bioinspired 2D energy nanomaterials. a) The photographs show a natural honeycomb and the inspired real-world construction with excellent structural stability and ventilation capacity; the schematic illustration shows the self-assembly route of honeycomb-inspired $CoMoO_x$ nanostructure, SEM images show the bioinspired $CoMoO_x$ microspheres composed of 2D nanosheets, and the comparison of rate capability and the capacity retention ratio of honeycomb-inspired $CoMoO_x$ nanostructure in a current density range between 0.2 and 5.0 A g^{-1} was summarized. Reproduced with permission.[67] Copyright 2019, Wiley-VCH. b) The photographs show the *Electrophorus electricus* and the electrocytes in its organs; the right picture shows the artificial electric organ in its printed implementation and the inset shows the flexible artificial organ that produced an open-circuit voltage of 80 mV. Reproduced with permission.[68] Copyright 2017, Nature Publishing Group. c) The schematic demonstrates the fabrication process of the silk spider web, the photograph shows the transparent leaf venation network, and the diagram illustrates the sheet network resistance oscillations. Reproduced with permission.[77] Copyright 2014, Nature Publishing Group.

demand for green and sustainable energy supplies. To meet the increasing requirements on materials for high-performance energy devices, nanomaterials with precisely tailored structures and interfaces inspired from natural species have been created.[66] Bioinspired nanomaterials have demonstrated some extraordinary properties, which are promising for enhancing the overall performance of energy conversion or storage devices.

Mei et al. proposed a honeycomb-inspired $CoMoO_x$ structures consisting of 2D nanosheets by learning from the robust mechanical stability and permeable channels and pores structures of natural honeycombs, which possess excellent structural robustness deriving from the elaborate structures.[67] The natural honeycomb or bee hive with cellular structures, composed of hexagonal channels and thin walls, as shown in **Figure 3**a, is well known for its excellent mechanical performances, highly ordered cellular structures, and excellent ventilation capacity arising from the interconnected pore/channel structure. In this work, owing to the structural advantages derived from the natural honeycombs, the bioinspired $CoMoO_x$ nanostructures consisting of interconnected pores and channels and ultrathin 2D walls exhibited much improved structural cycling stability and superior lithium storage capacity compared to electrode

materials with similar chemical compositions, but without the cellular nanostructure. This work confirms that bioinspiration offers new insights into the design of novel electrode materials for high-efficiency energy devices.

The electric eel *Electrophorus electricus* is an amazing aquatic animal, which can generate high voltages and currents from ion gradients through thousands of membranes with densely packed ion channels which separate long and thin electrically active cells stacked in parallel. Inspired by this power generation concept, a soft, flexible, and transparent power source composed with polyacrylamide hydrogel and a repeated sequence of ion selective hydrogel membranes, similar to the repeated 2D compartments of electric eel, has been designed. This has been shown to generate an open-circuit voltage of 110 V or a power density of 27 mW m^{-2} per cell under mechanical contact activation of serially stacked gel compartments.[68] This bioinspired power system indeed opens a new window to design sustainable electrical generators. In addition to the ion-gradient power generator inspired by electric eel, novel triboelectric generators based on 2D materials have also been inspired by human skin,[69] Hummingbird wing,[70] and other natural structures.

It is interesting that nacre has also inspired the integration of 2D nanofluidic devices, where the mass and charge transport are confined in the interspace between 2D layers. Cheng et al. reported a bioinspired 2D nanofluidic devices constructed from kaolinite-based Janus nanobuilding blocks. 2D Janus materials as a novel class of materials with distinct properties on each side have been considered as a promising material system in energy applications.[71] Unlike conventional 2D nanofluidics, the use of 2D Janus blocks in a nacre-mimicking assembly, delivers outstanding carrier transport mobility and almost perfect ion selectivity.[72]

The biological chain and the biocycle on the Earth all start from the energy supply from the sun, which have driven plenty of biological species to be extremely active for high-efficient solar energy harvesting or conversion. Recently, bioinspired solar energy harvesting and conversion technologies, such as bioinspired solar cells[73,74] and bioinspired carbon cycle processes,[75,76] have achieved significant progress.

