Energy Materials

Review

Structures, performances and applications of green biomass derived carbon in lithium-ion batteries

Wanran Lin[1,2,3,#], Shijiao Zhao[1,2,3,#], Binda Lu[1,2,3], Feng Jiang[1,2,4], Zhouguang Lu[2,3,*] iD, Zhenghe Xu[1,2,4,*] iD

[1]Shenzhen Key Laboratory of Interfacial Science and Engineering of Materials, Southern University of Science and Technology, Shenzhen 518055, Guangdong, China.
[2]Key University Laboratory of Highly Efficient Utilization of Solar Energy and Sustainable Development of Guangdong, Southern University of Science and Technology, Shenzhen 518055, Guangdong, China.
[3]Department of Materials Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, Guangdong, China.
[4]Advanced Materials Innovation Center, Jiaxing Research Institute of Southern University of Science and Technology, Jiaxing 314031, Zhejiang, China.
[#]Authors contributed equally.

*Correspondence to: Prof. Zhenghe Xu, Department of Materials Science and Engineering, Southern University of Science and Technology, 1088 Xueyuan Avenue, Nanshan District, Shenzhen 518055, Guangdong, China. E-mail: xuzh@sustech.edu.cn; Prof. Zhouguang Lu, Department of Materials Science and Engineering, Southern University of Science and Technology, 1088 Xueyuan Avenue, Nanshan District, Shenzhen 518055, Guangdong, China. E-mail: luzg@sustech.edu.cn

Abstract

Most countries worldwide have committed to reaching carbon neutrality by the end of the century, with the aim to achieve Net Zero before 2060. To reduce the dependency of energy demands on fossil fuels, use of renewable energy and its gradual transition into the overall energy portfolio becomes increasingly critical. The solar and wind energy accounts for a major part of renewable energy. Considering unstable and noncontinuous production of electricity from these two sources, energy storage becomes essential. Lithium-ion batteries (LIBs) have become the most favorable choice of energy storage due to their good electrochemical performance (high capacity, low charge leakage and good cycle performance) and safety, in particular for portable (3C products, electric vehicles and drones) and stationary applications as well as for emergency electricity supply. However, the specific capacity of graphite, the most common commercial anode material, is reaching its theoretical limit, posing great challenges for improving the overall capacity of LIBs. It is therefore necessary to develop anode materials of higher capacity and better cycle performance. Biomass-derived carbon materials are ideal candidates for further enhancing the performance of LIBs due to their special microstructures, functional diversity and easy structure regulation. Most of

these materials can reach capacities exceeding 500 mAh g^{-1}, even the best for more than 1,000 mAh g^{-1} combined with other anode materials. This review provides an in-depth analysis of diverse carbon sources derived from biomass, categorized based on their distinct structural characteristics, with the focus on evaluating the current roles and bottlenecks of carbon as a component of the electrode materials used in LIBs. The failure mechanisms associated with biomass-derived carbon in LIBs are summarized, with potential solutions to these issues being proposed. The potential challenges and prospects for biomass-based LIBs are identified and thoroughly discussed. Overall, this review aims to serve as a resource for the strategic design and advancement of carbon-based materials, to achieve next-generation LIBs of superior performance.

INTRODUCTION

As the global climate continues to change, the reduction of carbon emissions to the atmosphere is urgent. The concept of carbon neutrality has thus become the focus globally as it can promote global green production and sustainable living conditions. In recent years, an increasing number of countries and governments worldwide have made carbon neutrality a national strategy to achieve the goal of net zero carbon emissions[1,2]. The development of new energy sources, the implementation of energy transition, the reduction of fossil fuel-based energy production and consumption, and the construction of green and low-carbon energy systems are important measures to reduce carbon dioxide and other greenhouse gas emissions for carbon neutrality[3]. Renewable energy is one of the most important pillars of the energy transition to reach the carbon neutrality goal[4].

Biomass energy is gaining strong traction as the renewable energy with the most potential to disrupt the three traditional energy sources in China, i.e., coal, oil and natural gas[5]. Promoting the utilization of biomass resources with abundant reserves and green environmental protection is an effective technical way to meet the current needs of environmental protection, energy conservation and low-carbon economy for ultimate carbon neutrality[6].

Biomass materials mainly include plants that directly use photosynthesis to synthesize organic matter and also involve organic matter formed by indirect use of photosynthetic products[7]. The biomass materials have all kinds of natural structures, components and biophysical/biochemical properties[8-10]. At present, the efficient and comprehensive utilization of biomass is distributed in energy, ecological agriculture, environmental restoration and building materials[11-14]. In the future, the applications of biomass materials are expanding towards the direction of high-value propositions, especially biomass-based energy storage materials.

Lithium-ion batteries (LIBs), the most popular energy storage devices, play a crucial role in the energy transition and carbon neutrality. They have many attractive properties such as light weight, high energy density, small-scale size, few memory effects, long cycle life, and low pollution[15-20]. The performance of LIBs has thus been widely recognized in the market. At present, commercial LIBs can normally reach more than 1,000 cycles, with an initial Coulombic efficiency (ICE) of more than 90% and an energy density of 250 Wh kg^{-1}, which is projected to reach 300 Wh kg^{-1} by 2030[21]. In addition to the improvement of LIBs performance, significant cost reductions have also helped LIBs to increasingly dominate the energy storage market. Although early LIBs were very expensive and only used in high-value communication electronics, LIBs today are used across a wide range of portable and stationary energy storage applications. Taking the price of LIBs used in the electric vehicles as an example, the average price of LIB packs decreased from

1,124 $/kWh in 2010 steadily to 152 $/kWh in 2020, representing an overall decrease of 86% in price. The sharp drop in price proves the maturity and progress of LIB technology[22].

Graphite, which is by far the most commonly used commercial anode material in LIBs, is facing the bottleneck of limited theoretical capacity (372 mAh g^{-1}), making it unable to meet the current and future market demands. New types of carbonaceous materials of diverse structures have been proposed to improve electrochemical properties, opening up new ideas for the future development of LIBs. Biomass materials, as an abundant natural carbonous source, can be used to prepare many kinds of carbon materials[23]. Biomass-derived carbon materials (BCMs) have been used in anode and cathode materials of LIBs due to their diverse structures and shapes, such as spherical, tubular, flaky, fibrous, *etc.*[24,25].

At present, some reviews have introduced BCMs used in energy storage, energy conversion, catalysts, and other applications[26,27]. Most of them focused on lithium-ion and sodium-ion batteries, supercapacitors, and oxygen reduction reactions. Jin *et al.* comprehensively summarized recent advances in the structural design and fabrication of BCMs, providing insights into developing high-capacity biomass-derived supercapacitors[28]. Zhang *et al.* compared various BCMs, focusing on the advancement of biomass-derived carbon nanomaterials[29]. The advantages and disadvantages of various synthesis methods and the mechanism of biomass conversion to carbon were discussed to develop high-quality carbon materials. Moreover, new applications of BCMs in energy and environmental applications were reviewed. A systematic, specific review on the use of biomass-derived carbon in battery electrodes is lacking, and only a few articles are available in open literature on the working principles and failure mechanisms of biomass-derived carbons in LIBs[30,31]. Analyzing and understanding these mechanisms play a key role in promoting the development of biomass-derived carbons in batteries and supercapacitors. This review aims to summarize and provide potential solutions to failure modes of biomass-derived carbons used on LIBs.

Specifically, this review presents an in-depth analysis of the latest trends in the application of BCMs in LIBs. The review includes an illustration of the role and bottlenecks that carbon plays in electrode materials and a detailed introduction of BCMs based on their different structures, as illustrated in Figure 1. The classification of BCMs in Figure 1 and in part "STRUCTURES AND PERFORMANCES OF BCMS IN LIBS" is arranged as porous, fibrous, spherical and layered structures, according to the number of related researches. The review highlights the recent advances and performance metrics of different types of BCMs. A key emphasis is placed on the impact of the micromorphology and composition of biomass-derived carbons on their electrochemical performance. The operational principles of biomass-derived carbon, along with an analysis on its failure mechanisms and potential solutions, are discussed to provide directions in further improving the performance of LIBs. The review also addresses the existing challenges and future prospects for biomass-based LIBs. The relationship between biomass precursors and the resulting structures of biomass materials is highlighted, providing a valuable reference for the rapid identification and utilization of biomass precursors to achieve specific structures.

CURRENT ROLES AND BOTTLENECKS OF CARBON IN LIBS

To realize green, low-carbon and sustainable development goals, LIBs are of great significance and considered to be one of the most promising and greenest energy storage devices, with extensive uses in portable electronics, power tools, electric vehicles and more recently in eVTOLs (electric vertical take-off and landing aircraft)[25,32-35]. LIBs consist of anodes, cathodes, electrolytes, separators, binders and additives[36]. With the electrodes playing a decisive role, electrode materials have, over the past three decades, been continuously studied and optimized to improve energy density, cycle life and safety and to achieve higher charge and discharge currents. Carbon plays an important role in the anode and cathode
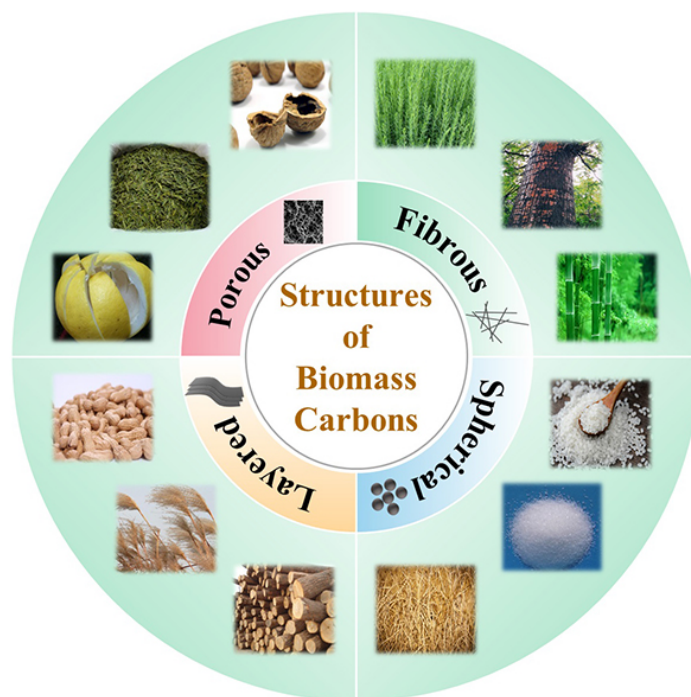
Figure 1. Structures of biomass carbon from different biomass sources.

electrodes, as it is usually used as a modified additive to protect the active material and increase electric conductivity. However, at present, there are still some bottlenecks that must be overcome to improve energy density, safety and cycle life in order to meet the market requirements of the future development[37-39]. Therefore, identifying the roles and bottlenecks of carbon in the electrode materials is of great importance to further improving the performance of LIBs.

