

# Research Progress of Lithium-ion Battery with Biomass Carbon as Anode

ZiHan Wu

University of Science and Technology of China, Hefei, China

wzh040617@mail.ustc.edu.cn

**Abstract.** Biomass materials are widely concerned because of their raw materials are rich and environmentally friendly, and their rich structure and high carbon content perfectly cut into the demand for electrode materials for lithium-ion batteries (LIBs). Nowadays, biomass-derived carbon materials (BCMs) have attracted more and more attention, and LIBs with BCMs as anode material have also developed rapidly. In this paper, the main sources and classification of biomass, as well as the commonly used modification methods when used as LIBs anode materials, are summarized, and their principles are briefly analyzed. The research progress of lithium-ion batteries with biomass carbon as anode is reviewed and summarized, and its future research and development are prospected.

**Keywords:** biomass, lithium-ion batteries, anode materials.

## 1. Introduction

Environmental problems such as the greenhouse effect and air pollution caused by the extensive use of fossil fuels such as coal and oil have aroused the vigilance of all countries in the world. Lithium-ion battery (LIBs) has the characteristics of high energy density, long life, and environmental friendliness. In the process of charging and discharging, lithium-ion stores and releases energy by embedding and stripping between positive and negative electrodes. For this reason, the green energy storage mode of LIBs is regarded as the key to getting rid of the dependence on traditional fossil fuels, and it is an important part of modern energy storage technology. In recent years, with the popularization and development of electric vehicles and various portable electronic devices, the market demand for lithium-ion batteries has increased, which further promotes the rapid development of this field.

At present, graphite is the main anode material of LIBs. Graphite anode has excellent cycle performance, high first charge-discharge efficiency and low cost, but its gram capacity is relatively low, only  $352 \text{ mA h g}^{-1}$ . Therefore, we are also looking for new anode materials and modifications of graphite materials to find materials with higher energy density. Materials such as hard carbon and soft carbon are all within the scope of our consideration.<sup>1</sup> Hard carbon has excellent surface chemical properties and some unique structures and has good lithium storage capacity. There are mainly three precursors, namely biomass, resins, and fossil fuels. This study will not discuss resin and fossil fuel precursors. The former has relatively limited material sources and high cost. The latter is not suitable as a precursor because of its high impurity content. Biomass carbon material is undoubtedly a better choice.

Biomass carbon material (BCMs) has a wide range of raw materials, including bamboo, coconut shells, various animal and plant tissues, etc., and its cost is low. At the same time, BCMs are rich in structure, which determines the diversity of their surface properties and functions. The structures of BCMs include carbon fiber, carbon nanosheets, carbon microspheres, etc. They represent the typical structures of carbon materials in one-dimensional, two-dimensional, and three-dimensional respectively (Figure 1). Because of the different surface properties brought by structures, their application fields are different. Next, we mainly discuss the layered structure obtained by stacking two-dimensional BCMs. Due to the short solid diffusion distance of lithium-ion in the closed space between battery packs, two-dimensional flaky BCMs are considered an ideal framework for the electrode design of lithium-ion batteries.<sup>2</sup> Nowadays, more and more BCMs are used as electrode

materials for lithium-ion batteries. Wu et al.<sup>3</sup> prepared a new type of hollow carbon with biomass lotus root as raw material by one-step carbonization. Lotus root material has many pores, so it has a rich mesoporous structure. This provides excellent lithium storage performance, and the carbon anode obtained at 900°C shows the best electrochemical performance.

When BCMs are used as electrode material, their mechanical properties are often limited due to their inherent heterogeneity.<sup>4</sup> And today's rich modification technology provides a higher limit for its development. The modification methods used for BCMs mainly include activation, element doping, metal material compounding, and so on.<sup>5</sup>

As a green environmental protection material, it is undoubtedly a promising research direction to apply BCMs to LIBs. In this paper, the working principle of LIBs, the types of BCMs used in LIBs, and the modification methods of BCMs are introduced. It aims to provide a reference for researchers in this field and further promote the research of BCMs in electrode materials.

