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Improve Lithium-Ion Battery Performance: Amorphous Carbon, Graphene, and Carbon Nanotube-Modified Silicon Anodes

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Abstract:

With ultra-high theoretical capacity, silicon is seemed as a potential anode for the next generation lithium-ion batteries. Its extremely huge volume expansion (~300%) of Si anode upon lithiation-delithiation processes, however, causes particle pulverization and unstable SEI, hinders its practical application. To solve these drawbacks, related Si-C composite strategies have been proposed by researchers. This review systematically covers typical three types of carbon materials (including amorphous carbon, graphene, and CNTs) that can be employed to modify nanosilicon anodes. Amorphous C has a uniform coating, and economical preparation, which improves stability but it is not sufficiently conductive. Graphene provides high conductivity and mechanical strength, but it is featured by high cost and aggregation. CNTs can be used as a conductive and flexible networks to enhance the cycle life, however, they are difficult to disperse and interfaced. Each carbon population also exhibits unique merits and limitations in improving the mechanical strength, conductivity, and chargeability of silicon anodes. Further work is desired for investigating hybrid carbon structures, tunning interfacial bonding, and scalable, green synthesis. These developments will promote the commercialization of high-energy-density silicon−carbon anodes for advanced Li-ion batteries.

Keywords: Si anodes; Carbon coating; Lithium-ion Battery.

1. Introduction

Lithium-ion batteries are very common to use in electronic devices, automobiles and energy storage systems, thus demanding high requirements for their capacity, cycle life and safety[1]. Traditional graphite anode materials, due to their limited capacity, are unable to meet the demands of batteries with high energy density. As the demand for high-performance batteries in new energy vehicles and portable electronic devices continues to grow, lithium-ion batteries are widely used because of its their high energy density, long cycle life and low self-discharge rate. However, the theoretical specific capacity of existing graphite anode materials only 372 mAh/g, which is not able to meet the demands of high energy density batteries[2]. Therefore, developing high specific capacity anode materials become one of the key research directions.

The theoretical specific capacity of silicon is up to 4200 mAh/g, which is more than ten times that of graphite, and has abundant reserves in the earth's crust and good environmental friendliness, making it one of the ideal alternative materials. However, silicon undergoes significant volume changes during charging and discharging leading to electrode pulverization increased electrolyte side reactions and unstable electrode interfaces which limit its practical application[3]. Nanosilicon, with its extremely high theoretical specific capacity and overcome the weak conductivity , has become a popular candidate material to replace graphite. However, during charging and discharging, silicon undergoes a volume expansion of over 300%, leading to structural pulverization and rapid capacity degradation. Therefore, researchers have combined nanosilicon with conductive and flexible carbon materials to enhance its structural stability and conductivity, making it a research hotspot in recent years[4].

Researchers have adopted carbon composite strategies to combine nanosilicon with different carbon materials (such as amorphous carbon graphene, carbon nanotubes) to prepare composite anode materials. These carbon materials can not only effectively buffer volume expansion but also provide a good electronic conductive network, improving structural stability of the electrode. For example, improved the cycling performance of silicon by coating graphene maintaining over 82%capacity after 1200 cycles[5].

This article reviews the research progress of nanosilicon-carbon composite materials as anode materials for lithium-ion batteries analyzes the influence of different carbon-based structures (for example amorphous carbon reduced graphene oxide, carbon nanotubes) on the composite performance introduces typical research cases, and proposes suggestions for improvement. And will compare the structural advantages and electrochemical performance from three typical carbon materials combined with nanosilicon, list key cases, and propose improvement suggestions to provide reference for the improvement of high-performance silicon-based anode materials. Finally, it summarizes that current research trends and looks forward to future development directions.

2. Case

2.1 Amorphous Carbon-Modified Silicon Anode

Amorphous carbon is a structurally disordered carbon formation. Typically it combined with nano-silicon through polymer pyrolysis or mechanical ball milling to form coated or embedded composite structures. The composite can retain about 80% of its capacity after 500 cycles at a current density of 0.1C, demonstrating fantastic stability and long cycle life.