Carbon for cathode materials
Several cathode materials such as $LiCoO_2$, spinel $LiMn_2O_4$, $LiNi_{0.8}CO_{0.15}Al_{0.05}O_2$, $LiNi_{0.33}CO_{0.33}Mn_{0.33}O_2$ and olive $LiFePO_4$ have been widely used. However, the specific capacity of all these materials can only reach 120-200 mAh g^{-1}[40]. There are also problems for these cathode materials, including transition metal dissolution, low conductivity, poor cycle stability and fast voltage fading[41-44]. Therefore, researchers tried to solve these problems through surface coating of carbon materials on corresponding active cathode materials[42,45-47]. In particular, carbon as the coating can not only protect the cathode materials but also improve the electronic conductivity and rate capabilities of LIBs[48]. Zhang *et al.* demonstrated that carbon nanotube coatings (CNTs) on $LiMn_2O_4$ exhibited a better electrochemical performance and longer cycle life than $LiMn_2O_4$ without a coating[49]. The coating layer decreased the contact area between the cathode material and electrolyte, which prevented the dissolution of Mn. Moreover, the coating also increased conductivity. Luo *et al.* stated that $LiFePO_4$ nanoparticles coated with CNTs exhibited good conductivity due to the formation of a conductive network by CNTs through intimate connections with $LiFePO_4$ nanoparticles[50]. LIBs assembled with such cathode materials exhibited an excellent cycling performance with a capacity loss of less than 10% after 450 cycles at 10 C. In addition, graphene is a desirable additive that can be divided into single-, double- and multi-layer types. Regardless of the type, the number of layers, in general, is less than 10. For this reason, we collectively referred to them as graphene. These graphene materials share common features, including large specific surface areas (SSA), high ion and electron carrier mobility, and unique physical and chemical properties. As an additive, these characteristics make graphene

highly desirable for improving the performance of anode materials. Yang *et al.* used reduced graphene oxide as the coating material to improve the electrochemical properties of $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ (RGO-NCM)[51]. However, as the number of cycles increases, the graphene falls off the surface of the active material powders, resulting in the failure of the coating. Therefore, in order to make graphene more tightly and evenly wrapped around the positive electrode material and increase the interface conductivity, epoxy-functionalized silane (KH560) was added between the graphene and $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ particles to build a dual-function coating (RKNCM1), which improved the capacity retention rate, rate performance and high-temperature performance of cathode materials, as illustrated in Figure 2.

Carbon for anode materials

Graphite is one of the most widely used anode materials, and its layered structure ensures the smooth and stable insertion and desertion of lithium ions. It has a wide range of sources and is relatively easy to produce industrially while being low-cost, safe, and environmentally friendly[52-55]. However, LIBs with graphite-based anodes have inherent shortcomings of low specific capacity, low Coulombic efficiency (CE) and poor rate performance[56-58]. Although graphite is still being further developed and optimized, its electrochemical performance has almost reached the theoretical limit[59]. Scientists have developed alternative anode materials of higher theoretical capacity, such as graphene, silicon, germanium, metal oxide, alloy, metal-organic frameworks, and so on, along with changing the structure and size of anode materials to further improve the electrochemical performances of LIBs[60-66]. However, carbon materials are still often used as additives to protect active anode materials and/or form a conductive framework to increase the electric conductivity.

Some new anode materials have such problems as volume expansion and poor electrical conductivity[67]. Carbon materials are often combined with these anode materials to improve electrical conductivity and reduce stress inside the electrode by providing a buffer volume, thus enhancing electrochemical characteristics. Lee *et al.* prepared anode materials by combining ultrafine TiO_2 nanocrystals (TiO_2 NP) with a macroporous three-dimensional graphene network (3D PG) to achieve a faster electron transfer [Figure 3A and B][59]. The binder-free 3D PG, on the other hand, has a low sheet resistance and can be made in direct contact with the current collector for ultra-fast electron transfer, providing a good cycle performance of a CE value close to 100% after 10,000 cycles [Figure 3C]. Zhang *et al.* proposed a hierarchical tubular structures constructed by carbon-coated ultra-thin tin oxide nanoplates (SnO_2@C-HTs) [Figure 3D], combining the advantages of low-dimensional ultra-thin nanoplates, hollow tube structures and nanocarbon coatings[68]. Anodes of SnO_2@C-HTs exhibited a higher specific capacity and better cycle stability than SnO_2 without ultra-thin carbon coatings [Figure 3E and F].

Silicon is seen to be the most promising alternative to graphite as an anode material because its high specific capacity is more than ten times that of graphite. However, the problem of large volume changes of silicon during charging and discharging needs to be solved. Structural design or carbon coating are the most popular methods and can greatly alleviate the problem. Liu *et al.* designed anode materials of pomegranate structure, using a conductive carbon layer encapsulating a single silicon nanoparticle and then a thicker carbon layer encapsulating the ensemble of these nanoparticles [Figure 3G and H][69]. Such design resulted in a high CE, high volumetric capacity (1,270 mAh cm^{-3}) and stable cycling over 1,000 cycles [Figure 3I].

STRUCTURES AND PERFORMANCES OF BCMS IN LIBS

After understanding the roles and bottlenecks of carbon in the electrode materials of LIBs, the importance of BCMs is more incisively reflected. In recent years, many studies on the application of biomass-derived carbon in LIBs have shown its superiority. These carbons display higher capacity and better cycle stability
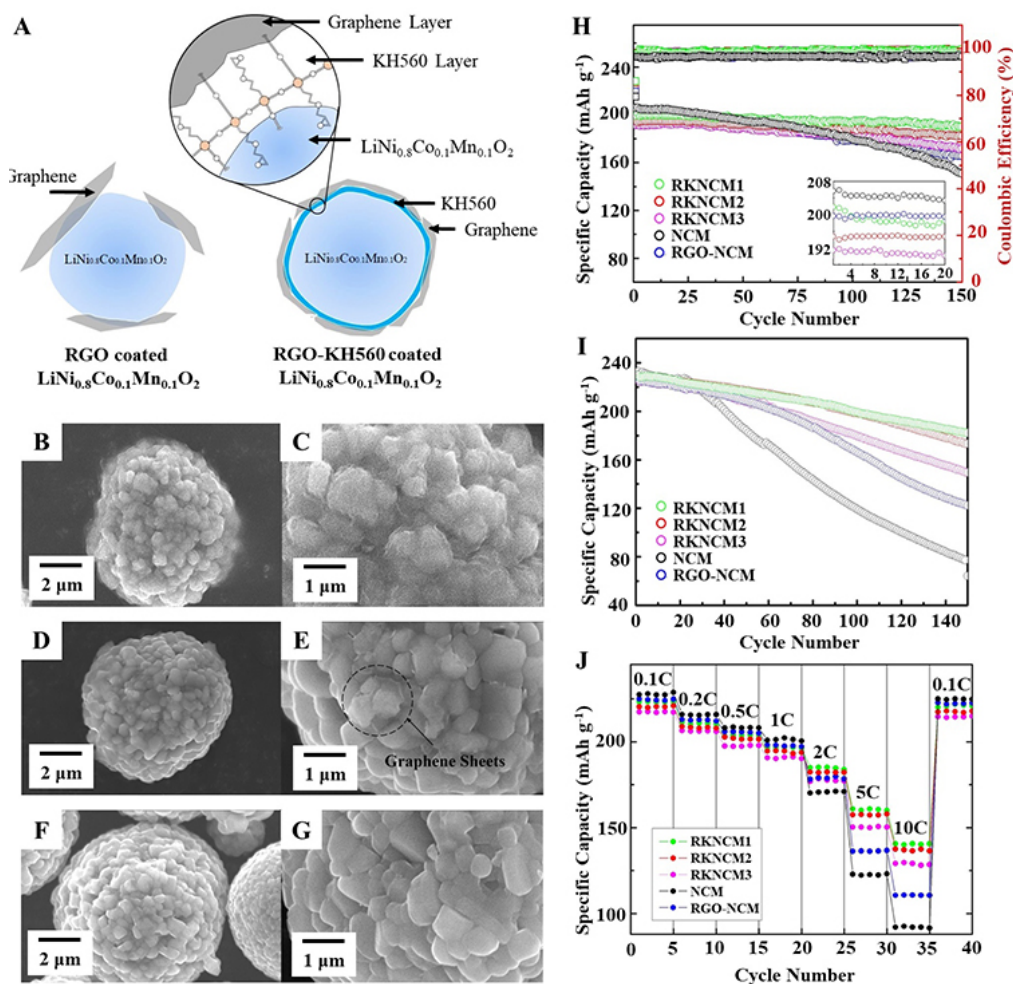
Figure 2. (A) Structure schematics of RGO- and RGO-KH560-coated $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ particles; (B and C) SEM images of RKNCM1 particles; (D and E) SEM images of RGO-NCM particles; (F and G) SEM images of NCM particles; (H) Cycling performance at room temperature of RKNCM1, RGO-NCM and NCM particles; (I) Cycling performance at 55 °C of RKNCM1, RGO-NCM and NCM particles; (J) Rate performance of RKNCM1, RGO-NCM and NCM particles. These figures are quoted with permission from Yang *et al.*[51].

due to their rich and diverse structures, more lithium-ion storage sites and channels for fast lithium-ion transport. Table 1 summarizes the application of various BCMs in the electrodes of LIBs and their electrochemical properties.

This section delves into the intrinsic macroscopic and microscopic characteristics of BCMs employed as electrode materials for LIBs, focusing on the latest developments in utilizing biomass-derived carbons of diverse structural forms. The influence of various processing techniques on the resulting structures of biomass-derived carbon will be highlighted. An in-depth discussion on how the structural attributes of different types of biomass-derived carbons directly correlate with their electrochemical performance will also be presented, aiming to elucidate the intricate relationship between structural design and the functional capabilities of these carbons in the context of LIBs.

