

Carbon-Based Silicon Composite Anodes for High-Performance Lithium-Ion Batteries: Structural Design Strategies and Electrochemical Improvements

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Abstract. Because of the theoretical high specific capacity, silicon has attracted much attention as a potential anode material for lithium-ion batteries. However, the large volume expansion generated in the process of repeated charging and discharging will lead to particle cracking, unstable solid electrolyte interface and rapid capacity loss, making silicon difficult to be used in practical batteries. The combination of silicon with carbonaceous materials can overcome these problems and improve the performance of silicon anodes. This paper reviews three main kinds of silicon/carbon composite anodes. This paper reviews three main types of silicon/carbon composite anodes, including silicon/graphite, silicon/carbon nanotubes and silicon/graphene composites. Their structure, preparation methods, advantages, limitations and possible applications are compared. The review shows that graphite provides good structural stability and practical performance and carbon nanotubes and graphene can form conductive networks and help deal with silicon expansion. However, these materials also face limitations such as reduced overall capacity, higher cost and more complex preparation methods. Therefore, the performance of silicon/carbon composite anode depends on the balance between capacity, conductivity, structural stability, cycling performance and practical feasibility. In general, silicon/carbon composites provide a promising way to improve silicon-based negatives. Further optimizing the composite structure, preparation method and long-term cycle stability is expected to promote the development of high-energy density and long-life lithium-ion batteries.

Keywords: lithium-ion batteries, anode materials, silicon-carbon composites, electrochemical performance, cycling stability

1. Introduction

Electric vehicles and portable electronic devices are becoming more common, creating a greater need for better energy storage technology. Renewable sources such as solar and wind also depend on reliable storage because their energy supply is not always stable. Lithium-ion batteries (LIBs) are now widely used for energy storage because of their mature technology and high theoretical capacity. However, with the continuous improvement of battery performance requirements, new challenges are also faced in improving electrode materials and achieving higher energy density [1].

Because of its outstanding cycling stability and suitable operating potential, graphite has become a common choice for commercial lithium-ion battery anodes. However, its theoretical capacity is only 372 mAh g⁻¹ limits further improvements in the energy density of LIBs [2]. Therefore, researchers have explored some alternative anode materials with higher storage capacity. Among various candidates, silicon has gained significant attraction due to its extreme high theoretical specific capacity of approximately 4200 mAh g⁻¹, making it a promising replacement for conventional graphite anodes [3].

However, despite its outstanding capacity, the practical application of silicon anodes remains limited by several real-life challenges. During charging and discharging, the volume of silicon changes significantly. This leads the silicon experience huge volume expansion, which can reach nearly 300% of the original size. Repeated mechanical stress can damage the silicon particles and make the SEI unstable. It can also weaken electrical contact between particles and lead to a gradual loss of capacity. Furthermore, silicon's electrochemical performance is further limited by its low electrical conductivity.

Silicon-carbon composite anodes are widely studied because silicon and carbon can make up for each other's weaknesses. Silicon provides a large lithium storage ability, while carbon helps improve conductivity and keeps the electrode structure more stable. Carbon components can improve electron transport, buffer silicon expansion, and stabilize the electrode structure during repeated charge-discharge cycles. Recent studies have further introduced advanced architectures, such as yolk-shell structured Si/C composites, where internal void spaces provide additional room for silicon expansion while carbon shells maintain structural integrity. These designs have demonstrated improved cycling stability and electrochemical performance compared with conventional silicon electrodes [4].

