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Topical Review

Plant-inspired surfaces and interfaces for sustainable technologies

Binodhya Wijerathne[1,3], **Ting Liao**[2,3,*], **Xudong Jiang**[1], **Juan Zhou**[1,4] **and Ziqi Sun**[1,3,*]

[1] School of Chemistry and Physics, Queensland University of Technology, 2 George Street, Brisbane, QLD 4000, Australia
[2] School of Mechanical Medical and Process Engineering, Queensland University of Technology, 2 George Street, Brisbane, QLD 4000, Australia
[3] Centre for Materials Science, Queensland University of Technology, 2 George Street, Brisbane, QLD 4000, Australia
[4] School of Mechanical Engineering, University of South China, 28 West Changsheng Road, Hengyang, Hunan 421001, People's Republic of China

E-mail: t3.liao@qut.edu.au and ziqi.sun@qut.edu.au

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Abstract
The flora and fauna in nature endow the Earth with a flourishing scene of prosperity with their diverse appearances, colors and patterns, constituting common biopolymers and biominerals. The principles of construction of manifold structures and functionalities from fundamental building blocks in flora and fauna have inspired materials scientists to innovate artificial materials with superior properties and performance. Specifically, florae present numerous minute structures established from elementary blocks of lignin, cellulose, pectin and hemicellulose to induce extraordinary demands to survive in extremely diverse environments on Earth. In this review, we introduce the robust material properties and thought-provoking functionalities of plants, such as super-wettability, liquid/ion transport properties, actuation properties, etc. Then, we summarize the intriguing inspiration in the development of artificial superstructures, self-cleaning surfaces and responsive structures for applications in energy harvesting and generation, electrochemical energy storage, environmental cleaning and remediation, and strong and tough mechanical components. It is expected that some principles of how minute structures and functionalities of plants construct extraordinary capabilities and properties that could be extracted from the current progress and some insight could be offered for future material innovations by learning the best from nature.

Keywords: plant bioinspiration, multifunctional surfaces and interfaces, energy harvesting, conversion and storage, environmental technology, structural strength and resilience

* Authors to whom any correspondence should be addressed.

1. Introduction

Nature, as a flourishing source of inspiration for material innovations, has led to the development of novel materials, which we call 'bioinspired materials', with enhanced structural and functional properties from existing compositions and materials [1, 2]. The term 'bioinspiration' originates from the methodology to draw inspiration from natural structures and functionalities to develop superior artificial materials [3–5]. The targeted natural materials or biological structures are either flora or fauna that provide the basis of bioinspiration to construct artificial materials with mimicking or stimulating properties or functions [6–8]. Some typical biological structures have been explored for bioinspired materials, such as nacres, spider silk, lotus leaf, fly-eyes, butterfly wings, etc [3, 9–15]. Plants appeared around 400 million years ago and have evolved greatly to sustain life during severe eras [16]. Even though their elementary construction blocks are very limited, plants display manifold diversity in both structures and functions, which have inspired the design of superior advanced surfaces and interfaces, such as the *Nepenthes*-inspired directional liquid transport properties [17]. It is also commendable to see that the application of the plant bioinspired materials and structures has dramatically enhanced the performances of energy harvesting, conversion and storage devices, water desalination and decontamination devices, separation and collection devices, structural reinforcement and toughening components, etc [18–23].

With increasing attention being paid to the unique compositions and properties of plants, more and more interesting functions and properties are being discovered. In terms of composition, plants are made up of major organic compounds, such as cellulose, hemicellulose, lignin, and some inorganic components made of nitrogen, phosphorus and potassium [24]. Overall, the organic components play a major role in sustaining life, such as through rapid electrolyte transport and rigidity [25–28], and the inorganic compounds are responsible for some functionalities, such as CO_2 capture and photosynthesis [29]. Both organic and inorganic components synchronize to collate into well-defined tissues and structures to realize extraordinary structural toughness and rigidity, a variety of coloration, rapid humid/temperature/mechanical responsivity, and directionally confined wettability and transport properties. While plenty of structures and mechanisms have been investigated to develop bioinspired multifunctional materials, the discovery of natural structures and the establishment of structure-function relationships are still at the very beginning stage, and extensive effort in this regard is still needed.

In this review, we summarize the recent advancements in the discovery of multifunctional surfaces and interfaces in plants and the corresponding bioinspired structures and materials for sustainable environmental and energy applications. According to the application fields, we categorize the plant-inspired materials into the types of bioinspired materials for energy harvesting and conversion, such as solar energy harvesting and conversion, electricity generation and energy storage; bioinspired materials for sustainable environmental technologies, such as water harvesting and water–oil separation; bioinspired responsivity and actuation, and bioinspired materials for advanced structural applications with enhanced strength and toughness (figure 1). We expect that this review will bridge the knowledge gap between natural plant structures and the manufacturing of advanced artificial materials as well as the real-world demands for superior and efficient novel materials.

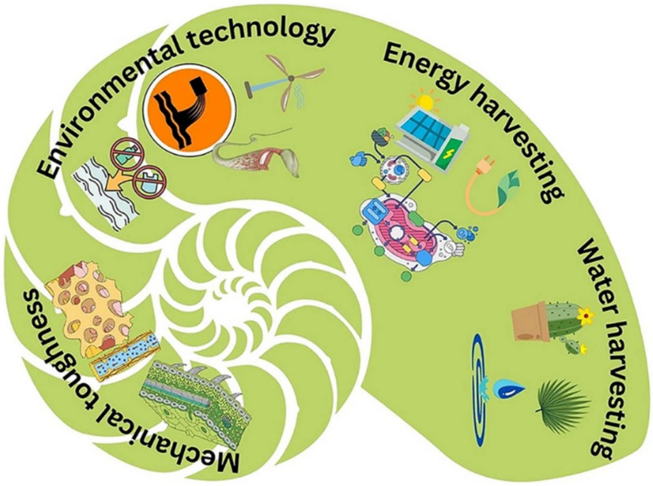

Figure 1. Graphical illustration of plant-inspired surfaces and interfaces for various applications.

2. Composition of plants

As the earliest and largest biological species on Earth, there are about 380 000 known species of plants, ranging from single cells to tall trees. Plants are dominantly photosynthetic, which produce sugars from carbon dioxide and water using the green pigment chlorophyll and generates a substantial proportion of molecular oxygen, feeding most of Earth's ecosystem and other organisms. Plants play vital roles in human life and civilization. People use plants for many purposes, such as building materials, ornaments, writing materials and medicines. Overall, the major components of plant structure are lignin, cellulose, pectin and hemicellulose [24]. It is noteworthy that the remarkable hierarchical structures and characteristics of plants are always available to offer inspiration in developing advanced materials. Due to the biocompatibility, chemical stability and good processability of plant compositions, innovations based on individual plants or combining compositions of plants have been employed in various applications directly or indirectly, such as in engineered wood [25–28].

2.1. Lignin

The functions of lignin in a plant are to maintain its rigid upright structure, strength and stability under various environmental conditions. Given the plant phenomena, such as evaporation, transpiration, osmotic pressure and capillary forces, having the ability to withstand adverse envrionmental conditions is imperative. The availability as a physical and chemical barrier for protection of some of the plant structure is another function of lignin. Lignin is mainly polymerized from p-coumaryl alcohol, coniferyl alcohol and sinapyl alcohol, through radical coupling, into lignin units of p-hydroxyphenyl (H), guaiacyl (G), and syringyl (S) [30]. The properties and functions of lignin are variable according to the ratios of these units, such as anti-UV activity, antioxidant activity, antibacterial activity, etc.

These inherent qualities of lignin have given rise to versatile biomass-based or bioinspired materials for a number of applications such as energy harvesting, generation and storage, environmental technology and structural reinforcement [31]. Typically, lignin is directly used as a sustainable biomass for energy harvesting. For example, lignin was used as a dispersant for carbon nanotubes (CNTs) and promoting the thermoelectric properties in thermoelectric devices, which delivered the maximum power factor as 223.1 μW m^{-1} K^{-2}, higher than that with only CNTs [32]. Lignin has also been used as a membrane for ionic thermoelectric energy harvesting, which enhances the presence of channels and thereby increases the ionic movement. With a membrane composed of 69.2 wt% of lignin, an exceptional ionic Seebeck coefficient of 5.71 mV K^{-1} and an ionic figure of merit of 0.25 were achieved [33].

2.2. Cellulose

Cellulose is known to be the most prevalent renewable and natural resource that is sustainable and reliable for enhancing material properties. Cellulose exists in various shapes and forms, such as cellulose chiral nematic-liquid-crystalline structures and cellulose-based micro/nanoscale fibers [34]. Using the characteristics of cellulose, such as toughness, crack resistance, plastic deformation capability, cellulose has been used in various applications such as membranes, mechanical reinforcement, flexible electronics and responsive sensors [35, 36].

By taking advantage of the toughness of cellulose nanocrystals (CNCs), cellulose was employed as a sustainable polymer block to build crack-resistant nacre-like CNC-epoxy composite. The submicrometer CNC aggregates crosslinked with epoxy and formed a nacre-analogous lamellar structure, which significantly enhanced the toughness and bulk ductility [35]. In addition, cellulose can be fabricated into bioinspired water-harvesting systems after proper surface engineering. Inspired by cactus spines and beetle elytra, an asymmetric amphiphilic surface is constructed from amphiphilic cellulose ester coatings and laser-engraved spines of fluorinated ethylene propylene (FEP) for a dual bionic system for both water harvesting and triboelectric generation, which exhibits a fog-harvesting efficiency of 93.18 kg m^{-2} h and the power generation to light up 400 commercial LED bulbs [37].