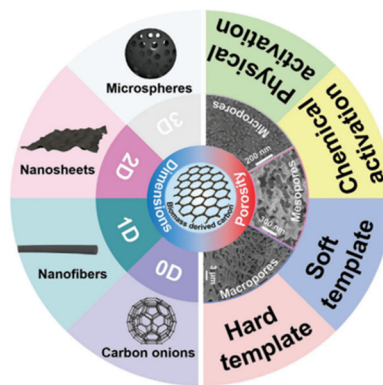


Figure 1. Schematic Diagram of the Diverse Structures of BCMs.<sup>2</sup>

## 2. Biomass Carbon in LIBS

### 2.1 LIBs working principle:

The working principles of all kinds of LIBs are similar, and the working principles of anodes are basically the same. Ferrous lithium phosphate, a common cathode in the market, is taken as an example (Figure 2). LIBs move back and forth between anode and cathode by  $\text{Li}^+$  in the battery and realize the conversion and storage of electric energy through current in the external circuit. During the discharge process, when the battery is connected to an external power supply and starts to use power,  $\text{Li}^+$  moves from the anode to the cathode through the electrolyte and the diaphragm and is embedded in the cathode material. At this time, the stored chemical energy is released and converted into electrical energy for external equipment. The charging process is just the opposite. The voltage applied by the external power supply reverses the chemical reaction of the battery, forcing  $\text{Li}^+$  to deintercalate from the cathode material, move to the anode, and embed in the anode material. At the same time, electrons flow from the anode to the cathode through the external circuit to maintain the charge balance formed by lithium ions in the cathode.

Charging process:  $\text{LiFePO}_4 - x\text{Li}^+ - xe^- \rightarrow x\text{FePO}_4 + (1-x)\text{LiFePO}_4$

Discharge process:  $\text{FePO}_4 + x\text{Li}^+ + xe^- \rightarrow x\text{LiFePO}_4 + (1-x)\text{FePO}_4$

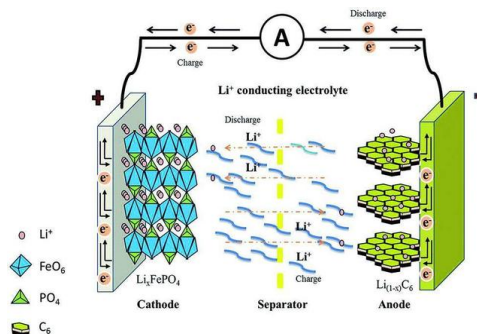


Figure 2. Working Principle of Rechargeable Lithium-ion Battery

In this process, the conventional carbonate solvent is unstable on the surface of the negative electrode with low potential, and reductive decomposition occurs, and then a layer of organic and inorganic mixed decomposition products, that is, SEI film, is produced on its surface. This film can inhibit the further decomposition of the negative electrode surface, thus playing a protective role.<sup>6</sup> However, it will also slow down the transmission efficiency of lithium ions and increase the loss of lithium ions to a certain extent, thus greatly reducing the first coulombic efficiency of LIBs. Therefore, it is also an important research direction to obtain dense and thin SEI films by improving electrolyte and electrode materials.

For the hard carbon anode materials focused on in this paper, due to the diversity of their precursors, they often have different microstructures. The research shows that lithium ions are mainly stored in hard carbon in two ways: adsorption and embedding.<sup>7</sup> Therefore, when choosing hard carbon materials, the specific surface area and other parameters that can reflect its adsorption and embedding ability will be taken into account. In addition, experiments have also proved that these materials with large specific surface areas often have better lithium storage capacity.

## 2.2 Biomass carbon materials in LIBS

When used in the field of lithium-ion batteries, we can roughly divide them into two categories according to their precursors, namely plant biomass and animal biomass. Plant biomass mainly includes cellulose and lignin, while animal biomass mainly includes chitin and chitosan 5 (Figure 3).

