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Applications of Carbon-Based Materials in Battery Anodes: Classification, Preparation Methods and Comparisons of Electrochemical Performances

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Abstract. Batteries have become indispensable in the modern energy storage field. However, the performance of current lithium-ion batteries is largely limited by the anode materials of the batteries. Graphite is most commonly used and most stable, but it has limited theoretical capacity and insufficient charging capability. To solve this problem, graphene and hard carbon are receiving increasing attention from research and experiments for becoming the alternative carbon-based anode materials. At the same time, carbon nanotubes are mainly applied into conductive frameworks, and enhance the performances of batteries. Each of these materials has its own advantages and disadvantages. Graphene achieves a balance between stability and properties, hard carbon is conducive to ion storage, and carbon nanotubes have the best conductivity and mechanical properties. This review summarizes the classification and development of carbon-based anode materials, describes the preparation methods of these materials: temperature-variable aerobic calcination, chemical vapor deposition, and pyrolysis. This emphasizes how different processing methods will affect various electrochemical properties. However, each material also has unavoidable drawbacks, such as cost, cycle stability, and environmental issues. In conclusion, a reasonable design that combines optimized synthesis, composite structures, and sustainable methods is crucial for promoting carbon electrodes towards high-performance and scalable energy storage systems.

Keywords: Carbon anode material, Graphite, Graphene, Hard carbon, Carbon nanotube.

1. Introduction

Batteries have become an important component in the modern energy storage system. From small electronic devices such as mobile phones and computers used by people in their daily lives, to the increasing number of electric vehicles on the roads, and even large-scale power transmission grids that are crucial for national development, batteries are needed and strongly supported in all these scenarios. With the advancement of technology, the world's demand for batteries with higher energy density, faster charging speed, and longer cycle life is constantly increasing. This makes it urgent to improve batteries to surpass the current state-of-the-art level. Among the various components of batteries, the anode plays a crucial role in determining the overall capacity, rate performance, and cycle stability.

Currently, due to low cost, high stability, and good reversibility, graphite dominates the anode market for lithium-ion batteries. However, its theoretical capacity is limited to 372 mAh/g [1], which is insufficient to meet the requirements of fast charging and high energy density systems. At this time, other carbon-based anode materials, such as graphene and hard carbon (HC), have received increasing attention. In addition, although carbon nanotubes (CNTs) cannot be directly served as anode materials, they can be widely used as a kind of conductive additives or structure supports of anodes. These materials have better properties, making them strong contenders and alternatives for the new generation of anode materials.

This study aims to integrate several carbon materials and review the development of anodes, focusing on the basic properties of various materials and some current preparation methods, as well as the relationship between structure and performance. Through comparison methods, this paper can correlate the preparation methods with the electrochemical properties of each material, and also more

clearly display the factors and solutions that affect capacity, cycle stability, and rate performance in each material. In summary, the purpose of this review is to summarize the current research progress, point out the existing limitations, and offer insights on the rational design of carbon-based anode materials. Through comparative analysis, this study can provide some insights and is expected to provide guidance for the development of sustainable and high-performance battery materials.

2. Carbon Material Anode Classification and Research Status

2.1. Graphite

Graphite is currently the most widely used anode material in the lithium battery market and manufacturing, accounting for 98% of the market share. Graphite is one of the most common forms of carbon allotropes and is also the most stable form of carbon. Graphite is formed by the sp^2 hybridization of carbon atoms, creating a hexagonal lattice, which undergoes planar layering. The weak van der Waals forces between the delocalized electron orbitals of carbon atoms connect each layer [2]. This structure has proven that graphite has a capacity potential of 372 mAh/g under conventional lithium intercalation [1].

This structure is very conducive to the ion insertion into graphite, forming graphite intercalation compounds (GIC), which can be well fabricated into battery anode materials and used. Graphite-alkali metals compounds are a relatively common type. In the alkali metal-GIC, lithium-GIC is the simplest one. Multiple studies have shown that reversible formation of LiC or Li_2C_2 materials GIC can even achieve a high capacity of over 1000 mAh/g [1].

However, after this material undergoes multiple charge-discharge cycles, it may cause the phase separation of lithium, resulting in uneven diffusion of lithium ions. This will lead to a decrease in rate capability, irreversible loss of capacity and power density, and an increase in initial charge loss, and even potentially trigger safety issues for the battery [3]. The solutions to this problem include reducing the graphite particle size, using Si-SiO_2-C composite materials to form amorphous carbon coatings, and pre-lithiation strategies, etc [1, 3].