2.3. Pectin

Apart from the above two major building blocks, pectin, primarily composed of repeating units of α-(1–4)-linked D-galacturonic acid units, is another polysaccharide that commonly exists in plants. Pectin is categorized as a flexibility determining factor in plant cells, where the gel-like pectin provides molecular interaction between rigid cellulose and branched hemicellulose [38]. Pectin has been regarded as an exceptional natural polymer, due to its unique functionalities and excellent properties, such as biocompatibility, biodegradability, low-cost and simple gelling capability, and has been extensively employed in the food industry, medical materials and advanced materials.

The elasticity nature of pectin has endowed its direct and indirect applications in different fields. For example, the direct addition of extruded pectin into inks has significantly enhanced the 3D printability for fabricating cell-friendly scaffolds, where the addition of pectin to the ink benefitted the improvement of the shear thinning property and the viscosity adjustability [39]. With the rapid development of hydrogel, pectin has been widely used as a critical material for hydrogel preparation, due to the unique properties of gelation, solubility and rheological behavior exhibited by different pectins. Pectin-based hydrogels possessing unique structural/textual stability have been widely applied in food technology, drug delivery and tissue engineering.

2.4. Hemicellulose

Another abundantly found plant polysaccharide is hemicellulose, which has several types, such as branched, complex, and diverse structures based on the plant species. Not only can regular sugars be found in hemicellulose, but also their acidified forms, such as glucuronic acid and galacturonic acid. Unlike cellulose, hemicellulose has shorter chains at 500–3000 sugar units, while cellulose comprises 7000–15 000 glucose molecules. Due to its inherent properties, hemicellulose is amorphous and has lower stability compared to cellulose, along with a lower degree of polymerization, which has benefited its industrial applications in sustainable technologies.

Regarding the sustainable energy-related applications, hemicellulose has contributed as a flexible precursor, for example, in nanogenerators [40]. The altered chemical hydrogel incorporated with hemicellulose has enabled electrical conductivity together with other enhanced properties. The nanocomposite comprising nano-polydopamine and hemicellulose was self-powered at movement, and the flexibility made a good attachment onto the skin of this hydrogel-based sensor. In environmental technologies, hemicellulose has acted as an absorbent of heavy metals [41]. Amalgamating the biomasses with hydrogels developed from water-soluble hemicellulose and O-acetyl galactoglucomannan, enhanced absorption of As(V) and Cr(VI) was realized in solutions with different pH values.

3. Plant-inspired sustainable energy harvesting, conversion and storage

Demand for sustainable and environmentally friendly energy conversion technologies is drastically growing as a solution to cope with the global environment and conservation crisis and reach the goal of net-zero emissions by 2050. Plants exhibit the highest efficiency in solar energy harvesting through their leaves, CO_2 adsorption and conversion through the natural photosynthesis process, and ultrafast transport of electrolytes through their stems, which provides inspiration to scientists in designing materials and devices to mimic the structural and functional features of plant tissues and cells.

Plant-inspired materials have been successfully employed in almost all sections of sustainable energy technologies. For example, plant-inspired materials have been used in developing solar cells and other photovoltaic devices, by which the absorption of sunlight and the conversion efficiency into electricity have been dramatically enhanced. By studying the process of photosynthesis in plants, photocatalytic devices for converting captured CO_2 into biofuels, such as ethanol, starch and other hydrogen carbonate fuels under solar irradiation have been innovated. Different from direct solar energy harvesting and conversion processes, plant-inspired energy generation has been remarkably integrated with kinetic and mechanical energy. Plant-inspired hydro-voltage devices convert water diffusion energy into electricity by learning from the water diffusion gradient effect of plants. By taking inspiration from the nanofluidic transport properties and selective cell structures, bioinspired materials have been developed for advanced energy storage devices for real-world scenarios, such as batteries and supercapacitors. For instance, increased ion conductivity and water retention were observed in solid-state electrolytes incorporated with natural bamboo cellulose nanofibers, which enhanced the electrochemical performance of the flexible Zn-air battery [42]. A plant-like battery (FlowER: an evaporation flow redox) with prolonged battery life was eco-designed to power the wireless sensor networks for aiding precision in agriculture [43]. Similar plant-inspired designs have also been applied to other active metal-based batteries, including zinc-ion batteries [5, 44]. To showcase how bioinspiration paves the way to further enhancing the performance of sustainable energy technologies, we categorize plant-inspired materials in terms of the types of emerging technologies.

3.1. Plant-inspired materials for solar energy harvesting and conversion

The main challenges of harvesting and converting solar energy into electricity or any sustainable energy are the effective adsorption of solar irradiation and conversion efficiency of solar energy. Solar cells are a typical class of well-proven sustainable energy conversion devices that directly convert solar irradiation into electricity and have been widely installed almost everywhere in the world. However, the conversion efficiency of solar cells has reached a threshold, which needs to be further significantly improved, while over 70% of the incident energy is dissipated as waste heat, which not only brings safety concerns but also lowers the device efficiency. By taking inspiration from trees, which rapidly transport water from soil to the tip to keep the leaves cool and moist via the transpiration effect, a bioinspired hybrid multigeneration photovoltaic-leaf (PV-leaf) was designed with the functionality to drive water flow passively from a tank to cover the cell surface for evaporation and to recapture the evaporated water into clean water (figure 2(A)). Contributed by the evaporation cooling to remove 75% of the heat and reduce the temperature by $\sim$26 °C, the bioinspired PV-leaf presented a further 13.6% gain in solar energy conversion efficiency under an irradiance of 1000 W m^{-2} [45]. On the other hand, the boosted solar utilization efficiency is imperative to co-generate electricity, heat and clean water by using this pumpless 'hybrid' solar collector. The principal mechanisms inspired by the structural generation are osmotic pressure and capillary forces along the vascular bundles and the channels in the leaf for nutrient and water distribution.

Throughout billions of years of evolution, natural organisms have learnt how to efficiently collect energy from the sun and store it in the form of hydrocarbons, such as sugar and starch, to support their energy needs in growth and survival. It has been found that there are light-harvesting antenna structures (chloroplasts) on the surface of plant leaves, which provide a wide-spectrum range of solar adsorption. This antenna structure also works as a long-range channel to effectively transfer the generated excitons from the excited electronic states of chlorophyll with the absorption of photons to the reaction centers for subsequent photosynthesis reactions [46]. Inside a natural leaf, there are also interconnected 3D porous and channel networks for optimizing light absorption and facilitating mass flow. By learning from natural leaf structures and natural photosynthesis reactions, artificial solar-to-fuel conversion from CO_2 to multi-carbon products or solar-driven generation of hydrogen have been extensively reported [47].

By mimicking the chlorophyll structure or the photosynthesis of the functionality of plant leaves, artificial leaves have been designed for bioinspired solar energy harvesting and conversion [48–50]. To improve the gas diffusion and light-harvesting properties, the 3D artificial photosynthetic system (APS) usually selects a hierarchical architecture in the electrodes like that of a natural leaf. For example, the artificial leaves were synthesized from synthetic nanomaterials enclosed within plant-derived frameworks to create bio-hybrid systems [51]. As shown in figure 2(B), via a bio-templating method, perovskite titanates ($ATiO_3$, A = Sr, Ca, and Pb) that mimic natural leaf venation networks were fabricated followed by the loading of co-catalysts to aid the CO_2 reduction reactions. During the synthesis, the leaf architecture and the venation systems were directly extracted and then embedded into the formation of the APS to create a 3D hierarchical macropore network. These artificial leaves made from man-made

