Proceedings of the 6th International Conference on Materials Chemistry and Environmental Engineering
DOI: 10.54254/2755-2721/2026.KA30374

Advanced Carbon-Based Negative Electrode for Energy Storage of High Efficiency Lithium-ion Batteries

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Abstract. In recent years, the ongoing advancement of society and technology has placed increasingly higher demands on battery performance. Currently, in commercial lithium-ion batteries (LIBs), graphite serves as the most widely used anode material. However, its specific capacity remains relatively low, making it difficult to satisfy evolving development requirements. However, carbon-based materials have a better theoretical specific capacity. When used as anode materials, they can significantly improve the battery performance, showing great development prospects. This article describes a detailed review of the research progress of carbon-based anode materials of LIBs. Firstly, we introduced the principle of LIBs, the overall classification of carbon-based materials and their characteristics. Furthermore, various modification methods of carbon-based anode materials for LIBs are discussed, including activation, doping, and composite with metal materials. Finally, the future research directions of carbon-based anode materials for LIBs are prospected.

Keywords: Lithium-ion Batteries, carbon-based materials, modification methods

1. Introduction

With the extensive use of fossil fuels including coal, petroleum, and natural gas, the energy structure has become irrational, leading to serious environmental pollution, energy shortages, as well as issues like global warming and the deterioration of the ecological environment. In order to reduce humanity's dependence on fossil fuels and address environmental pollution problems, the development of clean and renewable energy sources such as nuclear energy, solar energy and wind energy is an important means to solve the energy crisis. As a clean and renewable secondary energy source, lithium-ion batteries (LIBs) possess advantages such as high long cycle life, energy density, rapid charging capability, low self-discharge rate, diverse sizes and shapes, and environmental protection property [1]. It is currently the most widely used and most promising high-efficiency secondary battery, and also the chemical energy storage battery with the fastest development.

At present, although LIBs have unique advantages and are widely used in various fields such as the military, industry, transportation, and all kinds of electronic products, there are still a large number of problems in their practical applications. These mainly lie in the following three aspects: (1) Rate performance. The rate performance directly determines the charging and discharging efficiency of LIBs. Although the performance of LIBs has made great progress, when charging and

DOI: 10.54254/2755-2721/2026.KA30374

discharging at high rates, [2] there are problems such as excessive capacity fading and a decline in safety performance. (2) Cycling performance. The cycling performance is a core indicator for measuring the service life and reliability of LIBs. It determines the battery capacity retention rate and the degree of battery degradation as the number of charging cycles increases. For instance, high-quality battery cells, such as lithium iron phosphate batteries, typically maintain a capacity retention rate of over 90% after 1,000 charge-discharge cycles, whereas conventional battery cells retain only approximately 70% of their initial capacity under similar conditions. For example, Zou [3] and his colleagues proposed that by modifying the current collector with perforations, better cycling performance can be achieved.(3) Low energy density. Currently, the capacity density of mainstream lithium batteries is below 200 Wh/kg, while that of ternary cathode LIBs is between 200 and 300 Wh/kg [4]. Relatively speaking, such densities are too low for industrial scale applications. Therefore, the battery energy density is currently the biggest bottleneck in the development of LIBs.

Good anode materials can significantly improve the power density, capacity density, cycle life, and safety performance of LIBs. When choosing anode materials, three conditions should be met: (1) Lithium ions (Li^+) should be easily intercalated into and deintercalated from the anode material, and it should have a high Coulombic efficiency, so as to ensure a relatively stable charging and discharging voltage during the deintercalation and intercalation process of Li^+. (2) It should have good electronic and ionic conductivity, as well as good stability, and have a certain compatibility with the electrolyte. (3) The material should be sourced from abundant resources, be low in cost, have a simple manufacturing process, and be safe, green, and free of pollution.

Today, the mainstream anode materials are mainly carbon-based [5], silicon-based [6], and alloy-based[7–9].So far, carbon-based anode materials are the best overall performance LIBs anode materials that can meet the above requirements, and they are most widely used. At present, the more researched carbon-based anode materials are biomass carbon, graphite carbon, carbon nanotubes and so on. These anode materials have their own strengths and weaknesses, for instance, in recent years, with the in-depth study of carbon-based materials, it has been found that the performance can be improved by various modifications, such as activation (physical activation, chemical activation), doping of particles such as N, S, P, B, Co, Fe, and many composite alloys.

