Recent progress on biomass-derived ecomaterials toward advanced rechargeable lithium batteries

Jia Liu1,2 | Hong Yuan3 | Xinyong Tao4 | Yeru Liang5 | Seung Jae Yang6 | Jia-Qi Huang3 | Tong-Qi Yuan1 | Maria-Magdalena Titirici7 | Qiang Zhang2

1Beijing Key Laboratory of Lignocellulosic Chemistry, Beijing Forestry University, Beijing, China
2Beijing Key Laboratory of Green Chemical Reaction Engineering and Technology, Department of Chemical Engineering, Tsinghua University, Beijing, China
3Advanced Research Institute of Multidisciplinary Science, Beijing Institute of Technology, Beijing, China
4Department of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou, China
5College of Materials and Energy, South China Agricultural University, Guangzhou, China
6Department of Chemical Engineering, Inha University, Incheon, Republic of Korea
7Department of Chemical Engineering, Imperial College London, London, UK

Correspondence
Tong-Qi Yuan, Beijing Key Laboratory of Lignocellulosic Chemistry, Beijing Forestry University, Beijing 100083, China.
Email: ytq581234@bjfu.edu.cn

Qiang Zhang, Beijing Key Laboratory of Green Chemical Reaction Engineering and Technology, Department of Chemical Engineering, Tsinghua University, Beijing 100084, China.
Email: zhang-qiang@mails.tsinghua.edu.cn

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Abstract
Biomass materials are of great interest in high-energy rechargeable batteries due to their appealing merits of sustainability, environmental benefits, and more importantly, structural/compositional versatilities, abundant functional groups and many other unique physicochemical properties. In this perspective, we provide both overview and prospect on the contributions of biomass-derived ecomaterials to battery component engineering including binders, separators, polymer electrolytes, electrode hosts, and functional interlayers, and so forth toward high-stable lithium–ion batteries, lithium–sulfur batteries, lithium–oxygen batteries, and solid state lithium metal batteries. Furthermore, based on the multifunctionalities of bio-based materials, the design protocols for battery components with desired properties are highlighted. This perspective affords fresh inspiration on the rational designs of biomass-based materials for advanced lithium-based batteries, as well as the sustainable development of advanced energy storage devices.

KEYWORDS
biomass, ecomaterials, lithium batteries

1 | INTRODUCTION

The flourishing development of electric vehicles and portable electronics, and further expansion of their commodity markets advocate low-cost, high-energy-density, and safe energy storage devices. Among state-of-the-art energy storage systems, rechargeable batteries, especially lithium–ion batteries (LIBs) based on intercalation
In this contribution, we highlight how biomass-derived materials (e.g., natural biological polymers and bio-derived oriented carbonaceous materials) with special properties improve the interfacial and bulk problems in lithium batteries, as well as promote the creation of energy devices from alternative, abundant, and renewable sources for increasing their overall sustainability. An appropriate guideline of the combination between sustainable natural materials and protective strategies are provided, highlighted especially for advanced Li batteries. Finally, perspectives and recommended research directions for the further development of biomass materials in Li batteries are proposed.

2 | NATURAL PRIMORDIAL BIOLOGICAL POLYMERS

2.1 | Functional aqueous binders

Since the commercialization of LIBs in 1991, the Li battery industries have flourished by the increasing development of 3C electronics and electric vehicles. In general, fluorinated vinyl polymers (such as polyvinylidene fluoride, PVDF) as binder and organic solvents (such as N-Methyl-Pyrrolidone, NMP) as dispersant are adopted in electrode fabrication of Li batteries, which cause not only the considerations on the health and environment by the poisonousness of organic solvents but also the safety hazards by their inflammability at elevated temperature.

Natural biological polymers have attracted numerous scientific attentions due to their structural and functional diversities. Particularly, abundant hydrophilic polar functional groups in the skeletons intensify the affinity with water molecules, which endows natural polymers with a good water-solubility and therefore improves the compatibility with aqueous solutions. With the increasing concerns on human health and environment protection, natural polymers have been integrated into green manufacturing of batteries, in which harmful organic solvents are seldom or even not used. To date, plenty of natural polymers (for instance, cellulose, chitosan, alginate, gum, cyclodextrin, lignin and their derivates) have been served as water-soluble binders for fabricating green electrodes toward advanced Li batteries.

