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Research and Development of Silicon-Based Anode Materials for Lithium Ion Batteries

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Abstract. Silicon, known for its high capacity, has become a key material in developing anodes for lithium-ion batteries (LIBs). However, the substantial increase in volume that silicon-based anodes undergo during the process of charging and discharging presents a significant obstacle. This expansion can lead to structural disintegration and loss of electrical conductivity. It also initiates undesirable parasitic reactions, all of which significantly reduce the cycle life of the battery. Consequently, these issues hinder the wider adoption of silicon in the commercial LIB sector. This comprehensive review synthesizes key advancements in silicon-based anode research, highlighting the latest trends and innovations. This investigation examines the intricate engineering of silicon lattices and the development of silicon-based composite materials, illuminating the advancement in the field of silicon anode technologies, encompassing hybrid materials like silicon integrated with graphite and those amalgamated with various metals. Additionally, it scrutinizes the lithium storage characteristics and the mechanisms at play in silicon/graphene and other silicon composites. The review concludes with a forward-looking exploration of the future trajectory of silicon-based materials, considering their potential for growth and integration within the field.

Keywords: Silicon-based anodes, Lithium-ion batteries, Structural stability.

1. Introduction

In the field of lithium batteries, The theoretical maximum energy storage capacity of graphite-based anode materials is 372 mA/g, and the actual specific capacity of commercial high-end graphite materials has reached 360-365 mA/g, which indicates that Its performance approximates closely to the theoretical maximum. Therefore, the capacity for enhancing the energy density of lithium-ion batteries through advancements in graphite anode materials is considerably constrained.

As a potential negative electrode material, silicon (Si) is considered to boast an exceptionally high theoretical specific capacity, reaching 3579 mA/g at ambient temperatures. In contrast to graphite, silicon is plentiful in the Earth's crust, extensively available, and characterized by its low cost and accessibility, which makes it expected to become an ideal substitute for graphite negative electrode.However, silicon also presents some challenges when used serving as the anode in lithium-ion batteries. In the process of lithium removal, the silicon anode is subject to substantial volumetric changes during the charge and discharge processes, which will produce huge mechanical stress. After repeated use, silicon particles may fracture and powder, which will seriously affect the transmission of lithium ions inside the negative electrode.To solve this problem, the researchers developed silicon-carbon composites. This composite material can not only provide a higher specific capacity for the negative electrode, but also significantly improve the conductivity of the electrode. Additionally, the silicon-carbon composite material acts as a buffer layer to reduce the drastic volume changes of silicon during charge and discharge cycles. This helps protect the structure of the silicon material and maintain the integrity of the electrode.

Based on this, the research progress of silicon anodes is primarily discussed in two main parts. First, the development of various robust silicon nanostructures is introduced, including silicon nanoparticles (NPs), hollow nanoparticles, a specific Vinami wire material, two-dimensional thin-film silicon. Second, a variety of silicon-based anode materials for lithium-ion batteries have been investigated, with a review of the current status and existing issues. Additionally, the discourse encompasses an initial perspective on the prospective trajectory for the evolution of polar materials tailored for lithium-ion batteries that utilize silicon anodes.

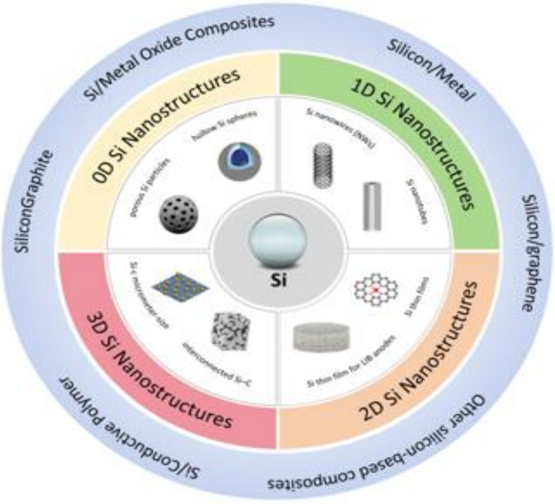

Figure 1. Different types of the Si nanostructures and Silicon-Based Anodes in Lithium-Ion Batteries.