2.2. Graphene

Graphene is a single layer of graphite, and it is also composed of carbon atoms that are sp^2-hybridized and arranged in a honeycomb-like hexagonal lattice. It is worth noting that graphene is a two-dimensional material consisting of only one layer of carbon atoms. This means that each atom in graphene can be regarded as a surface atom. If applied in lithium battery anodes as well, this special structure gives graphene a specific surface area of up to 2600 m^2/g and an electron mobility of up to 15000 $cm^2/(V·s)$ [4]. Meanwhile, graphene also possesses the excellent stability of graphite. This has made graphene a new potential material for composite materials used in making battery anodes.

Due to the excellent properties of graphene, it is currently more commonly used as a substitute material for traditional graphite. For instance, the modified tin-based oxide made from graphene has shown excellent performance. At a current density of 50 mA/g, its initial charge and discharge capacities are 1588 mAh/g and 1240 mAh/g respectively. After cycling 40 times at different current densities, the reversible discharge capacity still remains at 730 mAh/g [4]. Similar improvements can also be seen in modified silicon-based materials, where this compound has a theoretical charging capacity of up to 4200 mAh/g [4].

However, all graphene composite anode materials face a similar problem, which is that during the lithiation and delithiation process, the volume of the electrode material expands significantly and the internal stress of the battery also becomes large. This leads to the material being prone to cracking after repeated charging and discharging， resulting in poor cycling performance. Moreover, the initial Coulomb efficiency of graphene itself is also relatively low [4]. As of now, graphene cannot directly replace graphite as the anode material.

2.3. Hard Carbon

HC refers to carbon materials that do not undergo graphitization even at high temperatures. This substance can be produced through the pyrolysis of large-molecule polymers [5]. Since the superior performance of graphite materials' anodes is achieved under low current density conditions, to address this application challenge, using HC materials is a feasible solution [5].

The main structure of HC materials is randomly dispersed and bent graphene sheets. These sheets cannot be expanded or flattened, so they do not further stack to form graphite. Unlike the simpler structure of graphite, the spacing of graphene sheets, the type and concentration of defects affect the performance of HC [5, 6].

Currently, the three latest types of HC anode materials are produced from biomass, polymers, and soft carbon precursors [7].

Biomass-derived HC has a wide range of sources, low cost, is renewable, environmentally friendly. Moreover, it has a high porosity and is very suitable for storing ions [6, 7]. However, the natural growth of biomass materials is complex, so it is difficult to directly obtain the ideal structure for anodes. Meanwhile, the major drawback of biomass-derived HC is its high ash content and low carbonization rate, which will seriously affect the cost and electrochemical performance of the anode [7].

The advantages of using polymers to prepare HC anode materials include controllable synthesis process and easy microstructure design. Different polymer precursors and polymer methods can be selected to prepare various HC materials with special structures to obtain different properties [7]. However, the drawbacks are also obvious: the platform capacity, initial Coulomb efficiency, and overall energy density of polymer-based HC materials are very low [7].

The soft carbon precursor-based HC is produced from products with high carbon content and low cost from the petrochemical and coal chemical industries, such as asphalt, anthracite, and needle coke. A wide variety of HC materials can be produced. Moreover, this production method is currently the most commercially viable, meaning it has a low cost [6, 7]. However, by using this method, HC easily forms a graphite structure, resulting in smaller interlayer spacing and pore structure, which consequently leads to a decrease in electrochemical performance. Additionally, the actual performance of HC from different batches varies significantly, and it requires more precise screening before being used [6, 7].

2.4. Carbon Nanotubes

CNT refers to a nanoscale cylindrical structure composed entirely of carbon atoms. These carbon atoms are arranged in a hexagonal lattice, similar to rolled-up graphene sheets. CNTs come in two types: single-walled carbon nanotubes (SWCNT) and multi-walled carbon nanotubes (MWCNT). SWCNTs are composed of a single layer of graphene, while MWCNTs are made up of two or more layers of graphene, with van der Waals forces existing between adjacent layers [8].

CNTs possess excellent mechanical properties, good electrical conductivity and thermal conductivity, an ideal one-dimensional geometric shape and an extremely high surface-to-volume ratio, which are highly conducive to the formation of a continuous conductive network at extremely low penetration thresholds. These excellent properties make CNTs an especially suitable additive for enhancing conductivity and mechanical strength in composite anodes [8, 9].