2.2 Graphene-Modified Silicon Anode

Graphene features a two-dimensional structure with excellent electrical conductivity and flexibility. It can effectively construct a three-dimensional conductive network by wrapping around silicon particles to form flexible composite structures. The Graphene/Si composite anode retains 92% of its capacity after 200 cycles and exhibits good rate capability.

2.3 CNTs (Carbon Nanotubes)-Modified Silicon Anode

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3. Analysis & Challenge

3.1 Amorphous Carbon-Modified Silicon Anode

Amorphous carbon-modified silicon anode materials are mainly synthesized by coating silicon particles with amorphous carbon to enhance the electrical conductivity and chemical stability. Common synthesis methods contain chemical vapor deposition , sol-gel process, high temperature carbothermal reduction, and mechanical ball milling. CVD method decomposes carbon gase at high temperature to deposit a uniform amorphous carbon layer with silicon particle surface, forming a dense coating, enhances electrical conductivity [6]. The sol-gel process mixes a carbon-containing sol with silicon, which after decomposition forms an amorphous carbon layer for large-scale production [7]. The high-temperature carbothermal reduction method refer to mixing carbon and silicon sources followed by high-temperature calcination to produce carbon-coated composite materials,this method is simple and cost-effective[8]. Mechanical ball milling achieve carbon and silicon composite materials by physical mix-

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ing; it is convenient but the result lack of continuous and uniformity of the coating layer[9]. A typical Amorphous Carbon-Modified Silicon Anode was shown in Figure 1.

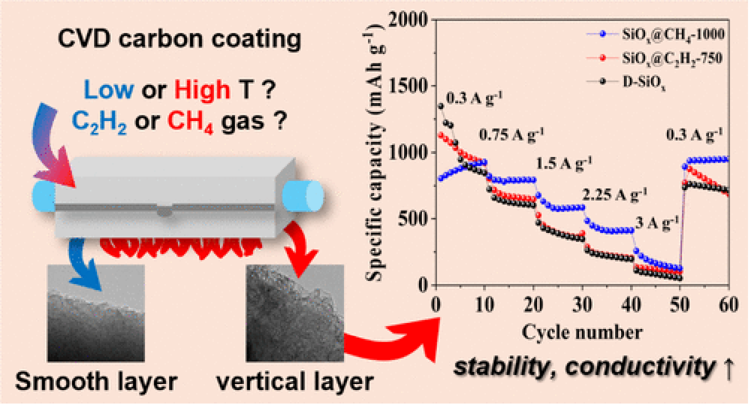

Fig. 1 Synthesis of Amorphous Carbon-Modified Silicon Anode by CVD [10]

Amorphous carbon coating remarkably improve the electrochemical performance of silicon anodes. The carbon layer not only provides a good conductive network but also effectively buffer the volume expansion of silicon during lithiation (around 300%-400%), reducing mechanical stress and prevent particle pulverization and electrode structure collapse[11]. Additionaly, the coating aids in forming a stable solid electrolyte interphase (SEI), reducing side reactions with the electrolyte, and significantly enhance the cycle stability and rate[12]. Related studies have shown that amorphous carbon-modified silicon anodes maintain a much higher capacity retention after multiple cycles compared to uncoated silicon, demonstrating better cycle life[13].

Although amorphous carbon-modified silicon anodes exhibit excellent performance, challenges remain such as difficulty controlling the thickness and uniformity of the coating layer, mechanical stress caused by volume expansion, insufficient interface stability, and high preparation costs. Future research should focus on optimizing carbon layer design, enhancing interface stability, reducing preparation costs, and promoting practical applications.

3.2 Graphene-Modified Silicon Anode

Graphene-modified silicon anodes are usually prepared by mixing graphene sheets with silicon particles in order to increase electrical conductivity and mechanical stability. Typical syntheses are chemical vapor deposition (CVD), hydrothermal/solvothermal synthesis, solution blending plus freeze-drying, and ball milling. In CVD process, the graphene is grown/transfer directly on the silicon surface in order to obtain a consistent, conductive modified surface. Both hydrothermal and solvothermal routes reduce graphene oxide (GO) in the presence of silicon, to get graphene-silicon composites. 144-146 Solution blending causes a homogeneous mixing of graphene and silicon dispersions and it is often followed by freeze-drying or spray drying to fabricate the composite powders. Graphene and silicon are powder-like materials, and ball milling can physically mix them, providing a facile and large-scale method, but the distribution quality could be different. Ball milling process for graphene-modified silicon anode was shown in Figure 2.