Biomass materials have various natural structures that are highly desirable for design of tailored carbon materials. These structures can be preserved in the biomass-derived carbon after processing, including porous, fibrous, spherical and layered carbon. Numerous studies have explored biomass-derived carbons of

Table 1. Electrochemical properties of various biomass carbon materials as electrodes in LIBs

	Products	Biomass materials	Cell types	MR* (wt.%), loadings (mg cm^{-2})	ICE (%)	ICDC** (mAh g^{-1})	Capacity retention	Year, references
Cathode	LFP/rGO	Coconut shell	CR2016	90:04:06, NA	NA	131	CR$_{70}$*** 98%	2021[70]
	LVP-NC	Waste reed	CR2016	80:10:10, NA	NA	132	CR$_{100}$ 94%	2020[71]
	C4Q/PPL	Physalis Peruviana L	CR2032	90:00:10, NA	100	437	CR$_{100}$ 228 mAh g^{-1}	2019[72]
	Biomass-carbon@FeS$_2$	Auricularia auricula	CR2032	80:10:10, NA	92	949	CR$_{80}$ 850 mAh g^{-1}	2018[73]
	3DMCN-LFP/Fe-NSs	Alginate	CR2032	80:10:10, NA	100	177	CR$_{70}$ 176 mA h g^{-1}	2016[74]
Anode	L-900	Lotus root	NA	80:10:10, NA	80	560	CR$_{500}$ 355 mAh g^{-1}	2023[75]
	CuCo$_2$O$_4$ nanowires/ corn pod	Corn pod	CR2032	NA, NA	85	1,314	CR$_{250}$ 887 mAh g^{-1}	2023[76]
	PTA-700	Tannic acid	CR2023	80:10:10, NA	53	603	CR$_{100}$ 535 mAh g^{-1}	2023[77]
	NP-BC	Bagasse	CR2025	80:10:10, NA	51	2,348	CR$_{50}$ 816.36 mAh g^{-1}	2021[78]
	UAWP char	Wastewater-treatment filamentous algae	CR2025	80:10:10, 1.2	49	789	CR$_{100}$ 358 mAh g^{-1}	2021[79]
	Biocarbon	Tamarind plant seeds	CR2032	80:10:10, 0.9-1.3	40	1,037	CR$_{100}$ 86%	2021[80]
	WGF-KOH	Wax gourd flesh	CR2032	83:00:17, NA	NA	777	CR$_{200}$ 64%	2021[81]
	PSCN	Mustard seed husk	CR2032	80:10:10, 2.0 mg	75	617	CR$_{550}$ 112%	2021[82]
	GP-CMTs	Hair	CR2032	80:10:10, 0.6-0.9	69	1,006	CR$_{1000}$ 658 mAh g^{-1}	2018[83]
	SPC-700	Soybean	CR2032	80:10:10, 1.0	NA	1,516	CR$_{100}$ 360 mAh g^{-1}	2018[84]
	HTB	Cladophora glomerata	CR2032	75:15:10, NA	NA	700	CR$_{100}$ 350 mAh g^{-1}	2017[85]
	600 °C	Raw wheat flour	NA	80:10:10, NA	56	728	CR$_{100}$ 217 mAh g^{-1}	2017[86]
	Porous carbon	Waste shrimp shells	CR2032	80:10:10, 1.0	51	1,843	CR$_{100}$ 1127 mAh g^{-1}	2017[87]
	CNP	Coconut oil	CR2032	70:20:10, 1.5 mg	50	1,330	CR$_{20}$ 63%	2016[88]
	BDC-K	Bean-dreg	CR2025	85:05:10, NA	66	1,215	CR$_{100}$ 93.4%	2016[89]
	GT-700	Green tea leaves	CR2016	85:05:10, NA	NA	530	CR$_{50}$ 471 mAh g^{-1}	2014[90]

*MR represents the mass ratio (active materials: conductive agent: binder); **ICDC stands for initial charging/discharge capacity; ***CR$_x$ refers to capacity retention after x cycles; x indicates the cycle number.

specific structures from different biomass materials[91-94]. Many cellulosic plants, such as flax, wood, bamboo, cotton, *etc.*, are excellent sources of fibrous carbons[95]. Organic matter or plants that contain sucrose or glucose can be processed into spherical carbons[96]. Petals, oatmeal, maple and other biomass materials are ideal for production of layered carbons[97]. Most biomass materials such as fruit peels, nut shells, mushrooms, tea leaves and even animal feces can be prepared by diverse methods to obtain porous carbon, which is one of the most useful structures for many applications[98-103]. Presented below is an overview of the latest advances in a categorized form of BCMs, organized according to their structural features.
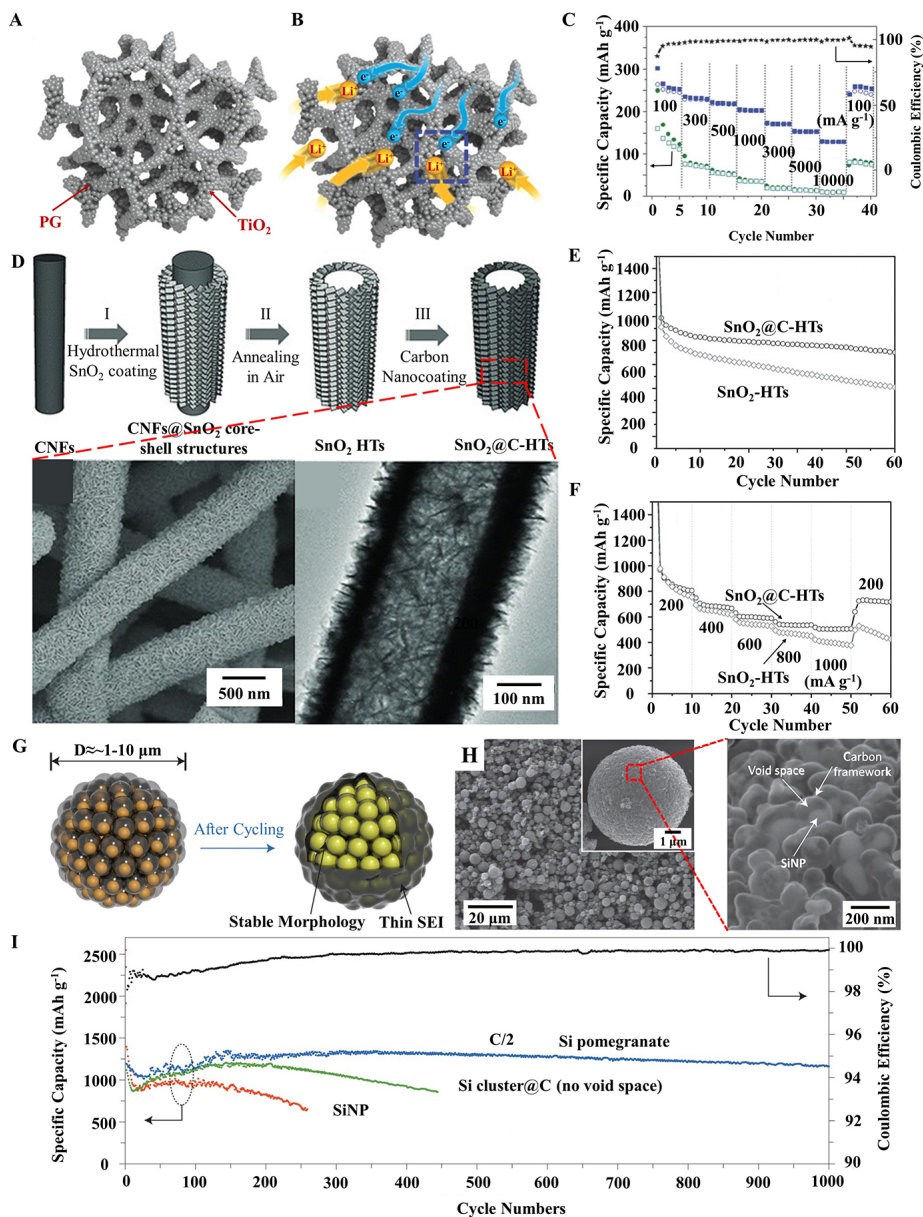
Figure 3. (A) Structure of TiO$_2$ NP-PG; (B) Working mechanism of TiO$_2$ NP-PG; (C) Discharging (circle)/charging (square) specific capacities of TiO$_2$ NP-PG (blue) and TiO$_2$ NP (green) at different current densities along with the CE (black). (D) Schematic illustration of the formation and TEM images of SnO$_2$@C-HTs; (E) Cycling performance of SnO$_2$@C-HTs and SnO$_2$ without ultra-thin carbon coatings; (F) Rate capacities of SnO$_2$@C-HTs and SnO$_2$ without ultra-thin carbon coatings; (G) Three-dimensional structure of Si pomegranate; (H) SEM images of Si pomegranate; (I) Reversible capacities for 1,000 cycles of the Si pomegranate and other structures tested at C/2. (A-C) are quoted with permission from Lee *et al.*[59]; (D-F) from Zhang *et al.*[68]; and (G-I) from Liu *et al.*[69].

Porous biomass-derived carbon

Porous structures are complex and changeable but easy to access. Porous biomass-derived carbons (PBCs), in general, have a high SSA, which increases the loading of active substances. The pore size and structure can be adjusted according to target applications and functions, facilitating ion transport and allowing the electrolyte to penetrate into the whole electrode.

In general, a two-step method of hydrothermal carbonization and KOH activation strategy is used to prepare porous carbons. Qiang *et al.* synthesized a porous form of biomass-derived carbons through a two-step process: hydrothermal carbonization of cypress coats, followed by an activation step using KOH as a porogen. By modulating the KOH to carbon mass ratio during the activation phase, it is possible to regulate the morphology and porous characteristics of resulting carbons[104]. Recently, a simple, low-cost and green one-step method has been proposed by Gong *et al.* Such a method gave carbon products abundant microporous structures, a high degree of graphitization and many oxygen-containing groups[105]. The 3D porous carbon (3DPC) was synthesized from bamboo char using this one-step method. The porous carbon synthesized through a one-step method was found to have more abundant micro/meso pores, higher degree of carbonization and better electrochemical performances than the carbon synthesized using a traditional two-step method.