CNTs are usually manufactured as independent nano-porous carbon films, allowing lithium ions to be inserted into the tubes. Even smaller and more ordered CNTs can be fabricated within the membranes of the tubes, providing good electrochemical activity for lithium-ion insertion on both the inner and outer layers [9]. Through this method, various studies have produced many composite materials, such as $Li_4Ti_5O_{12}$/MWCNT with CNTs added, transitional metal oxide/MWCNT composite anode materials, and overall nano-sized TiO_2/MWCNT, SnO_2/MWCNT and Si/MWCNT composite anode materials. Furthermore, MWCNTs can be modified through methods such as

chemically etching, ball milling, and changing their length. Subsequently, for example, compared to the original graphite anode, after chemically etching, the anode material containing MWCNTs acquires a large number of defects and pores, and can have a charging rate of up to 422 mAh/g and a discharging rate of 2087 mAh/g, showing a significant improvement [9].

However, there are also challenges in using CNTs to create composite materials. The adhesion of the nanoscale particles to the CNTs is relatively poor, making it difficult to achieve a uniform distribution. Moreover, the process of manufacturing CNTs itself has significant issues in terms of environmental protection, cost, and output, and these problems urgently need to be addressed [9, 10].

3. Preparation Methods

3.1. Temperature-variable Aerobic Calcination of Graphine

Researchers have discovered that by subjecting coal containing bitumen to long-term aerobic heating in a tube furnace, graphite can be obtained [11, 12]. This process can be called "temperature-variable aerobic calcination". Among them, in order to produce the final product graphite, it involves two processes: carbonization and graphitization. Graphitization is the most crucial step [12].

Graphitization is an effective step in the artificial synthesis of graphite. Graphitization can be defined as a process where a disordered graphite structure (i.e., "carbon") transforms into an ordered graphite structure [11].

Firstly, at 1000℃ for 4 hours, carbonization was carried out. Then, at 2000℃ to 2800℃ for 2 hours, graphitization was performed. The resulting graphite material, after electrochemical measurement, was determined to be suitable for use as a negative electrode material [12]. At the same time, there are cases indicating that coal tar, coke, asphalt, etc. can also be synthesized into HC through a very similar process, further becoming graphite [12].

After the carbonization step in the first part, the products were subjected to X-ray diffraction, and the results indicated that the fat chains and functional groups of the carbonized coal were decomposed, and a graphite-like microcrystalline structure was initially formed [12].

In the subsequent higher-temperature graphitization process, the graphite crystal structure increased, and the graphite crystalline layer spacing decreased. After increasing the heating temperature, it was found that the microcrystal size and layer stacking height increased sharply. This indicates that the increase in graphitization temperature can gradually intensify the growth of graphite, and the degree of graphitization gradually increases. It is worth noting that at 2600℃ and 2800℃, the data of the obtained products are increasingly close to the theoretical value of graphite, indicating that this is the production temperature for good crystallization [12].

The graphitization method can use low-value bituminous coal to synthesize graphite and has a highly ordered layered structure with a relatively large surface area [11, 12]. However, this method requires long-term high-temperature heating, and the energy cost may need to be considered.

3.2. Pyrolysis of Hard Carbon

Pyrolysis is a type of thermochemical transformation process, which refers to the chemical changes that occur when materials are heated in the absence of oxygen [13].

HC is usually prepared by thermal decomposition of organic polymers or sugars at high temperatures. At this time, the amorphous regions inside the HC mix with more graphite layers, and this structure prevents HC from continuing to graphitize [14].

The most economical method currently is to obtain HC materials through pyrolysis of biomass. It is worth noting that, through XPS analysis, the raw materials of biomass will have different platform temperatures due to the presence of different functional groups. Overall, within the range of 300℃

to 1000℃, the higher the carbonization temperature required to form HC, the greater the total surface area and pore volume of the formed HC [15].

Recently, various methods such as plasma sintering, Joule heating, carbon shock, and microwave carbonization have emerged one after another, all of which have enhanced the efficiency of the pyrolysis process. At the same time, they can also improve the internal structure of HC materials. For instance, carbon shock can instantly reach a high temperature of 2800℃, removing potassium from organic substances and making the structure more porous, resulting in a significant increase in capacity [15]. Additionally, the pyrolysis rate also has an impact on the structure and performance of HC materials, but further research is needed to understand exactly how the pyrolysis rate affects the carbonization process [15].