This paper firstly introduces the mechanism of LIBs and the role of carbon-based materials, and then reviews how all kinds of carbon-based materials can be modified to improve the performance, analyzes their role in the mechanism and effect, and describes the use of different carbon-based materials in LIBs. Finally, the development of carbon-based materials as anode in LIBs is summarized and prospected, with a view to providing reference for realizing the large-scale use of carbon-based materials in LIBs.

2. Mechanism introduction

2.1. The mechanism of LIBs

It is commonly understood that the operation of LIBs through reactions involving lithium ions as charge carriers, which continuously undergo intercalation processes at both the cathode and anode. A fully assembled lithium-ion battery comprises four key components: the cathode, anode, separator, and electrolyte. Both the cathode and anode materials are capable of supporting the insertion and removal of Li^+. Fig. 1 introduces the carbon-based materials of different configurations at present. The operational mechanism resembles that of a pendulum. During charging, lithium ions migrate from the cathode to the anode, while during discharging, they travel in the reverse direction—from

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the anode back to the cathode. In essence, lithium ions shuttle between the two electrodes in a cyclical manner, enabling the battery's charge and discharge functions [10].

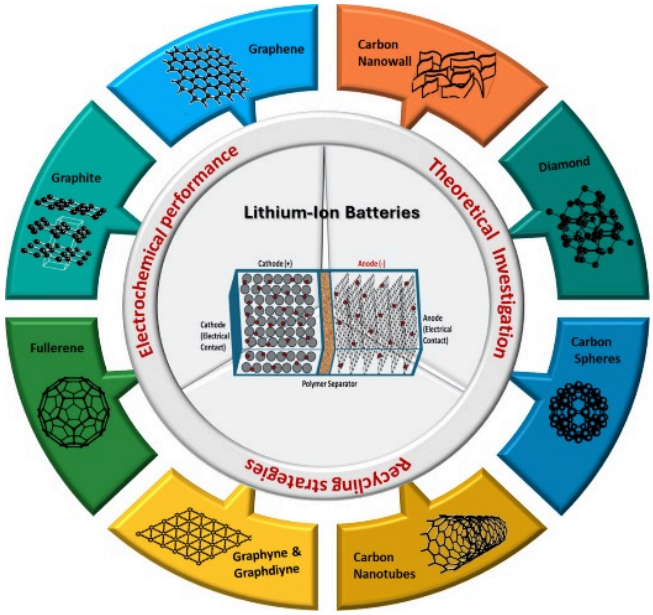

Figure 1. Carbon-based materials of different configurations

2.2. Carbon-based materials

According to the storage mechanism of Li^+ in carbon negative electrode materials, when using hard carbon as the negative electrode material, its capacity mainly comes from the Li insertion into the carbon layers as well as the adsorption of lithium on the surface of pores and defect sites. Currently, many hard carbon materials such as Polymer, Phenolic Resin, Epoxy Resin have been found to have high intercalation capacity, which is far higher than the theoretical capacity of graphite. For these hard carbons, a large number of studies and developments have been triggered.

For example, Liao and his group designed and tested spherical hard carbon negative electrodes for lithium-ion batteries. They mixed hard carbon with graphite powder at different mass ratios. Due to adding hard carbon, the mixture has a highly disordered structure, which is conducive to increasing the reaction rate [11]. Rao et al. aimed to develop a high-performance advanced carbon anode material. They prepared a new organic carbon material, hard carbon, by pyrolyzing polyacrylonitrile at 1050°C. The Coulombic efficiency, cycle stability and rate performance of this material were significantly superior to those of graphite electrodes [12]. Nurbolat Issatayev et al. proposed an economical and efficient cathode material, which is to use modified walnut-core hard carbon with lithium fluoride as the anode material for lithium-ion batteries. They found that after 100 cycles in a -20°C environment, its capacity remained at a high level of 280 milliampere-hours, solving the problem of performance degradation of traditional LIBs when operating below zero degrees [13].

Through research, it was found that it is quite different from the layered structure of graphite. Hard carbon mainly consists of single-layer carbon atoms that are closely connected in a disordered manner. Li^+ can be embedded into the structure formed by the combination of these single-layer carbon atoms.