In addition, as a critical component in electrode, the primary responsibility of polymer binders is to maintain the structural stability and integrity, which is the premise of constructing effective conductive framework and achieving lithiation/delithiation of active materials. Natural polymers are generally insoluble in organic electrolyte solutions. The implementation of water-soluble
natural polymer-based binders can mitigate and even avoid the instability and collapse of electrode structure resulted from the dissolution and swelling of conventional binders in organic electrolyte. Furthermore, sufficient polar groups that widely presented in natural biological polymer structure, such as \(-\text{OH}, -\text{COOH}, -\text{NH}_2\), also propel the intramolecular or intermolecular interaction between them and themselves. The formed intermolecular cross-linking framework could contribute to the structural or interfacial stability and the electrolyte solution reservation in electrode.\(^{40-42}\)

Especially in a working Li–S battery, sulfur conversion chemistry involves phase transform and migration. The dissolution and migration of polysulfide intermediates lead to low active species utilization and reduced Coulombic efficiency, seriously limiting the practical application of Li–S batteries.\(^{43-46}\) In 2013, Cui and coworkers demonstrated that the electronegative heteroatoms or groups have the ability of trapping electropositive Li ions.\(^{47}\) Due to the presence of abundant polar heteroatom-containing groups, natural polymers used as functional binders exhibited great superiority in surface electronegativity (Figure 1A-D). The polar functional groups can be served as affinity sites to anchor dissolved polysulfides, thus significantly mitigating the shuttle effect and improving energy efficiency in a working Li–S battery (Figure 1E,F).\(^{40}\) Meanwhile, the natural polymers are also enriching with many electronegative functional groups. They have the ability to coordinate with electropositive multivalent metal ions, which act as efficient chemical binding agents for immobilizing intermediates.\(^{48,49}\) Owing to the electrostatic interaction between sodium alginate (SA) and Cu\(^{2+}\) ions, an ionically cross-linking network binder with robust mechanical propriety was recently proposed to not only trap polysulfide anions through strong chemical coupling of electropositive Cu\(^{2+}\) ions but also accommodate electrode stress during cycling.\(^{48}\) By incorporating the dual-function binder, a highly stable sulfur electrode with superior cycling stability and capacity retention were observed.

Recently, a facile in situ self-polymerization of bioderived polymer monomers to form polymer resin serving as functional binders have attracted fascinating interests.\(^{50}\) On one hand, the in situ polymerization process of monomers was triggered inside electrode, which can be served as electrode additives to fill in the inner gaps and pores of electrode, and thus reduced the amount of electrolyte required for the electrode. On the other hand, this strategy offered the skinned binders with remarkable mechanical strength and long-range order structure, which not only averted the electrode structure destruction but facilitated electronic/ionic transportation, consequently rendering an excellent electrochemical performance of battery systems. The electrode fabrication based on in situ self-polymerization strategy opens a new way on the revolution of electrode process from environmentally benign raw biomass to biopolymer monomers and eventually to polymeric bio-derived electrode materials.

**FIGURE 1** A and B, Chemical structures of natural guar gum (GG) and xanthan gum (XG). C and D, Schematic of the intermolecular interaction between GG and XG. E and F, Mechanical property and cycling stability of the sulfur cathode with GG-XG binder. Reproduced with permission. Copyright 2017, Royal Society of Chemistry\(^{40}\)
2.2 Gel polymer electrolyte or separator

To date, most of commercial Li batteries are based on organic electrolyte. The fluidity and low flash point of organic solution easily result in the leakage and volatilization of electrolyte as well as the burning of battery devices, which deviate from the tenet of pursuing higher security of Li battery in future development direction.\(^{51,52}\) Especially in Li metal batteries, Li dendrites are one of major risks for battery safety. Li dendrites can react with and consume limited liquid electrolyte, resulting in iner-tile and finally dry electrolyte condition in a working Li metal battery. Moreover, the generation and growth of Li dendrites have the possibility of permeating across separator and lead to battery short circuit, causing thermal runaway and final combustion or explosion of batteries.\(^{37}\)