Although the silicon carbon nanostructures have huge advantages, key issues such as cycle life, conductivity and high lithium storage capacity still face major challenges. Therefore, this article focuses on the application of nanomaterials in lithium-ion battery silicon-carbon composite anode, analyzes its structural design and performance improvement, and explores its potential role in the next-generation energy storage system. An in-depth understanding of these nanostructured silicon carbon materials is conducive to the development of new lithium-ion batteries with higher energy density and better cyclic stability. This research is of great significance for extending the range of electric vehicles and supporting the practical application of large-scale renewable energy storage systems

2. Silicon-carbon composites

2.1. Silicon/graphite composite anodes

Graphite can be combined with silicon to make a more practical anode material. Silicon provides high capacity, while graphite helps improve conductivity and keeps the electrode structure more stable. Although silicon has attracted much attention as an anode material because of its much higher capacity than that of commercial graphite, its huge volume expansion in the process of lithiation and delithiation will lead to particle cracking, solid electrolyte interphase (SEI) instability and gradual loss of electrical contact. Therefore, the introduction of graphite into silicon-based electrodes can not only provide mechanical support, but also increase the conductive pathways, thus helping to improve the cycling stability of silicon anodes. In silicon/graphite composites, graphite mainly acts as a stable carbon skeleton to buffer the mechanical stress generated by silicon expansion. The graphite structure can not only improve electron transport inside the electrode, but

also reduce the direct mechanical damage caused by repeated volume changes. In this structure, silicon acts as an active lithium storage component, while graphite acts as a reinforcing material to improve the strength and conductivity of the structure. The surrounding carbon matrix also works as a bonding phase and helps keep the whole composite together after repeated charge and discharge. In this concrete-like design, silicon nanosheets can be compared to "sand", graphite particles can be compared to "stones", and the pitch-derived carbon matrix can be compared to "cement". This arrangement allows the components to support each other, thus building a more stable structure than simply using silicon particles. Fine graphite also provides additional conductive pathways between different parts of the composite material, helping electrons to transfer more efficiently through the electrode. This description closely follows the structural design reported in the paper, where graphite and waste silicon nanosheets all have distinct reinforcing functions.

The preparation method is another key factor affecting the properties of silicon/graphite composites. Spray drying, chemical vapour deposition, and ball milling are common techniques for creating silicon-carbon composite materials. Among these, spray drying has garnered a lot of attention because of its relatively simple process, high efficiency and large-scale production potential. Li et al. [5] fine graphite and pitch were first ball-milled and mixed in water. Waste silicon powder was mixed in, and the mixture was ball-milled again until a uniform suspension was formed. The suspension was subsequently spray-dried to produce Si/pitch/graphite particles. Finally, these particles were heated to 900 °C under an argon atmosphere to convert the pitch into a carbon matrix, and an outer carbon layer was formed through an additional pitch coating and heat-treatment process.

These preparation processes have multiple advantages in practical battery production. Ball milling is a simple method of mixing silicon, graphite and carbon precursors, while spray drying can convert the suspension into relatively uniform micron-sized particles. The formation of spherical particles is also useful because they can achieve better particle contact and packing during electrode preparation. In addition, pitch is suitable as a carbon precursor because it can effectively connect silicon particles with graphite particles after carbonization. The final composite material contains both an internal carbon matrix and an external carbon coating. This dual structure provides additional protection during repeated lithiation and delithiation, effectively reducing mechanical degradation. The hierarchical structural design of silicon/graphite composites can further improve their electrochemical properties. The carbon matrix and graphite particles form a solid skeleton, which effectively binds silicon nanosheets and slows down structural degradation. Another important advantage of silicon/graphite composites is their practical application potential in existing lithium-ion battery systems. Silicon-containing composite materials can be mixed with artificial graphite instead of replacing commercial graphite with silicon completely. This method allows the electrode to obtain part of the high-capacity advantage of silicon while maintaining the stability of graphite. Li et al. [6] combined the silicon/graphite-carbon composite with artificial graphite. The initial Coulombic efficiency reached 94.1%, close to that of commercial graphite. The material was also tested in a full lithium-ion cell with LiFePO_4 as the cathode. The full cell retained 85.1% of its capacity after 100 cycles, indicating that the composite material has potential not only in laboratory half-cell testing, but also in more practical battery configurations.

