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Application of Silicon-Carbon Anode Materials in Lithium-ion Batteries

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Abstract. In the trend of global electrification, Li-ion batteries play a vital role in energy storage systems. However, low capacity of graphite anodes has become a critical constraint limiting further development of Li-ion batteries. Silicon has emerged as a promising alternative anode material due to its high theoretical capacity—approximately 11 times that of graphite. However, its practical application is hindered by inevitable drawbacks, including large volume expansion of 300%-400% during charge-discharge cycles and poor long-term durability. To achieve a compromise, integration of silicon and carbon materials (such as graphite, graphene, and CNTs) offers a new idea, because they can buffer silicon's volume expansion and enhance electron/ion conductivity. In this paper, silicon-carbon composite fabrication will be analyzed as a novel approach to balance the energy capacity and working stability. The composition design of silicon-carbon materials, the synthesis mechanisms, and the factors for commercialization are discussed. This paper also examines current technical challenges and proposes promising outlooks for future development of next-generation lithium batteries with high energy density, holding great potential for advancing sustainable development and global energy transformation.

Keywords: li-ion battery, carbon-silicon composite, silicon anode, electrochemical performance

1. Introduction

Despite technological advancements and investments, humanity is undoubtedly still facing intensifying climate change driven by increasing human emissions of greenhouse gases from fossil fuel combustion, which are already having widespread negative impacts on the environment: more frequent heat waves, global warming enhancement, melting glaciers, etc. However, the global economy continues to be powered by up to 80% of fossil fuel-based energy. High energy dependence and consumption of traditional non-renewable resources are major obstacles to achieve carbon neutrality and sustainable development.

To address current challenges, electrification will probably be the only technologically viable option for revolutionary transformation. Electrification is a systematic endeavor covering numerous sectors, including transportation, buildings, and industry. At the point of use, electric technologies do not produce any emissions; thus, they transfer the emissions to the power sector. This would allow

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policymakers to focus more on decarbonizing power plants and the electric grid for emission reduction, rather than struggling to decarbonize every single emission point.

In the process of electrification, it is necessary to develop technologically advanced energy storage systems for the efficient utilization of renewable energy resources. The most common energy storage systems are electrochemical energy storage systems—rechargeable batteries, such as nickel-cadmium batteries, lead-acid batteries, lithium iron phosphate batteries, lithium-ion batteries, and sodium-ion batteries. These batteries feature high safety, energy density, long-term stability, and relatively low cost, making them widely used across various fields in modern society [1]. The key to these exceptional properties lies in unique structural design and material options.

Among these five aforementioned batteries, lithium-ion batteries are the most popular choices and have been widely deployed across mobile phones, digital cameras, laptops, electric vehicles, etc. Its working principle is based on the ion migration between the positive and negative electrodes and electron conduction on an external circuit. The basic structure comprises five components: cathode, anode, separator, electrolyte, and external circuit. The cathode of a lithium-ion battery is typically made of lithium-containing transition metal oxides or phosphates, which provide lithium ions during discharge. The mainstream anode material is usually carbon-based, such as graphite, graphene, CNTs, etc. The most popular material is graphite, a carbon allotrope with a layered structure. This unique structure provides large interlayer spacing for lithium ions to intercalate and deintercalate without damaging its original structure after repetitive charging-discharge cycles. Another factor realizing its widespread application is cost-effectiveness. As graphite is abundantly deposited in nature and the purification and processing techniques are well-established, cost can be reduced and mass production can be achieved.

Apart from carbon-based materials, silicon has also attracted significant attention for its high potential as an anode material due to its high theoretical specific capacity—4200mAh/g, which is about 11 times that of graphite. However, silicon also has a significant drawback. When used as an anode, lithium-ion intercalation into silicon will lead to a volume expansion of approximately 300%, while the value of graphite is only about 10%. The solid electrolyte interphase (SEI) film is a layer formed on the anode surface during the first charge-discharge cycle to allow lithium ions to pass through while preventing reactions of other substances. After long-term, repeated charge-discharge cycles, the SEI film will generate cracks or even break, resulting in instability of battery performances.

To find a balance between the high capacity of pure silicon and the structural durability of graphite, Si-C composites could offer the ideal solution to anode materials for high-performance lithium batteries, which would be a promising alternative to the current pure graphite anode. In this paper, the focus will be on working principles, synthesis approaches, and commercialization analysis of three major Si-C anode materials, and giving possible advice for futuristic anode material development, serving as a reference for the invention of next-generation lithium batteries with enhanced energy density and long cycle lives.