By learning from the 2D networks of leaf venation system of White Jade Orchid Tree Magnolia alba and spider web networks of a common spider *Agelena labyrinthica*, metallic networks featuring superior current delivery, minimal optical shading, and outstanding strength and flexibility have been designed for electro-optical devices, such as solar cells, light sources, touch screens, or flexible displays.[77] By directly mimicking the shape of a leaf, a bioinspired flexible $Zn_{0.5}Cd_{0.5}S$@polyacrylonitrile photocatalyst in 2D leaf-like structure has been fabricated. It shows enhanced photocatalytic hydrogen evolution activity compared to the pure $Zn_{0.5}Cd_{0.5}S$ nanoparticles with improved electrochemical cycling and structural stability in the photocatalytic hydrogen production processes, owing to the flat and flexible leaf-like structure and the superior light efficiency engendered by the hierarchical pore channels.[78] Also inspired by a leaf, 2D AgS_2 nanosheets with a leaf-like morphology with needle-like secondary structures have shown promise in solar cell applications.[79] Targeting a very different component of solar cells, Shams et al. extracted a transparent crab shell-inspired chitin nanofiber sheet by removing the inorganic calcium carbonate particles, proteins, lipids, and pigments from a real crab shell, and then impregnated a monomer to the sheet, followed by polymerization. This material has good transparency and very low coefficient of thermal expansion and is an interesting candidate as a transparent substrate materials for flexible displays and solar cells.[80]

4. Bioinspired 2D Superwetting Materials

Bioinspired surfaces with superwettability have been intensively explored in past decades by investigating the superwetting phenomenon in nature and this has stimulated the applications of superwettability in liquid-proof textiles, oil–water separations, super/antiadhesion surfaces, etc.[3] For example, water birds, both fresh water and salt water birds, such as the duck, goose, sea gull, pelican, etc., have superhydrophobic feathers to prevent wetting by water when they feed on lakes, rivers, beaches, etc.[17] Some birds, such as the cormorant, can even dive tens of meters deep into water to prey on fish while their feather remain unwetted by maintaining a thin layer of air on the surface of the feather. This superhydrophobic feature is due to both a 2D configuration of "quasi-hierarchical" microstructures and a specific greasy surface coating or preening oil secreted by the bird itself. Fish scales also exhibit self-cleaning and antifouling properties under water. Fish scale is an important organ as its role in protecting fish from contamination in the water environment.[17] By mimicking the natural 2D scale-like structure of the Asian *Arowana* fish scales, Sun et al. developed bioinspired nanostructured coatings with tunable wettability in response to surface modification and scale-like structural orientation variation (**Figure 4**a).[81] In this study, the bioinspired materials were fabricated in ZnO and the orientation of the scale-like nanostructures has been tailored to present different responses to water contact. Thin films with highly tilted scale structures have the capability to be tuned to exhibit either superhydrophilic or superhydrophobic. This interesting feature gives the bioinspired materials the potential for many applications, such as marine antifouling, self-cleaning material, microfluidic regulation, bio-adhesion, etc.

Another example for natural superwettability is the compound eye of flies. The unique surface structure gives the compound eye a superhydrophobic antifogging property, which allows them to maintain good functionality even in extreme environments. By learning from the biological structures and antifogging properties of the fly eye, Sun et al. designed bioinspired superhydrophobic antifogging ZnO nanomaterials with a low adhesion force to water drops, which effectively prevent the adhesion of water droplets on a tilted or curved surface.[20] Figure 4b shows artificial hierarchically ordered ZnO microspheres that have a similar structure to the anatomical structure of the compound eye. Bioinspired ZnO coatings can be easily fabricated by either brush-painting or spin coating of the fly-eye-inspired nanomaterials. As shown in the fogging test in Figure 4b, the fogging droplets on the bioinspired coatings were in perfect spherical shapes due to the high contact angle and fully slid off a tilted substrate due to the low adhesive force. This bioinspired antifogging coating material is fabricated from low-cost material and is easily adapted for scalable production and large-scale painting, and thus has great potential

