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Silicon-Carbon Composite Anode Materials in Lithium-ion Batteries

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Abstract. As new energy and electric cars become more popular, the need for better lithium-ion batteries is rising. Traditional graphite anodes in lithium-ion batteries don't store enough energy to meet today's high demands. Because silicon can store much more energy, it has become a great alternative material for battery anodes. However, it has problems such as volume expansion and low electrical conductivity during charging and discharging. To address these deficiencies, scientists have designed silicon-carbon composite materials as the anode material for lithium-ion batteries. Silica graphite composites strike a balance between capacity and stability, among which the spray-dried core-shell structure demonstrates the best performance and industrial potential. Silicon–graphene composites build a three-dimensional conductive network that improves conductivity and reduces swelling; after treatment, which gives them higher charging efficiency though they are expensive and difficult to make. Silicon–carbon nanotube composites form one-dimensional conductive networks that improve electron transfer and reduce expansion. However, they are costly and difficult to produce on a large scale. In conclusion, silicon-carbon composites combine the high capacity of silicon with the excellent electrical conductivity and stability of carbon, enhancing the performance of batteries of electric vehicles, portable electronic products, and renewable energy storage. Although there are still challenges in terms of cost, stability and cycle life, improvements in design and production may make it a key anode material for the next generation of high-energy lithium-ion batteries.

Keywords: lithium ion batteries, anode materials, silicon carbon composite materials

1. Introduction

Since the world's technology and industries have continuously improved a lot, the need for high-efficiency energy and renewable energy has increased. For a long time, the development of human society has been highly dependent on fossil energy. In modern society, most industries and transportation rely on non-sustainable energy sources like coal, natural gas, and oil. In 2024, natural gas, coal, and oil accounted for a 54% share of the total rate of growth in energy supply [1]. However, these non-renewable energy sources will eventually run out one day. In addition, the application of fossil fuels can also cause environmental pollution. Therefore, finding an alternative renewable energy source that can mitigate environmental pollution is really important.

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To solve these problems, electric transportation like electric cars, electric motorcycles, etc., was produced. This has resulted in an increasing demand for improved batteries. Lithium-ion batteries are the most popular batteries nowadays. Compared to other batteries, they have improvements in their capacity, efficiency, and duration, meanwhile reducing environmental pollution. However, in the practical application of lithium-ion batteries, there are problems such as volume expansion and capacity attenuation, which still cannot meet the demands [2].

At present, graphite is the most commonly used anode material for lithium-ion batteries; nevertheless, its theoretical capacity is only 372 mAh/g [3], which is quite far from meeting the current requirements. Therefore, choosing high-capacitance materials to replace graphite is considered the best solution [4]. For example, the theoretical capacity of silicon combined with lithium is 4200 mAh/g. Additionally, silicon is still one of the most abundant maters on Erath and is non-toxic and pollution-free. These properties of silicon make it the most likely to replace graphite in batteries. However, silicon also faces significant challenges: during the discharge process, silicon and lithium ions undergo alloying reactions, with the size reaching three times the initial size, while during the process of charging [5], lithium ions are released from the alloy, and the volume of the silicon anode material shrinks again. Silicon, as a semiconductor material, has poor electrical conductivity, which is not beneficial to the transport of electrons and the diffusion of lithium ions, leading to low rate performance and cycling stability [6].

To respond to these limitations, researchers have looked for ways to combine silicon with other materials. From the perspective of the periodic table, silicon and carbon belong to the same group of elements. Compared with carbon, carbon is more prone to losing electrons, while silicon has a better ability to attract electrons. If the material is replaced with silicon, it will increase the capacity of the battery. On the other hand, silicon has poor electrical conductivity, which is unfavorable for lithium-ion diffusion and electron transport. Therefore, combining silicon with carbon composite materials that have better electrical conductivity can greatly enhance the electrical conductivity of silicon, thereby enhance the rate performance and cycle stability of silicon-based materials.

The classification, advantages, and disadvantages of silicon-carbon composites used as anode materials for lithium batteries were discussed here, mainly focusing on the preparation methods, advantages, and disadvantages, and application scopes of silicon-graphite, graphene, and carbon nanotube composites. By comparing their structures, advantages, and disadvantages, the aim is to gain a clearer understanding of their application prospects in the future creation of lithium-ion batteries and the challenges they face. The primary goal of this study is to promote the advancement of electric vehicles and renewable energy. By improving the negative electrode materials, the safety, energy density, and cycle life of lithium-ion batteries can be enhanced, making them more adaptable to the increasingly high demands, closer to the future of clean energy, and helping to solve the future energy crisis.