PBCs can be used in both anodes and cathodes. However, their impacts on the performance of cathodes and anodes are different. In cathodes, porous carbons derived from biomass are introduced to cathodes as a coating, improving electrical conductivity and stability, which generally increases the cycling stability. Noerochim *et al.* utilized, for example, sucrose as a carbon precursor to fabricate a porous and conductive coating for $LiFePO_4$ cathodes, aiming to enhance the electrical conductivity and stability of the cathodes when used with an aqueous electrolyte[106]. Their findings indicated that a coating with a 9 wt% carbon yielded a product of the best performance, achieving an initial specific discharge capacity of 13 mAh g^{-1}, with only a 2% capacity loss after 100 cycles. Zhang *et al.* designed a high-rate performance cathode material (FF@3DPC) by embedding $FeF_3 \cdot 0.33H_2O$ nanoparticles in nori-derived nitrogen-oxygen double-doped 3DPC [Figure 4A and B][107]. FF@3DPC had high reversible capacities of 134 and 104 mAh g^{-1} at 5C and 20C, respectively [Figure 4C]. After 500 cycles, it maintained a reversible capacity of 101 mAh g^{-1} at 5C, as shown in Figure 4D.

PBCs applied to the anode are often doped with other elements to further improve the rate capacity and cycling stability. Ou *et al.* proposed to use chrysanthemum as a novel carbon source for creating a hierarchical porous nitrogen-doped carbon (HPNDC) anode, as depicted in Figure 4E and F[108]. This HPNDC demonstrated a superior high-rate performance, as shown in Figure 4G, with a specific capacity of 347 mAh g^{-1} at 5 A[108]. Furthermore, the anode exhibited a commendable electrochemical performance, maintaining a reversible capacity of 975 mAh g^{-1} after 100 cycles at 100 mA g^{-1}, as illustrated in Figure 4H. Xu *et al.* explored the use of sulfur and nitrogen-doped porous carbon derived from bean shells as an anode material for LIBs[109]. At 1C, this material sustained a capacity of 262 mAh g^{-1} even after 100 cycles. Zheng *et al.* reported the synthesis of porous nitrogen-doped carbon (PNDC) from duckweed, concluding that the material could support LIBs with a capacity of 1,071 mAh g^{-1} after 100 cycles when subjected to a current density of 100 mA g^{-1}[110]. Yan *et al.* developed an anode material from bamboo leaves, resulting in a HPNDC[111]. This material not only showcased an impressive rate capability but also exhibited significant long-term stability. After 500 cycles at a current density of 0.2 A g^{-1}, the optimized carbon could deliver a discharge capacity of 450 mAh g^{-1}.

To sum up, natural substances with an intricate network of interconnected channels and pores are the ideal precursors of PBC. PBC is easy to prepare using two- and one-step methods, including the activation method, template method or direct carbonization. Moreover, it has been proven to have high capacity and excellent cycling ability due to its pore structure which provides electroactive sites to contribute additional capacity, fast diffusion channels for ions to enhance kinetics and a buffer zone to decrease interfacial pressure. If the preparation method can be further simplified and more environmentally friendly, PBC may become a viable substitute for graphite.
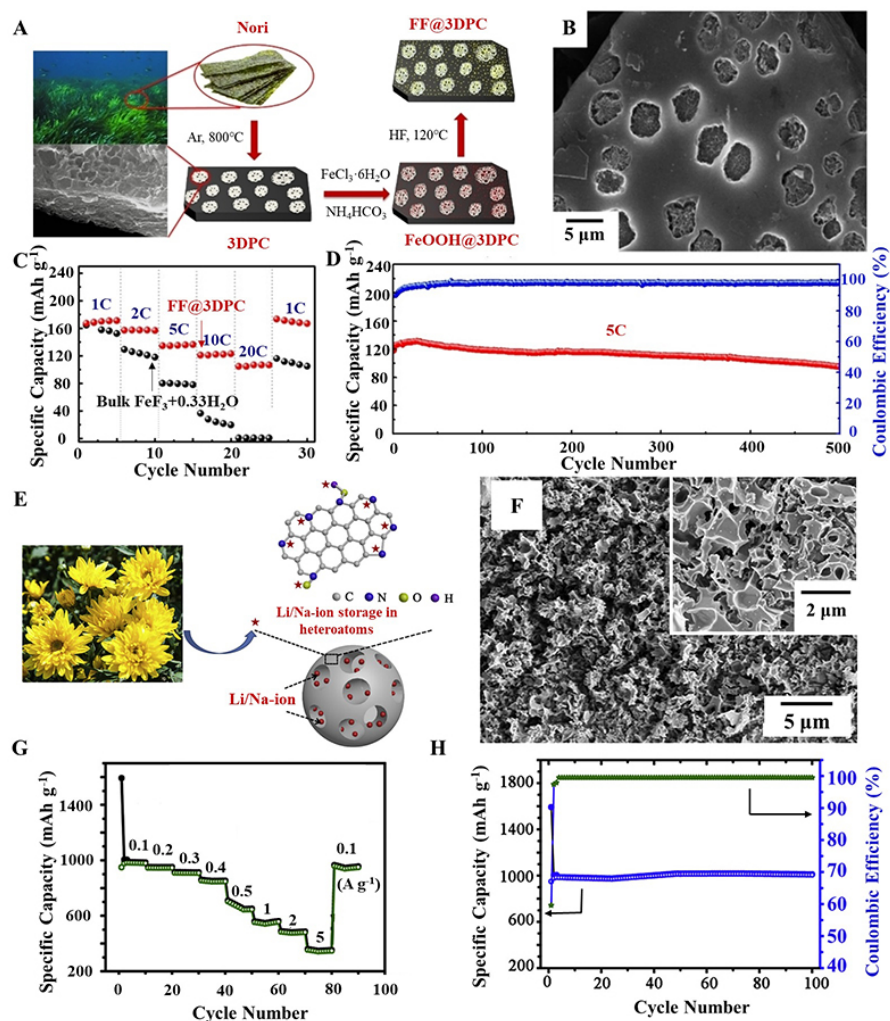
Figure 4. Schemes, structure and performance of porous biomass-derived carbons. (A) Synthesis process of 3DPC and FF@3DPC; (B) SEM images of 3DPC; (C) Rate capability of FF@3DPC electrode and bulk FeF$_3$·0.33H$_2$O electrode at different current density; (D) Cycling performance of FF@3DPC electrode at 5C. (E) Schematic illustration of the porous structure and working mechanism of HPNDC; (F) SEM images of HPNDC; (G) Rate capabilities of HPNDC at different current densities; (H) Cycling performance of HPNDC at 100 mA g^{-1}.(A-D) are quoted with permission from Zhang *et al.*[107]; and (E-H) from Ou *et al.*[108].

Fibrous biomass-derived carbon

The potential of fibrous biomass-derived carbon (FBC) materials applied to the anode has been discovered in recent years. These materials are usually from cellulose, silk and chitin, which have complex hierarchical structures composed of micro- and nano-fiber. Therefore, the fibrous carbon of various shapes can form from these FBCs. It is often fabricated by dissolution and regeneration, followed by carbonization. Cellulose is dissolved and separated in the solution, and then regenerated fibers are processed into a new dimension. Finally, FBC is obtained by carbonization. This processing method is simple and low-cost.

FBC used in LIBs provides fast transport channels for ions and electrons as such that the electrochemical performance of LIBs is improved significantly. Zhang *et al.* transformed bamboo, pine, poplar and paulownia into uniform hollow fibers of micrometer sizes by delignification and carbonization[112]. FBC had a larger interlayer spacing of about 0.39-0.40 nm which was calculated by Bragg's law using the results of X-ray diffraction (XRD) characterization. The anode made from the fibrous carbon exhibited a better

electrochemical performance than commercial graphite anodes. LIBs had a high reversible capacity, 435 mAh g^{-1} at 50 mA g^{-1}, competitive rate capacity of up to 150 mAh g^{-1} at 2 A g^{-1} and good cycling stability of 76% capacity retention after 500 cycles. Scientists have discovered that wild fungus is also an ideal source for fabricating FBC. Tang *et al.* reported synthesis of carbonaceous fibers from Tyromyces fissilis wild fungus [Figure 5A] by controlling the carbonization process[113]. The carbon produced as such had a larger interlayer spacing of 0.386 nm as compared with 0.335 nm of graphite. It exhibited longer-term cycling stability, as shown in Figure 5B. Its specific reversible capacity was 340 mAh g^{-1} at C/10 and 300 mAh g^{-1} at a fast 1C charge/discharge rate [Figure 5C]. Seaweed is also an ideal carbon source for the preparation of carbon materials. Lv *et al.* harnessed the templating ability of the natural structure of alginate molecular chains to produce a high-performance fibrous anode [Figure 5D][114]. Fe_2O_3 particles are embedded in the fibers to form a yolk-shell structure through coordination (C@Fe_2O_3-F). This fibrous anode demonstrated excellent cycling performance with no capacity decrease after 200 cycles (1,035 mAh g^{-1}) and good rate performance [Figure 5E and F].

In summary, natural products with fibrous, filamentary or directional textures are more likely to produce FBC. Though such an approach has requirements for the structure of biomass materials, its fabrication process is relatively easy and environmentally friendly, only using carbonization. In addition, it has delivered good performance for LIBs because fibrous carbon forms a cross-linked conductive network. Such a network increases the overall conductivity and provides a rapid transmission channel for lithium ions, increasing the overall rate performance. Therefore, FBC has a good future of commercial utilization in the anode.