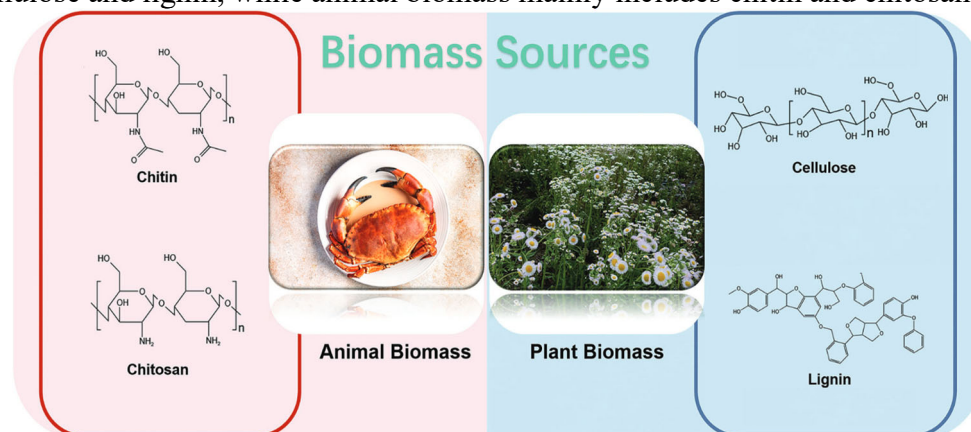


Figure 3. Summary of Typical Animal and Plant Biomass Precursors for BCM Preparation. Animal Biomass Precursors Include Chitin and Chitosan. Plant Biomass Precursors Include Cellulose and Lignin.<sup>1</sup>

### 2.2.1. Plant Biomass Carbon Materials

Cellulose and lignin are the main components of the plant skeleton. Plant biomass commonly used as electrode materials mainly consists of cellulose or lignin, such as jute, lotus root, and sisal. For cellulose in plants, we often separate it by alkali extraction<sup>8, 9</sup>, and other methods. Hard carbon materials made from these materials usually have high carbon content and a large specific surface

area.<sup>10</sup> This property has an excellent positive effect on the storage of lithium ions. Zhu et al.<sup>11</sup> absorbed  $\text{Mg}(\text{NO}_3)_2$  solution with absorbent cotton, and then dried and carbonized it to obtain the carbon material introduced into the MgO template. This realized the application of cotton with 90-95% cellulose content, one of the richest and most environmentally friendly biomass materials in nature, in LIBs electrode materials.

Dou et al.<sup>12</sup> activated jute fiber by  $\text{CuCl}_2$  to obtain carbon with a high specific surface area and porous structure of  $2043.5 \text{ m}^2 \text{ g}^{-1}$ . By comparing the activated carbon prepared from the mixture of jute fiber and copper chloride with the mass ratios of 1:0, 1:6, 1:8, and 1:10, it can be found that  $\text{CuCl}_2$  etches the carbon surface of jute fiber, resulting in a porous structure (Figure a-d), thus improving its lithium storage performance. It was used as the anode material of LIBs, and after 100 cycles, it showed an excellent specific charge capacity as high as  $580.4 \text{ mA h g}^{-1}$  at a current density of 0.2 C. Similarly, Yu et al.<sup>13</sup> pyrolyzed sisal fiber, and then the carbon material prepared by hydrothermal activation also achieved excellent results when used as anode material. This carbon material has a specific surface area as high as  $616.4 \text{ m}^2 \text{ g}^{-1}$ . After five cycles, the coulombic efficiency has reached nearly 100%, showing excellent cycle stability (Figure 4e).

Rice husk, straw, corn shell, peanut shell, and walnut shell all contain more lignin. Li et al.<sup>14</sup> extracted the carbon source from alkali-treated rice husk black waste liquid by acid precipitation, and the porous carbon material (Figure 4f) was prepared by one-step method at different temperatures with zinc chloride as the activator. When it was used as the cathode material of LIBs, it showed excellent performance far higher than the theoretical capacity of graphite. The results show that the porous carbon material obtained by one-step calcination at  $500^\circ\text{C}$  has the best cycle stability and high cycle-specific capacity. After 100 cycles, the capacity remains at  $469.0 \text{ mA h g}^{-1}$  (Figure 4g), which is better than most anodes reported in previous studies and much higher than the theoretical capacity of graphite. This excellent electrochemical performance is attributed to the existence of a large number of micropores and mesopores in the material and the specific surface area as high as  $7752.4 \text{ m}^2 \text{ g}^{-1}$ .