2. The Exploration of Si Nanostructures

2.1. 0D Si Nanostructures

Materials made of nanoscale silicon exhibit improved ability to handle volume expansion because of their higher specific surface area and increased average binding energy per atom on the surface. This helps prevent material breakage and enhances cycling performance. A common approach involves using porous and hollow silicon structures [1]. Hollow silicon spheres and porous silicon particles with zero-dimensional properties have better capacity to accommodate volume changes compablue to solid silicon particles. The reason for this superior capability lies in their larger specific surface area and stronger resistance to tensile stress. One technique to produce hollow structures is through the Kirkendall effect, a traditional method that relies on the different diffusion rates of two materials to form holes at the material interface.

Bao Zhihao's research group at Tongji University has introduced a technique for producing multi-porous hollow silicon spheres via the aluminothermic reduction of SiO2-encapsulated carbon spheres within a molten salt medium(figure 2(a)) [2]. This process is characterized by the swifter outward transport of silicon and oxygen elements relative to the inward movement of aluminum, which results in the creation of a void at the interface. The silicon, once reduced, initiates nucleation and expands on the carbon shell's surfaces, culminating in the synthesis of multi-porous hollow silicon spheres. These spherical structures exhibit remarkable capabilities, including outstanding energy storage, sustained performance through numerous cycles, and enhanced charge-discharge rates. More specifically, at a current density of 1 A/g, these spheres have maintained a consistent specific capacity of approximately 1770 mAh/g after 200 cycles. Furthermore, at a higher current density of 8 A/g, they have exhibited a steady capacity of about 800 mAh/g post 600 cycles of operation.

Compared with the single hollow layer sphere, the exterior of the bilayered hollow structure has more abundant pore structure, which can induce electrolyte diffusion to the hollow cavity of the hollow sphere while cooperally mitigating the volumetric strain of silicon, and improve the contact between electrolyte and elemental silicon.The research team led by Wang Hui from Northwest University has employed a magnesiothermic reduction and hard templating approach to fabricate double-shelled hollow carbon spheres encapsulating silicon nanoparticles (as depicted in Figure 2(b)) [3]. These double-shelled hollow carbon spheres, which exhibit a high mesoporosity rate, serve as a containment vessel for the silicon nanoparticles, effectively alleviating the expansion of silicon's volume and bolstering the material's structural integrity. The material exhibited a specific capacity of 1350 mAh/g after enduring 400 cycles at a current density of 0.3 A/g. Additionally, when subjected to a more intense current density of 1 A/g, it sustained a capacity of 750 mAh/g through an extended period of 2000 cycles.

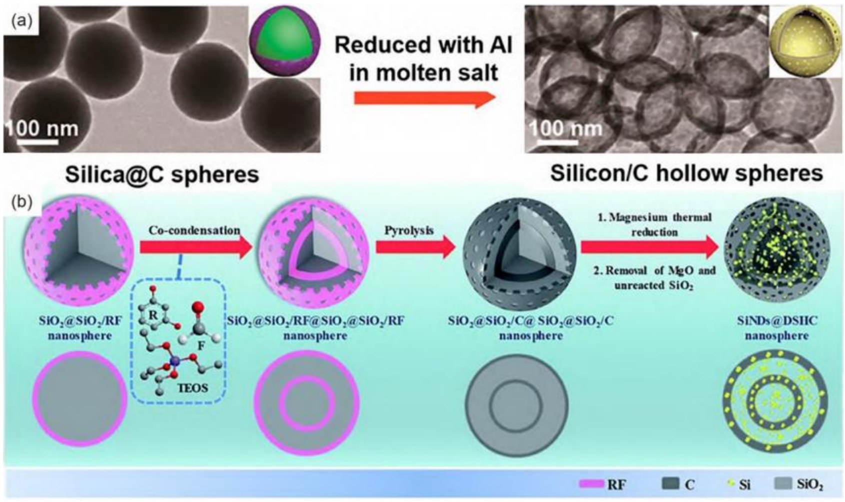

Figure 2. A visual representation of common silicon-derived hollow sphere/cage anode materials. (a) the creation procedure of cavitated, porous silicon spheres utilizing a molten salt reduction technique [2]. (b) The accompanying diagram depicts the creation of SiNDs@DSHC nanospheres, following the methodology [3].