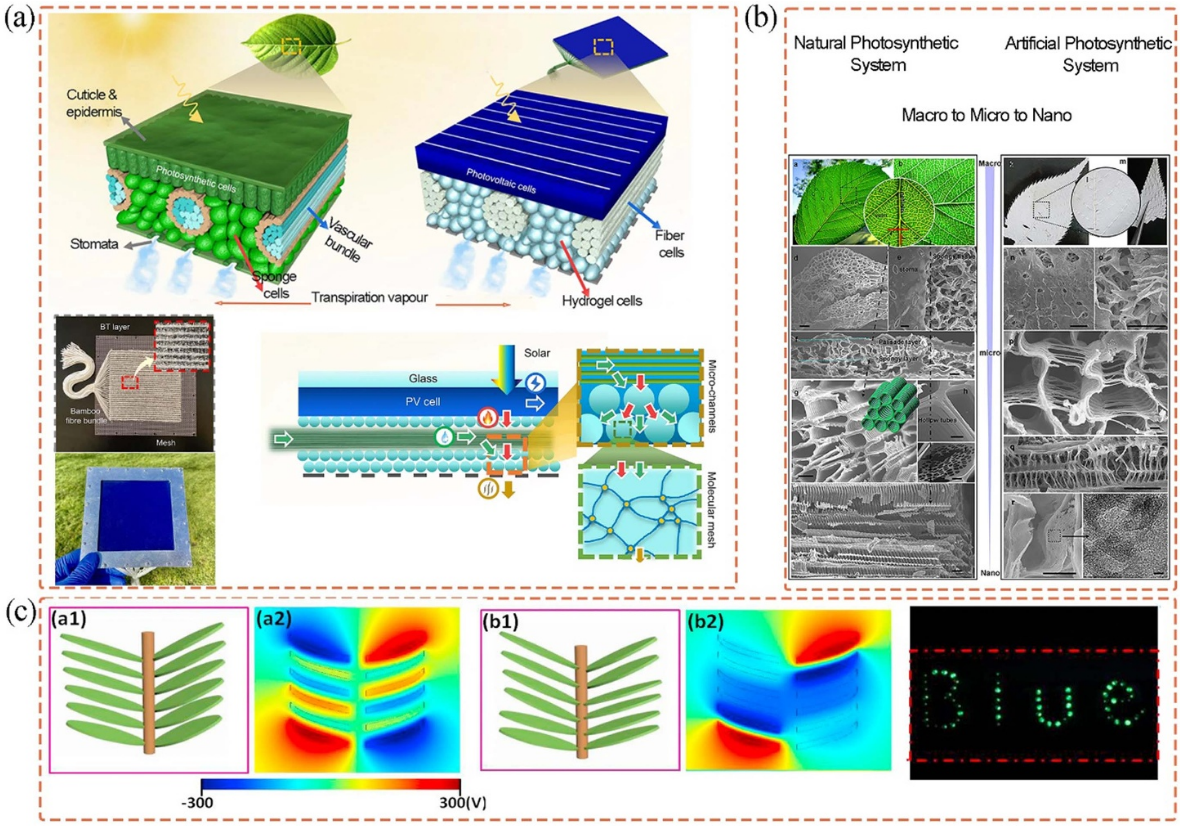

Figure 2. Plant-inspired designs in energy harvesting and conversion. (A) Structural design and performance of natural leaf and leaf-inspired photovoltaic cell with biomimetic transpiration structure to produce clean water and cool the surface for higher solar conversion efficiency. Reproduced from [45]. CC BY 4.0. (B) Natural photosynthesis-inspired artificial photosynthetic device with 3D hierarchical macropore network for effective CO_2 reduction. Reproduced with permission from [51]. CC BY-NC-ND 3.0. (C) Kelp-forest-inspired tribological nanogenerator for harvesting wave energy with output energy lighting up 60 LEDs [61]. Reprinted from [61], © 2018 Elsevier Ltd All rights reserved.

catalysts achieve efficient light-harvesting and CO_2 photoreduction performance through a natural mimicking photochemistry process to convert CO_2 and water into high-value hydrocarbons under natural or simulated sunlight.

In addition to bioinspired CO_2 capture and conversion photosynthesis devices, solar water splitting using bioinspired leaf structures has been investigated over the past decade. As the main alternative energy source without carbon dioxide emission, H_2 may play a key role in future energy supply, especially that produced from water splitting reactions. The water splitting induced hydrogen production has also received inspiration from the natural photosynthesis processes [52, 53]. For example, under anaerobic conditions, hydrogenase can accept electrons from reduced ferredoxin molecules and use them to reduce protons to H_2 [54]. Via the design of a CoWO|ITO|3jn-a-Si|Steel|CoWS monolithic artificial leaf device, in which the CoWO layer works as an oxygen evolution reaction catalyst, and the CoWS acts as a hydrogen evolution reaction catalyst, a solar-to-hydrogen conversion yield of 1.9% was achieved [55]. Very recently, Gwon *et al* reported the direct conversion of swimming green algae into a single cellular photovoltaic power station to generation hydrogen via a bioinspired artificial photosynthesis process [56]. In this study, by mechanical insertion of carbon nanofibers (CNFs) into algal cells as electron transfer highways connecting the cytosol and the extracellular space and deposition of Pt as a hydrogen-forming catalyst, alga-CNF/Pt composite power stations were formed for photosynthetic hydrogen production. It is remarkable that photosynthetic algae can be used for naturally powered hydrogen production, where the alga-CNF cell is used as a cellular photovoltaic power station to supply eco-friendly power. The algal bio-electrogenic reactor self-regulates the oxygen levels during photosynthesis and outputs hydrogen directly into the fuel cell.

Even though significant progress has been achieved in mimicking the solar harvesting and charge separation mechanisms in green leaves and plants for artificial energy production, the photosynthesis within the plant photosystem is too complicated to be replicated. Functional and chemical processes inspired by artificial photosynthesis in converting solar energy into chemical fuels have been widely investigated. However, we are still a very long way from reaching the dedicated multiscale structures and the most energy saving efficiency modes of natural leaves and plants.

3.2. Plant-inspired materials for hydrovoltaic and tribological electricity generation

Different from generating electricity from hydropower by building dams, the production of electricity from water by utilizing the electrokinetic phenomena under osmotic pressure has been proposed very recently. In a typical electrokinetic phenomenon, hydrodynamic pressure induced by gradient transporting counterions between the two ends of a capillary or porous medium can generate an electrical potential difference. This can be harnessed by various energy-harvesting techniques as an alternative form of hydropower generation. In addition to hydrovoltaic electricity generators, triboelectric nanogenerators (TENGs) have also been widely reported to be able to collect mechanical energy from the environment, such as wind, tide waves, compressions, tensions, etc, and convert it into electricity via piezoelectric materials [57–59].

It is very interesting that, by combining with some inspiration from natural species processing exceptional mass and charge transport properties, the performance of these emerging electricity conversion devices can be significantly enhanced. For instance, by learning from natural plants, the lotus-leaf-inspired droplet-based electricity generator [60], plant hierarchical multibranched structure inspired silicon hydrovoltaic device [45] and kelp-inspired TENG [61] showcase well how the inspiration obtained from plant structures contributes to efficient electricity generation. In water kinetic energy harvesting devices, slippery surfaces that allow water droplets to slide off the surface to generate electricity are the key components, and this is referred to as 'liquid–solid contact electrification' [62]. It has been revealed that the slippery surface of the lotus leaf composed of nanoscale wax and micropapillae facilitates the sliding of spherical water droplets on the superhydrophobic surface. In a droplet-based electricity generator with raindrop energy harvesting and self-cleaning properties, the application of a lotus-leaf-inspired hydrophobic dielectric layer introduces a wide range of droplet volume operation and an efficiency enhancement to 13.7% [60]. In this design, the principle of sufficient hydrophobicity to boost the electricity output is assisted by the hindrance of the rolling of the water droplets on an inclined surface. While the lotus-leaf-inspired slippery surface is very attractive for water kinetic energy harvesting, the reduced liquid–solid contact area compromises the energy-harvesting efficiency. However, this issue can be solved by the introduction of a porous substrate, the elimination of unwanted wetting transition and the choice of lubricant liquid with proper viscosity and dielectric constant [35].

TENGs are a family of very delicate devices that convert mechanical energy into electricity based on the coupling effect of triboelectrification and electrostatic induction. Inspired by the structures and functionalities of plants, such as petiole porous structures, hierarchical structures and tapered structures, bioinspired microstructures or nanostructures have been developed into the friction layers that achieve high output power and other functional properties, such as self-cleaning [63]. In a kelp-inspired TENG to harness the wave energy by mimicking the motion of the kelp along with the wave (figure 2(C)), stable and efficient energy harvesting at a vibration frequency as low as 1 Hz was realized with an output peak current of 10 μA and voltage of 260 V in a single unit, which is sufficient to power LED arrays [61]. The device uses a kelp-like electrode array consisting of soft polyethylene terephthalate and FEP as the substrate to mimic the gentle movement of kelp with wave vibrations to produce power and a layer of copper film at the backside for current collection.

Based on the above examples of plant-inspired energy-harvesting and conversion devices, it is very clear that the bioinspired design by learning from the natural morphologies, structures and functionalities has significantly enhanced the performance of energy devices. The research pathway that integrates bioinspired designs with energy materials and device innovations thus paves the way towards more efficient and sustainable energy conversion and harvesting.

3.3. Plant-inspired materials for sustainable electrochemical energy storage

Plant-inspired materials mimicking the ability to store energy in their leaves, stems and roots have provided promising solutions to the design of emerging energy storage devices to meet the requirements of high energy density, low cost, environmental sustainability, etc. Some biomass materials directly extracted from plants have been successfully employed in sustainable energy storage devices. One of the most promising natural materials from plants for energy storage is cellulose, which is a type of naturally occurring polymer that makes up the cell walls of plants. Cellulose is abundant, renewable, and can be easily processed into various forms, such as fibers, films and nanofibers. This natural material has high mechanical strength, high surface area and excellent electrical conductivity, making it a suitable material for energy storage devices, such as supercapacitors and batteries [64, 65]. Another plant-derived material for energy storage is lignin. Lignin is a complex polymer that gives plants their structural support and rigidity [66]. It is also an abundant and renewable biomass resource that can be easily processed into various forms with the characteristics of high thermal stability, high mechanical strength and good electrical conductivity, and have been employed into supercapacitors and batteries [67–69]. Interestingly, a new type of battery called a 'plant battery' directly uses natural photosynthetic microorganisms composed of a natural algae cell, an aluminum anode and a commercial open-air cathode, which can power an Arm Cortex M0+ microprocessor continuously for several months (figure 3(A)) [70]. When the plant cell is exposed to light, it generates a flow of electricity to power a microchip without any additional energy storage device, artificial lighting or organic feeding. This technology has the potential to provide a sustainable, low-cost and environmentally friendly source of energy.