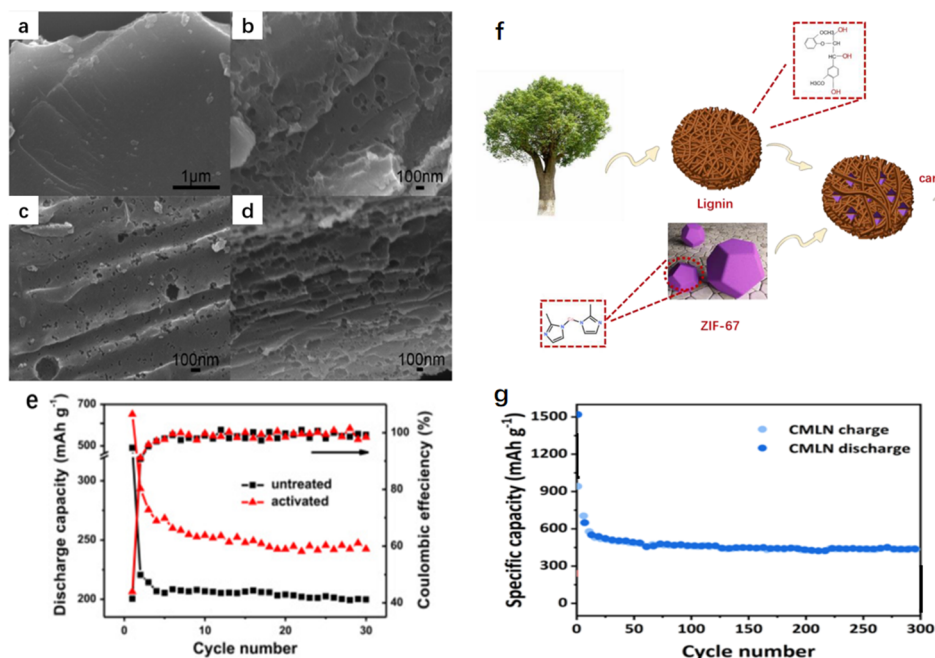


Figure 4. SEM Images of (a) JFC-0 (b) JFC-6 (c) JFC-8 and (d) JFC-10 Samples.<sup>12</sup> (e) Cycling Performance Curves and Coulombic Efficiency Plots during Cycling of the Electrodes.<sup>13</sup> (f) Fabrication Process of the CMLN Porous Carbo. (g) Cycling Performances of and CLMN at Current Density of  $100 \text{ mA g}^{-1}$ .<sup>8</sup>

### 2.2.2. Animal biomass carbon materials

Chitin and chitosan obtained from the cell walls of fungi and the exoskeleton of arthropods are the main substances that can be used as electrode materials in animal biomass. Chitin is similar to cellulose in structure, with high carbon content and a large specific surface area<sup>10</sup>, which gives it a structural basis for anode materials. Wang et al.<sup>15</sup> successfully prepared three-dimensional honeycomb nitrogen-doped carbon nanoplates/graphene (N-DC/G) nanonetwork films (Figure 5a-d) by dissolving chitin and graphene oxide in NaOH/ urea aqueous solution and carbonizing at high temperature in Ar atmosphere. This interconnected honeycomb structure with high electronic conductivity is beneficial to provide an electrolyte channel for active materials that is not limited by charge transfer has a short diffusion distance, and shows good electrochemical lithium storage performance. Even after 300 cycles at 500 mA g<sup>-1</sup>, it still has a high reversible capacity (798.86 mA h g<sup>-1</sup>) with a capacity retention rate of 94.21%.