Gel polymer electrolytes that combine the advantages of highly interfacial infiltration of liquid electrolyte and high solidification strength of solid electrolyte have been regarded as a promising strategy to mitigate safety hazards.\(^{53}\) Liquid organic electrolyte can be fixed and reserved in micro gel polymer framework structure, which can refrain from the leakage of electrolyte and alleviate their exhaustion with Li metal. In addition, high mechanical flexibility and elasticity of gel electrolyte can also function as a physical barrier to not only separate from the positive and negative electrodes but also prevent the growth of Li dendrites, thus enhancing battery safety.

Natural polymers have intrinsic superiority in forming ionically cross-linked networks owing to intramolecular and intermolecular interaction in water solution. Generally, the interaction between polar hydrophilic functional groups is dynamic and changeable. In water electrolytes, carboxymethylcellulose can be gelled to form a self-healing aqueous gel polymer network.\(^{54}\) The aqueous Li battery with carboxymethylcellulose gel electrolyte exhibited better safety, stability, and reliability in comparison with organic electrolytes.\(^{55}\) Other biological polymers, such as tamarind seed polysaccharide, can also be solidified by gelation in aqueous solution.\(^{56}\) However, owing to the insolubility in organic solvents, natural polymers are generally difficult to use directly as gel polymer skeletons to reserve organic liquid electrolytes in conventional Li batteries. Modification strategies through grafting or combining with other macromolecules have also been demonstrated.\(^{51,57-63}\) For instance, by combining starch with γ-(2,3-epoxypropoxy)propyltrimethoxy silane, organic electrolyte can be gelled via the chemical substitution reaction between −OH groups and −Si−(OCH\(_3\))\(_3\) (Figure 2A,B).\(^{57}\) Recently, Dong et al reported a new kind of bio-based poly (methyl vinyl ether-alt-maleic anhydride) composite electrolyte layer supported by natural bacterial cellulose and demonstrated its effectiveness in Li metal anode protection.\(^{58}\) In addition, electrolyte gelation can be also achieved by the doping oleic acid and glycerol plasticizer into carboxymethyl cellulose,\(^{60}\) the reinforcement of gelatin protein toward polyethylene oxide (PEO) electrolyte,\(^{61}\) the cross-linking polymerization of functionalized lignin-derivatives, vanillyl alcohol, and gastrodigenin with multifunctional thiol monomers,\(^{62}\) and the recombination of inorganic molybdenum disulfide (MoS\(_2\)) nanoflakes with surfactant-oxidized cellulose nanocrystal (OCNC).\(^{64}\) Particularly, beneficial from the well-designed three-dimensional ion transportation channels and high mechanical modulus enabled by well-dispersed MoS\(_2\) and OCNC, this plastic crystal composite polymer electrolyte displayed excellent capability of suppressing Li dendrites in Li metal batteries (Figure 2C,D).

Similar to most of conventionally industrial polymer macromolecules, naturally biological polymers have the same ability of casting membranes and electrospinning fibers to serve as a separator in Li batteries.\(^{65,66}\) Owing to the tensile strength and thermal stability, cellulose-based microporous membranes have been wildly investigated.\(^{67-69}\) Cui and coworkers prepared cellulose-based nonwoven nanofiber membranes as an advanced LIB separator with low cost, renewability, and environmental benefits.\(^{68}\) Compared to the commercialized polyolefin separator, the cellulose-based nanofibrous separator displayed not only higher ionic conductivity, but also improved thermal-resistance property with no shrinkage up to 200 °C, rendering the LIBs higher rate capability and better capacity retention. After that, the cellulose/polysulfonamide composite membrane was obtained by the same research group via a facile papermaking process, exhibiting excellent electrolyte wettability and thermal endurance.\(^{69}\) Besides, a series of cellulose-based battery nanofiber membrane enhanced with alkali-treated polysulfonamide fibers or functionalized with polypryrole and polyaniline have been successfully implemented and exhibit improved electrolyte wettability and superior electrochemical performances.\(^{43,70,71}\) Notably, in addition to excellent heat tolerance, cellulose-based separators hold one of prominent advantages in narrow nanopore distribution. When used for Li protection in a Li metal battery, cellulose-based membranes can effectively regulate the Li ion flux on the surface of Li anode and contribute to the safe Li plating with a dendrite-free morphology.\(^{72,73}\)