2. Silicon-carbon anode

Various types of high-capacity silicon materials have been investigated for the production of carbon-silicon anodes. Here are some proposed examples of the graphite-blended silicon anode, aiming at alleviating intense volume changes [2].

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2.1. Si/graphite anode

2.1.1. Graphite/SIOx

Due to adjustable oxygen content and relatively superior electrochemical performances demonstrated by effective mechanical stress alleviation of Li_2O and Li, numerous scientific researches in terms of SiOx have been conducted. In the random mixture model, silicon monoxide is a mixture with little silicon and silicon dioxide mingled together randomly in multiple phases [3].

Additionally, Hohl et al. [4] suggested an interface clusters mixing model based on data from angstrom-beam electron diffraction, wherein silicon and oxygen coexist as clusters of silicon and silicon oxide, surrounded by various suboxide-type tetrahedral coordinates. In contrast to the approximately 3-fold volumetric expansion of a pure silicon anode, the one based on SiOx exhibited a diminished volume change, decreasing from 300% to around 160%. Though there is a tremendous capacity decrease of more than 2000 mAh/g, the remaining capacity is more than enough compared with the capacity of a typical cathode (200 mAh/g).

2.1.2. Graphite/nanosilicon

There are several types of combinations of nanosilicon particles and graphite, including watermelon-like structures, core-shell structures, and quasi-sandwich structures [5].

For watermelon structure, nanosilicon particles are uniformly distributed like "watermelon seeds" and embedded in graphite (“watermelon flesh"), in which nanosilicon can be buffered effectively in the carbon matrix during lithiation. Qian et al. [6] conducted an experiment to prove the feasibility of watermelon structure. They prepared a hierarchical graphite-scaffold Si/graphite composite. The hierarchically structured anode was determined to provide a reversible capacity of 445 mAh/g at the current density of 372 mA/g after 300 cycles with only 21% of capacity fading. When the current densities were increased to 2976 and 3720 mA/g, there were still good capacity retention performances, which were 236 and 186 mAh/g, respectively. Therefore, “watermelon-like” structured anodes truly have excellent capacity stability after volume expansion for numerous cycles.

The core shell is also a typical nanosilicon/graphite structure. The graphite is at the core, providing an adhesion surface for silicon-based layers. The silicon coating layer comprises silicon and carbon on the base of polymer. The role of the silicon coating is a "binder". It was reported that a kind of carbon-enhanced hard nanosilicon/graphite composite had outstanding performance. The anode with 10% binder and 11.4% Si showed a relatively high capacity retention of 92% after 150 cycles but a relatively low reversible capacity of only 638.6 mAh/g. However, the anode with same binder content and higher silicon content of 14.6% could store more lithium ions and exhibited a highly reduced capacity retention (less than 80%). When the binder content was increased to 20% but with silicon content unchanged, the capacity retention could be further increased to over 80%. The key to this core shell structure is the contents of the binder and silicon, which could impact anode stability and battery performances significantly.

Quasi-sandwich structure is a carbon-silicon-graphite structure with three layers at the micron scale, in which the silicon layer is adhered to the bottom graphite layer and covered by the carbon layer for buffering on the outside. A study showed that a quasi-sandwich material with silicon in the middle of graphite and a carbon layer was achieved after heat treatment. An anode with 10.5% carbon was put into experiments, and a reversible capacity of 821 mAh/g and an initial coulombic efficiency of 87.6% were obtained. The two values exceeded those of pure silicon. If the proportion

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of carbon was increased to around 22%, the lithium storage capacity slightly decreased to less than 800 mAh/g and ICE increased to about 89%. Hence, the key factor to the quasi-sandwich structure is the carbon layer, which provides enhanced surface protection to Li-ion batteries [7].

2.1.3. Graphite/Si-metal alloy

The silicide alloy could reduce the concentration of active silicon and alleviate intense volume expansion. For instance, a graphite-blended Si anode with the Si-based alloy 3M alloy, referred to as V6, was investigated. Though its cycle efficiency was high and the microstructure of V6 was stable, calendaring would lead to particle fractures and low capacity retention. The graphite-infused anode utilizes supplementary graphite to mitigate calendering effects, therefore avoiding particle breakage, reducing volumetric expansion, and enhancing capacity retention. Furthermore, the incorporation of graphite with an average particle size of 3μm significantly enhanced the overall cycling performance.