Chitosan is the product of the deacetylation of chitin. Among all kinds of hard carbon materials with biomass as the precursor, chitosan has no obvious advantages when used as anode materials, but its carboxyl group can form hydrogen bonds (Figure 5e, f) with the surface of anode materials mainly composed of C or Si, thus reducing the damage caused by volume expansion and improving the cycle stability, that is, it can be used as an anode material binder for LIBs. Chen et al.<sup>16</sup> used cross-linked chitosan as an efficient adhesive for the silicon negative electrode of lithium-ion batteries, which makes the battery obtain higher reversible capacity.

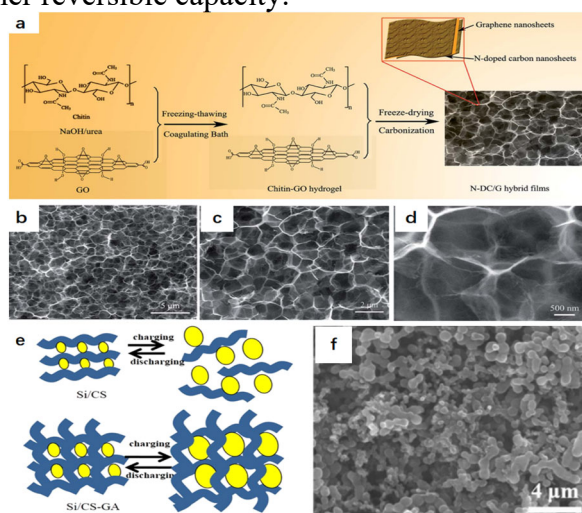


Figure 5. (a) Schematic Illustration of the Synthesis Process of Honeycomb-like N-DC/G Films. (b-d) Photograph and SEM Images of Cross-sections of the N-DC/G Nanohybrid Films at Different Magnifications.<sup>15</sup> (e) Illustration of the Charge/discharge Process of the Si/CS and Si/CS-GA Anodes. (f) Surface Morphology of Si/CS-GA Film.<sup>16</sup>

## 3. Carbon Modification of LIBs Biomass

As mentioned above, because BCMs are directly or indirectly derived from organisms, their inherent heterogeneity will greatly limit their electrochemical performance, and today's rich modification methods will solve this problem well. The commonly used modification methods in this field include activation, heteroatom doping, metal composite, and so on.

### 3.1 Activation

The main methods of activation include physical activation, chemical activation, and template activation. Through the reaction of biomass raw materials or precursors with activators, some carbon atoms are ablated and gasified, resulting in a large number of pores. Activation is the key to regulating the pore structure of carbon materials, which will directly affect the electrochemical and adsorption properties of porous carbon materials<sup>17</sup>.



Physical activation and chemical activation are traditional activation methods. The physical activation method refers to the method of using oxygen-containing gas such as water vapor, flue gas, or air as an activator to react with atoms inside carbon materials at high temperatures. Chemical activation means that chemical activators (such as sodium hydroxide, potassium hydroxide, phosphoric acid, etc.) react with raw materials at high temperatures. The separation of carbon atoms and the escape of gases produced by the reaction lead to the formation of rich pore structures in the framework of activated carbon. The mechanism of chemical activation and the performance of products depend largely on the type of activator, which mainly dehydrates or erodes raw materials and then produces pores. Guo et al.<sup>17</sup> prepared porous carbon materials by chemical and physical activation of chestnut seeds with KOH and CO<sub>2</sub> at 800 °C. In terms of specific surface area, compared with that before activation (17.1 m<sup>2</sup>/g), the specific surface area and total pore volume increased significantly after physical activation (105.7 m<sup>2</sup> g<sup>-1</sup>) and chemical activation (1221.2 m<sup>2</sup> g<sup>-1</sup>).

Conventional physical and chemical activation methods can produce a large specific surface area and pore volume, but it is difficult to obtain an ordered nano-pore structure. Template carbonization activation strategy can effectively control the morphology and pore size, and can be used to develop pore size and balanced and orderly microstructure in the range of 1-200 nm.<sup>18</sup> Li et al.<sup>19</sup> selected boric acid as a new activator and agaric was used as raw material. After soaking, the volume of dried agaric can expand to 3.5 times its original size, and boric acid can well enter the interior of agaric under the action of hygroscopicity. During carbonization, boric acid firstly melts into molten Be<sub>2</sub>O<sub>3</sub>, which has high viscosity and excellent wettability and adheres to the agar matrix, and then the Be<sub>2</sub>O<sub>3</sub> template is removed by KOH. The prepared material has a typical cellular porous structure with a specific surface area of 2279.5 cm<sup>2</sup> g<sup>-1</sup>.