2.3 Artificial solid electrolyte interphase layers

Li metal has been regarded as the most promising anode alternative owing to its ultrahigh theoretical capacity and
negative electrode potential. However, instable solid electrolyte interphase (SEI) as well as the resultant Li dendrite growth seriously cause safety concerns, once Li metal is implemented as an anode in a battery. In the view of being able to manipulate the mechanical properties by macromolecular chains as well as regulate Li ion flux and distribution by lithiophilic functional groups, biomass materials (eg, cellulose, starch, protein, lignin, chitin, polysaccharide) have been integrated into artificial SEI protective films for restraining Li plating and stripping to suppress the dendrite propagation. Recently, a natural agarose biopolymer film (AG) was reported as an enabling SEI protective layer. The assembled Li–Cu half cell with AG protective layer displayed higher Coulombic efficiency of 98% even after 100 cycles, much better than that with bare Cu foils (< 40% after 50 cycles), indicative of the excellent structure stability of bio-based AG films against Li metal anode. More importantly, the AG film was also capable of high ionic conductivity and good elasticity, which significantly promoted fast Li-ion transfer and effectively accommodated Li dendrite growth during repeated discharging/charging processes (Figure 3A,B). A kind of cellulose fiber paper membranes was also successfully adopted to protect Li metal. In most cases, due to the evident protuberances on electrodes surface, the electric field intensity around these tips was enlarged, intensifying the inhomogeneous Li ion distribution, which usually was called “tip effect” (Figure 3C). For the cellulose paper protective layer, there were abundant lithophilic polar groups such as –OH and –C=O at the cellulose backbone, which inhibited the movement of Li ions toward the protrusions of electrode, mediating the distribution of Li ion and finally the homogeneous electric field on the surface of Li metal (Figure 3D). Owing to eliminated nonuniform electric field, Li deposition at local area as a result of tip effect was suppressed and thus smooth Li plating without Li dendrite growth was achieved.

Owing to the advantages of improving the mechanical properties and enhancing the electrolyte reserve by intermolecular or intramolecular cross-linking, natural polymers with low cost, renewability, and environmental friendliness have been widely used as binders, gel electrolyte matrix, or separators in advanced Li batteries. Particularly, when considering the regulating distribution of Li ion and thus the electric field by surface polar groups, natural polymers have the ability of mediating the Li deposition behavior and inhibiting the growth of Li dendrites. However, the compatibility of natural polymers with organic electrolyte solutions should be considered. The use of functional natural polymers, natural polymers-inorganic composites, electrolyte additives, and concentrated electrolytes, are considered to be effective strategies to enhance compatibility and thus stabilize the natural polymers-based protective layers/electrolyte interface. Furthermore, the relative chemical/electrochemical stability of natural...
polymers to Li metal need to be considered due to the high reactivity of Li metal.

3 | BIOMASS-DERIVED CARBONACEOUS MATERIALS

3.1 | Conductive carbon host

The fundamental challenges, including huge volume deformation of electrode, low electrical conductivity, and dissolution/diffusion of electrochemical intermediates, as well as dendrite propagation on metallic Li anode, plague advanced Li batteries with severe electrode pulverization, poor electron conduction, active materials loss and unstable electrolyte/electrode interface, which lead to poor Coulombic efficiency and rapid capacity decay, and therefore limit their practical realization. Nanostructured carbonaceous materials reveal significantly potentials to mitigate the above problems in a Li battery owing to their unique structural merits in nano spatial confinement, long/short-range conductivity, as well as surface/interfacial property.84-87