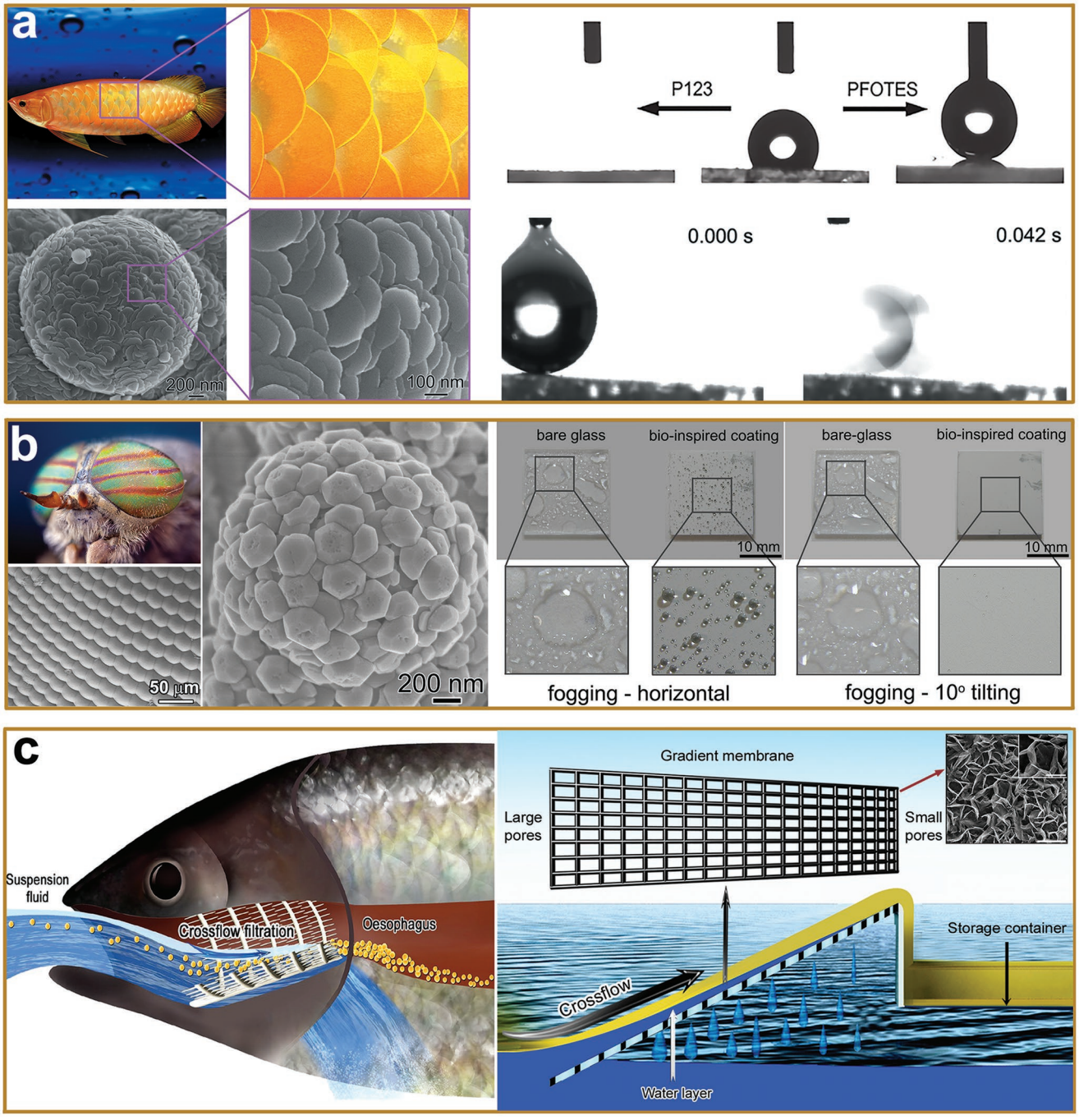

Figure 4. Bioinspired 2D superwetting nanomaterials. a) Optical and SEM images of Asian Arowana fish scales, and the surface wettability modulation of the fish-scale-inspired nanostructured coatings showing the hydrophobic–superhydrophilic–superhydrophobic modulation of the water response. Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/).[81] Copyright 2015, The Authors, published by Springer Nature. b) Optical and SEM images of fly compound eyes, the microstructure of the fly-eye bioinspired ZnO nanostructures shows the ommatidium lens-like structures; the right picture demonstrates the fogging test of the bare glass and bioinspired coatings. Reproduced with permission.[20] Copyright 2014, Wiley-VCH. c) Schematic illustration of crossflow filtration in fish gills and the demonstration of the bioinspired crossflow collection of spilled oil; the inset shows the ultrathin 2D-Co_3O_4 nanosheets coated on the wire surface. Reproduced with permission.[82] Copyright 2017, American Chemical Society.

for anti-fogging or anti-icing applications in some extreme environments.

Oil/water separation is another important application of bioinspired superwetting nanomaterials. It has been proved that the utilization of functional surfaces with both superoleophilic and superhydrophobic properties is an effective way to separate an oil-water mixture.[3] Dou et al. demonstrated a gradient porous separation mesh for one-step spilled oil collection