To meet the growing demand of batteries for electric vehicles, large-scale energy storage systems, and wireless gadgets, bioinspired materials are being used for solutions to problems of low energy density and capacity of electrodes,

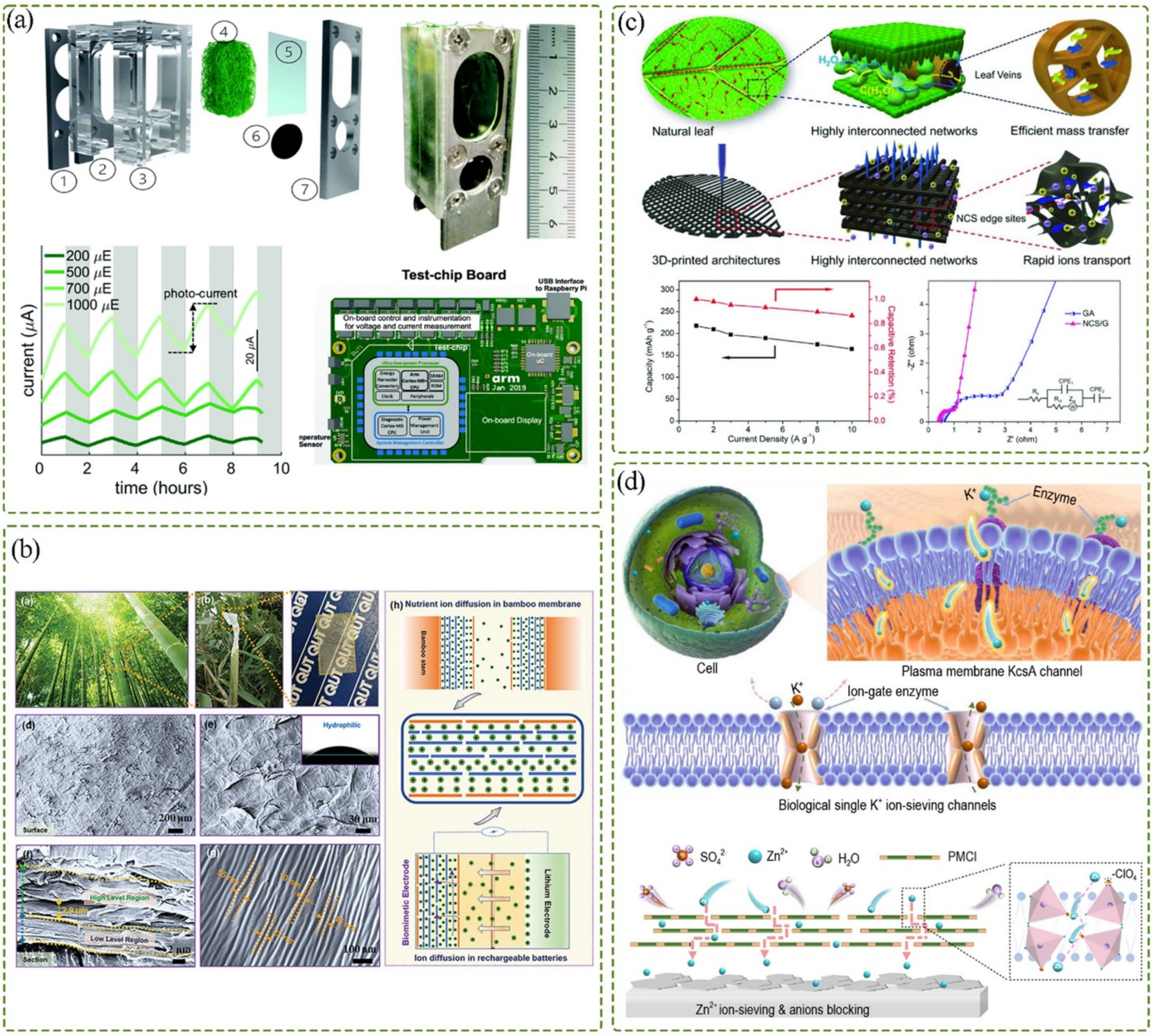

Figure 3. Bioinspired energy storage materials. (A) Photosynthetic microorganisms incorporated efficient energy bio-photovoltaic energy harvesting for powering a 'plant battery' composed of a natural algae cell, an aluminum anode and a commercial open-air cathode, which can supply energy for an Arm Cortex M0+ microprocessor continuously for several months. Reproduced from [70] with permission from the Royal Society of Chemistry. (B) Bamboo-membrane inspired Li-ion battery anode with ultrafast ion transport properties in a 2D Co_3O_4-graphene electrode for reaching superb volumetric storage performance. [79] John Wiley & Sons. © 2021 Wiley-VCH GmbH. (C) 3D printed $Ni_{0.33}Co_{0.66}S_2$/graphene aerogel (3DP-NCS/G) plant-leaf-inspired architectures with high-speed channels for electron/ion transport enabling high current density and capacitive retention of a hybrid electrochemical energy storage system. [80] John Wiley & Sons. © 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (D) Biological cell plasma membrane-inspired artificial electrode top layer with ultrafluidic Zn-ion transport and ion sieving properties for ultralong lifespan aqueous Zn-ion batteries. Reproduced from [76]. CC BY 4.0.

unsatisfactory stability of major components, and concerns regarding the eco-friendliness of spent batteries [71]. To date, bioinspired structures and materials have been developed as almost all the key components of batteries, including cathodes, anodes, solid electrolytes and robust interfaces, majorly by taking inspiration from plants with regard to their ultrafast mass transport channels [72, 73], robust mechanical properties [74], unique surface properties [75] and perfect ionic selectivity [76, 77].

It has been reported that the ion transport property of the electrode materials governs the high-rate performance of batteries, and the battery performance can be enhanced by proper engineering of the ion transport channels [78]. Bamboo has been deemed as a plant with the rapidest growing rate (91 cm per day or 0.000 02 mph), which requires a huge amount of liquid to be transported from the root to the tip via the bamboo stem and the inner bamboo membrane [79]. The study demonstrated that the bamboo inner membrane has a 2D stacking structure with a multilevel distribution of interlayer distances varying from tens of micrometers facing the cavity for facilitating the contact and wetting and bulk water and a few nanometers for the nanofluidic transport property of ions (figure 3(B)). By learning from the features of bamboo membrane, a thin-film bamboo-membrane-inspired electrode consisting of 2D graphene oxide, cobalt oxide nanosheets and 1D CNTs as a restacking separator was constructed with mimicking multilevel interlayer spaces. The bioinspired thin-film electrode reached a volumetric capacity (∼1500 mAh cm^{-3}) that outperformed other thin-film electrodes and possessed high-rate performance and stability contributed by the ultrafast interlayer transport of the electrolyte and ions.

As described above, the plant leaves have interconnected mass transport channels that could also inspire the design of analogous mass transfer processes towards superior electrochemical performance of energy storage devices. Via a 3D printing approach, a $Ni_{0.33}Co_{0.66}S_2$/graphene aerogel (3DP-NCS/G) was printed with a plant analogy of a hierarchical leaf vein structure, where the interconnected graphene framework provides high-speed channels for electron/ion transport and the uniformly dispersed $Ni_{0.33}Co_{0.66}S_2$ nanoparticles supply massive exposed edge sites for effective electrochemical storage (figure 3(C)) [80]. This bioinspired architecture drastically enhanced the electrochemical performance of a hybrid electrochemical energy storage device by coupling it with a 3D-printed multiwalled CNT/graphene (3DP-MWCNT/G) aerogel electrode, by which the device reached energy densities ranging from 32.16–22.92 Wh kg^{-1} and power densities from 443.69–4128.64 W kg^{-1}.

In the design of aqueous electrolytes, the suppression of hydrogen/oxygen evolution reactions at a high-voltage window, the retardance of dendrite growth and the avoidance of corrosion of metallic electrodes are necessary, which can be approached by proper electrode structure and electrolyte design [81–84]. The plasma membrane of biological cells in plants and beyond presents both perfect ion selectivity and ultrafluidic transport properties, which are the desired properties for rechargeable batteries. Zhang *et al* developed a plasma-membrane-inspired top layer for aqueous Zn-ion batteries to reach ultrafast Zn^{2+} transport but with the rejection of water and SO_4^{2-} groups from the passivation and the corrosion of the Zn anode [76] (figure 3(D)). With the grafting of $-ClO_4-$ onto a metal-organic-framework structure as a gating group, a 'gate on' to allow Zn^{2+} to pass through the channels while an off state for anions by electrostatic repulsion is achieved. As a result, the bioinspired membrane achieved a Zn^{2+} flux of 1.9×10^{-3} mmol m^{-2} s^{-1} and a $Zn^{2+}/SO_4{}^{2-}$ selectivity of 10, which contributed to an ultralong lifespan of over 5400 h at 10 mA cm^{-2}/20 mAh cm^{-2} without the significant decay of capacity, the growth of dendrites and the corrosion of the Zn anode. By using ideas from the methylation of biological peptides to suppress intracellular phase separation at high concentrations *in vitro* mediated by H-bonding, a hydrogel electrolyte with a salt fraction as high as 44 mol%, while retaining a $Na+/H_2O$ ratio of 10 without phase separation, was achieved via bioinspired methylation [85]. This bioinspired electrolyte endowed the aqueous Na-ion battery with ∼89.6% capacity retention after 400 cycles and 82.8% capacity after 580 cycles at 2 °C.

Given the above examples, it is notable that the bioinspiration obtained from either the structures or the functionalities of the biological species has significantly enhanced the performance of various types of sustainable energy devices without altering the materials' compositions. These successful cases provide us with motivation to further enhance the energy density and efficiency of emerging energy devices by optimizing the microstructure and mass/charge transportation behavior. Rather than discovering fully conceptual new materials to increase intrinsic energy storage and conversion properties, the creation of ultrafast mass/charge transport channels and new ion/electronic storage sites by learning from natural structures should be a more tangible approach to push the performance of sustainable energy devices to real-world applications.