Biomass, holding the traits of high carbon content has been regarded as a very suitable raw precursor to prepare porous carbon materials for advanced Li-based batteries.88-95 In fact, biomass materials are endowed with unique structures, which consequently confer biomass-derived carbonaceous materials with versatile microstructures and special characteristics.4,96

3.1.1 | Structure-oriented host

Host materials featuring highly porous structure and large surface area can allow sufficient space to encapsulate the electrode active phase and abundant active electrode/electrolyte interface for promoting reversible electrochemical conversions. Meanwhile, high porosity can enhance Li ion transport kinetics within the electrode, which should also be emphasized in the design of conductive carbon matrix.96-99 Zhong and coworkers developed a puffing process to produce porous carbon (PRC) as a sulfur cathode host derived from rice (Figure 4A).100 The resultant materials exhibited a unique porous microcellular structure, providing enough space to homogeneously confine active
electrodes, which endowed the PRC/S electrode with high reversible capacity and excellent cycle life (Figure 4B,C).

More interestingly, the inherent structure and morphology in biomass sources can always be well retained after treatment. In nature, some special biomaterials, such as cotton, kapok, bacterial cellulose, and so forth, intrinsically preserve their 3D cross-linked fibrous structure. When used as raw materials, their carbon-derived analogues can inherit the interconnected fiber structure, and thus benefit from high porosity as well. Recently, 3D hollow carbon fibers (3D-HCF) containing metallic lithium hosts were demonstrated inherited from fibrous cotton (Figure 4D,F). Such special structure of 3D-HCF effectively regulated Li ion deposition behavior and confined Li growth within the interspace among the fibers, consequently realizing smooth deposition without discernable dendrites at high deposition capacity of 4 mAh cm$^{-2}$ (Figure 4G).

Tortuosity of pores in carbon hosts also play a critical role for electrolyte permeation or ion transportation in the electrodes. The randomly distributed pores without interconnected and perforative structure give rise to a long ion diffusion distance or decreased ion transport channel, which lead to high ion migration resistance. Especially in electrodes with high loading, this adverse effect will be amplified owing to the higher response to ion transportation. Therefore, the oriented design of interconnected architecture with low...
tortuosity electrode should be strongly encouraged. Enlightened by well-aligned channel structure within natural wood for water and ion transport, Hu and coworkers developed a 3D aligned porous carbon matrix derived from natural wood to accommodate cathode electrocatalysts in Li–O2 batteries (Figure 5A).108 This carbonized wood (CA-wood) perfectly inherited the well-distributed channel structure with low tortuosity from wood, which considerably shortened the ion transport pathways and facilitated oxygen diffusion into electrode (Figure 5B). With such design, the assembled Li–O2 battery with wood-based cathode demonstrated a high specific area capacity of 8.58 mAh cm\(^{-2}\) at 0.1 mA cm\(^{-2}\) and superior cycling performance. Besides, oriented design strategy of porous electrode has also been implemented in a working Li metal battery, which not only alleviates volume expansion and enables a uniform Li ion flux, but also guides the Li deposition within channels and impedes dendrite growth.109

To further rationally construct electrodes with interconnected conductive network, bio-derived carbon frameworks combined with other low-dimensional (1D and 2D) conductive fragments (such as graphene, carbon nanotube, etc.) are desired.110-114 The 2D nanosheet or 1D nanotube morphology are beneficial for fast electron transfer; and meanwhile, the interconnected framework promotes rapid ion transport, thus facilitates reversible electrochemical redox.

### 3.1.2 Affinity-oriented host

The dissolution/diffusion of electrochemical intermediates into electrolyte, such as the shuttle effect of Li polysulfides in a working Li–S battery, generally results in low Coulombic efficiency and poor cycling stability. The shuttle of electrochemical intermediates can be hindered by the confinement inside a porous carbon host. Factually, the diffusion of intermediates driven by concentration gradient cannot be completely avoided thermodynamically. Thus, reducing the concentration of intermediates in the electrolyte should be one of effective strategy to mitigate the shuttle effect. However, in conventional Li–S batteries, the host material of sulfur is usually a conductive carbon. The great difference in polarity leads to the weak chemical interaction between nonpolar carbon and polar Li polysulfides, which reduces the retention effect of conductive skeleton on Li polysulfides and consequently strengthens the shuttle effect of polysulfide within electrolyte. Recently, researchers found that introducing heteroatoms (eg, nitrogen, oxygen, or sulfur) into graphene can render tunable electronic properties and thereof enhance the affinity of carbon surface toward polar polysulfides.115-117 Therefore, the regulation of surface affinity by heteroatom-doped has been widely implemented to the design of host materials for trapping the dissolved electrochemical intermediates.