and separation inspired by the crossflow filtration behavior of fish gills (Figure 4c).[82] The key innovation of this technique is the growth 2D ultrathin cobalt oxide (Co_3O_4) nanosheets on metallic meshes, which turn the separation mesh from hydrophobic/oleophilic to hydrophilic/oleophobic. By using the innovative fish-gill-inspired separation membrane, crossflows formed on the membranes, in which the oil/water flows parallel to the bioinspired 2D-Co_3O_4 membrane surface, and water is gradually filtered through the membrane, while oil is repelled and transported on the surface of membrane to the collection tank. Via this unique separation mechanism, the fish-gill-inspired technology enables high-efficient and continuous spilled oil collection and separation. This fish-gill-inspired technology is therefore very promising for the clean-up of large-scale oil spills (Figure 4c). A shape-memory graphene film with electrothermally controlled surface wettability inspired by natural *Nepenthes* pitcher plant has been proposed by Wang et al. This bioinspired thin film exhibited good electrical conductivity, superelasticity, and high strength. The bioinspired material possesses controllable slippery properties due to the reversible surface wettability, and thus can be used in various applications, including liquid harvesting devices, microfluidic channels, medical instruments, liquid handling robotic systems, etc.[83]

5. Conclusions and Outlook

In summary, the well-evolved biological world, with its biodiversity and fascinating environmental responsive behavior is a supremely effective material-development laboratory to inspire our material and technology innovations. When science meets nature, a magic door opens to stimulate us to develop novel multifunctional nanomaterials beyond the present material systems. The combination of 2D nanomaterials with the concept of bioinspiration, as we demonstrated above, has stimulated the development of novel materials and technologies.