Another Si-metal-based anode was proposed. In this model, the metal silicide made of iron, copper and silicon exhibited a spherical shape, where silicon nanoparticles were attached to the surface and well encapsulated. The FeCuSi/graphite combination, when combined with graphite in a conventional anode, has exceptional cycling stability, achieving a capacity retention almost unchanged after 50 cycles [8]. These works directly show the advantages of the graphite/Si-metal alloy anode.

2.1.4. Synthesis of silicon-graphite anode

The production of silicon-graphite anodes may be achieved by several viable processes, including mechanical grinding, spraying, chemical vapor deposition, and wet processing, all of which hold considerable potential.

2.2. Silicon-graphene anode

2.2.1. Composition of Si/graphene composite anode

The overall performance of anodes is highly related to graphene mass ratio. Hwang et al. [9] investigated the impact of a high graphene weight ratio in Si/Graphene nanocomposite. The study made a comparison of four groups of different weight ratios, which are 3:1, 2:1, 1:1, and 1:2, respectively. The results indicate that the Si/graphene composite with a weight ratio of 3:1 exhibited the optimal equilibrium between initial capacity and cycle performance.

Loading is also an important part of the battery performance, as it's closely related to energy density, conduction efficiency, and mechanical stability. Chabot et al. [10] investigated the loading ratio of silicon nanoparticles to graphene. Nanocomposite ratios of 1:3, 1:2, 1:1, and 2:1 were examined. And the result indicated that the combination with a loading ratio of 2:1 exhibited the best initial capacity, and the composite with a ratio of 1:1 had the maximum retention capacity. Ji and Ruoff et al. [11] demonstrated a really thin graphite foam anode encapsulated with graphene with silicon mounted on. As tested, the retained capacity at the 100th cycle was 370 mAh/g. Loading density should be higher, as low loading density would increase the overall weight while decreasing energy density, which makes batteries much bulkier.

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2.2.2. Synthesis of silicon-graphene anode

Kung et al. [12] first reported a type of silicon-graphene composite for lithium-ion battery anodes. This research involved pre-treating silicon nanoparticles. They must be fully exposed in air for a whole night to ensure the place of silicon oxide production was their surface. After that, the silicon oxide layer was mixed with graphene oxide derived from graphene oxidation. The mixture underwent a reduction using 10% H_2 in a heated argon atmosphere to form Si/Graphene nanocomposites. The synthesized nanocomposites showed a high initial discharge capacity of about 1950 mAh/g and roughly 50% retention at 1000 mA/g after 120 cycles.

After two years, Liu et al. [13] fabricated a silicon-graphene nanocomposite with a three-dimensional porous structure. Compared with typical random mixtures of silicon nanoparticles and graphene nanosheets, this nanocomposite exhibited enhanced electrochemical stability due to the robust adhesion of silicon nanoparticles to the nanosheets via a thin SiOx layer. This three-dimensional porous architecture delivered a reversible capacity of approximately 900 mAh/g with little capacity loss when the charge rates increased by 10 times. Even when the current rate was up to 5 A/g, the 3D nanocomposite still demonstrated capacity retention over 400 mAh/g.

Yu et al. [14] proposed a method of silicon/graphene composite fabrication, which is a vertical structure made of silicon, graphene sheets, graphite, and carbon (VGSs). The preparation is to adhere silicon nanoparticles to graphite particles, followed by the growth of vertically aligned graphene sheets in place using thermal chemical vapor deposition. This nanocomposite exhibited a reversible capacity of about 780 mAh/g after 1000 cycles and a capacity retention of 91.5% after 500 cycles, attributed to the buffering function of this vertical structure and the connection between silicon and graphite particles.

2.3. Silicon-CNTs anode

2.3.1. Composition of Si/CNTs composite anode

A study indicated that scientists fabricated a type of Si/CNTs composite by single-step heat reduction of a mixture of SiO_2 nanoparticles and CNTs at 650°C [15]. The composition of the resulting sample is contingent upon the mass ratio of SiO_2 to CNTs in the reactants. To figure out the impact of mass ratio, Si/CNTs-1, 2, 3 were separately tested. For Si/CNTs-1 composite, a sparse and random distribution of Si nanoparticles was observed. The silicon nanoparticle distribution of the Si/CNTs-2 composite was uniform and of high content, because interfacial silicon carbide stabilized silicon nanoparticles on CNTs. Si/CNTs-3 also showed an even silicon dispersion, but the particle size decreased to 10 nm as a result of the reaction between the two materials. In electrochemical performance tests, the three materials were used as anodes of coin cells. It was found that Si/CNTs-2 exhibited the highest initial reversible discharge capacity and charge capacity due to its highest Si content. After 200 cycles, the capacity retention was still high, up to 83.3%. By contrast, Si/CNTs-1's capacity retention was relatively low at 32.4%. For Si/CNTs-3, though the capacity retention was the highest, about 90%, the capacity was only about 730 mAh/g after 200 cycles. Overall, the composition with the best cycling performance is Si/CNTs-2. The mass ratio of silicon dioxide to carbon nanotubes, their content, and particle sizes are all key factors in terms of battery anode performances.