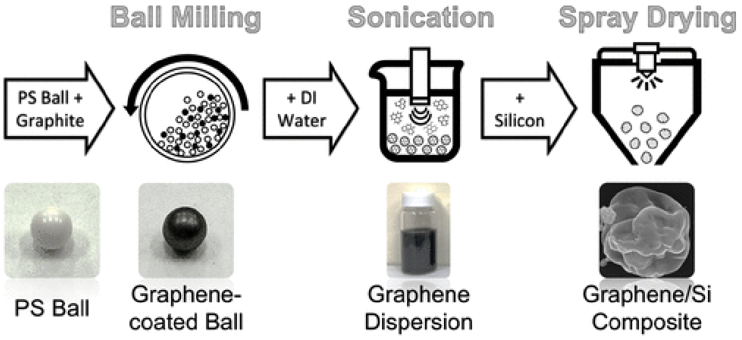

Fig. 2 synthesis of Graphene-Modified Silicon Anode[14]

Functionalization of silicon anode with graphene makes a great enhancement for electrochemical properties. The outstanding electrical conductivities of the graphene form electron pathways, which together with these efficient electron transfer stations to improve charge transfer. It has good mechanical strength and flexibility to accommodate the large volume change (~300%) of silicon during lithiation, which minimizes the particle pulverization and preserves the electrode integrity[15]. In addition, graphene layers can deposit a stable SEI and protect the side reaction of silicon surface, reflected in the cycle stability and rate capability improvement. The experimental results demonstrate that graphene-decorated silicon anodes present more than 85% capacity retention after 100 cycles, and the performance is better than that of naked silicon electrodes [16].

Despite the good performance, graphene-silicon anodes are confronted with a number of challenges. However, obtaining graphene with uniform silicon nanoparticles at large scale is still challenging, which affects the uniformity. Moreover, the interfacial contact between graphene and silicon may be poor, resulting in debonding during cycling and leading to poor cycling stability. Additionally, the price of premium graphene and complication of some synthesis processes cautions the commercial scalability. Subsequent research needs to focus on optimization of the graphene combining method, strengthening the interfacial adhesion, lowering the cost of production and scaling up the production to make them practical.

3.3 CNTs Modified Silicon Anode

CNTs-modified silicon anode materials are prepared by the coattaching of CNTs on the silicon particles for improved electrical contact and mechanical stability. Synthesis routes are solution mixing with vacuum filtration or freeze-drying, chemical vapor deposition (CVD) growth of CNTs on silicon and mechanical ball mill. Solution blending shown in Figure 3: A homogenized mixture of CNTs and silicon nano-particles in solvent is prepared to form a composite band, which is then dried to form composite powder. By employing CVD yield CNTs can grow directly on the surface of Si, so that the interfacial force is strong. The process of ball milling physically combines CNTs with silicon powders for easy composite fabrication, but is less uniform.

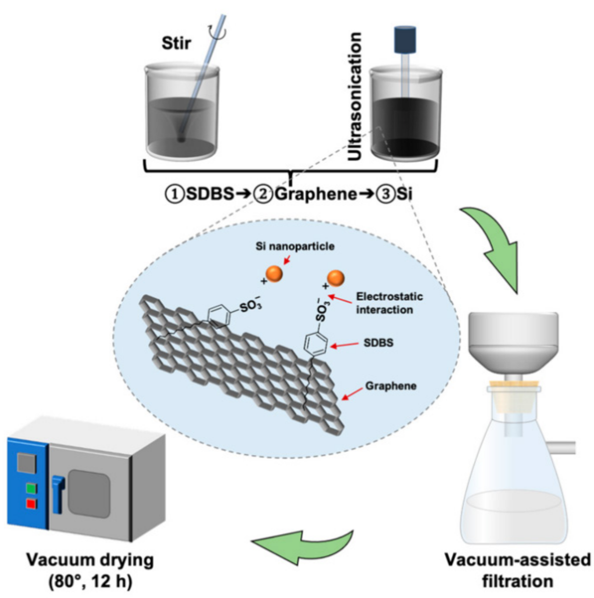

Fig. 3 synthesis of CNTs Modified Silicon Anode[17]