However, silicon/graphite composites still face many challenges. Since the theoretical capacity of graphite itself is much lower than that of silicon, excessive graphite content may reduce the overall energy density of the electrode. In addition, when the silicon loading is increased to obtain higher capacity, the buffering capacity of the traditional graphite skeleton may not be enough to fully adapt to the volume expansion of silicon. Therefore, researchers have developed advanced structures such

as porous structures and additional carbon coatings to provide extra space for silicon expansion and maintain the stability of the interface between silicon and carbon materials.

Therefore, there is an important trade-off between capacity and stability. Although increasing the silicon content can improve electrode capacity, it can also lead to more severe volume expansion and faster performance degradation. In contrast, increasing the graphite content can enhance stability, but it weakens the capacity advantage provided by silicon.

The manufacturing process also needs to be further optimized. Although methods such as ball milling and spray drying have good large-scale production potential, the preparation of this concrete-like structure still involves multiple processes such as mixing, spray drying, high-temperature carbonization and additional carbon coating. In addition, the distribution of silicon particles, graphite and carbon must be carefully controlled. If silicon is unevenly distributed, local areas may experience greater expansion stress, which can lead to structural failure of the electrode during long-term cycling. Therefore, future research should focus not only on improving electrochemical performance, but also on simplifying the production process and reducing manufacturing costs [7].

Silicon/graphite composite anodes can help bring silicon-based batteries closer to practical use. Compared with pure silicon anodes, this kind of anode has advantages in conductivity, structural stability and manufacturability, making it more suitable for large-scale battery systems. The possibility of mixing silicon-carbon composite materials with existing commercial graphite is particularly important because it provides a gradual transition path from traditional graphite anodes to higher-capacity silicon-based materials. However, in order to achieve higher energy density while maintaining long-term cycling stability, it is still necessary to further optimize the structural design and silicon loading.

2.2. Silicon/carbon nanotube composite negative pole

Carbon nanotubes (CNTs) are another important carbon material used to improve the performance of silicon-based anodes. Compared with graphite, carbon nanotubes have a one-dimensional tubular structure, which has high conductivity, high mechanical strength and good flexibility. These characteristics make it particularly suitable for building a conductive network inside silicon carbon composite electrodes. In the process of repeated lithium embedding and lithium removal, silicon particles will undergo huge volume changes, which may cause damage to the electrode structure and block electron transmission. Carbon nanotubes can not only maintain electrical contact between active particles, but also provide mechanical support to reduce structural degradation.

One of the advantages of carbon nanotubes is that they can form a continuous conductive path throughout the electrode. Kim et al. prepared a silicon-carbon composite material containing single-walled carbon nanotubes (SWCNTs). The carbon matrix contains both silicon nanoparticles and SWCNTs spread throughout the material. Single-walled carbon nanotubes enhance the structure of composite materials and establish conductive connections between different regions of the electrodes, so that electron transmission can be maintained even under a high surface load of active substances. The surface capacity of the obtained Si@C@CNT composite material reaches 3.20 mAh cm⁻², and still maintains 85% of the capacity after 100 cycles. The material also shows stable cycling performance in soft-pack whole batteries, indicating that silicon-carbon composites containing carbon nanotubes have potential in actual battery applications [8].

By building a three-dimensional network, the conductive effect of carbon nanotubes can be further enhanced. Wang et al. [9] prepared porous Si/MWCNT@C microspheres containing a 3D network of multi-walled carbon nanotubes. This structure provides paths for electron transport and

helps lithium ions move through the electrode. After 300 cycles at 1 A g^{-1} , a discharge capacity of 536.6 mAh g^{-1} remained, equal to 80.5% capacity retention. The study also found that using a 3D network of single-walled carbon nanotubes in self-supporting electrodes can improve cycling stability. This suggests that the arrangement of carbon nanotubes can affect the performance of silicon-carbon composites.