Normally, biomass raw materials possess abundant heteroatom-containing functional groups in macromolecular backbone, which endow their carbonaceous derivative with the possibilities of in situ doping of functioned heteroatoms during carbonization.118-120 Such a self-doped strategy alters the surface properties of the carbon host, which ensures strong electrochemical affinity toward electrochemical intermediates and therefore increases the utilization of active phase and final the

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**FIGURE 5** Structure-oriented design of bio-based hosts. A, Schematic diagram of the Li-O2 batteries with the CA-wood/Ru cathode. B, The cross-section SEM image of CA-wood after carbonization. Reproduced with permission. Copyright 2018, Wiley-VCH108
capacity retention. To date, a mass of biomass-based heteroatom-contain carbon materials derived from hair (Figure 6A), soybeans, red algae (Figure 6B,C), coffee waste, chrysanthemum, and so forth, have been developed and exhibit enhanced electrochemical performances. Additionally, in a working Li metal battery, the heteroatom-containing polar carbon surface demonstrates also affinity toward metallic Li, which can guide a uniform nucleation of lithium.

Besides heteroatom-doping, the incorporation of biomass-derived carbon hosts with affinity species (such as metal, metal oxide, and metal sulfide) is also an effective strategy to trap electrochemical intermediates or Li ions within the electrolyte. In a typical example, Tao et al demonstrated a bamboo-derived 3D hierarchical porous carbon host decorated with lithiophilic ZnO quantum dots (ZnO@HPC), which can induce preferential Li deposition within the porous scaffold, rendering a dendrite-free Li metal anode (Figure 6D-F).

Such multifunctional carbon host derived from biomass are anticipated to be the most available avenue for advanced Li-based batteries. However, different energy devices have different critical issues and challenges, thus the requirements of carbonaceous hosts with respect to structure or physiochemical properties may be completely different. Therefore, understanding of the basic relationships between the electrochemical performance of batteries and structure and functions of conductive carbon hosts is imperative, which is the precondition for guiding the rational design of the multifunctional-oriented biomass-based carbon hosts in advanced Li-based batteries.

### 3.2 Functional interlayers

Except the aforementioned electrode hosts, bio-based carbonaceous materials can also be used as interlayers to address the critical issues in Li-based batteries, especially to retard the diffusion of polysulfide intermediates in Li–S systems. In general, carbonaceous interlayers are established to contact with cathode, which function as not only conductive matrixes to improve the conductivity of whole cathode but also active surfaces to facilitate the reversible electrochemical conversion of active species in electrolyte. As the bio-based interlayers indicate similar effects as electrode conductive scaffolds, the oriented strategies toward electrode hosts can also be implanted into the interlayers engineering.

**FIGURE 6** Affinity-oriented design of bio-based hosts. A, Schematic illustration of sulfur/porous N-doped carbon derived from hair. Reproduced with permission. Copyright 2015, The Royal Society of Chemistry. B, The synthesis process and (C) cyclic performance of sulfur-doped carbon from red algae. Reproduced with permission. Copyright 2019, Elsevier. D, Schematic diagrams of ZnO@HPC for uniform Li deposition. E, TEM image of ZnO@HPC material. F, SEM image of ZnO@HPC scaffold after 80 cycles at 1 mA cm⁻². Reproduced with permission. Copyright 2017, Elsevier.
Enlightened by inherent moisture retention properties of natural leaves, Manthiram group reported a separator interlayer based on a carbonized leaf (CL, Figure 7A). After carbonization, the CL inherited the pore-size gradient structure: The macroporous network composed of pores and stomata can serve as reservoirs for electrolyte (Figure 7B,C); the dense layer with micro/mesopores can function as a blocking layer to impede the diffusion of dissolved polysulfides across separator (Figure 7D). In view of the superiorities of bio-derived functional materials in immobilize polysulfides, lotus plumule, fructose, banana peel, and so forth have also been highlighted as promising resources of producing porous carbonaceous interlayer to suppress polysulfides intermediates in Li-S batteries.