In summary, in hard carbon materials, due to the unique carbon structure, such as single-layer carbon atom structure, disordered and irregular structure, lattice defects and doping of some other

DOI: 10.54254/2755-2721/2026.KA30374

atoms, etc., the lithium storage position is much larger than that of graphite materials, and thus the lithium intercalation capacity is particularly high [14].

Although the theoretical maximum lithium intercalation capacity of graphite materials is 372 mAh/g, the majority of the actual intercalation capacities are lower than this theoretical value. For lithium/graphite batteries, the voltage change is relatively stable, and the reaction of Li^+ intercalating into graphite materials mainly occurs below 0.2V. During its first charge and discharge process, there is usually a 15% to 20% irreversible capacity loss, part of which is used to decompose the solvent on the surface of the exfoliated graphite structure, while the other part is used for the initial reaction of Li^+ with the organic solvent on the carbon surface to form the SEI film.

In summary, although both natural graphite and artificial graphite have high capacities, due to the destruction of the graphite structure during charging and discharging, their re-production for practical use is rather difficult.

Carbon nanotubes are approximately one-dimensional, seamless hollow tubes formed by curling single or multiple layers of graphite sheets along the central axis at a certain helical angle. Their diameters are at the nanometer level, while their lengths are at the micrometer level [15].

Duan and colleagues employed mobile chemical vapor deposition to fabricate lignin-based carbon nanotube/carbon (L-CNT/C) nanocomposites, wherein carbon nanotubes were grown in-situ on a lignin-derived carbon substrate. Notwithstanding the advantages of the lignin-derived carbon- such as low cost, an innate porous architecture, environmental benignity, and tunable electrochemical properties, its development is constrained by a low mesopore ratio and limited conductivity, which are cardinal constraints on rate capability and power density. The in-situ integration of carbon nanotubes mitigates these issues by augmenting the mesopore fraction and bolstering the electrical conductivity of the hybrid material [16].

In recent years, there have been numerous studies on using carbon nanotubes as anode materials for LIBs. For instance, Erviani Rusman team successfully created a composite material through a hydrothermal method, using the affordable and environmentally friendly GeO_2 precursor. In this material, the nitrogen-doped carbon nanotubes provided more active sites and enhanced the structural stability, thereby improving their electrochemical activity. At a ampere density of 100 mA g^{-1}, after 100 cycles, the capacity was 1017 mA h g^{-1}, maintaining a Coulombic efficiency of 98.15% and a capacity retention rate of 71.15% (compared to the second cycle discharge capacity) [17].

3. Materials classification

3.1. Activation

Supercapacitors store energy through a synergy of double-layer capacitance and pseudocapacitance. Their performance depends on electrode materials, electrolytes, and separators, with electrode materials being the decisive factor for capacitance. Consequently, the properties of the electrode material directly dictate the device's overall performance. Given this critical role, an effective electrode material requires a large specific surface area, a well-tuned pore structure, excellent conductivity, light weight, and low cost. The activation process is key to engineering the pore structure, thereby directly determining the material's efficacy in electrochemical and adsorption applications. Therefore, the research on the activation mechanism during the material preparation process is of great significance [18].

Physical activation method: Agricultural biomass waste (ABW) can be used as raw materials to produce various products, such as biofuels, composite materials, carbon nanotubes and activated carbon. It has significant social and economic benefits in creating sustainable processes and

DOI: 10.54254/2755-2721/2026.KA30374

generating employment and income. Through precise control of parameters like heating rate, heat treatment temperature (HTT), and steam conditions, the Gilberto Maia Brito team prepared activated biochar (AB) with high surface area and pore volume. Their one-step carbonization/activation process utilized steam at 800 °C and precursors including SB, CS, and EB [19].

To boost hydrogen storage capacity, the team of Pargai Neema employed Pekala's sol-gel method to synthesize carbon aerogels, followed by physical activation. Fig. 2a displays the schematic design illustrating the synthesis process of CAs, and Fig. 2b shows the RF resin formation by polymerization. The activated aerogel achieved a specific surface area of 799.68 m^2/g-significantly higher than the pristine material-along with a pore volume of 0.47 cm^3/g. This structural optimization resulted in markedly enhanced hydrogen uptake [20].

