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Application of Silicon-Carbon composite materials in the Lithium-Ion Battery Anode

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Abstract. The energy crisis has become a focal issue that the entire world is currently facing together. In that case, electricity, as an important renewable energy source, has become the main development target in the current technological context. With the development of electrification, mobile electrical devices, such as electric vehicles and mobile phones, have increased significantly, thereby increasing the demand for high-performance batteries, especially lithium-ion batteries. Due to their high energy density, the ability to be recharged, and broad development prospects, lithium-ion batteries have attracted much attention. The performance of lithium-ion batteries is greatly influenced by the performance of their negative electrodes. Therefore, the development of the negative electrode of lithium-ion batteries has long been on the agenda. The silicon-carbon composite negative electrode not only utilizes the strong performance of the silicon negative electrode material but also solves the common problems faced by all the silicon-based negative electrodes by adding carbon materials. Therefore, its application potential is extremely high. This paper conducts a systematic study of the characteristics and application prospects of three types of silicon-carbon composite negative electrodes based on literature research and summary. The results help lay a certain foundation for further research directions and large-scale production for lithium-ion batteries which based on silicon-carbon composite negative electrodes in the future.

Keywords: Silicon-carbon composite material; lithium-ion battery; carbon-based material.

1. Introduction

In today's world, the major energy consumption still relies on the fossil fuels, such as coal, gasoline, diesel, etc. However, fossil fuels are not renewable. As human consumption increases, the total reserves of fossil fuels on Earth are rapidly decreasing. a great number of oil fields have been depleted, and the world has already fallen into the energy crisis. Additionally, burning fossil fuels also generates a large amount of greenhouse gases and pollutants. For example, burning coal generates a lot of coal ash and carbon dioxide, which exacerbates the greenhouse effect and is extremely unfavorable to the current situation of global warming. Therefore, people have begun to vigorously develop electricity to replace fossil fuels. For batteries, as the important devices for storing electricity, market surely has an increasing demand to both their quantity and their performance.

At present, the most mainstream battery types include lithium-ion batteries, lead-acid batteries, nickel-cadmium batteries, etc. Among them, lithium-ion batteries have attracted much attention due to their durability, high energy density, and the ability to be repeatedly charged and discharged. Lithium-ion batteries are mainly composed of positive electrode, negative electrode, separators, and electrolyte. The negative electrode material is an important factor affecting their performance. Currently, the negative electrode of lithium-ion battery is mainly composed of graphite, but the rate capability, capacity, and service life of this type of battery are relatively poor [1]. Although using silicon material as the negative electrode can increase the theoretical specific capacity, low-temperature working ability, and rate capability of the battery, it also leads to severe expansion and contraction of the negative electrode material during lithium ion intercalation and deintercalation, eventually causing electrode pulverization and battery damage. Therefore, researchers have added carbon material to significantly improve its electrochemical properties. This kind of negative electrode made of silicon-carbon composite material has high conductivity and high capacity, significantly raising the upper limit of future battery performance. Carbon materials can provide buffering, helping to maintain the integrality of the negative electrode when it expands and contracts. For example, Zhang et al. [2] used the spray drying method to synthesize silicon-carbon composite materials with porous carbon coating. Lithium-ion batteries, which using this material as the negative electrode, exhibit extremely superior electrochemical properties, far exceeding the performance of lithium-ion batteries which made with traditional graphite negative electrodes. This article will focus on the silicon-carbon composite negative electrodes in lithium batteries, aiming to summarize the characteristics of different types of silicon-carbon composite materials and explore their application scope. This article will review several types of silicon-carbon composite negative electrodes, such as silicon/graphite, silicon/carbon nanotubes, and silicon/graphene, and propose suggestions for the future development of silicon-carbon negative electrodes for laying the foundation for the development of lithium batteries with higher energy density and longer service life.

2. Classification, advantages, and disadvantages of silicon-carbon composites

With the development of electric and electronic devices, lithium-ion batteries have become the dominant force in the current battery market. Improving the performance of their negative electrode is an urgent need. Using graphite as the negative electrode of lithium-ion battery is the current mainstream, but, nowadays, this kind of battery have almost reached the theoretical specific capacity of graphite negative electrode (372 mAh·g-1). The theoretical specific capacity of silicon-based negative electrode(4200 mAh·g-1) is approximately ten times that of graphite negative electrode, which is fully sufficient to meet the current market demand [3]. However, we cannot easily transform silicon negative electrodes into widely applicable battery negative electrodes because during charging and discharging, with entrance and exit of lithium ions, the volume of the silicon material undergoes drastic changes. In that case, the battery negative electrode will powderize and fall off, resulting in the destruction and thickening of the solid electrolyte interphase layer (SEI). Ultimately, the discharge efficiency of the battery will rapidly decrease, and the actual capacity will significantly reduce. In addition, the low conductivity of silicon-based negative electrodes also limits their application. Therefore, the scientific community modifies silicon-based negative electrodes by introducing carbon materials for solving the common problem faced by silicon-based negative electrodes. In this way, the practical specific capacity of this composite anode far exceeds the theoretical specific capacity of graphite anode.