Spherical biomass-derived carbon

Spherical carbon has good structural stability, high packing density and a high surface-to-volume ratio. Therefore, it shows great potential for the use in LIBs. Spherical carbon is often prepared by carbonization of spherical biomass which is referred to as spherical biomass-derived carbon (SBC). Kim *et al.* synthesized micrometer-diameter SBC particles as the anode of LIBs using coffee oil as raw material[115]. It was found that the spherical carbon retained its initial structure after 250 cycles. These LIBs had good cycling stability as the capacity remained 290 mAh g^{-1} after 200 cycles. Song *et al.* obtained a robust carbon microsphere negative electrode material (MES600) by carbonizing the maleic anhydride esterified starch [Figure 6A and B][116]. Compared with starch-derived carbon without esterification modification (PS600), MES600 had better rate and cycling performance [Figure 6C and D]. To promote lithium-ion transport, carbon is processed into porous carbon spheres. Shi *et al.* utilized sucrose to prepare carbon microspheres with hierarchical micropores using an impregnation-carbonization method[117]. The electrode with hierarchical porous carbon microspheres presented good rate capacities (1,141, 650, and 348 mAh g^{-1} at the current densities of 0.05, 0.2, and 1 A g^{-1}, respectively). Chen *et al.* used corn starch to fabricate microporous carbon spheres through enzymolysis, pre-oxidation, and carbonization[118]. The microporous carbon spheres showed an exceptional rate capability (150 mAh g^{-1}) at an ultrafast charge/discharge current density of 20 A g^{-1}, and as a result, LIBs were fully charged in 27 s. Ming *et al.* used glucose to prepare porous carbon balls as Fe_3O_4 coating layers (Fe_3O_4@porous-C) to improve the electrochemical performance of Fe_3O_4 [Figure 6E and F]. Fe_3O_4@porous-C demonstrated a better rate capacity and cycle stability [Figure 6G][119].

In general, natural substances of spherical or regular shapes are more likely to form spherical carbon materials. SBC is usually obtained through carbonization of spherical biomass. The resulting SBC has high structural stability and a large specific area, which can improve cycle life. When combined with the porous structure and fabricated into a porous carbon sphere, the SBC can supply channels to transport ions quickly, promoting the fast charge and discharge performance and improving the rate capacity of LIBs.
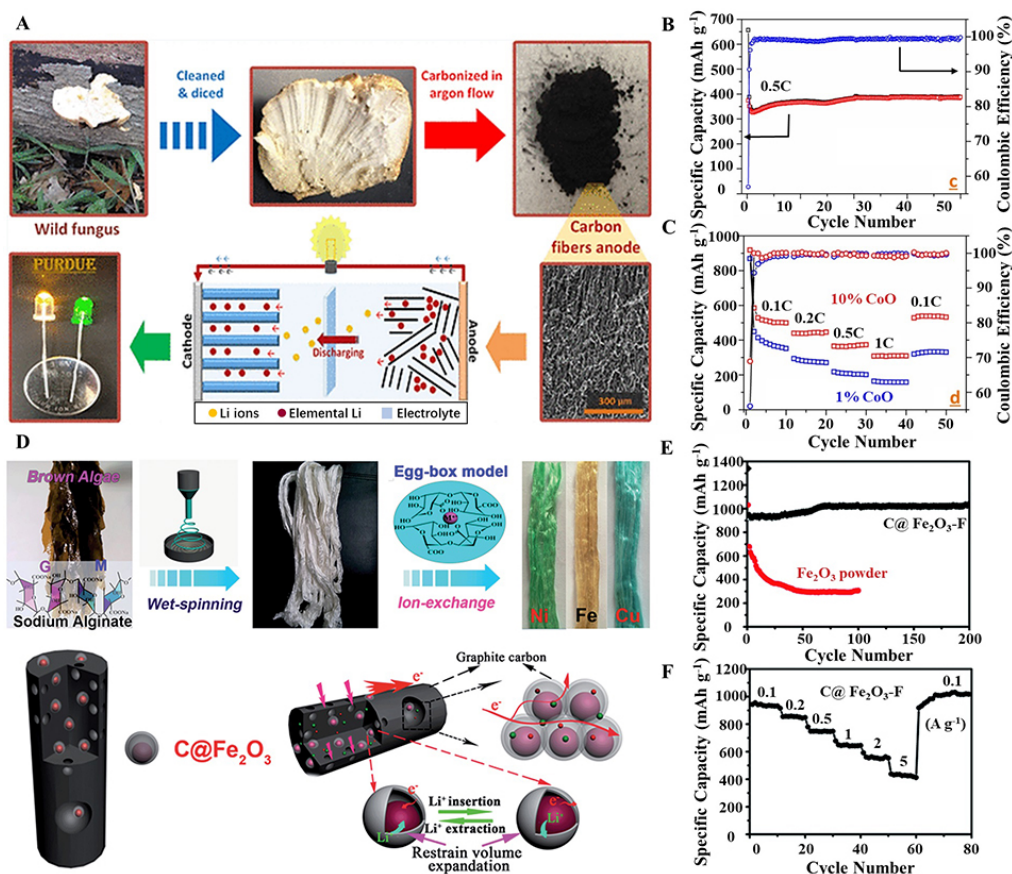
Figure 5. Schemes, structure and performance of fibrous biomass carbon. (A) Synthesis process of carbonaceous fibers from Tyromyces fissilis wild fungus; (B) Cycling performance of carbonaceous fibers from Tyromyces fissilis wild fungus; (C) Specific discharge capacities of carbonaceous fibers from Tyromyces fissilis wild fungus. These figures are quoted with permission from (D) Synthesis process of C@Fe$_2$O$_3$-F; (E) Cycling performance of C@Fe$_2$O$_3$-F; (F) Specific discharge capacities of C@Fe$_2$O$_3$-F. (A-C) are quoted with permission from Tang *et al.*[113]; and (D-F) from Lv *et al.*[114].

Layered biomass-derived carbon

Raw biomass materials often contain fibers, which can be arranged into a layered structure similar to that of graphite anodes in LIBs. Layered biomass-derived carbon (LBC) can be synthesized by hydrothermal treatment, followed by graphitization and KOH activation. This process is similar to the fabrication of PBC. For the application in LIBs, the layered carbon often replaces graphite as the anode. It can provide a larger interlayer spacing to help fast transport of electrons and ions, containing more positions for the insertion and withdrawal of Li$^+$ ions.

Zhou *et al.* synthesized complex and interconnected highly graphitic carbon nanosheets (HGCNS) using wheat stalks as the precursor[120]. The HGCNS consists of ultrathin nanosheet frameworks with a 0.3362 nm interlayer spacing [Figure 7A]. The LIBs made with HGCNS were reported to have a high reversible capacity of 502 mAh g^{-1} at 0.1 C [Figure 7B]. In addition, such LIBs had an excellent rate capability and superior cycling performance, providing 215 mAh g^{-1} at 5 C after 2,000 cycles [Figure 7C]. Mondal *et al.* used eucalyptus tree leaves to prepare interconnected and porous carbon nanosheets which were used in the anode of LIBs[121]. The anode showed a high specific capacity (819 mAh g^{-1} at 100 mA g^{-1}) and good cycling stability (513 mAh g^{-1} after 500 cycles). Gao *et al.* used oyster shells to prepare two-dimensional (2D) hierarchically porous carbon (HPC) [Figure 7D][122]. Sheet-like HPC electrodes displayed an excellent
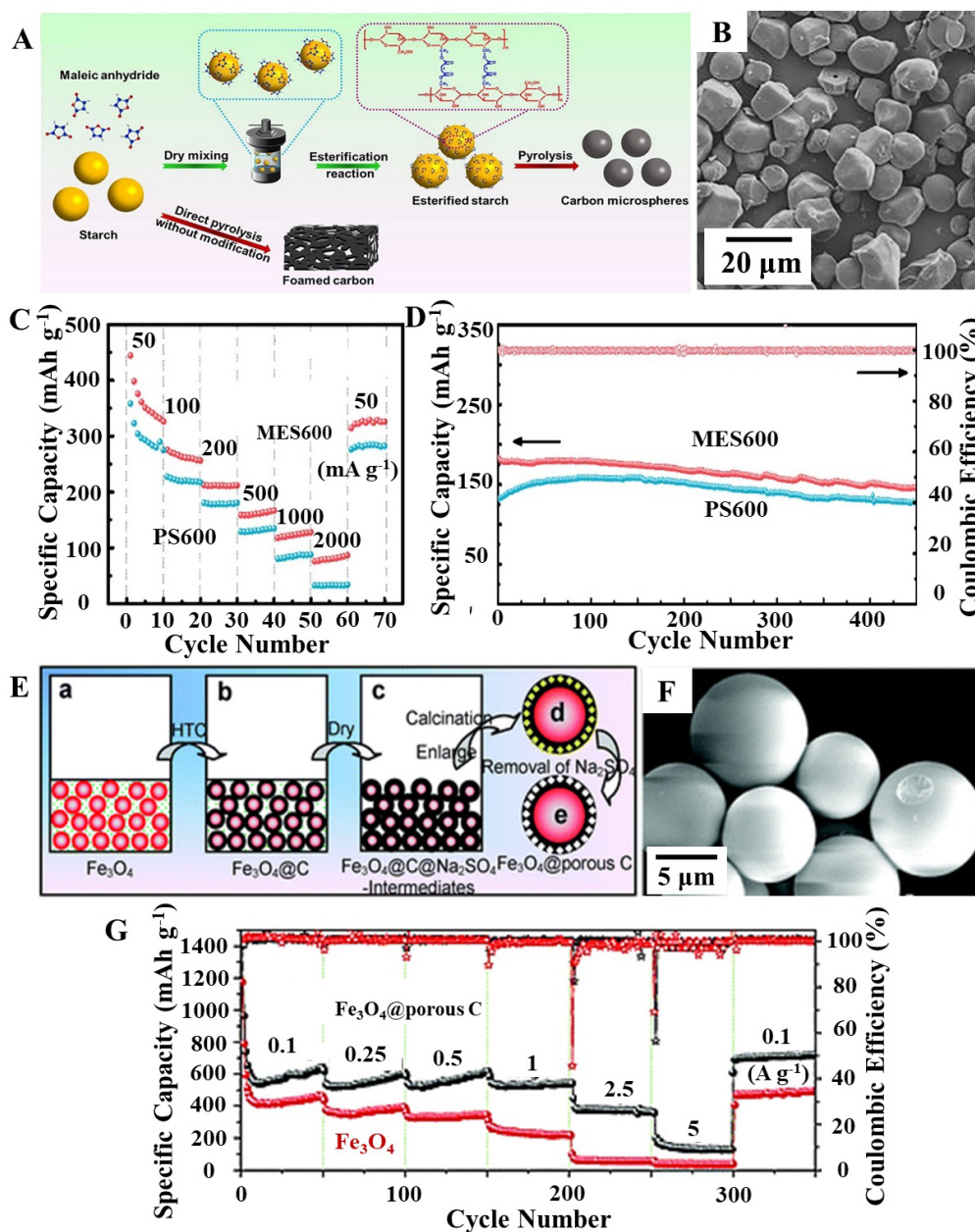
Figure 6. Schemes, structure and performances of spherical biomass carbon. (A) Schematic illustration of the route to fabricate MES600 and PS600; (B) SEM image of MES600; (C) Rate performance of MES600 and PS600; (D) Cycling performance of MES600 and PS600; (E) The synthesis route of Fe_3O_4@porous-C; (F) SEM images of glucose-derived carbon microparticles; (G) Comparative cycling performance and rate capability of primary Fe_3O_4 and Fe_3O_4@porous-C. (A-D) are quoted with permission from Song *et al.*[116]; and (E-G) from Ming *et al.*[119].

capacity retention at both 0.5 and 1 A/g after 400 cycles [Figure 7E]. The HPC-3-3 exhibited a high reversible capacity of 1,151 mAh g⁻¹ at 0.1 A g⁻¹ because of its cross-linked porous carbon network, more than three times higher as compared with pure carbon, along with good rate capacities [Figure 7F].