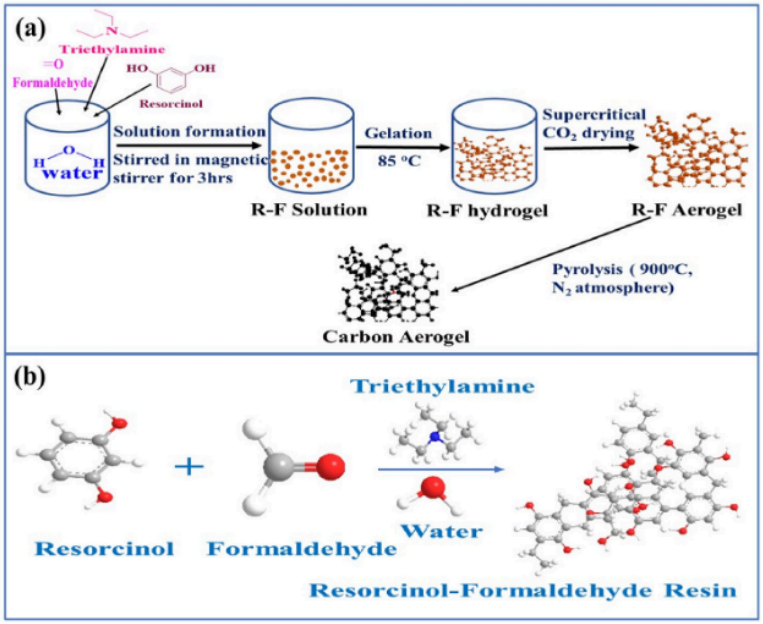

Figure 2. (a) Schematic diagram depicting the synthesis of RFA and its subsequent conversion to CA. (b) The RF resin, which is formed by the polycondensation of resorcinol and formaldehyde

For the chemical activation method: The M. Auezov team applied chemical activation to overcome the low yield problem in traditional phosphorus fertilizer production. This strategy enhanced the assimilable phosphorus content and facilitated a direct route to phosphorus-containing fertilizers, circumventing the need for acid and avoiding waste [21]. A two-step method was developed by Suhdi and Wang to synthesize carbon nanofibers from rubber shells (RFS). The process involved pyrolysis of this renewable precursor, followed by phosphoric acid activation via a hydrothermal treatment at 90 °C to produce RFSAC. This approach is notably simple and economical, as it dispenses with the need for any catalyst, vacuum, or gas [22]. The morphology is shown in Fig. 3.

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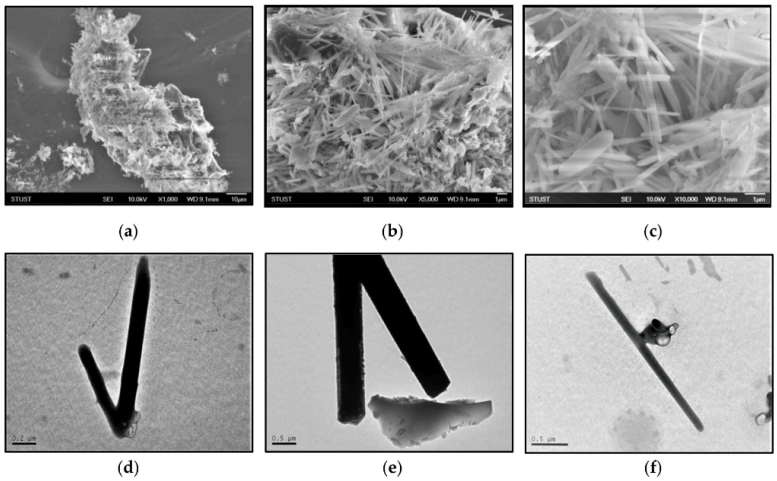