The electrochemical performance of silicon anodes is greatly enhanced by the introduction of CNTs. The CNTs demonstrated superior electrical conductivity and mechanical flexibility to maintain the robustness of the electrode during the volume expansion (~300%) of the Sis during the lithium intercalation[18]. A CNT network is formed as an electron conduction path and for buffering the volumetric change of silicon particles, resulting in low particle pulverization and electrode degradation. CNTs also help form a quite stable SEI, for a superior cycling stability, rate performance. CNT-coated Si anodes are reported to have a capacity retention of above 80% after 100 charge-discharge cycles [19].

However, CNTs-modified silicon anodes still have some issues to be addressed, including uniform distribution of CNTs, strong interface bonding between CNTs and silicon, and high expenses and complicated synthetic methods of CNTs. The CNTs aggregating and no uniform covering may prevent them working uniformly. In addition, there are many constraints for the scalability of CNT fabrication and integration technologies. It is necessary to further enhance dispersion of CNTs, interfacial adhesion,

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simplicity and cost of CNT synthesis, scalability of the production process, and etc., for future operations in industrial applications

4. Summary & Suggestion

Table 1. Comparation of various Carbon-modified anodes

Type	advantages	disadvantages
Amorphous Carbon	Have good electrical conductivity High chemical stability	Difficult to spread envenly
Graphene	Have good electrical conductivity Ductile	High cost Tendency to aggregate
CNT	Good corrosion resistance	Interfacial adhesion issues

The 3 kinds of material mention in works were compared and concluded in Table. 1.

Amorphous Carbon modified Si as anodes have advantages likes good electrical conductivity, it increase the efficiency of battery charging, and due to high chemical conductivity, it can help reducing the negative reaction between electrolysis and material ; on the other hand, it have disadvantages like difficult to spread evenly, the thick part will increase resistance but the thin part will not buffer the volume expansion efficiency.

Graphene-modified Si as anodes have advantage likes ductile, it can buffer the volume expansion more efficiency and enhance the stability; on the other hand it have disadvantage like it is easily to aggregate together, it will decrease the surface area of reaction and negatively affects its conductivity.

CNT -modified Si as anodes have advantage likes good corrosion resistance. It can reduce a lot of negatively effects, and improve the safety and lifespan of battery; on the other hand, it have disadvantage likes interfacial adhesion issues, it may cause interface delamination during cycling and cause safety problem as well..

Conclusion

In conclude, this work be concerned about applying of nano-structured silicon-carbon (Si–C) composite materials as the prospective anode material of LIBs. Three typical carbon materials, including amorphous carbon, graphene, and CNTs, have been highlighted in improving the electrochemical performances of the silicon anodes.

Conclusion 1: Amorphous carbon provides good uniform casting characteristics, medium conductivity, and is easy to scale up synthesis. It can well accommodate with volume expansion of silicon and stabilize the solid electrolyte interphase (SEI) film. Its low conductivity and interfacial adhesion, however, need to be further improved.

Conclusion 2: The addition of graphene in composite increases the conductivity, strength and capacity, and the large surface area character of graphene also develop the rate performance of modified composite material. But, the high price, agglomeration tendency, and weak combination with silicon are the main bottlenecks.

Conclusion 3: CNTs constitute a conductive and flexible 3D network, which can enhance the cyclic life and the structural integrity of the electrode. However, problems, including the difficulty of dispersion, the interfacial bonding and the environmental protection, still restrict the practical application at the step of mass production.

To develop silicon-carbon composite anodes we need the following future works. Developing hybrid carbon structures (such as amorphous carbon combined with graphene or CNTs) that take advantage of the synergistic effects; Optimizing interface engineering to increase the bonding strength, long-term stability, To establish low-cost, scalable fabrication processes with the high performance; Search for environment-friendly and safe carbon sources for sustainable development. The solutions to these issues will bring silicon-carbon composite anodes one step closer to practical use in the next-generation high-energy lithium ion batteries.

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