In a word, natural substances of cellulose can be used to synthesize LBC. However, LBC needs to be treated first with a strong alkali and then carbonized. Such a processing method is similar to that used for graphite but the layered morphology can improve the capacity and cycling performance of LIBs. The layered carbon
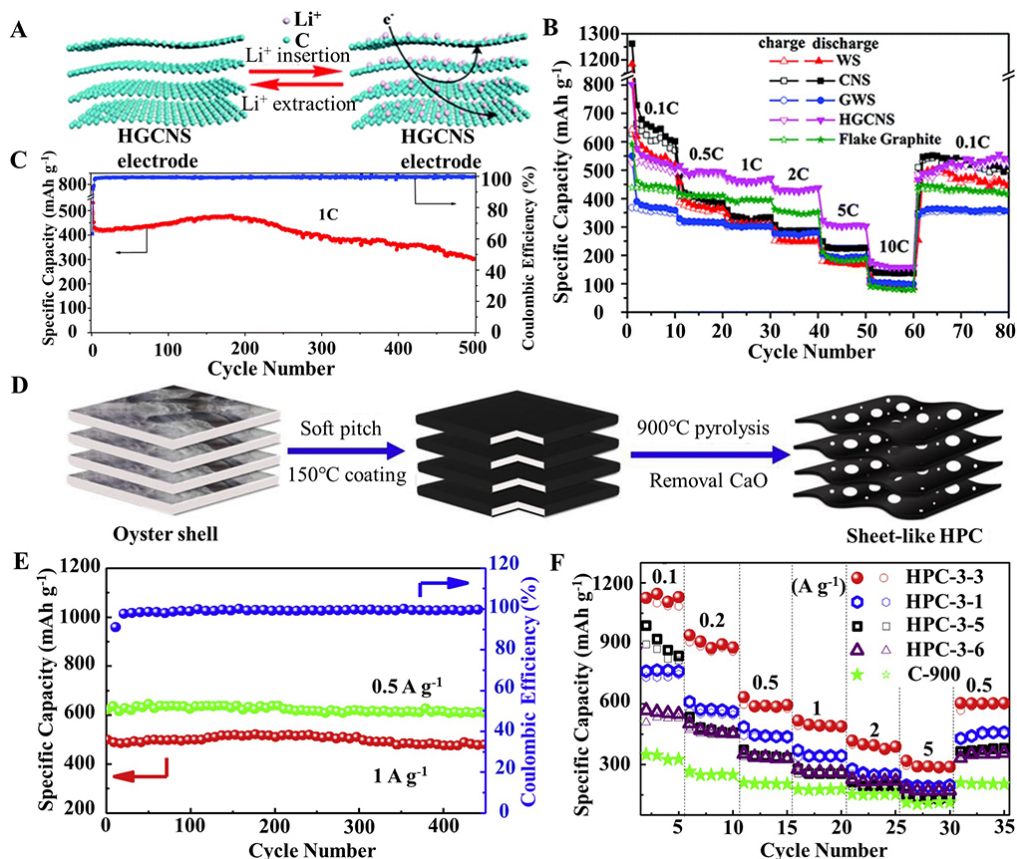
Figure 7. Schemes, structure and performances of layered biomass carbon. (A) Schematic diagram of lithium-ion transport during charging and discharging in HGCNS nanosheet frameworks; (B) Rate capacities of HGCN and flake graphite electrodes at different rates; (C) Cycling performance of HGCNS electrode at 1 C; (D) Schematic illustration of synthesis of sheet-like HPC; (E) Cycling performance of sheet-like HPC electrode at 1 and 0.5 A g^{-1}; (F) Rate capacities of sheet-like HPC. (A-C) are quoted with permission from Zhou *et al.*[120]; and (D-F) from Gao *et al.*[122].

nanosheets can supply sites containing lithium ions and facilitate rapid transport of both electrons and lithium ions, which have the potential to replace the commercialized graphite in the future.

FAILURE MECHANISMS AND SOLUTIONS

Compared with graphite, biomass-derived carbons are often hard materials with a highly disordered carbonaceous structure. They have higher defect concentrations, higher heteroatom contents, greater distances between graphite layers (d$_{002}$) and more closed pore structures[123,124]. Such features are beneficial for shortening the transport distances of Li$^+$ ions and providing abundant active sites for charge transfer reactions. However, low CE and high irreversible capacity are the shortcomings that restrict BCMs from being widely used. The failure mechanisms of biomass materials are summarized and analyzed in this section, including low conductivity, low ICE, high irreversible capacity, and continuous voltage hysteresis during cycling. Meanwhile, current solutions proposed are also presented in this section.

Failure mechanisms

Hard carbon is mostly prepared using polymer precursors, and the precursors of biomass-derived carbons are certain types of polymer substances, such as glucose, cellulose, starch, *etc*. As a result, the prepared and obtained BCMs are mostly hard carbon materials. Since the precursors of BCMs are polymers, the reactions in the pyrolysis process are complex, including dehydrogenation, condensation, hydrogen transfer and

isomerization occurring simultaneously and releasing H_2, CH_4, CO, CO_2 and other gases during the pyrolysis process. During the pyrolysis, the precursors evolve gradually from a 3D amorphous phase to short-range ordered graphite microcrystals in the form of a 2D regular graphitized carbon layer. Due to the existence of molecular cross-linking and covalent C-O-C bonds in biomass materials, it is easier to form rigid cross-linked structures. The ordered regions are interconnected by twisted layers of graphitized carbon, which inhibits the growth and orientation of graphite layers during the carbonization stage, and hinders the directional movement of electrons. As a result, the electrical conductivity of hard carbon materials derived from biomass is lower than that of graphite[125,126].

Table 1 shows that most biomass materials used in LIB electrodes exhibit low ICE, which is not even mentioned in many studies. Compared to graphite carbon, hard carbon has low crystallinity and poor conductivity. Although its high SSA improves the specific capacity, its ICE and reversible capacity are low as a result of the irreversible decomposition of the electrolytes and the excessive formation of the solid electrolyte interface (SEI) layer, which requires a large amount of lithium [Figure 8A and B]. During cycling, the SEI layer formed on the anode is unstable, causing the thickness and composition of the SEI layer to change constantly. The thick SEI layers increase ohmic resistance and hinder ion diffusion, resulting in a continuous increase in high voltage hysteresis and capacity decay[127]. In addition, the pyrolyzed hard carbon materials have a small amount of oxygen elements and some residual fragments of terminal aromatics as a result of the precursors being rich in aromatic compounds and oxygen-containing functional groups which inhibit the graphitization of the precursor[128]. Therefore, the hard carbon exhibits less graphitization and more holes and defects, causing the Li^+ ions to bond to these sites and produce deposits [Figure 8C][129]. In this case, the removal of Li^+ ions from these locations shifts the potential toward a higher voltage, resulting in a voltage hysteresis [Figure 8D][130].

Despite its great importance, there are few studies on failure mechanisms of BCMs. The analysis of BCM failure mechanisms here employs a method similar to that used for hard carbon because both types of carbon share a similar lithium-ion storage mechanism, involving a combination of intercalation and adsorption. The intercalation/deintercalation of lithium ions occurs at the low voltage, and the adsorption/desorption and possible additional intercalation take place at the higher voltage. Therefore, the failure mechanisms can be analyzed from these two aspects. On the one hand, the principle of intercalation and delamination is similar to that of graphite, but most of the BCMs have a porous structure and hence a larger SSA than that of graphite. As a result, more lithium ions are consumed when the SEI layer is formed, eventually leading to the depletion of lithium ions and the failure of the material. On the other hand, excessive lithium ions adsorbed in the pores after multiple cycles will block the pores, resulting in less lithium ions being able to adsorb and desorb, causing shortening and tilting of the electrochemical platform of biomass-derived carbons, as shown by increasing voltage hysteresis. In addition to these two aspects, there may be other undiscovered causes[131]. Therefore, future research on BCMs should focus more on analyzing failure mechanisms to provide a scientific basis for designing viable solutions to these problems and further improving the electrochemical performance of BCMs.

Solutions

Based on the two reasons summarized from the above failure analysis, there are two prominent pathways to improving BCMs: enhancing the degree of graphitization and prelithiation, which will boost ICE and reduce irreversible capacity.