4. Plant-inspired environmental technology

Increasing environmental issues associated with the soaring global population and the upsurge of emissions and discharges from human activities are other critical topics that have to be addressed urgently alongside the energy crisis. By leveraging the functionalities of natural species in balancing the carbon and nitrogen cycles, bioinspired materials have started to contribute to the innovation of environmental technologies, such as for environmental remediation and restoration [86]. For emerging environmental applications, the surface states of functional materials, such as wettability, chemical affinity, adsorption selectivity, etc, are critical to realize the well-defined functionality [87–90]. Specifically, by learning from the unique wettability of plants, super-wetting surfaces, ultrarapid and directional liquid transportation structures, self-cleaning surfaces, etc, have been developed for applications in water and air purification, water–oil separation, air quality sensing and controlling, etc [91–94].

Based on the contact with water and the four intrinsic states of hydrophilicity, oleophilicity, hydrophobicity and oleophobicity on flat substrates in air, up to 64 wetting states combining in air, water and oil have been identified after introducing micro or nanoscale roughness [1]. These findings have greatly contributed to the development of new surface technologies and increased our understanding of fundamental principles of wettability.

Nature has evolved countless mysterious life forms with a variety of remarkable wettability in plants [95], such as the superhydrophobic, low-adhesive and self-cleaning lotus leaf [96], the anisotropic superhydrophobic rice leaf [3], the superhydrophobic and anti-reflective poplar leaf [97], the superhydrophobic, high-adhesive and structural color red rose petal, the superhydrophobic and air-retaining *Salvinia* leaf [98], the superhydrophobic, high-adhesive peanut leaf with fog-capturing abilities [99], the anisotropic, slippery and amphiphilic *Nepenthes* leaf, etc [17]. These findings provide unique strategies for designing bioinspired interfacial materials with superwettability for effective water harvesting, water–oil separation, and self-sensing and actuating demanded in sustainable environmental technologies.

4.1. Plant-inspired water harvesting

Freshwater is a vital resource for all living organisms. It is essential to maintain proper hydration as well as for growing crops, cooking and cleaning. In addition, freshwater is necessary for a wide range of industrial processes, including power generation and manufacturing. However, freshwater is a finite resource, and its availability is becoming increasingly limited in many areas of the world, due to factors such as climate change, population growth and overuse of water resources. To address this issue, especially in arid areas, it is important to take steps to harvest freshwater efficiently from rain and mist [100–102]. Over the years, numerous water-harvesting methods have been developed to collect freshwater from fog, dew and steam. It has been reported that the use of tip spines, meshes and nets to capture water droplets from fog, where the surfaces or tips complete the coalescence of fog to water droplets, and the water is then transported and stored for later use. For these existing technologies, the utilization of inspiration from plants, such as cacti and *Stipagrostis sabulicola* leaves, have significantly contributed to enhancing the efficiency and durability of water collection and harvesting devices [103, 104]. For example, cacti have been well investigated for their inbuilt structure and proven mechanisms in efficient water collection [105–107], whose special conical structure aids in achieving a high water collection rate (figure 4(A)). In general, by studying the natural water-harvesting species, a few critical factors, such as Laplace pressure gradient [108, 109] wettability, gravity, and morphology of grooves, are found to govern the water collection/harvesting performance [110]. Interestingly, a 2D kirigami structure made from wax-infused paper simplified from the 3D cactus spines of grooves and barbs can produce effective fog harvesting at a rate of ~4000 mg cm^{-2} h^{-1} [111]. By applying these understandings, such as the collection, ultrafast transport, and storage of water of the lotus leaf, pitcher plant and cactus, etc, water collection/harvesting devices have been developed for efficiently collecting and storing water in arid regions or drought conditions [37, 112].

In a typical case, inspired by green bristlegrass (*Setaria viridis*), a biomimetic hydrogel with conical spines, a groove structure and aligned vertical channels inside was developed using a combining method of 3D printing and ice template for all-day harvesting [113]. At night, through the directional thorns, groove structure and superhydrophilic surface, the bioinspired hydrogel facilitates rapid transport of water through the super-spreading of the liquid film to achieve a fog collection rate of ~5 g cm^{-2} h^{-1} in fog harvesting (figure 4(B)). During the daytime, the long-range aligned vertical channels inside the hydrogel effectively pump water to the surface with a large evaporative area with an evaporation rate of 3.5 kg m^{-2} h^{-1} for solar steam generation. In addition to the harvesting and collection properties, the transport of the harvested water is a critical parameter for effective water harvesting. During further studies on the ultrafast transport in spider silk and cactus spine [114, 115], an ultrafast water transport process was observed on the surface of a pitcher plant, *Sarracenia* trichome, which is three orders of magnitude higher than those of spider silk and cactus spine, attributed to its hierarchical microchannel organization modes of the trichome (figure 4(C)) [116]. The corresponding bioinspired microchannels, by mimicking the pitcher plant surface, were fabricated and it was verified that ultrafast liquid transport properties can be successfully obtained on artificial surfaces by mimicking groove patterns. Very recently, directional liquid transport properties in natural plants have also attracted great attention [117].

In addition to direct water harvesting and collection, freshwater can also be obtained from salty water or polluted water via solar desalination technologies [118, 119]. In this process, the desalination efficiency can be improved by tailoring the functional surfaces for water transport, light absorption and water evaporation, from the design of bioinspired structures or interfaces [120–122]. Specifically, plants have the capability to transport large amounts of water through their internal channels and transpire water through their leaves under solar radiation. These natural processes have inspired the development of bionic designs to obtain pure water through solar steam generation and thermal management systems [123–126]. For example, figure 4(D) indicates a sophisticated state-of-the-art model development to produce solar steam together with anti-fouling properties, which improves the evaporation rates [126]. The best performance was recorded as 2.22 kg $m^{-2}\cdot h^{-1}$ with an overall evaporation efficiency of 139.18% achieved under 1 sun illumination. This study was inspired by the water lily in which the petals swell in light conditions and contract in darker conditions due to the addition of tannic acid–Fe^{3+} complex, which is inspired by plant compositions. This has enhanced the efficiency of freshwater production cycles by accumulating an amount of 62.99 g of salt at maximum. Furthermore, the recovery has been assessed and the evaporating performance has not decreased over time.

Natural plants showcase their superior properties in water storage. For instance, baobab trees grow in a water-scarce environment, but these have strategies to retain water for survival during harsh seasons. These mostly grow in drought-prone areas, such as Africa, and are known as the tree of life due to their versatility of uses [127]. Even without a thoughtful study on the water storage properties of this interesting plant, we believe that some inspiration for bioinspired water storage in arid areas will be obtained in the near future.

Overall, plant-inspired water-harvesting technologies can be an effective way to increase the availability of fresh drinkable water in areas where it is scarce, and they may also have the potential to reduce the need for irrigation and other forms of artificial watering. Unquestionably, plants that display unbeatable survival capability by harvesting every tiny drop of water or even vapor from both the air and the soil in those extremely dry areas via their