Figure 3. The carbon nanofibers images of (a–c) SEM and (d–f) TEM

3.2. Doping

Although carbon-based anodes for LIBs exhibit good performance, their capacity is notoriously difficult to improve. Heteroatom doping, such as with silicon, is a promising strategy. A study by Wu et al. on silicon-doped carbon demonstrated that within a concentration range of 1.56% to 15.6%, increasing the Si-C bond concentration enhances the bulk modulus. Consequently, the doped structure's volume expansion during cycling is suppressed, an effect that is more pronounced at higher carbon concentrations. While this enhancement improves structural stability, it also increases the Li+ diffusion energy barrier. Ultimately, the overall effect of carbon doping leads to a higher anode capacity [23]. Zhang et al. developed a $PbSnO_3$-doped carbon nanocomposite anode for LIBs, which addressed the issue of volume expansion during cycling, thereby enhancing stability. The composite exhibited outstanding electrochemical performance, delivering reversible capacities of 1200 mAh g^{-1} at 476 mA g^{-1} and 100.1 mAh g^{-1} at 770 mA g^{-1}. These results confirm the significant promise of $PbSnO_3$-based nanocomposites as high-performance anode materials [24].

3.3. Recombination

The ongoing advancement of new energy technologies has motivated the search for alternative anode materials beyond traditional graphite, which is limited by its theoretical specific capacity of 372 mA h g^{-1}. In parallel, composite strategies involving graphite have gained significant research interest. One example is the SiOx/C composite anode developed by Zhou et al. for high-capacity LIBs. The microscopic morphology structure is shown in Fig. 4. When evaluated as an anode, this material showed stable cycling performance at a current density of 100 mA g^{-1}, delivering an initial reversible capacity of 1213.9 mAh g^{-1} with a Coulombic efficiency of 65.7%. After 100 cycles, it maintained a high reversible capacity of 1292.4 mAh g^{-1}, while the Coulombic efficiency increased to 99.7% [25]. Su and his team successfully fabricated a multi-layer silicon-based composite material, which was formed through a two-step in-situ carbon coating process and consists of multicomponent carbon. According to SEM and TEM images, the nano-silicon particles are uniformly dispersed within the multilayered carbon matrix, which is built from resin pyrolysis carbon and reduced graphene oxide. When applied as an anode in lithium-ion batteries, this composite exhibits outstanding electrochemical performance. Test results indicate that the material

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delivers a high initial charging capacity of 1474.9 mAh g^{-1}, along with an initial Coulombic efficiency of 74.57% [26].

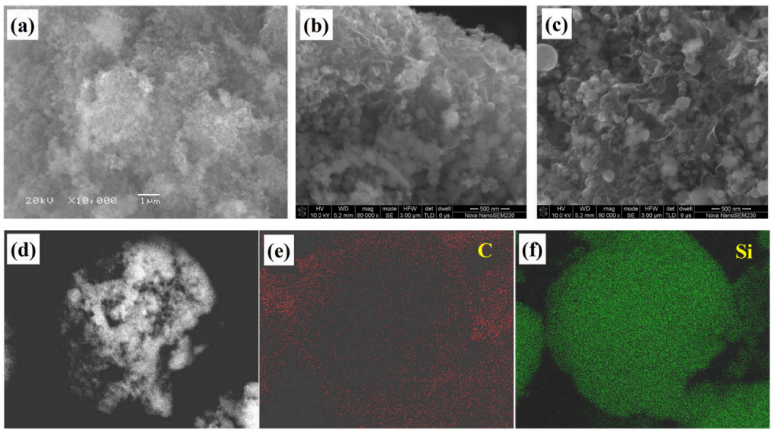

Figure 4. SEM images of (a) Si@C composite, (b-c) Si@C@RGO composite, (d-f) EDAX spectra dots of silicon and carbon

4. Conclusions and future directions

This article reviews the application of carbon-based materials as negative electrode materials in lithium-ion batteries. Based on literature research, it is found that carbon-based materials have been widely used as anode materials in LIBs. However, because of the continuous progress of new energy technologies, traditional graphite anode materials can no longer meet the demands, and thus need to be modified to improve their performance. In the future, research can be conducted from the following aspects:

(1) Using activation techniques, such as physical activation, chemical activation, and template activation, to regulate the pore structure of the materials, thereby enhancing their electrochemical and adsorption performance.

(2) Doping, introducing other substances into carbon-based anode materials to reduce the volume expansion caused by the charging and discharging processes, and thereby enhancing the cycling performance of the materials.

(3) Compositing to improve material performance, which can be achieved by combining with high-conductivity materials to construct efficient electron transmission channels, reduce the internal resistance of the materials, alleviate the volume expansion during cycling, and thereby enhance their rate performance and achieve fast charging and discharging.

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