Other structure-oriented biomass-based carbonaceous materials are also considered as effective functional interlayers toward polysulfide immobilization. Gu et al proposed an interwoven carbon fiber (BCF) membrane obtained from carbonized bamboo as a functional interlayer (Figure 7E). The BCF membrane offered ample macro/micropore structures for fast transportation of electrolyte and ion, and sufficient active surface and efficient conductive networks for the deposition of sulfur active materials (Figure 7F,G). A series of conductive bacterial cellulose-based nanofibers membrane, cassava-derived high conductive carbon sheet, and porous carbonized graphene-embedded fungus membrane were also implemented for achieving high stable Li–S batteries. Meanwhile, heteroatom-doped carbon within interlayer can also play an important role in improving sulfur electrochemical behavior as the heteroatom dopants function as active sites for polysulfide adsorption and electrocatalysis. Thanks to high-content sulfur- and oxygen-containing groups, Zhou and coworkers reported a sulfur-doped microporous carbon (SMPC) interlayer by the carbonization of luffa sponge. Unique microporous framework and in situ S-doping in SMPC effectively enabled rapid ion transport and powerful adsorption for dissolved polysulfides, consequently rendering superior rate capability and cycling stability in Li-S batteries.

4 CONCLUSIONS AND PERSPECTIVES

Sustainable biomass materials hold advantages in terms of versatile microstructures, diverse composition, ample

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**FIGURE 7** A, Schematics of the leaf structure and the CL polysulfide diffusion interlayers. SEM displayed the pore distribution gradient in CL interlayer of (B) surface of electrolyte reservoir, (C) cross-section of the CL, and (D) surface of polysulfide blocking-film. Reproduced with permission. Copyright 2014, Wiley-VCH. C, Schematic illustration of the Li-S battery with BCF membrane as interlayer; (F) SEM image and (G) conductive properties of BCF interlayers. Reproduced with permission. Copyright 2015, The Royal Society of Chemistry.
function groups, water-solubility as well as environmental benefits, which render multifunctional biopolymers and carbonaceous derivatives suitable for high-energy Li-based batteries.

In this perspective, on account of the special properties and functionalities of biomass materials, we highlighted the design principles of biological derivates in addressing critical problems in Li battery systems and promoting the sustainability of energy devices: (a) accommodating volume changes; (b) suppressing dendrite growth; (c) improving charge transportation; and (d) capturing electrochemical intermediates. Based on the breakthrough progresses achieved, future research directions on biomass materials for boosting advanced Li-based batteries are suggested herein (Figure 8).

1. Fundamental understanding of biopolymers: Owing to the complexity of composition in raw biomass precursors, the knowledge of biological materials parameters such as macromolecular structure, molecular weight, and polymerization degree as well as composition are key to achieve reproducible materials.

2. Consistency of raw biomass material: Biomass is a mixture of analogues with different molecular weights, so it is difficult to guarantee the consistency of biomass materials used in each experiment. Until now, the composition, structure, and properties of raw materials can only be qualitatively analyzed due to the difficulties in obtaining a pure substance. As such, determination and quantitative analysis of the key components should be particularly important. In addition, physicochemical stability of raw biomass materials and their detection methods also need to be taken into consideration.

3. Rational design of functional materials: Different battery systems need different functional structured materials. Rational design of appropriate functional structure materials is key to boost the electrochemical performance of advanced Li-based battery systems. However, current scientific research mainly relies on empirical strategies of trial and error, which is insufficient and time-consuming. More importantly, due to the species diversities of biomaterials, the screening criteria of available bio-based precursors is still difficult to be defined accurately, and become one of the restrictive problems in this field. Therefore, artificial intelligence assistance on achieving an efficient screening of structure-oriented, morphology-oriented, surface/interface chemistry-oriented, and function-oriented biomass precursors, and their derived functional materials should be considered.