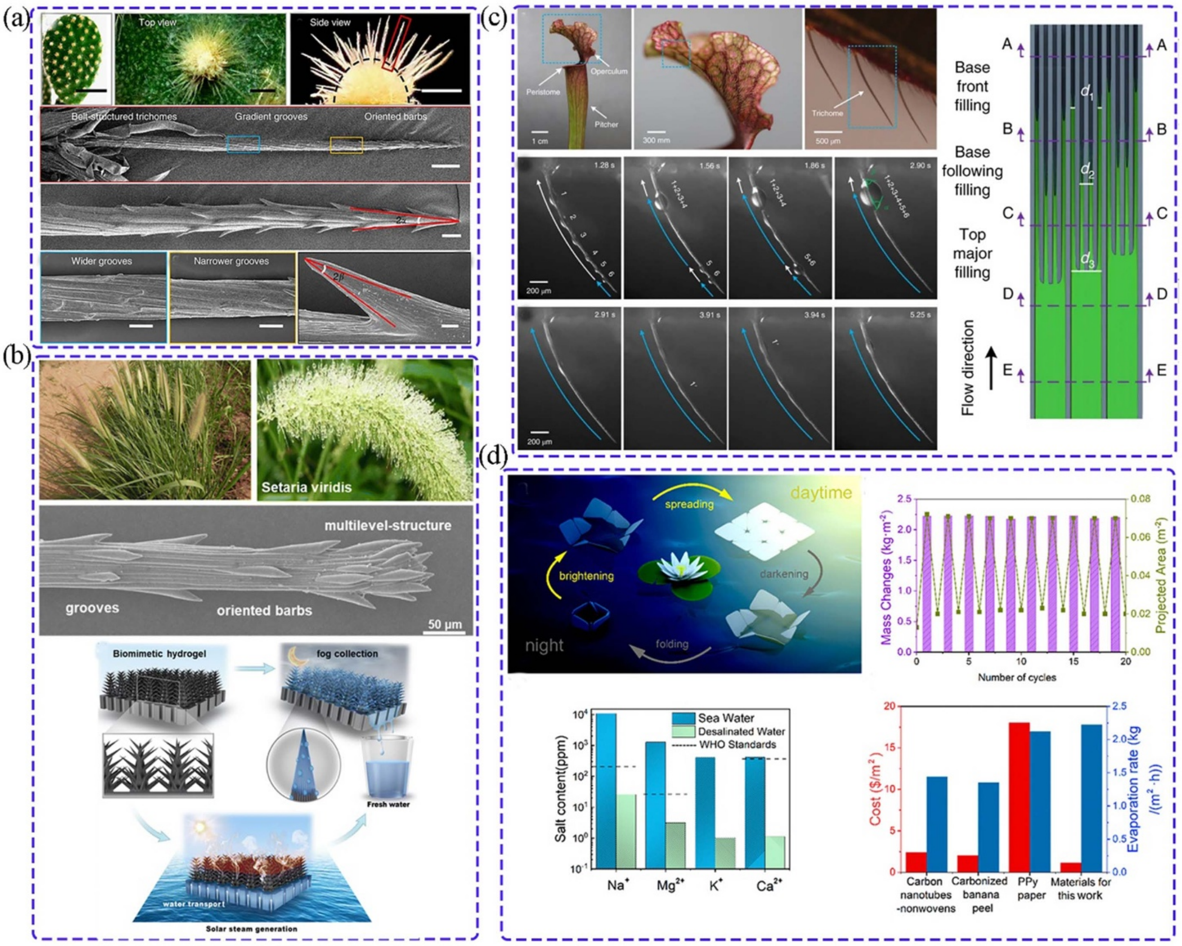

Figure 4. Plant-inspired water harvesting. (A) *O. microdasys* stem with grooves and barbs along the spine for fog collection for inspiring the design of bioinspired water-harvesting devices. Reproduced with permission from [105]. CC BY-NC-SA 3.0. (B) *Setaria viridis* fog harvesting structures and mechanism and the bioinspired fog-capturing structure made from hydrogel for 24 h solar steam generation. Reproduced from [113] with permission from the Royal Society of Chemistry. (C) Ultrafast and directional transportation of water droplets along the microchannels of *Sarracenia* trichome, which were first condensed on the trichome surface and collected into large water droplets and then directionally transported along the trichome, together with the bioinspired hierarchical microchannels on photoresist fabricated via a three-step ultraviolet lithography. Reproduced from [116], with permission from Springer Nature. (D) Solar-driven steam generation device for freshwater production inspired by a water lily, by which fresh water is produced during the daytime at a spread state and then folded and sunk into water to remove the accumulated salt at night. Reprinted with permission from [126]. Copyright (2024) American Chemical Society.

fascinating leaf and stem structures that possess amazing superwetting, ultrafast and directional water transport, and super-storage capacity have astonished researchers, motivating them to fabricate mimicking structures and properties to achieve effective artificial water-harvesting and storage systems. Most importantly, these natural structures realize effective water harvesting and storage in very sustainable ways, which provide real sustainable approaches without additional energy consumption and secondary pollution to the environment. While bioinspired water-harvesting and storage technologies are still at a very initial stage, we believe that sustainable systems derived from bioinspiration will be a major pathway towards real zero carbon-emission environmental technologies.

4.2. Plant-inspired water–oil separation

Water–oil separation is a critical process in the chemical industry, especially in the petroleum industry. Oil spills are a major threat to marine ecosystems and coastal communities and have been deemed a major problem, causing harm to both aquatic animals and plants and further disruption to the food chain, habitats and breeding grounds. Consequently, humankind is also impacted by these sorts of hazardous contaminants and pollutants. Therefore, effective environmental technologies to separate and collect oily discharges or pollutants from the environment are imperative [128]. It has been reported that bioinspired membranes with multiscale levels of hydrophilicity-hydrophobicity and oleophilicity-oleophobicity are an energy-efficient and environmentally friendly solution for water–oil separation and collection, by obtaining ideas from natural multiscale surfaces and textures, such as the lotus leaf, *Salvinia molesta*, cactus, etc.

Salvinia molesta is a free-floating aquatic fern that has been widely studied for its unique '*Salvinia Effect*', which results from the eggbeater-shaped hierarchical trichomes on the surface of leaves (figure 5(A)) [79]. *Salvinia* leaves have complex eggbeater structures composed of hydrophilic patches of four end-terminating multicellular wax-coated hydrophobic hairs. Due to the co-existence of air-trapping hydrophilic patches and the overall hydrophobic surface, a stable and long-lasting thick air layer can be maintained on the leaf when it submerges into water. This unique superhydrophobicity strong attachment towards water and the air-retainability of the *Salvinia* leaf have inspired the innovation of bioinspired surfaces for oil–water separation, drag reduction, thermal insulation and water repellence [129, 130]. As summarized in a review by Bing *et al* [129], the bioinspired eggbeater structures and arrays have been fabricated by a series of methods, such as direct laser lithography, chemical vapor deposition, 3D printing, chemical/plasma etching, etc. Interestingly, the natural *Salvinia* displays excellent oil affinity and strong oil adhesion, which allow the adsorption of crude oil within only 20 s [131], and the *Salvinia*-inspired structure has also presented excellent water–oil separation performance. Typically, the *Salvinia* nano beater structures were fabricated via a 3D printing method to realize the point-adhesion hydrophilic petal effect and the superhydrophobic surface on a photocurable resin reinforced by the addition of MWCNTs for effective water–oil separation and other potential applications, such as droplet-based microreactors, lossless water transport, 3D cell culture, etc [132].

While there are many examples to date, it is notable to see the directional steering gear developed for oil–water separation by learning from the *Nepenthes* peristome cavities (figure 5(B)) [133]. Functional surfaces with spiky arrays, such as the cat-tongue surface or arrayed cavity structures, i.e. *Nepenthes* peristome, have the function to transport liquid directionally, and bioinspired dual-bionic gears fabricated via 3D printing with similar surface structures achieve rapid, continuous and lossless oil–water separation together with antifouling and effect flow ability from the superwettability and the complementary topology morphology. In this bioinspired gear pair, the cat-tongue biomimetic gear exhibited extremely low adhesion to various oil drops under a water environment (red plots), and the peristome-inspired cavity gear exhibited extremely low adhesion to water drops under various oil environments (blue plots), which allow the effective demulsification and separation of the well-dispersed phases from a water–oil emulsion solution.

Nevertheless, among significant developments in bioinspired water–oil separation functional surfaces, scientists are yet to seek advancements in discovering simplified strategies to adapt somewhat complex natural structures with extraordinary wettability or directional transport properties into reliable and feasible water–oil separation solutions. Compared with the numerous species in nature, the plants that were discovered and understood for effective water–oil contact, adhesion and transport are yet only very few of them. Further effects on the investigation of new targeting objectives for water–oil separation are one critical research focus, and the exploration of simplified fabrication approaches for low-cost and large-scale production of relatively complex structures, such as the eggbeater structure, should be another focus of bioinspiration/biomimicry research.

4.3. Plant-inspired environmental sensors and actuators

With the development of artificial intelligence and the Internet of Things, there are significantly soaring demands on extremely sensitive and responsive materials for all-scale sensors and actuators [134]. Biological systems show amazing responses to external stimuli in adaptive and active ways, while the mechanisms are far from being well understood. Plants have a well-evolved ability to respond to a wide range of signals and adapt effectively to suit the changing environmental conditions, even if their lifestyle is of a stationary nature, such as pinecones being responsive to humidity by opening and closing to different degrees, and Venus flytraps rapidly actuating for the touch of insects [135, 136]. This aspect has enabled us to explore plant-inspired sensors and actuators, and some well-demonstrated examples of these have been reported [137]. For example, by understanding the geometry, mechanics and dynamics of the cells, tissues and organs of the pinecones, bioinspired bilayer structures with a controllable blooming/closing response to humidity or humidity-responsive actuators based on papers and polymers have been fabricated [135, 138]. The ability of plants to change color or shape in response to stimuli is also inspiring the development of new smart materials that can sense and respond to their environment in similar ways [139].

Bio-actuation is an aspect of adapting the external stimuli responsiveness from biological structures, which is commonly observed in plants and has also inspired the development of novel artificial materials to enhance the cutting-edge artificial actuation applications [140]. Soft matter sources, such as polymers and hydrogels, are excellent materials for the fabrication of bioinspired actuators. By using hydrogels, for example, plant-tendril-inspired self-coiling microfibers [141], *Mimosa-inspired* touch-sensitive and multi-stimuli-responsive starch actuators [142] and Venus-flytrap-inspired self-powered voltage-triggered actuators have been designed [136, 143]. Plant tendril coiling has exceptional shape-morphing traction and climbing and has widely inspired the design of soft actuators. Specifically, an alginate and silica-containing alginate Janus hydrogel microfiber that is responsive to humidity gradient could mimic the capability of plant tendrils with reversible coiling and uncoiling under the change of humidity [141]. This coiling strategy has enhanced the lifting capacity of the micro lifter based on external stimuli. Shape memory actuators exhibit reversibly multi-responsive performance, if exposed to humidity, temperature or light irradiation. Taking the ideas from the tough sensitive mimosa leaves, multi-responsive starch actuators composed of heat gelatinizable starch, photothermal responsive liquid metal microparticles, and sodium alginate as the natural molecular analog were manufactured for reaching irreversible-to-reversible actuating capability by