On the other hand, there are still lots of grand challenges to be addressed, which limit the development of bioinspired nanomaterials or technologies beyond 2D bioinspired materials. First, we need further efforts on discovering the phenomena and functionalities existing in natural species and gaining a better understanding of their structure–function relationships, which are fundamental but challenging two steps. Second, sophisticated synthetic methods or high-precise manufacturing techniques to achieve multiscale-ordering structures down to nanoscale are urgently needed to mimic complex natural structures or functions. In fact, some fascinating structures and properties found in nature cannot be properly mimicked in our laboratories. For example, Gur et al. have revealed the unique tunable optical properties in the male sapphirinid copepods, which is very promising for advanced optical or light-harvesting devices.[61] However, due to the lack of proper material and fabrication method, such novel structures and properties have not been successfully mimicked yet. Third, given that some biological systems present interesting properties themselves, the direct use of such biomaterials in functional devices would be an eco-friendly and efficient way to develop bioinspired technologies. Recently, researchers discovered that biological materials directly derived from natural species exhibited outstanding performances in various energy applications such as photocatalysis, electrocatalysis, and biomass conversion.[84–87] However, the precise and advanced biotechnology required in this route turns to be another major challenge. The last challenge is how to integrate multiple materials to realize desirable functions, such as well-designed heterostructures, organic-inorganic hybrid materials, etc. Apart from these challenges, the advances in developing 2D bioinspired nanomaterials confirm that bioinspiration is an effective approach to take the full advantages of the potential of materials for the design of mutifunctional smart devices. The development in this promising area thus paves a new way toward a green and sustainable society.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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[1] A. R. Parker, C. R. Lawrence, *Nature* **2001**, *414*, 33.
[2] L. Wu, J. He, W. Shang, T. Deng, J. Gu, H. Su, Q. Liu, W. Zhang, D. Zhang, *Adv. Opt. Mater.* **2016**, *4*, 195.
[3] S. Wang, K. Liu, X. Yao, L. Jiang, *Chem. Rev.* **2015**, *115*, 8230.
[4] J. Aizenberg, P. Fratzl, *Adv. Mater.* **2009**, *21*, 387.
[5] X. Jin, B. Shi, L. Zheng, X. Pei, X. Zhang, Z. Sun, Y. Du, J. H. Kim, X. Wang, S. Dou, K. Liu, L. Jiang, *Adv. Funct. Mater.* **2014**, *24*, 2721.
[6] P. Westbroek, F. Marin, *Nature* **1998**, *392*, 861.
[7] J. H. Cartwrighta, A. G. Checa, B. Escribanoa,, C. I. Sainz-Díaz, *Proc. Natl. Acad. Sci. USA* **2009**, *106*, 10499.
[8] F. Barthelat, Z. Yin, M. J. Buehler, *Nat. Rev. Mater.* **2016**, *1*, 16007.
[9] W. R. Hansen, K. Autumn, *Proc. Natl. Acad. Sci. USA* **2005**, *102*, 385.
[10] Y. Zheng, H. Bai, Z. Huang, X. Tian, F. Q. Nie, Y. Zhao, J. Zhai, L. Jiang, *Nature* **2010**, *463*, 640.
[11] Y. Zheng, X. Gao, L. Jiang, *Soft Matter* **2007**, *3*, 178.
[12] S. Kinoshita, S. Yoshioka, K. Kawagoe, *Proc. R. Soc. London, Ser. B* **2002**, *269*, 1417.
[13] U. G. Wegst, H. Bai, E. Saiz, A. P. Tomsia, R. O. Ritchie, *Nat. Mater.* **2015**, *14*, 23.
[14] A. Finnemore, P. Cunha, T. Shean, S. Vignolini, S. Guldin, M. Oyen, U. Steiner, *Nat. Commun.* **2012**, *3*, 966.
[15] B. J. Bruet, J. Song, M. C. Boyce, C. Ortiz, *Nat. Mater.* **2008**, *7*, 748.
[16] A. H. Barber, D. Lu, N. M. Pugno, *Nature* **2015**, *519*, S14.
[17] M. Liu, S. Wang, L. Jiang, *Nat. Rev. Mater.* **2017**, *2*, 17036.
[18] H. Xie, H. X. Huang, Y. J. Peng, *Nanoscale* **2017**, *9*, 11951.
[19] K. Liu, L. Jiang, *Annu. Rev. Mater. Res.* **2012**, *42*, 231.
[20] Z. Sun, T. Liao, K. Liu, L. Jiang, J. H. Kim, S. X. Dou, *Small* **2014**, *10*, 3001.