### 3.2 Heteroatom doping

Heterogen doping refers to the process of introducing other elements (such as nitrogen, boron, oxygen, sulfur, etc.) into carbon materials to change their structure and properties. This doping can significantly affect the electrochemical properties of carbon materials.<sup>20</sup> For example, nitrogen doping can often change the electron structure of carbon materials to increase their conductivity, and at the same time, it can be used as the active site of electrochemical reaction to promote the charge transfer reaction, thus improving the intercalation and deintercalation energy of lithium ions in LIBs electrode materials.

Ou et al.<sup>21</sup> prepared nitrogen-doped porous carbon (NDPC) from Ginkgo biloba leaves. The synthesized NDPC was further studied as the anode material of lithium-ion batteries. The initial reversible capacity of carbon is 505 mA h g<sup>-1</sup>, which is 1.36 times the theoretical capacity of graphite (372 mA h g<sup>-1</sup>). After 10 cycles, the reversible capacity can still be maintained at 490 mA h g<sup>-1</sup>. Meanwhile, the initial coulombic efficiency of 58% can be achieved. The carbon anode shows good cycle performance and high capacity. The reversible capacity of the first cycle is 430 mA h g<sup>-1</sup>, and it gradually keeps at 410 mA h g<sup>-1</sup> after 500 cycles, which is still higher than the graphite's 372 mA h g<sup>-1</sup>. The battery was first cycled at 0.1 C 10 times, and then the discharge/charge rate was gradually increased to as high as 10 C for cycling. After 60 cycles at different rates, the reversible capacity of 486 mA h g<sup>-1</sup> was still achieved when the current rate returned to 0.1 C, which indicated that the NDPC electrode had excellent rate performance.

Zhao et al.<sup>22</sup> used rape pollen as a biomass carbon source and template and a porous micro-carbon ball framework composite titanium dioxide hybrid material was successfully prepared by a simple template-assisted sol-gel method and subsequent calcination process. The hybrid of N, PAC@TiO<sub>2</sub> has a hierarchical porous hollow structure, increased redox active sites, and specific surface area, which can provide high reversible specific capacity and improve the rate performance and cycle stability of Li<sup>+</sup> storage. As shown in Figure 7d, the first discharge and charge capacities of N, PAC@TiO<sub>2</sub> are 1417.7 and 998.2 mA h g<sup>-1</sup>, respectively. The rate performance of N, PAC@TiO<sub>2</sub> is shown in Figure 7e, and the specific capacities are 708.1, 643.8, 591.1, and 591.1 at current densities

of 0.1, 0.2, 0.4, 0.8, and 1.2, respectively. When the current density returns to 0.1 A g<sup>-1</sup>, the reversible specific capacity can still reach 687.9 mA h g<sup>-1</sup>, showing excellent rate performance and reversibility.