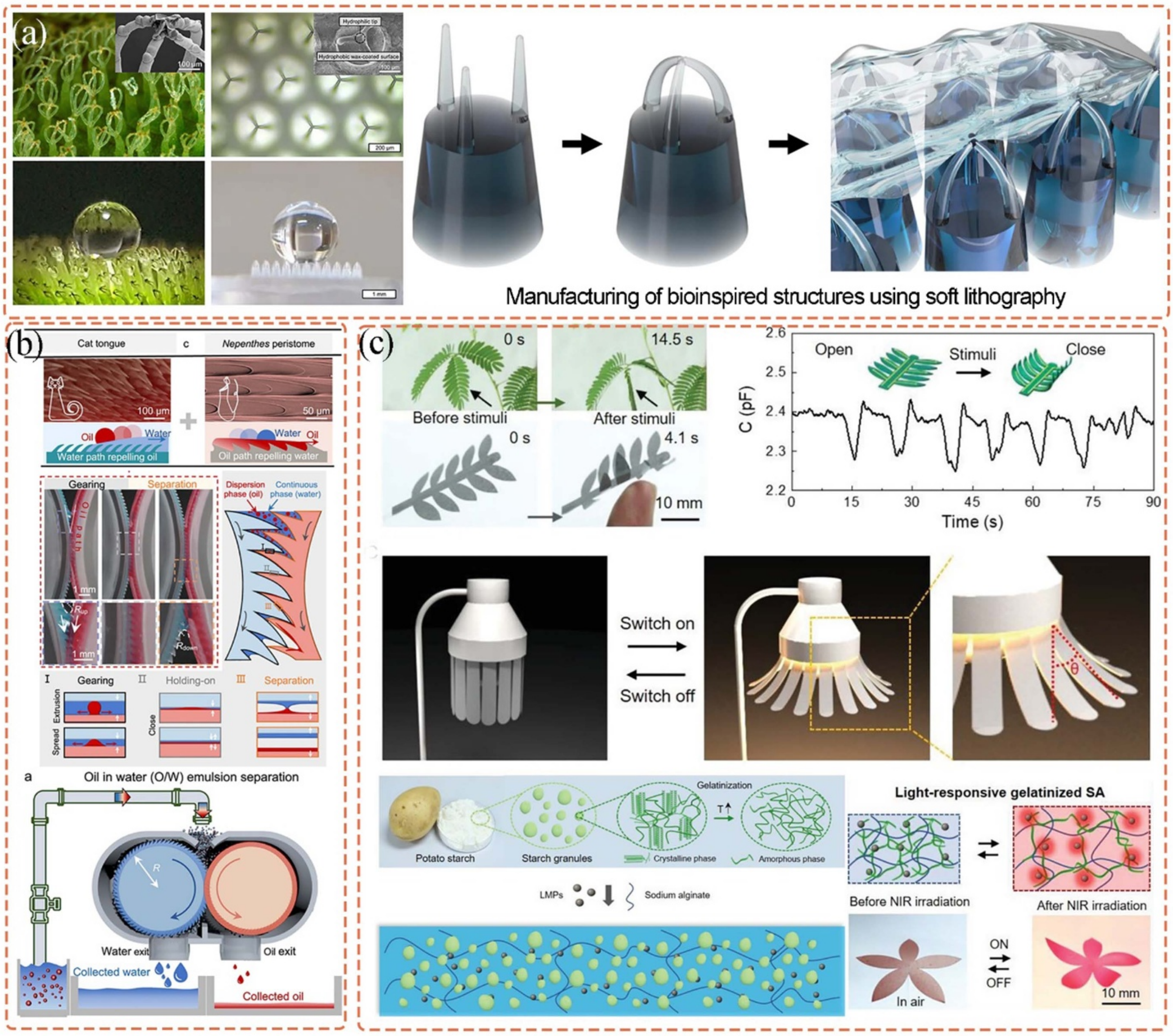

Figure 5. Plant-inspired environmental sensing and actuation. (A) Eggbeater-shaped hierarchical trichomes on a *Salvinia* leaf composed of hydrophilic patches of four end-terminating multicellular wax-coated hydrophobic hairs inspired the innovation of co-existence of air-trapping hydrophilic patches and the overall hydrophobic surface for effective water–oil separation. Reproduced from [98]. CC BY 4.0. (B) Low oil adhesion cat-tongue structure and low water adhesion *Nepenthes*/pitcher plant peristome structure inspired dual-bionic gears for highly efficient oil–water separation, where the rotating dual-bionic superwetting gears aid in collecting oil and water directional separation. Reproduced from [133]. CC BY 4.0. (C) *Mimosa*-leaf-inspired multi-responsive starch actuator, which is composed of starch with heat-induced gelatinization feature, sodium alginate, and micro-sized liquid metal particles (LMPs) with excellent photothermal behavior, with the responsivity contributed by the reversibility of the locking/unlocking hydrogen bonds upon stimuli of humidity, temperature, and low-energy near infrared irradiation. [142] John Wiley & Sons. © 2023 Wiley-VCH GmbH.

photothermal-induced locking/unlocking of hydrogen bonds (figure 5(C)). This mimosa-inspired actuator is highly sensitive, multi-responsive, exhibits irreversible-to-reversible actuation and has very promising applications in environmental monitoring, smart lampshades and smart food.

Sharing similar challenges with the study of other bioinspired materials, the exploitation of plant actuation and sensing structures as well as their mechanisms to execute the reactions under various stimuli are still to be uncovered. The superior sensitivity and actuation of natural plants can surely be employed in future innovative sensors and actuators, which have numerous possibilities to detect and indicate external differences and changes in the environment.

5. Plant-inspired mechanical toughness

Plants provide massive inspiration in mechanical superiority. Even though the major components of plants are cellulose, lignin and hemicellulose, they are assembled with each other to provide excellent rigidity and structural integrity. Special characteristics of plants that stand out are mechanical toughness, strength and moldability, which have enabled natural plants to play critical roles in buildings and constructions [144]. The robust and adaptive mechanical properties of plants inherited from the soft composites have significantly contributed to plant-inspired innovations, as we introduced in our previous study [19].

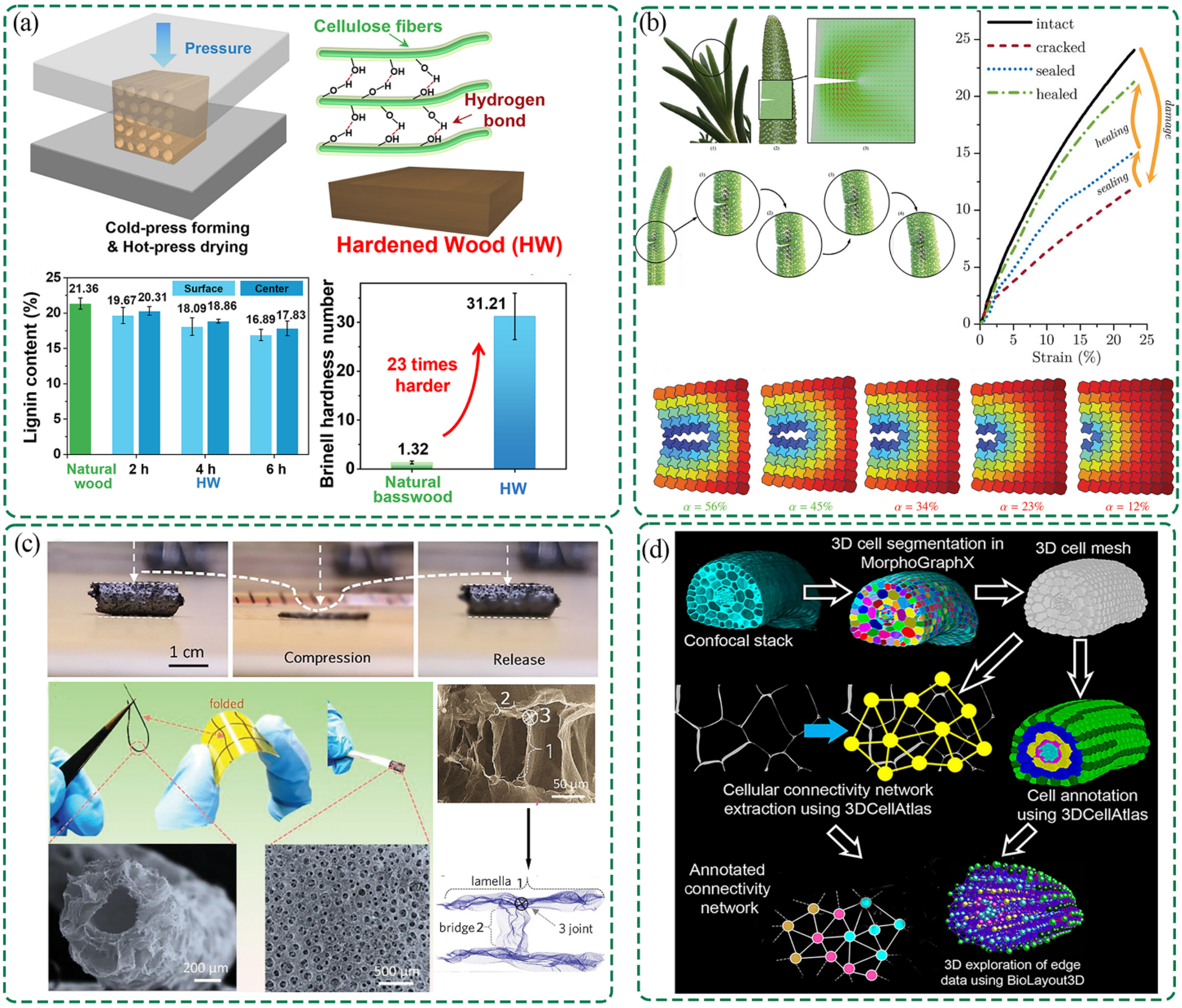

Figure 6. Plant-inspired robust mechanical properties and resilience. (A) Hardened wood prepared from natural raw wood after removal of lignin and hemicellulose via chemical treatment in 5 wt% NaOH/2.5 wt% Na_2SO_3 aqueous solution and then hot pressed for densification, presented 23-fold enhancement in hardness. Reprinted from [147],© 2021 Elsevier Inc. (B) Pressure-dependent programmable self-healing property achieved in a metamaterial inspired by the stem of *Delosperma cooperi*, in which permeable unit cells can change size depending on the internal pressure to close the cracks. Reproduced from [150]. CC BY 4.0. (C) Bamboo-fiber-inspired cellular graphene aerogel with superior recoverability up to ten cycles under 80% strain. Application in flexible electronics was acquired after etching the template and freeze-drying with further heating reduction. [151] John Wiley & Sons.© 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (D) Topological properties of complex organ architecture in a plant embryonic stem (hypocotyl) and the multicellular configuration within organs endowing higher-order functionality of robustness and plasticity. Reproduced from [152]. CC BY 4.0.