[21] Z. Han, X. Feng, Z. Guo, S. Niu, L. Ren, *Adv. Mater.* **2018**, *30*, 1704652.
[22] K. Liu, J. Du, J. Wu, L. Jiang, *Nanoscale* **2012**, *4*, 768.
[23] X. Jing, H. Y. Mi, Y. J. Lin, E. Enriquez, X. F. Peng, L. S. Turng, *ACS Appl. Mater. Interfaces* **2018**, *10*, 20897.
[24] Z. Yu, F. F. Yun, Y. Wang, L. Yao, S. Dou, K. Liu, L. Jiang, X. Wang, *Small* **2017**, *13*, 1701403.
[25] M. Cao, J. Ju, K. Li, S. Dou, K. Liu, L. Jiang, *Adv. Funct. Mater.* **2014**, *24*, 3235.
[26] R. A. Potyrailo, R. K. Bonam, J. G. Hartley, T. A. Starkey, P. Vukusic, M. Vasudev, T. Bunning, R. R. Naik, Z. Tang, M. A. Palacios, M. Larsen, L. A. Le Tarte, J. C. Grande, S. Zhong, T. Deng, *Nat. Commun.* **2015**, *6*, 7959.
[27] H. Zhou, J. Xu, X. Liu, H. Zhang, D. Wang, Z. Chen, D. Zhang, T. Fan, *Adv. Funct. Mater.* **2018**, *28*, 1705309.
[28] H. Kang, J. S. Lee, W. S. Chang, S. H. Kim, *Adv. Mater.* **2015**, *27*, 1282.
[29] R. Yan, M. Chen, H. Zhou, T. Liu, X. Tang, K. Zhang, H. Zhu, J. Ye, D. Zhang, T. Fan, *Sci. Rep.* **2016**, *6*, 20001.
[30] H. Chen, T. Ran, Y. Gan, J. Zhou, Y. Zhang, L. Zhang, D. Zhang, L. Jiang, *Nat. Mater.* **2018**, *17*, 935.
[31] K. Li, J. Ju, Z. Xue, J. Ma, L. Feng, S. Gao, L. Jiang, *Nat. Commun.* **2013**, *4*, 2276.
[32] N. Zhao, Z. Wang, C. Cai, H. Shen, F. Liang, D. Wang, C. Wang, T. Zhu, J. Guo, Y. Wang, X. Liu, C. Duan, H. Wang, Y. Mao, X. Jia, H. Dong, X. Zhang, J. Xu, *Adv. Mater.* **2014**, *26*, 6994.
[33] Z. Sun, T. Liao, Y. Dou, S. M. Hwang, M. S. Park, L. Jiang, J. H. Kim, S. X. Dou, *Nat. Commun.* **2014**, *5*, 3813.
[34] D. Xiang, T. Liu, J. Xu, J. Y. Tan, Z. Hu, B. Lei, Y. Zheng, J. Wu, A. H. C. Neto, L. Liu, W. Chen, *Nat. Commun.* **2018**, *9*, 2966.
[35] F. Zhang, J. Zhu, D. Zhang, U. Schwingenschlogl, H. N. Alshareef, *Nano Lett.* **2017**, *17*, 1302.
[36] P. Vukusic, J. R. Sambles, *Nature* **2003**, *424*, 852.
[37] P. Vukusic, J. R. Sambles, C. R. Lawrence, *Nature* **2000**, *404*, 457.
[38] J. Teyssier, S. V. Saenko, D. van der Marel, M. C. Milinkovitch, *Nat. Commun.* **2015**, *6*, 6368.
[39] Y. Zhao, Z. Xie, H. Gu, C. Zhu, Z. Gu, *Chem. Soc. Rev.* **2012**, *41*, 3297.
[40] S. Kinoshita, S. Yoshioka, J. Miyazaki, *Rep. Prog. Phys.* **2008**, *71*, 076401.
[41] C. Park, K. Koh, U. Jeong, *Sci. Rep.* **2015**, *5*, 8340.
[42] D. Wu, J. N. Wang, L. G. Niu, X. L. Zhang, S. Z. Wu, Q. D. Chen, L. P. Lee, H. B. Sun, *Adv. Opt. Mater.* **2014**, *2*, 751.
[43] M. Kolle, P. M. Salgard-Cunha, M. R. Scherer, F. Huang, P. Vukusic, S. Mahajan, J. J. Baumberg, U. Steiner, *Nat. Nanotechnol.* **2010**, *5*, 511.