4. Facile preparation processes: The refinement or conversion of biomass resources into available bio-based materials has low efficiency and yields. Generally, highly conductive bio-derived carbonaceous materials are obtained at an excessively elevated carbonization temperature (normally >800 °C), which is energy-consuming and environmentally inefficient. More importantly, the frequently applied methods can only generate materials with uncontrollable pore-size distribution. Therefore, exploiting facile, cost-effective, as well as controllable preparation methods is imperative. The incorporation of pretreatment process or template strategies, development of advanced thermal management systems with gradient heating protocols, and regulation of appropriate heat treatment atmosphere such as reducing or inert gases, are promising methods to manufacture the biomass-derived ecomaterials.

5. Advanced characterization techniques: At present, there are lack of powerful and effective characterization techniques to track the evolution behaviors of bio-based materials in a working battery that easily lead to inscrutable understanding of how the biological polymer or bio-derived materials functions in a working battery. In addition, the diverse structure and complex properties of biomass materials further exacerbate the difficulties in detecting their active state under battery operation. Advanced in situ characterization measurements, such as X-ray absorption, microscopy, and spectroscopy, are required to discover the working states and evolution behaviors of bio-based functional materials, which will give fresh insight to the oriented-design of biomass materials toward high-energy Li-based batteries.
6. Greenness and sustainability of energy devices: Green energy and devices are the ultimate goal of society development. It is worthy to consider the greenness degree of batteries including all battery components during the whole cycling life, which is of significance to improve the greenness in energy storage process. Moreover, artificial intelligence design of energy environmental materials is also an effective avenue for resourcilation of energy sources and devices.

To conclude, the implantation of biomass sources into battery systems is helpful to not only push forward green energy at the resource but address the critical issues in Li-based batteries, consequently rendering high-energy-density and high-safety battery systems. Future research efforts should be focused on the efficient bio-based precursors screening, multifunction-oriented biomass materials, as well as in-depth structure evolution and fundamental mechanism analyses. Furthermore, the development strategies of biomass materials from single battery component to whole battery components have received great interests for promoting sustainability of energy devices. In light of these, we hope that this perspective can guide an oriented-strategy of battery materials based on biomass resources, and also give new inspirations on rational design of functional biomass materials toward high-energy-density Li-based batteries and other advanced energy storage systems.

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ORCID
Qiang Zhang https://orcid.org/0000-0002-3929-1541

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AUTHOR BIOGRAPHIES

Jia Liu received B.S. degree from Shandong Normal University in 2010, and then obtained her M.S. and Ph.D. degree from Beijing Institute of Technology in 2014 and 2018, respectively. She currently is a faculty at College of Materials Science and Technology in Beijing Forestry University and a visiting scholar at Tsinghua University. Her research interests are focused on the design of biomaterials, as well as their applications in energy storage devices.

Tong-Qi Yuan graduated from Zhengzhou University with a B.S. in Chemical Engineering in 2007, and Beijing Forestry University with a doctorate in Chemical Processing of Forest Products in 2012. Since July 2012, he has worked at the Beijing Key Laboratory of Lignocellulosic Chemistry at Beijing Forestry University. He has focused on the pre-treatment, efficient fractionation, and high-value utilization of lignocellulosic materials for sustainable biofuels and biomaterials.

Qiang Zhang received his bachelor and Ph.D. degree from Tsinghua University in 2004 and 2009, respectively. After a stay in Case Western Reserve University, USA, and Fritz Haber Institute of the Max Planck Society, Germany, he was appointed as a faculty in Tsinghua University at 2011. He held the Newton Advanced Fellowship from Royal Society, UK and the National Science Fund for Distinguished Young Scholars. His interests focus on energy materials, includes Li-S batteries, Li metal anode, 3D graphene, and electrocatalysts. More details can be found in his Research ID (B-1799-2012) and ORCID (0000-0002-3929-1541).

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