Improving the degree of graphitization

Two common methods to improve the degree of graphitization are increasing the carbonization temperature and using catalysts. With the increase in the processing temperature, both the degree of
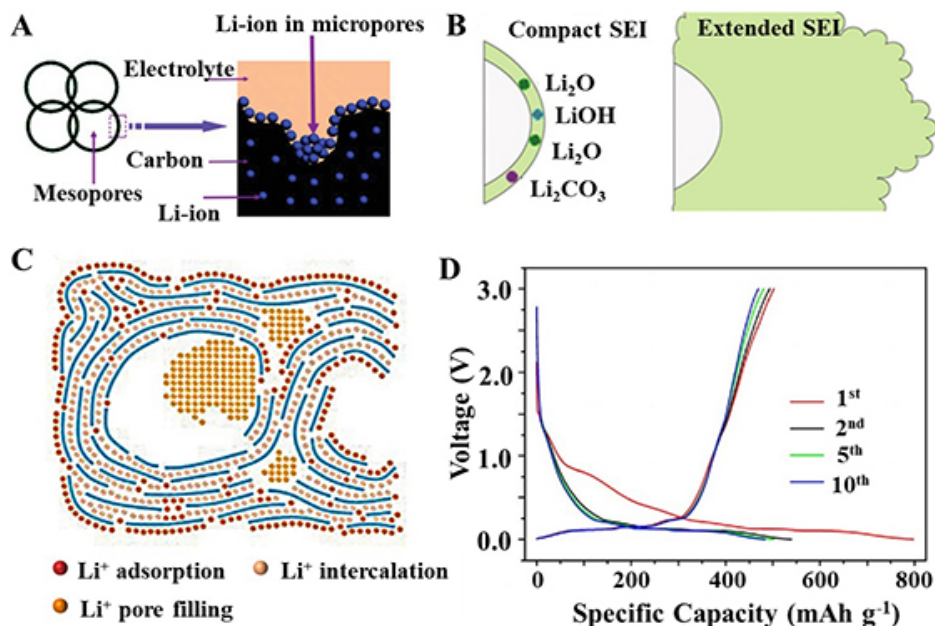
Figure 8. Schemes of lithium storage and failure mechanisms of biomass carbon. (A) Schematics showing bulk lithium storage in the mesopores and micropores; (B) Diagram of an unstable SEI film; (C) Schematic showing lithium storage mechanism of lithium ions in biomass carbon materials; (D) Discharge/charge curves of wheat straw-derived carbon. (A) is quoted with permission from Adams *et al.*[125]; (B) from Yao *et al.*[130]; (C) from Zhao *et al.*[126]; and (D) from Chen *et al.*[129].

graphitization of BCMs and the degree of long-range order grow significantly. Although the crystallites/ domains of graphite are small, they exhibit a considerable short-range order and have evolved into a more graphitized structure in a clearly identifiable domain. This feature is caused by the order enhancement and the increase of crystallinity in the nanoscale domain[132]. Wang *et al.* used a hydrothermal method to carbonize water chestnut shells at 900, 1,100, 1,300 and 1,500 °C (HT-X, X: carbonization temperature) [Figure 9A][133]. The length of the platform became shorter and the degree of graphitization was found to increase with the temperature of carbonization, indicating the generation of SEI film for samples carbonized at higher temperatures [Figure 9B]. ICE was found to increase accordingly from 53% to 67% [Figure 9C], with less change in irreversible capacity [Figure 9D].

In addition, the use of catalysts could reduce the temperature and improve the efficiency of reaction, while enhancing the degree of graphitization. The types of catalysts used in biomass-derived carbon for LIBs are roughly divided into three types: elemental, compound, and alloy catalysts. The catalyst and biomass are mixed to react and form carbides, which decompose at high temperatures to form graphite. The mechanism of the catalysis in the process of graphitization is the catalyst to participate in the C-C bond recombination in the conversion process[134]. Gomez-Martin *et al.* synthesized hard carbon anodes (MDF-Fe) by graphitizing the medium-density recycled wood of fiberboard using an iron catalyst at 2,000 °C [Figure 9E][135]. Compared with the hard carbon produced without an iron catalyst, the structural order and degree of graphitization of MDF-Fe were higher, with the degree of graphitization reaching 70% at 2,000 °C [Figure 9F]. The MDF-Fe formed as such exhibited a significant increase in the reversible capacity and a doubling of the ICE from 32% to 64%, resulting from the decrease in the amount of non-basal plane areas and SSA, which decreases the formation of SEI layers [Figure 9G].
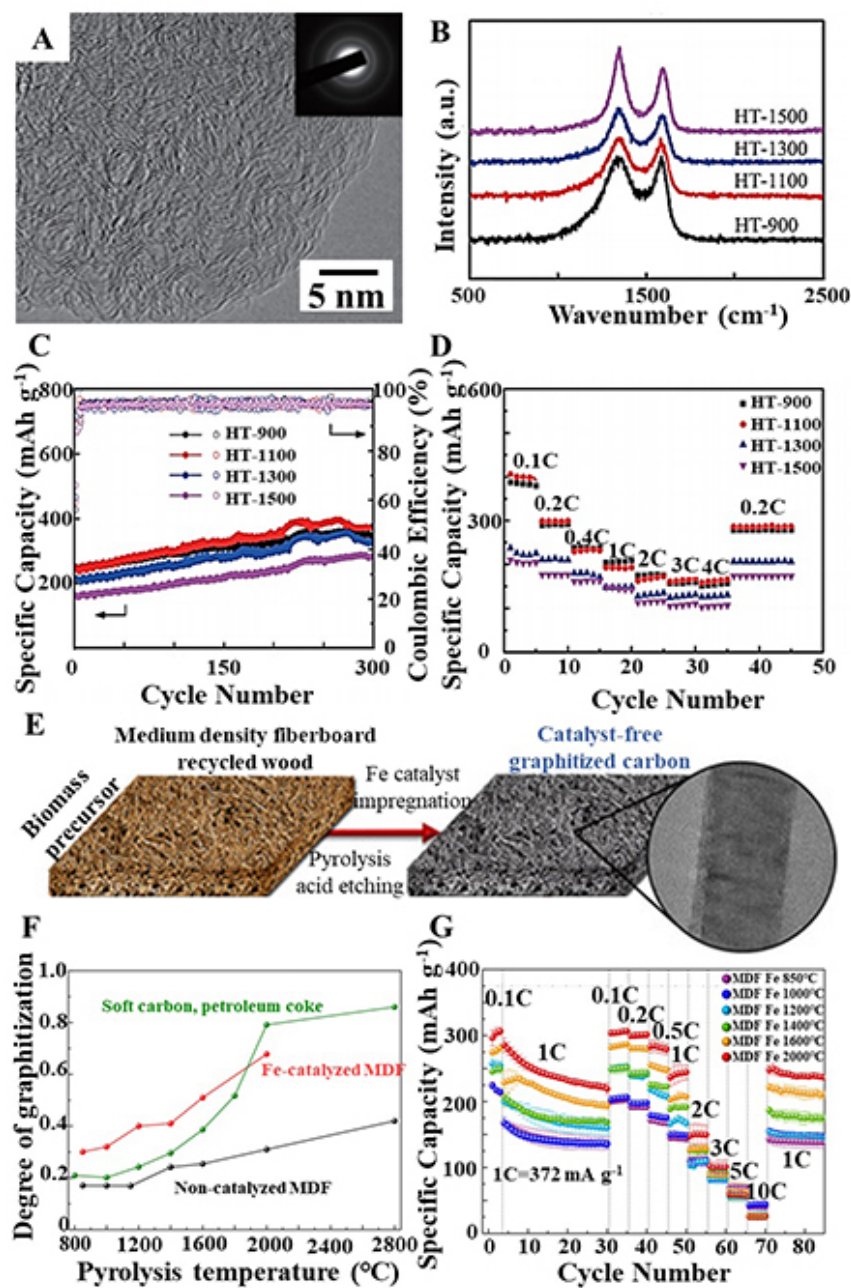
Figure 9. Solutions by improving the degree of graphitization of biomass carbon. (A) TEM and SAED images of HT-1500; (B) Raman spectra of the water chestnut-based hard carbon at different carbonization temperatures; (C) Cycling performance of the water chestnut-based hard carbon at various carbonization temperatures; (D) Rate performance of the water chestnut-based hard carbon at multiple carbonization temperatures; (E) Preparation progress of MDF-Fe carbon; (F) Degree of graphitization of Fe-catalyzed MDF and uncatalyzed MDF carbon; (G) Rate performance of MDF-Fe carbon by pyrolysis between 850 and 2,000 °C. (A-D) are quoted with permission from Wang *et al.*[133]; and (E-G) from Gomez-Martin *et al.*[135].

Prelithiation

Prelithiation refers to the addition of lithium ions prior to battery assembly. It aims to compensate for the loss of active lithium caused by SEI formation and other side reactions. After prelithiation of carbon materials, a complete and stable SEI film will form on the surface, thereby increasing the reversible specific capacity and ICE. Four major approaches for prelithiation include direct contact with lithium metal, Li-rich

material additives, electrochemical prelithiation, and chemical prelithiation[136]. Electrochemical prelithiation is one of the simplest and most common methods, which is a slow process but performs a precise and uniform prelithiation. This strategy not only ensures a more uniform SEI film, but also limits crack formation, which can effectively improve electrochemical performance[137].

Prelithiation can be performed on both cathodes and anodes. Huang *et al.* prelithiated waste feather-derived active carbon (prAC) [Figure 10A][138]. $Li_2C_2O_4$ was used as the source of lithium, which broke down into Li_2O nanowires. Compared with the waste feather-derived carbon material without prelithiation (AC) and the carbon material directly added to Li_2O (AC + Li_2O), prAC exhibited a higher energy density of 122 Wh kg^{-1} at 220 W kg^{-1} and a superior cycling performance [Figure 10B and C]. Drews *et al.* used spruce wood to synthesize hard carbon materials for LIBs and employed the electrochemical prelithiation method to develop a lithium-ion full cell of enhanced cycle life[139]. The electrode performance after prelithiation was significantly improved, showing a significant increase in ICE from 63% to 87%. After 150 cycles, the prelithiated full cell retained a capacity of 80% as compared with only 66% for the non-prelithiated full cell. In addition, great efforts have been made to dope graphene with P-type dopants, such as O, P, N, *etc.*, to promote the formation of an artificial SEI film[140]. These elements are more electronegative than carbon, adsorbing at the stable site of graphene and forming an artificial SEI film. Electrons are transferred from the graphene sheet to P-type dopants, resulting in a depletion of electrons and an increase in the hole concentration on the graphene plane, leading to an increase in the work function of the whole system and a decrease in formation of SEI films. Therefore, proper control of the work function is clearly one of the prelithiation methods, which can reduce the subsequent SEI film formation and improve ICE. Ren *et al.* doped Cl in hard carbon derived from cotton to obtain anode materials (Li//Cl-HC) that can inhibit the reduction of solvent molecules in the electrolyte and reduce the formation of SEI film [Figure 10D][141]. Compared with no Cl doping (Li//HC), the ICE of the prelithiated anode material increased from 65% to 78% [Figure 10E]. The cycle stability was also improved [Figure 10F].