2.2. 1D Si Nanostructures

Silicon with one-dimensional nanotube structure has a high axial to diameter ratio, which can shorten the carrier transport distance and reduce the longitudinal swelling during the charging and discharging phases, thereby avoiding material disintegration. In addition,1Dsilicon nanostructures are recognized for their enhanced electrical conductivity, which can be ascribed to the enhanced electrical conductivity between the active material and the conductive network or substrate. This leads to a decrease in electrical resistance at the boundary between the electrode and the electrolyte. Among the various 1D silicon materials that have been extensively investigated are silicon nanowires (NWs), silicon nanofibers, and silicon nanotubes [1]. Silicon nanowires, with their high surface-to-volume ratio and excellent electrical properties, are particularly useful in sensor technology and as components in high-speed electronics and optoelectronic devices. Silicon nanofibers, known for their flexibility and large surface area, are ideal for wearable electronics and energy storage devices, and can also be used to reinforce composite materials. Silicon nanotubes, which offer a combination of strength and light weight, are being explored for applications in nanoscale electronics and as structural components in various industries. These materials are all distinguished by their high surface area, which enhances their chemical sensitivity and interaction capabilities, making them valuable in a wide range of technological advancements.

2D Si Nanostructures Park et al. have detailed the synthesis of carbon-coated silicon nanotubes (C−Si NTs) through a template-assisted chemical vapor deposition process utilizing an alumina membrane [4]. These C−Si NTs exhibit an outer diameter of approximately 200−250 nm,a shell thickness of 40 nanometers, and are coated with a layer of non-crystalline carbon measuring around 1.50 nm in thickness. The electrode paste was mixed according to a precise weight ratio of 75:12:13, comprising C−Si NTs, superior P carbon black, and polyvinylidene fluoride binder. The C−Si NT electrode exhibited an initial discharge capacity of approximately 3650 mAh/g, with a corresponding charge capacity of around 3250 mAh/g, whichtranslates to a remarkable initial Coulombic efficiency, reaching approximately 90%. Such elevated initial Coulombic efficiency indicates minimal irreversible capacity loss during the primary period, which is crucial for long-term battery performance, which acts as a barrier to prevent direct interaction between the silicon active material and the electrolyte. This increased initial efficiency is due to the thin carbon coating, which aids in the establishment of a robust solid electrolyte interphase (SEI) layer on the surfaces of the silicon nanotubes (Si NTs). Additionally, the C−Si NT electrode has shown a commendable stability in cycling performance, retaining 89% of its capacity after 200 cycles at a 1 C rate withina complete cell incorporating a LiCoO2 cathode. It indicates that the electrode can undergo many charge-discharge cycles without significant degradation in its ability to store energy. This is particularly important when paired with a LiCoO2-based cathode, which is a common and high-performance cathode material known for its high energy density and voltage. The combination of a stable anode and a high-performing cathode can lead to a battery that offers both high energy capacity and long cycle life, which are key factors in ensuring the reliability and economic viability of energy storage systems. High cycle stability reduces the need for frequent replacements and extends the overall service life of the battery, making it a more sustainable and cost-effective solution for energy storage. It also displayed a robust rate capability, achieving a capacity of approximately 2900 mAh/g at a discharge rate of 5 C in a coin-shaped single-cell configuration.

2.3. 2D Si Nanostructures

Silicon thin films have garnered significant interest for using as anodes in lithium-ion batteries, a preference stemming from their markedly enhanced electrochemical performance and the simplicity of their electrode design [5]. The exploration of silicon thin films for LIB anodes can be traced back to the early 2000s [6].

One-dimensional silicon (1D Si) nanostructures, while demonstrating superior electrochemical characteristics, are not without their challenges. Notably, they encounter issues such as diminished Coulombic efficiencies, which is likely due to the the continuous development of the solid electrolyte interphase (SEI) layer. Additionally, these nanostructures suffer from a low tap density, which can impact their overall performance in energy storage applications. Furthermore, the repetitive volumetric expansion and contraction during each charge-discharge period can cause the SEI film to fracture. This damage triggers the development of a new SEI layer through a reaction between the electrolyte and the freshly exposed silicon surface (as illustrated in Figure 3c). Therefore, electrode structures that facilitate the creation of stable SEI layers are key to achieving durable cycle performance in silicon-based anodes [7].

Hertzberg and colleagues have proposed a solution to the issue of volume change in silicon anodes by introducing a porous silicon nanotube (Si NT) encased in a rigid carbon (C) outer shell [8]. This structure was fabricated using a method that involves an alumina membrane as a template (Figure 3a). This method allows for precise control over the nanotube dimensions, resulting in uniform and reproducible structures. The electrochemical interaction between silicon and lithium is contained to the robust carbon shell, enabling a highly reversible morphological transformation. The design has produced a silicon nanotube (Si NT) with a durable carbon (C) coating. This results in stable cycling over 250 cycles and a high Coulombic efficiency of above 99% in the first 50 cycles, which remains above 99.9% from cycles 50 to 250 (Figure 3b).