2. Silicon-carbon composite materials for lithium-ion batteries anodes

Graphite is commonly used as an anode material in commercial applications because it offers outstanding electrical conductivity and strong mechanical properties, and small volume change during cycling. It is relatively easy to obtain and has a controllable cost. However, the capacity of carbon-based materials is limited (372mAh/g), making it difficult to meet the high energy density requirements of lithium-ion batteries. While silicon offers much higher capacity (4200mAh/g) but has a significant disadvantage from severe volume expansion [7]. Silicon–carbon composites have been developed to balance these properties.

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At present, the anode materials for silicon-based lithium-ion batteries that have been extensively studied mainly involve the combination of silicon and materials with good electrical conductivity, such as graphite, graphene, and carbon nanotubes, to enhance electrical conductivity [8].

2.1. Silicon and graphite composite materials

One method (mixing method) to create silicon carbon composites is prepared through a mechanochemical milling process, where all Si particles are placed on the graphite surface. This method mixed up spherical graphite (SG), silicon powder, and pitch (used as both binder and carbon source) (the ratio of SG, Si, and pitch is 3:1:1 by weight). The mixture is then treated by a high-energy ball milling process. Afterward, the mixture was then heated at 900°C under N_2 atmosphere for an hour. During the heating procedure, the pitch carbonizes and deposits the silicon particles uniformly on the graphite surface, which forms a thin silicon layer, about 100nm, on the graphite particle.

Another method (embedding method) is made by embedding silicon into graphite with high-energy “rotational impact” mixing that pushes silicon into the gaps between graphite flakes. A small amount of pitch is then added, and the mix is heated at 900°C under N_2 atmosphere for an hour; the pitch turns into carbon during heating and helps lock the structure. The result of the process above is a graphite matrix with silicon particles embedded inside—but their distribution is ununiform.

In addition, there is another method called spray method (spray method) to make silicon-carbon composites. First, a slurry is made by mixing natural graphite, nano-silicon, and an organic additive like citric acid or pitch. This slurry is spray-dried into tiny precursor particles, then heated under a nitrogen atmosphere. During heating, the organic part breaks down and turns into porous carbon. The final material has a simple core–shell design: a sturdy, conductive graphite core wrapped in a porous carbon shell, with silicon nanoparticles spread evenly throughout that shell.

Comparing different preparation methods of silicon graphite composites, a subtle difference leads to obvious advantages and disadvantages. Mixing method silicon particles are evenly distributed around graphite, but some graphite becomes isolated and cannot fully react. For this reason, the initial capacity of lithium-ion batteries using mixing method composites, 613 mAh/g would be lower compared to embedding method and spray method; however, it would still double the capacity of lithium-ion batteries using carbon alone as an anode material. But that does not mean mixing method has no advantages. Because the silicon particles are evenly distributed on the surface of graphite, the graphite framework is less likely to be damaged, resulting in better cycling stability. After 30 cycles, the capacity is still as high as 78% [9]. For embedding method embedding method, it has a higher initial capacity, 657 mAh/g, compared with mixing method, since more silicon can participate in the reaction. However, due to its preparation method, the silicon particles are unevenly distributed within the internal pores of graphite. This uneven distribution causes silicon volume expansion, which damages the graphite structure and leads to capacity dropping. After 30 cycles, the capacity retention is only 73%. Compared with mixing method and embedding method, spray method has the best performance [10]. In spray method, graphite serves as the core, which not only provides conductivity but also improves mechanical strength. Silicon is uniformly embedded within gaps between the carbon layers, while the outer porous carbon shell ensures both good conductivity and effective buffering of volume expansion. As a result, spray method has an initial capacity of 724 mAh/g, and even after 100 cycles still has 83% of its capacity.