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2.3.2. Synthesis of silicon-CNTs anode

Fang et al. [16] proposed a method: anchoring carbon-coated silicon nanoparticles onto CNTs to boost electrochemical performances in Li-Ion batteries through polymerization and carbonization. The newly fabricated composite is denoted as Si@C@CNTs@C. The anchoring architecture could help control the aggregation of SiNP, and the CNTs as well as the carbon layer buffer large volume expansion. This design also exhibits great electrochemical performances: high initial discharge capacity of about 4162 mAh/g and coulombic efficiency of 84%.

Wang et al. [17] developed a type of three-dimensional composite coated with silicon, graphene, and CNTs. Firstly, silicon nanoparticles were modified by CTAB for dispersibility improvement. After modification, silicon nanoparticles were evenly distributed within graphene and carbon nanotubes and coated by a carbonized poly-dopamine carbon layer under a high-temperature condition, which enormously controlled volume expansion. This material, utilized for Li-ion battery anodes, exhibited exceptional electrochemical performances: a reversible capacity of 1946 mAh/g after 100 cycles and a high capacity retention of 69% at 1A/g.

3. Common factor analysis of carbon-silicon materials commercialization

3.1. Electrochemical performances

The short-term goal of building carbon silicon anodes is to attain superior electrochemical performance characterized by a higher volumetric capacity, higher initial CE, higher retention rate, longer cycle life, and lower volume expansion after repetitive charge and discharge cycles. All these parameters are supposed to be tested with relatively high standards to ensure their reliability and stability in real-life usage.

3.2. Carbon-silicon compatibility

As the anode includes both carbon and silicon materials, it is not enough to only focus on electrochemical features of the two materials separately. The compatibility is also crucial for the system to work as a whole, which hasn’t been deeply studied. Furthermore, it is also of enormous significance to conduct more research on the co-utilization of carbon and silicon materials.

3.3. Safety

As silicon-based materials undergo significant volume changes (up to 300%) during lithiation, which will probably lead to internal structural damage in the battery and in turn the severe safety hazards, structural design optimization methods such as adding buffering layers and using elastic binders are expected to be adopted.

Uneven lithium-ion intercalation in silicon-carbon anodes can cause lithium dendrites to form on the electrode surface. And the growing dendrites will probably pierce the separator and lead to a short circuit, which is a severe safety hazard. To deal with this kind of problem, dendrite inhibitors are crucial tools, requiring further scientific study and research.

In long-term uses, electrolyte faces the problem of depletion. The SEI layer on silicon makes charged silicon decompose binders, lithium salts, and organic solvents in the electrolyte, causing continuous electrolyte consumption. Innovation of electrolytes with higher chemical stability and compatibility is necessary.

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4. Conclusion

In the historical trend of global electrification, Li-ion batteries are widely accepted and adopted in all aspects. Silicon-carbon composite anode is a promising solution to simultaneously balance high structural stability provided by carbon materials and high capacity of silicon, giving a new insight into performance improvement of traditional Li-ion batteries. This review summarizes the current research progress of three major Si-C anode materials: silicon-graphite, silicon-graphene, and silicon-CNTs, focusing on their compositions, synthesis methods, and electrochemical performances. In the composition sector, material selection and proportions are analyzed and compared. In terms of synthesis, technologically advanced methods are demonstrated. And electrochemical performance evaluation is always on its way to offer an intuitive understanding of different techniques.

Key factors concerning mass production and commercialization include material cost, electrochemical performance metrics (cycle retention, initial coulombic efficiency), carbon-silicon compatibility (volume change control and interface stability), and safety (suppression of dendrite growth, electrolyte stability). Current challenges in terms of the aforementioned factors are identified, and potential solutions and outlook are proposed.

Future research will focus on developing next-generation batteries with better anode structural design, higher systematic stability, more advanced synthesis approaches, and more efficient electrochemical performances. In the innovation process, strict testing standards should be a basis for fabricating products with better quality.

Along the way from scientific research, production, commercialization to recycling and disposal, environmental friendliness is supposed to be a critical point. Only in this way will the electrification process make a difference to environmental protection fundamentally, putting the global call for sustainable development into practice.

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