CONCLUSIONS, CHALLENGES AND PROSPECTS

To sum up, we should accelerate the energy transition and use renewable energy to achieve the goal of carbon neutrality. LIBs are the most widely used secondary energy storage devices. In order to further improve the performance of LIBs, it is necessary to find anode materials of a higher capacity than graphite, among which BCMs are a great option. They have the advantages of a wide range of sources, lower price, easy access and environmentally friendliness, and possess outstanding electrochemical performances in energy density, cycling and rating performances and safety. BCMs can be used for the cathode or anode of LIBs according to their structures. The addition of BCM coatings in cathode materials can solve such problems as unstable structure, metal ion dissolution and poor conductivity. BCMs used in the cathode can help to improve the stability of the positive electrode and capacity retention rate. As for anode materials, BCMs have been widely studied and used directly as electrode active materials or modified additives to help improve electrochemical performance. Currently, the capacity of BCMs as anode materials can reach more than 500 mAh g^{-1}, significantly higher than the theoretical capacity of graphite. As green renewable resources, BCMs are, therefore, excellent candidates to replace graphite in next-generation high-performance LIBs.

Generally, the processing of BCMs includes hydrothermal carbonization, carbon activation, dissolution and regeneration, which is simple to operate and produces minimal pollution. Biomass materials of high carbon content with excellent natural structure and compositions can be used to prepare various BCMs. Different biomass materials can be used to prepare carbons of distinct structures. There are roughly four major structures of biomass: porous, fibrous, spherical and layered. Most biomass-derived carbon has a porous
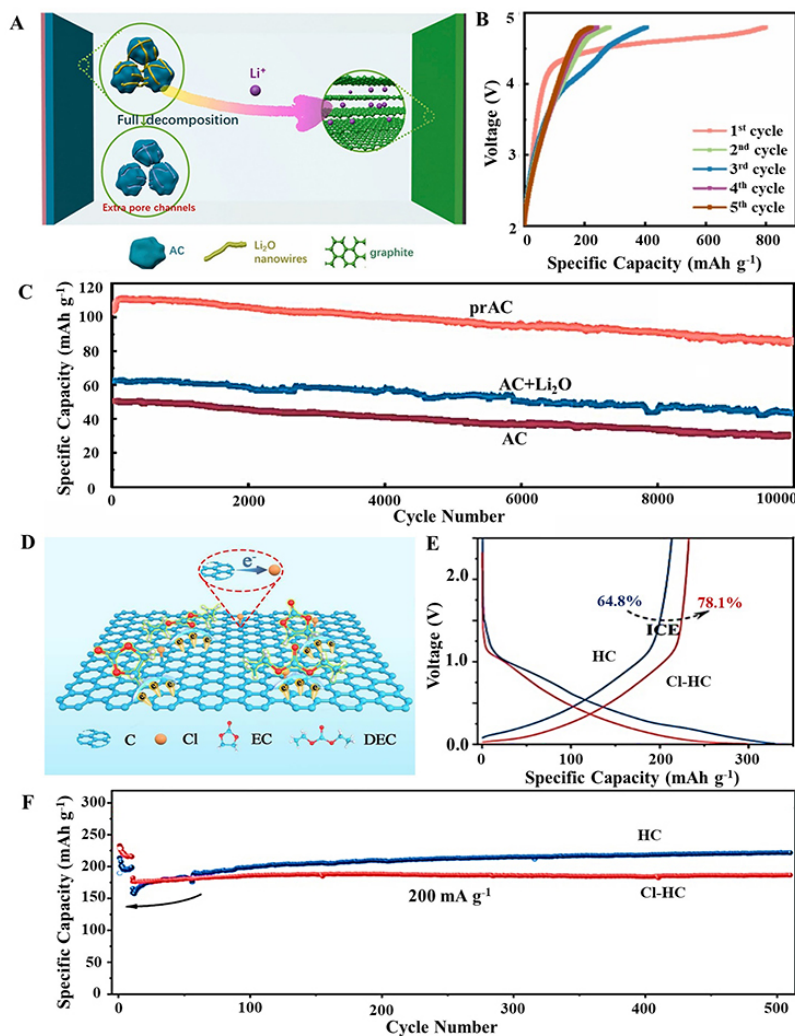
Figure 10. Solutions by prelithiation of biomass carbon. (A) Schematic illustration of the prelithiation mechanism of prAC; (B) Gas chromatogram of the prAC cathode over the first five cycles; (C) Rate performance of the prAC cathode after prelithiation, AC and AC + Li$_2$O cathode; (D) Cycle performance of the prAC cathode after prelithiation, AC and AC + Li$_2$O cathodes at a density of 1 A g^{-1} for 10,000 cycles. (E) Mechanism of Cl doping prelithiation; (F) Initial charge-discharge curves at 50 mA g^{-1} of Li//HC and Li//Cl-HC half-cells; (G) Rate performance of Li//HC and Li//Cl-HC half-cells; (H) Long cycle performance of Li//HC and Li//Cl-HC half-cells. (A-D) are quoted with permission from *Huang et al.*[138]; and (E-H) from Ren *et al.*[141].

structure, which provides more storage sites for lithium ions and increases the capacity. FBC and LBC provide conductive networks that increase the electrical conductivity, while also providing transport channels for fast lithium-ion shuttling, improving the rate capacity. SBC has a large SSA and a stable structure, which can improve cycle stability. If combined with the porous structure, it also shortens lithium-ion transport channels and improves the rate capacity. The advantages of each structure are summarized in Figure 11.

What is more, the working principles of lithium-ion storage in BCMs are intercalation and adsorption. The related failure mechanisms focused on the loss of lithium ions due to the high SSA and pores of BCMs. Besides, the low graphitization degree of BCMs caused low electron conductivity. Improving the degree of graphitization and prelithiation are two effective solutions to reduce the formation of SEI layers and improve the electrochemical performance of BCMs. In specific, increasing the carbonization temperature
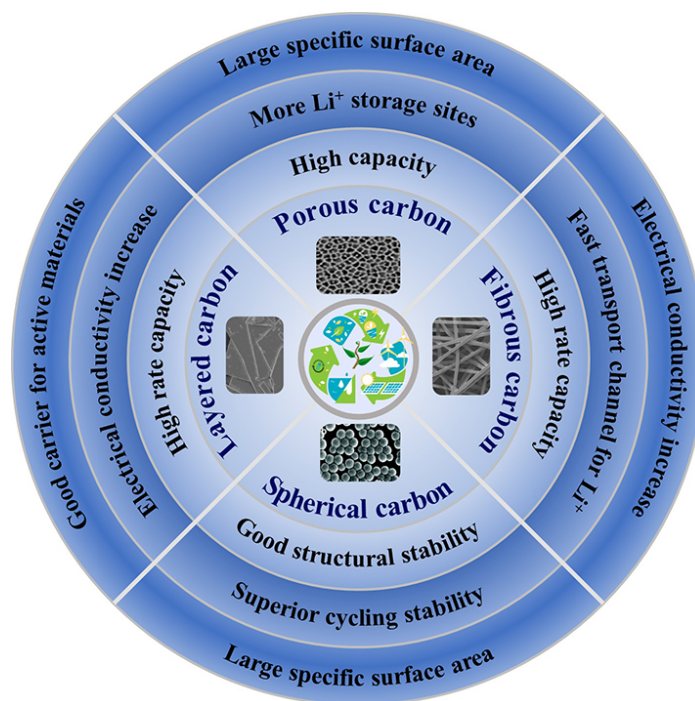
Figure 11. Pie chart describing advantages of various biomass carbon structures.

and adding catalysts are two important ways to improve the degree of graphitization. BCMs with a higher graphitization degree exhibit improved ICE. As for prelithiation, increase in ICE can be realized by adding additional lithium sources by direct contact with lithium metal, Li-rich material additives, electrochemical prelithiation, and chemical prelithiation. Doping is also useful in the prelithiation of BCMs by controlling the work function to attach a proper artificial SEI layer on the active substance. Though solutions are limited, it illustrates that the performance of BCMs can still be improved through a further and deeper understanding of work principles and failure mechanisms.

However, some challenges remain to be resolved. Firstly, the issue of low production yield of the above methods should be pointed out. The current yield is not satisfactory, hindering its commercial production. Furthermore, air pollution is inevitable in the process. Further study is therefore needed to determine how to increase production yield while capturing and treating the greenhouse gases released during the production process[142]. In addition, the current research on the working mechanism of BCMs in batteries remains far from well-established, and most of the literature on biomass-derived carbon does not mention the working mechanisms and failure analysis, which hinders how to further improve the performance of batteries. It is hoped that more studies in the future can further explore the role of biomass-derived carbon and the reasons for performance degradation and failure.

Moreover, we need to draw attention to the biomass carbon recycling. Since biomass materials as waste are recycled, we need to avoid those materials becoming an environmental burden. A lot of BCMs are as coatings or other mixed materials. It is therefore of critical importance to consider how to separate the mixture and achieve a reasonable recovery yield when we design BCMs in LIBs.

Finally, there remains a long way to go for the widespread commercial application of BCMs in LIBs. Whether we can continue to improve the performance of BCMs, help expand the production of BCMs and

excavate more biomass materials for applications in LIBs are problems that need to be urgently and timely studied and addressed. In general, there is a lack of research on structure of original biomass and the corresponding properties of the resulting biomass char in relation to processing and carbonization conditions. If a selection guide for biochar materials can be established, such as a database on an open platform, including sources of biomass, corresponding structures, preparation methods, the capacity of each biomass carbon material and electrochemical performance, *etc*., it would be helpful for researchers and users to quickly screen biomass for desirable applications. It would also contribute to the use of BCMs in LIBs, which can promote and accelerate the sustainable development of renewable energy portfolios and achieve the goal of carbon neutrality.

DECLARATIONS

Authors' contributions
Reviewed literature extensively, wrote and modified articles, drew the figures, and set the layout: Lin W
Contributed numerous ideas for the article, polished and modified the text, and designed the figures: Zhao S
Polished the article and designed the figures: Lu B, Jiang F
Revised the article and provided the research direction and funding support: Lu Z, Xu Z

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All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate
Not applicable.

Consent for publication
Not applicable.

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