In addition to improving electrical conductivity, carbon nanotubes also help alleviate the mechanical effects caused by silicon volume expansion. Carbon nanotubes (CNTs) have a high-to-length ratio and good flexibility. Even when the electrode undergoes repeated size changes, they can connect silicon particles as an enhanced network. A recent review of silicon carbon composites shows that carbon nanotubes can provide conductivity and mechanical support at the same time, making them an ideal buffer material for silicon anode. However, the effectiveness of carbon nanotubes largely depends on their dispersion inside the composite material. Due to the interaction between carbon nanotubes, they are prone to aggregation, which leads to uneven conductive networks and reduces their efficiency. Therefore, it is usually necessary to adopt surface modification, add dispersants or elaborately designed preparation processes to achieve the uniform distribution of carbon nanotubes.

Overall, silicon/carbon nanotube composite anodes can improve conductivity and mechanical stability at the same time. Their flexible conductive network also helps maintain electrical contact when silicon expands. However, carbon nanotube dispersion, cost and large-scale production are still important challenges that limit wider commercial use.

2.3. Silicon-graphene composite anodes

Graphene is another promising carbon material for improving silicon anodes in lithium-ion batteries. It is the two-dimensional layered structure of carbon atoms arranged in a hexagonal lattice, quite different to graphite. Graphene can form conductive paths around silicon particles and also give them structural support. Its large surface area and strong mechanical properties help improve the electrode structure. Silicon also expands and contracts repeatedly when lithium enters and leaves the material. As a result, the particles may crack, the SEI layer may become unstable, and electrical contact may weaken. Graphene can help electrons move through silicon-based electrodes and keep the structure stable.

Graphene has the significant advantage of being able to build a flexible, conductive scaffold around silicon particles. In comparison with conventional graphite particles, the graphene sheets form a continuous 2D conductive network, which promotes more efficient electron transport thru the electrode. Also, the flexibility of graphene can take up some of the mechanical stress from silicon expansion. A more even graphene coating on silicon particles can reduce direct contact with the electrolyte. It also helps improve SEI stability and cycling performance [10].

The structural design of graphene-based silicon composites is significant to their electrochemical performance. Porous graphene structures have attracted tremendous interest due to their additional space for silicon expansion. Lee et al. synthesized a composite of nanoporous silicon and reduced graphene oxide (rGO) thru a foaming agent-assisted approach. In this structure, the porous space between silicon and graphene acts as a buffer for silicon volume changes and the rGO framework enhances electrical conductivity and retains the mechanical stability of the electrode. The reversible capacity of the optimized composite remained at $1676.9 \text{ mAh g}^{-1}$ after 200 cycles. The results indicated that combining a porous silicon structure and a graphene network is an effective method to improve the performance of Si-based anodes [11].

Another effective strategy is to improve the interface of silicon and graphene, in addition to providing physical space for silicon expansion. A 3D silicon suboxide particle-embedded nitrogen-doped graphene framework was synthesized by Hong et al. In this structure, the silicon-containing material and the graphene framework formed strong Si-N-C bonds. These chemical bonds improved the bonding between silicon and carbon, which reduced the detachment of particles during cycling. The 3D graphene network meanwhile provided extra conductive pathways and mechanical support. The optimized composite displayed excellent long-term cycling stability with 91.3% capacity retention after 1,000 cycles at 2.0 A g^{-1} [12].

The particle size and distribution of graphene materials is another important factor that influences graphene-based composites. The oxygen functional groups of graphene oxide (GO) enable the interactions with silicon-based materials and the formation of composite structures. He et al. studied the influence of GO flake size on silicon-oxycarbide-derived anodes. Smaller GO flakes were found to be more porous and allowed for better lithium-ion diffusion, while larger GO flakes created a more continuous conductive network. Medium-sized GO demonstrated an improved balance of structural stability, electrical conductivity and ion transportation. As a result, medium-sized GO showed better overall results [13].

How silicon/graphene composites are prepared can also affect their final properties. Common methods include hydrothermal synthesis, spray drying, chemical reduction, and in-situ carbonization. Solution based approaches can be used to incorporate GO with silicon particles and then reduce to form a conductive graphene network. Spray drying is also widely applied because of its ability to produce uniform composite particles and the possibility of large scale manufacturing. These preparation techniques enable researchers to control the distribution of graphene, the structural porosity and the strength of the silicon-carbon interface.