In summary, silicon graphite composites offer a strong solution to the limitations of using pure silicon or pure graphite as anode materials for lithium-ion batteries. Mechanical and embedding methods show how different silicon distributions can affect performance, while spray drying

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methods highlight the potential of core–shell structures to overcome inherent challenges. Although some problems still exist, such as low initial efficiency and reduced cycling performance, progress in synthesis methods and structural design is expected to improve their performance. By combining the high theoretical capacity of silicon with the conductivity and stability of graphite, silicon graphite composites have become a key path to high-energy lithium-ion batteries. Their future use in electric vehicles, mobile devices, and mass energy storage is likely to speed up the transition to more sustainable and efficient energy technologies.

2.2. Silicon-graphene composite material

Silicon-graphene Composite Materials are another type of anode material for lithium-ion batteries that have shown notable improvements in increasing capacity and suppressing volume expansion. To manufacture silicon-graphene Composite Materials, the following steps are needed [11].

Porous silicon was prepared by chemically etching aluminum–silicon alloy powder using hydrochloric acid solution. Selective removal of the aluminum phase was caused by chemical etching. This step produced a three-dimensional porous silicon framework with abundant internal pores, which serves as the substrate for the vertical graphene growth next.

Vertical graphene was grown directly on the surface of porous silicon by plasma-enhanced chemical vapor deposition (PECVD). During the process, a mixture of CH_4, H_2, and Ar gases was added into the reaction reactor at 900 °C. The plasma gave the energy needed to break down methane, which helped graphene sheets start to form and grow upward on the porous silicon. The growth time was controlled between 10 and 30 minutes. At about 20 minutes, the graphene sheets grew uniformly, stood upright and connected with each other, forming a good three-dimensional network for transporting charged particles.

To make the battery work better at the start and lose less lithium in the first cycle, the porous silicon vertical graphene materials were treated with a liquid chemical method called prelithiation. A special solution (Li-DMB in THF) was used for this step. The electrode films were soaked in the solution for different times (1–4 hours). The best result came at about 2.5 hours, where the efficiency reached 99.7%. This treatment helped form a stable SEI layer, saved lithium in the first cycle, and made the battery last longer.

Using silicon-graphene composite materials as lithium-ion batteries anode materials has many clear advantages. The porous silicon framework keeps the sphere shape complete, and the spaces inside can absorb the volume changes during charging and discharging. At the same time, vertically grown graphene greatly improves the conductivity, so lithium ions and electrons can move quickly. The graphene grows upward in a three-dimensional way and forms a conductive network. This network provides many contact points, which improves the rate performance. Furthermore, as the graphene is grown in situ through PECVD, it sticks strongly to the porous silicon, which helps maintain the structural stability and makes it easier to form a stable SEI layer. Prelitigation also helps reduce the first-cycle capacity loss, reaching a coulombic efficiency close to 99.7%. However, PECVD needs a very high temperature of about 900°C, which uses a lot of energy and increases the cost, making large-scale production difficult. The process is also complicated and needs careful control of temperature, time, and gas conditions. It also requires advanced equipment, which is another reason why mass production is hard. Finally, if the growth conditions are not well controlled, too much graphene may block the pores in the silicon, thereby reducing the transport of lithium ions.

Jamaluddin et al. [12] reported a kind of silicon-graphene spherical composite material. This composite material was spray-dried without a post-annealing process. The anode's capacity and

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cycling stability were enhanced by its unique core–shell structure, which also provided excellent electrical conductivity and mechanical flexibility. The results show that the initial discharge capacity reaches 2882.3 mAh/g and the initial coulombic efficiency reaches 86.9% under the condition of 0.2A/g. Silicon-graphene composite materials could be used for high-performance lithium-ion batteries, especially for use in electric vehicles. In addition, their strong structure and long cycle life make them have a good chance for large-scale energy storage systems, such as renewable energy grids. Silicon-graphene composite materials could not only be used for normal lithium-ion batteries, but also provide ideas for the next energy storage devices that require stable and high-capacity anodes.