3.3. Chemical Vapor Deposition of Graphene and Carbon Nanotubes

Chemical vapor deposition (CVD) is a process that enables the desired solid material to be deposited from vapor through chemical reactions that occur on or near the heated substrate surface. The resulting solid is usually in the form of a film, powder or single crystal. CVD is widely used in industrial production, including the synthesis of graphene and CNTs [16].

There are many methods for producing graphene at present, such as CVD and mechanical exfoliation. This limits the manufacturing efficiency of devices and the application possibilities. The CVD method can directly produce large-area graphene films with average quality not too low and controllability [17]. Therefore, the following text mainly discusses the preparation of graphene by the CVD method.

Graphene films can be generated and collected on the surfaces of nickel or copper metals [18]. Firstly, use an argon-hydrogen mixed gas to evacuate the air, allowing the nickel or copper film to increase its grain size within the temperature range of 900℃ to 1000℃, and then expose it to a hydrogen/methane mixture. In this way, the methane serving as the carbon source will decompose, and the carbon atoms will dissolve into the nickel film to form a solid solution. Then, let it cool in argon gas to obtain a whole graphene film [18]. During the transfer step, apply a thin layer of polymethyl methacrylate (PMMA) to the graphene, then bake to evaporate the solvent. Next, use an etchant to remove the metal film, clean with deionized water, and bake again to evaporate the water. Finally, use acetone to remove PMMA and finally, obtain the graphene film [18].

When producing CNTs, there are other alternative methods available, such as laser ablation and arc discharge. However, the CVD method is still preferred due to its low cost, relatively high output, and good controllability [19].

A more specific method, the floating catalyst chemical vapor deposition (FCCVD) method was chosen. This method allows for more precise adjustment of parameters [19]. Hydrogen and argon are used as carrier gases to drive all substances to react. There are various options for the carbon source, such as hydrocarbons, carbon monoxide, and alcohols. Organic metal compounds such as ferrocene are used as precursors. Sulfur-containing substances, such as sulfuric acid and thiophene, are selected as growth promoters for CNTs and are in contact with the carbon source and react together with the carrier gas. There are many options for the feeding method, such as supersaturated vapor, injection, and hot wire generation, etc. [19]. There are many collection methods, such as directly pulling out the aerogel-shaped CNTs and rolling them into a film; after thorough cooling, using a specially designed rotating shaft to perform a similar spinning action to collect continuous CNT fibers; and so on [19].

CVD method is simple to handle and has good controllability, making it highly suitable for the production of thin film products. It is also very easy to scale up for industrial production. The output is also sufficiently pure, and there are multiple methods to obtain the desired form, making it very suitable for trade [16, 19]. However, the CVD method, especially the FCCVD method used for producing CNTs, also presents some significant challenges. Firstly, in actual production, the output

of the product is far less than the input of the carbon source and/or catalyst, which leads to waste and an increase in raw material costs. In the reactor system, the molar fraction of carbon is very low, indicating that the volumetric efficiency of the FCCVD process is very low. The growth mechanism of the film is also controlled because the growth mechanism of CNTs is not yet clear, so multiple experiments and precise calculation of the effective residence time are required. Even a slight change in parameters may lead to a significant reduction in efficiency [19].

4. Conclusion

This review summarizes recent progress in carbon materials for batteries anode, with a particular focus on their structural characteristics, electrochemical performance and preparation methods. This review systematically compares graphite, graphene, HC and CNTs, and discussed their typical synthesis strategies. Graphite is mainly used in anode due to its stability and low cost, but its limited theoretical capacity restricts future development. Graphene offers high conductivity and large surface area, but it has low cycling instability. HC is often derived from biomass, polymers or soft carbon precursor, and can provides large interlayer spaces and rich porosity that enable ion storage, though its Coulombic efficiency remains low. CNTs have excellent conductivity and mechanical strength and play an important supporting role as conductive networks, but the large-scale application is still limited by low output, high cost and potential environmental concerns.

Preparation methods such as temperature-variable aerobic calcination, pyrolysis and CVD impart distinct structural features that strongly influence electrochemical performance, such as crystallinity, porosity, and defect density. It is essential for further optimization to understand the correlations preparation-structure-property. Looking ahead, rational design integrating multiple strategies, including hybrid architectures and sustainable synthesis routes, is expected to drive the development of carbon anodes. For instance, biomass-derived carbons offer a promising pathway toward scalable, cost-effective, and environmentally friendly energy storage materials.

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