Silicon/graphene composite anodes can reduce several problems found in pure silicon anodes. Graphene provides good conductivity, mechanical strength, and a flexible structure. These properties help electron transport, reduce silicon expansion, and improve the stability of the electrode interface. Advanced silicon/graphene structures can remain stable after many charge-discharge cycles. But the commercial use of graphene-based lithium-ion batteries is held back by issues such as graphene dispersion, production costs and large-scale manufacturing.

3. Future perspectives

However, graphite, carbon nanotubes, and graphene have different advantages when combined with silicon. Therefore, their future development may focus on different areas. Currently, the silicon/graphite composites are more applicable to practical applications, because graphite has already been largely used in commercial lithium-ion batteries and the production cost is comparatively low. In future work on silicon/graphite anodes, the main emphasis should be placed on increasing the silicon content without causing an excessive volume expansion. Also, the preparation method needs to be simplified and the uniform distribution of silicon in the graphite-based structure needs to be improved for the large-scale production.

The main advantage of this for silicon/CNT composites is that it can form a flexible conductive network inside the electrode. Future work can be dedicated to improving dispersion of carbon nanotubes and interaction between carbon nanotubes and silicon particles. Achieving better distribution of carbon nanotubes with reduced material consumption can lead to similar conductivity and mechanical support capabilities at a lower production cost. Therefore, the design of 3D carbon nanotube networks could be an important research direction for high-loading silicon electrodes.

Graphene-based composites offer more possibilities in designing advanced structures. Porous graphene frameworks as three-dimensional conductive structures and the stronger silicon-carbon interface can provide more room for silicon expansion while maintaining electron transport. However, the aggregation of graphene and relatively high production costs are still limiting its commercial application. Thus future development should be geared toward a simple process of preparation and better control of the distribution of graphene.

In general, no carbon material can solve all the problems of silicon anodes completely. Graphite is more compatible commercially. Efficient conductive paths are available from carbon nanotubes (CNT), and graphene offers more flexible structural design. Thus, the silicon-carbon anode of the future may be able to use the advantages of different carbon materials and not be limited to one type alone. This hybrid structure can balance capacity, cycle stability, conductivity and production cost better.

4. Conclusion

This paper systematically reviews different types of silicon/carbon composite anodes in lithium-ion batteries. The results show that the current silicon-based anode research and development is mainly based on the combination of silicon and graphite, carbon nanotubes and graphene. As one of the most practical solutions, silicon/graphite composite materials can not only provide good structural stability and electrical conductivity, but also reduce the negative impact of large volume expansion of silicon materials. However, its overall capacity is still limited by the relatively low capacity of graphite. Silicon/carbon nanotube composites can form highly conductive networks and better maintain electrical contact during repeated charging and discharge, but their relatively high cost and complex preparation methods may limit their broader applications. Silicon/graphene composites also have good conductivity and mechanical strength. Their flexible structure can help deal with the volume change of silicon, although the preparation process is still complex and the material cost is high. Therefore, different silicon/carbon composites have different advantages. Future silicon anodes need to consider capacity, conductivity, structural stability, cycling performance, and practical use together.

Therefore, the future development of silicon/carbon composite negatives will depend on further improving these different composite structures while reducing their existing limitations. More research is needed to improve cyclic stability, maintain stable electrical contact, and reduce mechanical damage caused by repeated silicon expansion and contraction. At the same time, simpler preparation methods and lower production costs are crucial to the practical application of carbon nanotubes and graphene-based composites. Further research on the interaction between silicon and different carbon materials also helps researchers design more stable and efficient composite structures. Through continuous improvement in material design, preparation method and electrochemical properties, silicon/carbon composite anode is expected to play an important role in the development of the next generation of lithium-ion batteries with higher energy density and better long-term performance.

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