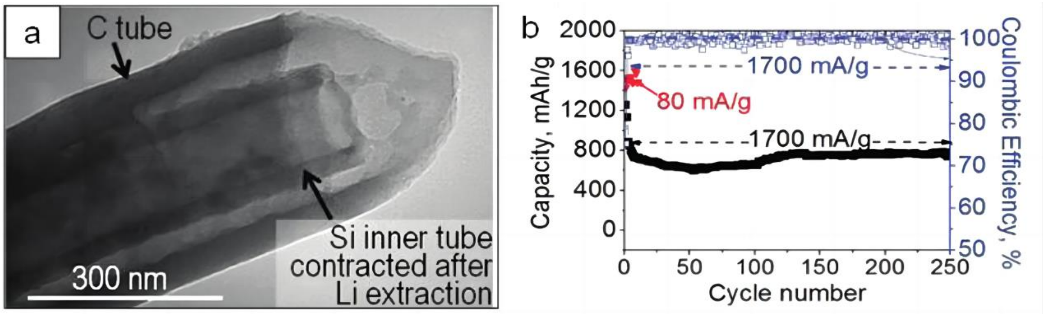

Figure 3. (a) The TEM image depicts the silicon-carbon tubular composite following lithium extraction at cycle 10. (b) This illustrates electrochemical properties of silicon-carbon composites, which contains 33 weight percent of silicon, with the capacity normalized to the combined weight of carbon and silicon [7].

Wu et al. have introduced a novel electrode design for lithium-ion batteries, the double-walled silicon-silicon oxide nanotube (Si-SiOx NT) electrode [9]. This structure features a SiOx shell that accommodates the volumetric dilation of the internal silicon stratum into the cavity within the tube during the charging phase. This design also facilitates the forming of a jarless solid electrolyte interphase (SEI) film by shielding the silicon active substance is shielded from immediate contact with the electrolyte. As a result, the duplex-shelled Si-SiOx NT electrode has demonstrated an elevated specific capacity reaching approximately 3000 mAh/g, along with an extended cycle life of 6000 cycles with an impressive 88% capacity retention. Additionally, it has maintained an exceptionally high Coulombic efficiency exceeding 99.9% throughout cycles, from the second through to the 6000th.

Karki et al. have investigated the electrochemical performance and morphological transformations of nickel-coated silicon nanotubes (Ni-coated Si NTs) during cycling, utilizing in situ transmission electron microscopy (TEM) [10]. In situ TEM is crucial for examining Ni-coated Si NTs as it provides real-time, high-resolution insights into the dynamic structural changes and morphological transformations that directly influence the material's electrochemical performance and stability. The Ni coating inhibit the lateral expansion of the inner silicon core and instead directs the expansion towards the hollow central region of the nanotubes. Notably, silicon nanotubes with a 16 nm thick nickel outer film exhibited increase of approximately 1% in their external diameters upon lithiation and demonstrated stable cycling performance.

3. Current Research Status of Silicon-Based Anodes in Lithium-Ion Batteries

Considering the numerous detrimental effects caused due to the considerable volumetric increase of silicon materials upon lithium intercalation, researchers have started to explore various carbon substrates as a buffering medium for silicon. This approach can significantly enhance its cycling performance. Typical uses encompass materials including graphite, carbon nanotubes/nanofibers, and graphene sheets.

3.1. Silicon/Graphite

Table 1. Comparison of properties of graphite, silicon base and silicon carbon anode [11-14].

Negative type	Advantage	Disadvantage
Graphite	High conductivity, excellent cycle stability	Low theoretical specific capacity (372mAh/g)
Silicon base	High theoretical specific capacity (420mAh/g)	Large volume expansion effect, low conductivity
Silocarbon	High specific capacity and excellent cycle stability	Scale production is not unified, the early stage of research and development

Silicon electrodes have garnered significant attention owing to their elevated theoretical capacity for charge storage, which is 420 mAh/g. However, the substantial volume expansion they undergo during the charge-discharge cycles may induce structural degradation, which can subsequently result in the rupture of the solid electrolyte interphase (SEI). The re-formation of the SEI layer consumes lithium ions, which reduces the overall performance of the battery. In addition, the silicon itself lacks electron conductivity, which hinders electron transport. In contrast, although graphite has a low theoretical capacity (372mAh/g), its excellent electron conductivity and stable cycling performance make it a good choice for electrode materials. Combining silicon with graphite and optimizing the electrode structure allows for the development of a silicon-carbon composite anode with superior electron conductivity and controlled volume expansion. As shown in Table 1.