[44] Z. Sun, T. Liao, L. Sheng, J. H. Kim, S. X. Dou, J. Bell, *Mater. Today Chem.* **2016**, *1-2*, 84.
[45] E. Fresta, V. Fernández-Luna, P. B. Coto, R. D. Costa, *Adv. Funct. Mater.* **2018**, *28*, 1707011.
[46] A. E. Seago, P. Brady, J. P. Vigneron, T. D. Schultz, *J. R. Soc., Interface* **2009**, *6*, 165.
[47] G. H. Kim, T. An, G. Lim, *ACS Appl. Mater. Interfaces* **2017**, *9*, 19057.
[48] J. Hwang, M. H. Song, B. Park, S. Nishimura, T. Toyooka, J. W. Wu, Y. Takanishi, K. Ishikawa, H. Takezoe, *Nat. Mater.* **2005**, *4*, 383.
[49] P. Tzeng, D. J. Hewson, P. Vukusic, S. J. Eichhorn, J. C. Grunlan, *J. Mater. Chem. C* **2015**, *3*, 4260.
[50] S. Y. Choi, M. Mamak, G. von Freymann, N. Chopra, G. A. Ozin, *Nano Lett.* **2006**, *6*, 2456.
[51] B. Auguié, M. C. Fuertes, P. C. Angelomé, N. L. Abdala, G. S. Illia, A. Fainstein, *ACS Photonics* **2014**, *1*, 775.
[52] G. M. Nogueira, D. Banerjee, R. E. Cohen, M. F. Rubner, *Langmuir* **2011**, *27*, 7860.
[53] M. Muallem, A. Palatnik, G. D. Nessim, Y. R. Tischler, *ACS Appl. Mater. Interfaces* **2015**, *7*, 474.
[54] P. Lova, C. Bastianini, P. Giusto, M. Patrini, P. Rizzo, G. Guerra, M. Iodice, C. Soci, D. Comoretto, *ACS Appl. Mater. Interfaces* **2016**, *8*, 31941.
[55] F. Fu, L. Shang, Z. Chen, Y. Yu, Y. Zhao, *Sci. Rob.* **2018**, *3*, eaar8580.
[56] E. V. Hooijdonk, S. Berthier, J.-P. Vigneron, *J. Appl. Phys.* **2012**, *112*, 114702.
[57] A. Levy-Lior, E. Shimoni, O. Schwartz, E. Gavish-Regev, D. Oron, G. Oxford, S. Weiner, L. Addadi, *Adv. Funct. Mater.* **2010**, *20*, 320.
[58] D. Gur, B. A. Palmer, B. Leshem, D. Oron, P. Fratzl, S. Weiner, L. Addadi, *Angew. Chem., Int. Ed.* **2015**, *54*, 12426.
[59] Z. Luo, B. A. Evans, C. H. Chang, *ACS Nano* **2019**, *13*, 4657.
[60] T. M. Jordan, J. C. Partridge, N. W. Roberts, *Nat. Photonics* **2012**, *6*, 759.
[61] D. Gur, B. Leshem, V. Farstey, D. Oron, L. Addadi, S. Weiner, *Adv. Funct. Mater.* **2016**, *26*, 1393.
[62] O. Berger, L. Adler-Abramovich, M. Levy-Sakin, A. Grunwald, Y. Liebes-Peer, M. Bachar, L. Buzhansky, E. Mossou, V. T. Forsyth, T. Schwartz, Y. Ebenstein, F. Frolow, L. J. Shimon, F. Patolsky, E. Gazit, *Nat. Nanotechnol.* **2015**, *10*, 353.
[63] O. Berger, E. Yoskovitz, L. Adler-Abramovich, E. Gazit, *Adv. Mater.* **2016**, *28*, 2195.
[64] Z. Sun, T. Liao, W Li, Y Qiao, K. Ostrikov, *Adv. Funct. Mater.* **2019**, *29*, 1901460.
[65] Z. Wang, J. Zhang, J. Xie, C. Li, Y. Li, S. Liang, Z. Tian, T. Wang, H. Zhang, H. Li, W. Xu, B. Yang, *Adv. Funct. Mater.* **2010**, *20*, 3784.