It has been revealed that the mechanical toughness and rigidity of natural plants, such as wood, is basically associated with the percentage of the major chemical composites. Unlike the normal direct use of natural wood, very recently the concept of engineered wood has been proposed, in which part of the components are modified or extracted to further optimize and enhance the mechanical performance of wood [19, 145, 146]. For example, wood that is engineered by partly removing lignin and hemicellulose from the natural basswood followed by hot-press drying processes presented a 23-fold hardness enhancement relative to the natural wood counterpart and comparable to steel and plastic (figure 6(A)) [147]. As a sustainable solution, the knife made from the engineered wood displayed higher hardness and better sharpness than the commercial knives and the wooden nails showed exceptional rust resistance and comparable mechanical performance to steel nails. Due to its light weight and mechanical strength, the engineered wood should also be an alternative renewable structural material to replace the current cement and concrete (figure 6(B)). Furthermore, moldable natural and engineered wood can be easily fabricated into complex shapes and mechanically strong structures [145, 148]. For example, decellularized plants have become favorable scaffolds for tissue engineering [149]. The method of fabricating strong structural and functional components will renew the development and utilization of tough and strong sustainable materials directly from natural resources with reduced environmental pollution.

Although numerous successful cases exist for directly utilizing amazing natural plants and engineered wood, the plant-inspired materials made from advanced materials further expand the performance limits and application areas. For example, lotus fibers have also inspired the design of green composite materials with superior strength and toughness [150]. By investigating the hydrostatically driven deformation-induced wound-sealing principle of *Delosperma cooperi*, a plant exhibiting rapid self-sealing of external wounds in 60 min, a programmable mechanical metamaterial was proposed by implementing hydraulic pressure-dependent unit cells for realizing the self-sealing function [150] (figure 6(B)). By mimicking the pressure-dependent volume change and permeability of the cell structure of the plant leaves, a perfectly sealed and inflatable metamaterial with hollow and permeable puzzle-like cell structure was manufactured from soft silica rubber to achieve successful crack closure during the self-healing process.

The hierarchical architecture of bamboo has inspired the careful development of flexible electronics, such as sensors and energy storage devices, due to the porosity observed at the micro-scale. The channels distributed across bamboo fibers have inspired the development of template-assisted highly ordered biomimetic hollow graphene fibers. Figure 6(C) shows the full recoverability tested upon the entire compression. Impressively, this graphene aerogel can support stable cycling loading-unloading mechanical tests up to ten cycles with strain up to 80% and engineering stress of 0.06 MPa [151]. When observing the mechanisms inside the aerogel, it is indicated that the linkage patterns in the unit cell are responsible for this superior recoverability. Thus, the architecture of bamboo fibers has inspired the design of graphene-based flexible electronics with superior mechanical properties.

The mechanical strength displayed in plant fibers, plant membranes, plant barks, roots, etc, has inspired the use of mechanical toughness in artificial materials. Apart from the chemical compositions and the constitutional substructures of plants, it has been revealed that surfaces and interfaces with specific topological structures play vital roles in their mechanical properties, which could further improve our understanding in designing mechanically robust bioinspired materials. For example, Jackson *et al* studied the topological properties of complex organ architecture in the plant embryonic stem (hypocotyl) by digitally capturing global cellular interactions and found that the multicellular configuration within organs endows higher-order functionality, such as robustness and plasticity, through a structure-function relationship [152]. A similar interfacial topological structure-mechanical property relationship has also been identified in fabricating polymer-based materials, in which the interfacial fracture toughness is correlated with the interfacial surface roughness [153]. Thus, the effect of topological structures of surfaces and interfaces on the structural and functional properties of both biological structures and bioinspired artificial materials will be a new research focus and shed light on designing advanced materials.

Compared with mechanically robust bioinspired materials produced by learning from the biological structures of animals, plants and the corresponding bioinspired advanced materials have been much less studied. One reason is that the composition of plants is relatively simple, and the other reason is that the structural and functional diversity in plants is significantly finite compared to animals, which leads to less attention being devoted to plants and plant-inspired materials. However, by performing an in-depth investigation on the structure-functionality relationships in plants, more interesting structures and functions in plants will be discovered to provide more ideas for designing mechanically robust advanced materials.

6. Conclusion

Bioinspiration has been a thriving trend in multidisciplinary research areas for developing innovative and superior structures and materials. In this realm, bioinspired materials produced by learning from various plants are important for designing greener sustainable materials, mechanically stronger structural materials, and functionally diverse multifunctional materials for emerging sustainable and intelligent technologies and innovations. This review discusses the progress of plant-inspired multifunctional surfaces and interfaces in adapting various aspects of plant functions and structures and pathways from structure-functionality relationship exploration to advanced materials design and manufacturing and finally real-world applications. Starting from the amazing function of plants to convert CO_2 and solar irradiation into nutrition and energy, plant-inspired materials and technologies for sustainable energy harvesting, conversion and storage, such as solar energy harvesting and conversion, hydrovoltaic electricity generation, tribological electricity generation and energy storage, were extensively discussed. Leveraging from the functionalities of plants in carbon and nitrogen cycle balances and living environment maintenance, bioinspired structures and materials for environmental cleaning and remediation were reviewed, such as the applications in water harvesting, water–oil separation, and environmental sensing and actuating. Based on the fact that plants have been directly used as structural and construction materials throughout history, engineered wood as a bioinspired advanced material is a sustainable alternative to concrete and steel and is also a promising option towards green structural materials.

7. Future perspectives

Despite substantial progress having been achieved in the study of plant-inspired materials, which have become an exciting field that leverages nature's efficient designs to create innovative, sustainable solutions, the inherent challenges associated with the structural design and advanced manufacturing of plant-inspired materials still exist. Understanding and revealing the natural structure-property relationships and the responsive mechanisms of plants for inspiring the innovation of advanced materials for both structural and functional applications are still at an initial stage. It is necessary to devotefurther effort to discover, through plant inspiration, new biological structures with enhanced efficiency and effectiveness in sustainable technologies.

First, the efficiency of plant-inspired designs in sustainable technologies should be further improved for potential practical applications. Unlike animals, plants are usually tiny and weak at their individual scale, leading to fascinating but low-efficiency modes of mass and energy harvesting, conversion and storage. For example, water harvesting inspired by plant-based phenomena could be a very promising solution for providing fresh-water in some arid areas in an environmentally friendly way. However, the amount of captured or collected fresh-water from fog and evaporation is still too small to support practical scale water usage. Therefore, optimizing the harvesting volumes and efficiency should be a future focus from an engineering perspective.

Second, further discoveries in structure-functionality relationships in plants are needed. Subtle improvements in sustainable energy harvesting, electricity generation and energy storage devices have been executed using the inspiration taken from the extraordinary properties of plants, such as the amazing CO_2 capturing and natural solar energy conversion structures in the plant leaves, the ultrafast mass and ion transportation in the stems and trunks, and the self-healing properties in numerous succulents. Moreover, multifunctional surfaces in plants with salient properties of superwetting, directional liquid transport, and environmental responsive and actuating have been successfully employed in sustainable environmental technology. As mentioned in this review, research on structure-functionality relationships in plants is still very rare. Further effort in plant discoveries to provide further inspiration is necessary to develop superior bioinspired materials.

Third, effective manufacturing technologies are required to mimic multiscale ordering and complex natural structures. For example, the eggbeater structures on the leaves of *Salvinia molesta* are critical to keep long-lasting air trapping together with the unique wettability for achieving the '*Salvinia Effect*'. However, the manufacturing of this type of complex structure, especially at micrometer and nanometer scales, is very challenging. Therefore, the large-scale fabrication of bioinspired materials and structures needs strong support from the advanced manufacturing sector.

Finally, it is crucial to exploit synergistic effects to understand fragile surfaces and interfacial structures to incorporate them into real-world applications through the employment of sophisticated techniques and advanced technologies. In particular, attention has been paid to how the topological structures of surfaces and interfaces contribute to the mechanical and functional performances of both biological and artificial structures. Advanced characterization equipment and facilities are critical to facilitate the discovery of topological architectural arrangements in natural structures without damage or alternation to the surface and interface structures of biological materials during the observation and characterization processes This must thus allow the incorporation of the actual biological structures and functionalities into the design of superior environmental devices having highly efficient sustainable energy with the expected level of effectiveness and performance.

Nevertheless, it does not matter how grand the challenges are, plant species differing in morphology and mechanisms to thrive in their adapting environments have provided us with fabulous inspiration in designing advanced materials with superior properties and performance compared to their conventional forms. This success will drive further exploration of novel structures and functions in plants, which will then offer vast opportunities for advanced material innovations. With endeavor and collaboration from cross-disciplinary researchers, bioinspired materials and designs will further expand the capacity of existing materials and contribute to the sustainability goals for a green and resilient future.

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ORCID iDs

Binodhya Wijerathne https://orcid.org/0000-0002-4314-1730
Ting Liao https://orcid.org/0000-0001-7488-6244
Ziqi Sun https://orcid.org/0000-0002-4777-4017

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