Holzafel et al. grew silicon on fine graphite particles by chemical vapor deposition technology, and the obtained silicon-graphite composite showed a starting specific capacity of 1350 mAh/g was observed at a current density of approximately 75 mA/g,this indicates that the composite material has a high charge storage capability,and sustained a capacity of 10 mAh/g post 10 cycles of operation, which represents a slow rate of capacity attenuation. This slow degradation can be attributed to the uniform dispersion of silicon nanoparticles on the graphite substrate, which likely prevents the agglomeration and uneven expansion that can lead to structural failure [15]. This slow capacity attenuation is mainly because of the consistent distribution of silicon nanoparticles across the graphite substrate. The graphite matrix here plays a role in limiting the silicon expansion, effectively improving the mechanical stability of the silicon base anode. Kumta et al. fabricated an innovative composite material of silica-graphite-PAN by integrating the reactive silicon phase into the graphite framework and applying a carbon coating derived from polyacrylonitrile (PAN) [16]. As a diffusion barrier, the PAN-based coating effectively inhibits the interaction occurring at the boundary between graphite and silicon. This process resulted in the composite achieving a reversible specific capacity of 60 mAh/g at a current density of 160 mA/g, with minimal capacity degradation observed after 30 cycles. Lee et al. synthesized a silica-graphite-carbon composite with spherical nanostructure by using high temperature solid phase reaction [17]. The material exhibited a specific capacity of 70 mAh/g and a Coulombic efficiency of 86% during its initial cycle, and had good long-term cycle stability, mainly due to its enhanced structural stability. Successfully mitigates the mechanical strain induced by silicon's volumetric fluctuations throughout the charging and discharging processes [16].

3.2. Silicon/Metal

The silicon/metal composite can effectively utilizes a combination of silicon with various metals to counteract the volume expansion and contraction experienced by silicon during the charge-discharge cycle. The silicon/metal composite is capable of effectively mitigating the volume expansion and contraction of silicon during the charge and discharge processes due to the synergistic properties of the combined materials. Additionally, metals can provide a stable and robust framework that withstands the mechanical stress resulting from the volumetric alterations of silicon throughout the charge-discharge process. The incorporation of metals into silicon composites creates a structurally robust and electrically conductive matrix that can accommodate the volumetric changes of silicon, thereby enhancing the mechanical integrity.

D.B. Puolat and colleagues have developed nano-columnar composite copper-silicon (Cu-Si) films through the oblique electron beam co-evaporation technique [18]. This nano-columnar architecture is beneficial for enhancing electrical conductivity, shortening the lithium ion (Li+) diffusion pathway, and prolonging the material's cycle life. The film's thickness can be modified by controlling the duration of the evaporation process. Experimental data reveal that the film's negative electrode demonstrates a significant specific capacity of 2933.7 mAh•g-1 during the the initial phase of discharging and maintains a commendable cycle performance, retaining high capacity for up to 100 cycles.

R.Y et al. synthesized MAX phase Ti3SiC2 ceramics followed by a ball milling oxidation process to create a TiO2/SiOx hybrid material with nanoparticles less than 10nm in diameter and a bicontinuous structure [19]. The material exhibited a gravimetric capacity of 370.7 mAh/g in the subsequent discharge cycle, evaluated at a current density of 1.0 A/g. The capacity gradually diminished to 355.3 mAh/g by the time of the 1000th cycle. The structural design ingeniously leveragesThe elevated capacity characteristic of SiOx coupled with the robust stability in TiO2, with the two active components interpenetrating each other during lithium insertion and extraction. This interaction mitigates and alleviates the volume expansion issues associated with each component, thereby enhancing the capacity and lifespan of the silicon-based anode material.

3.3. Silicon/graphene

The combination of silicon material and graphene markedly enhances the electrochemical characteristics. of silicon electrodes. The preparation of this composite substance not only facilitates the formation of a more perfect SEI film layer to enhance its stability, but also effectively avoids the agglutination of silicon particles, thus optimizing the electron transport performance of the electrode.

To date, graphite is still the commercial choice for lithium-ion battery anode materials. However, silicon, with its wide distribution in nature and theoretical lithium storage capacity of up to 420mAh1g, is increasingly being seen as a serious contender as a prospective anode material for forthcoming lithium-ion batteries. At the same time, Graphene's exceptional electron mobility allows for rapid charge transport, which is crucial for high-performance batteries. Efficient electron transport reduces the internal resistance of the electrode, leading to faster charging and discharging rates. The expansive surface area of graphene provides morereactive sites facilitating electrochemical processes, which is able to facilitate the capacity of the electrode to store charge. This is particularly important for energy storage applications where maximizing the utilization of available space is key. Because of its excellent electron conduction properties and broad specific surface area, graphene builds an efficient electron transport network in the electrode material and it is often used as a conductive reinforcement material in the electrode, a surface coating agent, or directly combined with the electrode material.