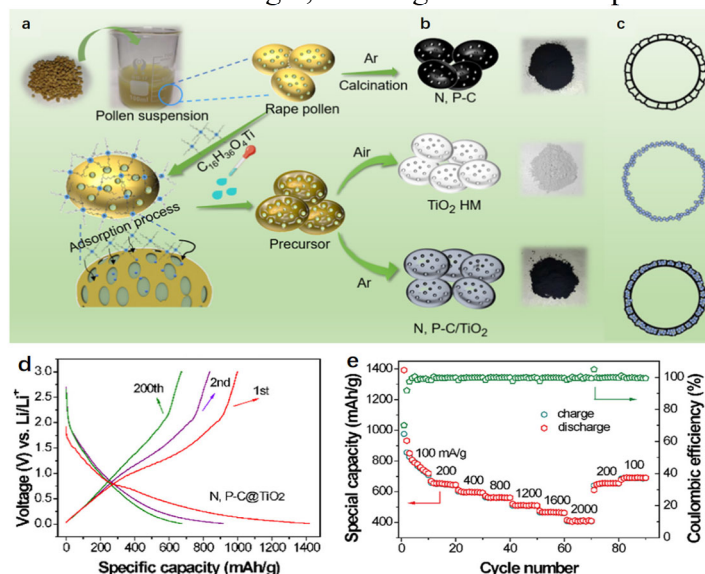


Figure 7. (a) Schematic Illustration of the Synthetic Route. (b) Digital Photographs of the N, PAC@TiO<sub>2</sub> HM and N, PAC@TiO<sub>2</sub> Samples. (c) Schematic Diagram of the Cross-section of the N, PAC, TiO<sub>2</sub> HM and N, PAC@TiO<sub>2</sub> Samples. (d) Charge/discharge Profiles at Current Density 0.1 A g<sup>-1</sup> of the First Three Cycles for N, PAC@TiO<sub>2</sub>. (e) Rate Performance of the N, PAC@TiO<sub>2</sub> Electrode.<sup>22</sup>

### 3.3 Metal composite

Metal composite carbon material is a composite material that is made by combining metal or metal oxide with carbon material. The existence of metal can enhance the mechanical strength and structural stability of composite materials, reduce the cracking or falling of materials caused by volume deformation during charging and discharging, and thus improve the cycle life. It can accelerate some electrochemical reactions, reduce the activation energy of reactions, and thus increase the reaction rate.

Liang et al.<sup>23</sup> successfully synthesized Ni/NiO/HHS-600 composites by hydrothermal method with folded porous carbon derived from hemp straw as a conductive matrix. The first cycle charge and discharge capacities of the composites are 2887.41 mA h g<sup>-1</sup> and 1258 mA h g<sup>-1</sup>, respectively. After 660 cycles, the charge-discharge capacity of the battery was stable at 1129.65 and 1124.94 mA h g<sup>-1</sup> and the coulombic efficiency increased from 43.57% to 99.58%.

Using carbonized waste pomelo peel as carbon substrate, Zhang et al.<sup>24</sup> successfully synthesized PPC/NiCo<sub>2</sub>O<sub>4</sub> composites by simple hydrothermal method. The prepared PPC/NiCo<sub>2</sub>O<sub>4</sub> composite has a carbon sheet with many wrinkles, on which NiCo<sub>2</sub>O<sub>4</sub> nanoneedles and nanoparticles are finely deposited. The composite material has mesoporous characteristics and a high specific surface area, and NiCo<sub>2</sub>O<sub>4</sub> is closely distributed on the carbon substrate. The reversible capacity of the composite anode is 473.7 mA h g<sup>-1</sup> after 210 cycles at a current density of 500 mA g<sup>-1</sup>, and it still maintains the capacity of 363 mA h g<sup>-1</sup> after 1100 cycles at a current density of 2000 mA h g<sup>-1</sup>.

## 4. Conclusion and Prospects

To sum up, LIBs with BCMs as anode have been precipitated and are still developing. By transforming biomass and other raw materials into anode materials of LIBs by activation, doping, metal compounding, and other modification means, the energy is efficiently utilized, and more importantly, they show excellent electrochemical performance, and they are not lost to common graphite materials in terms of cycle performance and capacitance. They have the potential to become a new generation of anode materials that are more environmentally friendly and excellent.

At the same time, there are still many problems to be solved in this field. Firstly, this study finds that doping is an excellent way to improve electrode materials, but in what way does doping of different elements, such as N and S, affect their electrochemical performance? Is it by affecting its binding energy or the arrangement of electron cloud density? Secondly, activation can improve the electrochemical performance of carbon materials by adjusting their pores, so how to balance the porosity, pore size, and pore connectivity of materials to get better electrode materials? Thirdly, the volume change of metal composite materials and the interface interaction between metal and carbon materials are all important factors that restrict the performance of the battery. What measures can we take to reduce its influence? Through the continuous study of these problems, we can better analyze the electrochemical properties and effects of biomass carbon materials, so as to seek more excellent materials.

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