[66] L. V. Thekkekara, M. Gu, *Sci. Rep.* **2017**, *7*, 45585.
[67] J. Mei, T. Liao, H. Spratt, G. A. Ayoko, X. S. Zhao, Z. Sun, *Small Methods* **2019**, *3*, 1900055.
[68] T. B. H. Schroeder, A. Guha, A. Lamoureux, G. VanRenterghem, D. Sept, M. Shtein, J. Yang, M. Mayer, *Nature* **2017**, *552*, 214.
[69] X. Wang, Y. Yin, F. Yi, K. Dai, S. Niu, Y. Han, Y. Zhang, Z. You, *Nano Energy* **2017**, *39*, 429.
[70] A. Ahmed, I. Hassan, P. Song, M. Gamaleldin, A. Radhi, N. Panwar, S. C. Tjin, A. Y. Desoky, D. Sinton, K. T. Yong, J. Zu, *Sci. Rep.* **2017**, *7*, 17143.
[71] Z. Wu, L. Li, T. Liao, X. Chen, W. Jiang, W. Luo, J. Yang, Z. Sun, *Nano Today* **2018**, *22*, 62.
[72] H. Cheng, Y. Zhou, Y. Feng, W. Geng, Q. Liu, W. Guo, L. Jiang, *Adv. Mater.* **2017**, *29*, 1700177.
[73] R. H. Siddique, Y. J. Donie, G. Gomard, S. Yalamanchili, T. Merdzhanova, U. Lemmer, H. Hölscher, *Sci. Adv.* **2017**, *3*, e1700232.
[74] R. Senthil, S. Yuvaraj, *Int. J. Energy Res.* **2019**, *43*, 1068.
[75] K. K. Sakimoto, N. Kornienko, P. Yang, *Acc. Chem. Res.* **2017**, *50*, 476.
[76] D. Kim, K. K. Sakimoto, D. Hong, P. Yang, *Angew. Chem., Int. Ed.* **2015**, *54*, 3259.
[77] B. Han, Y. Huang, R. Li, Q. Peng, J. Luo, K. Pei, A. Herczynski, K. Kempa, Z. Ren, J. Gao, *Nat. Commun.* **2014**, *5*, 5674.
[78] J. Fu, B. Zhu, W. You, M. Jaroniec, J. Yu, *Appl. Catal., B* **2018**, *220*, 148.
[79] I. Martínez-Ruvalcaba, J. F. Hernández-Paz, J. R. Mancilla, P. Ruiz, C. A. Pérez, P. E. García-Casillas, C. A. Rodríguez-González, *J. Alloys Compd.* **2014**, *586*, S526.
[80] M. I. Shams, M. Nogi, L. A. Berglund, H. Yano, *Soft Matter* **2012**, *8*, 1369.
[81] Z. Sun, T. Liao, W. Li, Y. Dou, K. Liu, L. Jiang, S.-W. Kim, J. H. Kim, S. X. Dou, *NPG Asia Mater.* **2015**, *7*, e232.
[82] Y. Dou, D. Tian, Z. Sun, Q. Liu, N. Zhang, J. H. Kim, L. Jiang, S. X. Dou, *ACS Nano* **2017**, *11*, 2477.
[83] J. Wang, L. Sun, M. Zou, W. Gao, C. Liu, L. Shang, Z. Gu, Y. Zhao, *Sci. Adv.* **2017**, *3*, e1700004.
[84] Z. Jiang, H. Sun, T. Wang, B. Wang, W. Wei, H. Li, S. Yuan, T. An, H. Zhao, J. Yu, P. K. Wong, *Energy Environ. Sci.* **2018**, *11*, 2383.
[85] H. Zhang, Y. Wang, D. Wang, Y. Li, X. Liu, P. Liu, H. Yang, T. An, Z. Tang, H. Zhao, *Small* **2014**, *10*, 3371.
[86] S. Tian, Z. Wang, W. Gong, W. Chen, Q. Feng, Q. Xu, C. Chen, C. Chen, Q. Peng, L. Gu, H. Zhao, P. Hu, D. Wang, Y. Li, *J. Am. Chem. Soc.* **2018**, *140*, 11161.
[87] W. Gong, Y. Lin, C. Chen, M. Al-Mamun, H. Lu, G. Wang, H. Zhang, H. Zhao, *Adv. Mater.* **2019**, *31*, 1808341.