Luo and colleagues used an a novel aerosol-assisted capillary deposition technique to successfully synthesize a composite structure of silicon nanoparticles (SiNPs) wrapped in a graphene shell [20]. The silicon nanoparticles are between 50 and 10 nanometers in diameter, while the graphene layer that wraps them is about 5 to 10 nanometers thick. The folded structure of the graphene layer can successfully accommodate the volumetric variations of silicon throughout the lithium intercalation process, ensuring the stability of the electrode structure. At a current density of 10 mA/g, the composite material was able to deliver a specific capacity of 940 mAh/g after 250 cycles, exhibiting a minimal capacity fade rate of 0.5% per cycle.

3.4. Other silicon-based composites

MXene is a novel category of two-dimensional materials akin to graphene, with titanium carbide ($Ti_3C_2T_x$) being the most extensively researched due to its high electrical conductivity. Y.C. UI and colleagues have functionalized silicon nanoparticles (Si-NP) with (3-aminopropyl) triethoxy silane, commonly known as Aminopropyltriethoxysilane (APTES) [21]. Through a combination of electrostatic interactions and self-assembly of $Ti_3C_2T_x$, they synthesized a composite material denoted as $NH_2Si/Ti_3C_2T_x$. The optimized $NH_2Si/Ti_3C_2T_x$ demonstrated an impressive reversible gravimetric capacity of 864 mAh/g following 100 cycles at a current rate in 0.1C.

M. Abdollahifar and colleagues have utilized chitosan, a non-toxic and biodegradable material, as the primary substrate for their study [22]. They crosslinked sulfonated chitosan (SCS) with glutaraldehyde (GA) to enhance its properties. The Si@Gr/C particles, which are silicon and graphene oxide composites, were then coated with the crosslinked SCS to form a versatile polymeric synthetic solid electrolyte interphase (A-SEI) shielding film, resulting in the Si@Gr/C-SCS composite anode material. This composite anode has exhibited a gravimetric capacity reaching up to 600 mAh/g. Notably, even without the optimization of the electrolyte, the composite sustained a capacity retention exceeding 67% post 1000 cycles.

4. Conclusion

In the past 25 years, LIBs have risen rapidly to become a key component of society's infrastructure, and they play a major role in portable electronics alongside extensive apparatus and initiatives. Still, existing LIBs are approaching the saturation point of their theoretical capacity, with diminishing opportunities for enhancing battery design to augment energy density to increase energy density through traditional materials science and methods. The escalating need for elevated energy density in sectors like electric mobility has prompted an urgent search for new LIB technologies that were previously considered impractical. Silicon has a theoretical lithium storage capacity of up to 4,200 mAh g^-1, a value that far exceeds the carbon-based negatives that currently dominate the market. Due to its abundant resources and good environmental adaptability, silicon is considered as an ideal substitute for carbon-based negative electrode materials and has attracted extensive research attention.In this paper, the theories and methods of preparing these compounds, as well as their physical properties and electrochemical properties are discussed. In the fabrication of composites that incorporate silicon, carbon is the most widely used element with diverse application forms. This encompasses straightforward carbon layering, blending with silicon, and crafting structures with core-shell or yolk-shell configurations, integrating with graphene and carbon nanotubes, and constructing multistage or three-dimensional silicon-carbon structures.In addition to the study of silicon morphology and its development in various silicon-based composites, there is also increasing research interest in auxiliary components of batteries, such as co-solvents for electrolytes, additives, and functional adhesives. These auxiliary components are instrumental in building a stable solid electrolyte interface (SEI) film and maintaining the electrode structure and conductivity. While nanosilicon is favored for its advantages in providing excellent capacity and stable cycling property, people need to recognize that nanotechnology is not the ultimate solution to all problems. The ultimate goal of silicon anode research is to develop commercially competitive products, which need to take into account all practical factors. The challenges associated with silicon negatives have no chance to be completely overcome by a simple strategy. Therefore, a reasonable combination of the aforementioned strategies to achieve overall superior performance is critical to driving widespread commercialization of silicon-based lithium-ion batteries.

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