2.3. Silicon-carbon nanotubes composite materials

Advances in nanoscience and nanotechnology provide promising opportunities to design new nanostructured electrode materials with improved performance for next-generation lithium-ion batteries [13]. Nanostructured materials are key to these advancements because they have feasible surface areas, small mass and charge diffusion spans, as well as volume change adaptation during the processes of charging/discharging. Carbon nanotubes feature a unique one-dimensional Tube-like structure, excellent conductivities of electrical and thermal, mechanical flexibility, and a larger surface area, and are regarded as ideal additives for enriching the chemical properties of electrodes [14]. At present, the global commercial production of carbon nanotubes (CNTs) exceeds several thousand tons each year [15]. Bulk carbon nanotube powders have found applications in numerous commercial products, including rechargeable batteries, automotive components, sports equipment, ship hulls, and water filtration systems. Progress in the synthesis, purification, and chemical functionalization of carbon nanotubes has further facilitated their use in thin-film electronics and large-area coatings. In making the carbon nanotube–reinforced silicon-carbon composite materials of lithium-ion batteries anode, the materials must be chosen carefully. Silicon powder is used as the main active material because it can store a very large amount of lithium ions, but it is prone to cracking due to expansion. To reduce this volume expansion, carbon nanotubes are added. They act like tiny wires, which can enhance electrical conductivity and thereby reduce the volume expansion of silicon. In addition, glucose, a carbon source, is used to form a thin layer on the surface, and this process can make the material become more stable. To fix this together and make sure they are in good contact, materials such as binders and conductive agents need to be used. By combining these materials, researchers would obtain a powerful and efficient anode material for lithium-ion batteries.

Lithium-ion batteries exhibit some significant advantages when carbon nanotube-reinforced silicon-carbon composites are used as anode materials. The main advantage is that the carbon nanotubes form a powerful network, which makes it easier to transfer charged particles, and they also help reduce volume expansion of batteries. The thin carbon layer from the carbon source protects the material and keeps the structure more stable, which improves the lifespan and maximum capacity of batteries. However, this material also have some disadvantages. For instance, carbon nanotubes are expensive, which increases the cost of manufacturing anode materials for lithium-ion batteries. In addition, the process of mixing and coating is not simple, and It can be challenging to manage during large-scale production.

The carbon nanotube–reinforced silicon-carbon composite shows great potential when it's used as an anode material in lithium-ion batteries. Because of the high performance the material has, relative to other anode materials. Experiments show that the initial capacity of carbon nanotube–reinforced silicon-carbon composites exceed 2000 mAhg-1, which is significantly higher than that of traditional lithium-ion batteries with graphite anodes. Even after 100 charge–discharge cycles, it

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could still keep about 1400 mAhg-1, showing good stability. It also works well at high current rates, meaning it can charge or discharge quickly without losing too much capacity. Because of these performances, carbon nanotube–reinforced silicon-carbon composite materials are suitable for applications that need both high energy and long cycle life, such as portable electronics, electric vehicles ,and large energy storage systems [16].

3. Prospects of silicon carbon composite anode materials

In the future, silicon–carbon composites are expected to improve as anode materials in lithium-ion batteries. Technically speaking, scientists are researching better designs for the anode materials of lithium-ion batteries, such as using nano-silicon, carbon coatings, and networks made of carbon nanotubes or graphene, all of which can make the anode materials stronger and more stable. To cut costs, waste silicon from the solar energy industry and cheap natural carbon sources, such as starch or glucose, can be used to make production more eco-friendly and less expensive. Because silicon–carbon composites have a higher capacity and longer lifespan than graphite, they may be applied more widely in electric cars, portable electronic devices, and energy storage systems. The research will also focus on making production simpler, larger in scale, and more environmentally friendly. In the long term, silicon-carbon composites may become one of the key materials for the next generation of safe and high-energy density batteries.

4. Conclusion

This study revealed that silicon–carbon composite materials serve as highly effective anodes for lithium-ion batteries. The combination of silicon's high storage capacity with carbon's excellent conductivity and stability enhances the batteries' energy density, cycle life, and overall performance. Each type of silicon–carbon material has its own strengths: silicon–graphite composites with spray-dried core–shell structures are stable and suitable for industry; silicon–graphene composites create three-dimensional conductive networks that boost conductivity and limit expansion, though they are expensive and hard to produce; silicon–carbon nanotube composites provide excellent electron transport and long cycle life, but large-scale production is costly. Overall, silicon–carbon composites has way better performances than graphite and show great potential for use in electric cars, portable electronics, and renewable energy storage.

The main contribution of this work is a clear summary of how different silicon–carbon composites are made, their advantages, and their challenges. This gives other researchers useful comparisons and helps them understand the trade-offs between capacity, stability, and cost. It also fills a gap in earlier studies by showing which methods may work better for industry. In the future, this knowledge could support the development of cheaper, scalable, and eco-friendly batteries, which are important for renewable energy, electric transportation, and large-scale energy storage systems.

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