Silicon-carbon composites mainly address the drawback of volume change in silicon-based anodes. As composite materials, their properties vary depending on the carbon materials they are combined with. Currently, the most mainstream silicon-carbon anode materials include: silicon/graphite composites, silicon/carbon nanotubes composites, and silicon/graphene composites. These materials exhibit different properties, which depend on the characteristics of the combined carbon materials, but their primary purpose is to utilize the outstanding advantages of silicon while addressing the defects of silicon-based anodes.

2.1. Silicon/graphite Composite material

Silicon/graphite composites are prepared by combining graphite— which possesses mechanical strength, high electrical conductivity, and excellent chemical stability—with silicon anode materials that possess high specific capacity. This type of composite material highly coordinates the advantages of silicon and carbon and complements the disadvantages of them. Nowadays, with lots of researches by scientists, It is also regarded as one of the most promising development directions for battery anodes. Silicon and graphite are abundant in the Earth's crust, resulting in low costs and high commercial value, giving them an advantageous position in large-scale industrial production and commercialization in the future. Owing to its certain mechanical strength and stability, graphite can effectively mitigate the volume change of silicon during lithium ion insertion and extraction when combined with silicon or silicon oxides. Although silicon has low electrical conductivity, graphite exhibits high conductivity; blending these two substances can make up for the drawback of low electrical conductivity of silicon-based anodes. In summary, the composite exhibits the advantageous characteristics of both materials, such as good conductivity, small volume, long cycle life, and high specific capacity. However, due to the significant differences in properties between the two substances, preparing them into a single integrated composite anode remains a major challenge [4].

The main preparation methods for silicon/graphite anodes include mechanical ball milling, spray drying, and CVD (Chemical Vapor Deposition). Among these, ball milling and spray drying have low requirements for process conditions and low operating costs, exhibiting high prospects for commercial application and value in large-scale industrial production. However, the performance of the anode materials produced by these two methods is not as outstanding as that of those produced by the latter. The silicon-carbon anode materials prepared by CVD possess higher Coulombic efficiency and longer cycle life, but the preparation conditions of this method are relatively harsh and the cost is high, making it unsuitable for large-scale production and commercialization. Fig. 1 shows a transmission electron microscopy (TEM) image of a kind of silicon/graphite composite at different magnifications [5].

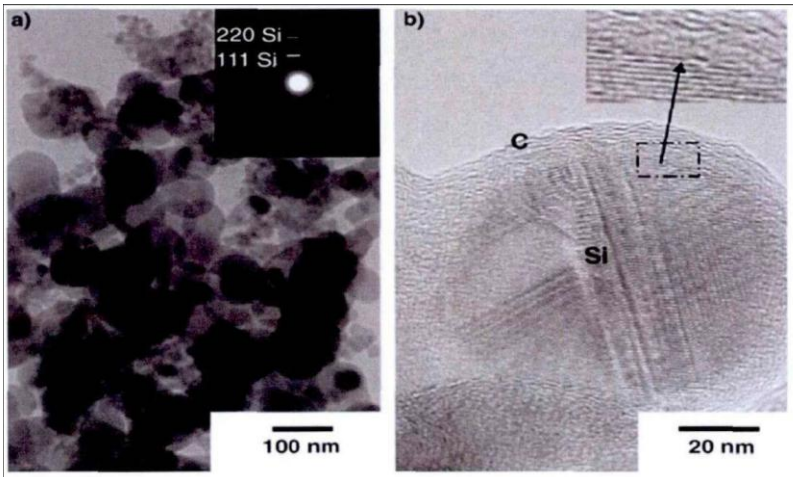

Fig. 1 TEM images of silicon-carbon composite nanoparticles prepared by spray drying method: a) Image at low magnification; b) Image at high magnification [5]

Owing to its excellent cycle life, high capacity, enhanced discharge efficiency, and high electrical conductivity, this silicon/graphite anode enables the further development of high-capacity, compact fast-charging batteries. Such batteries can be applied in fields such as electric vehicles and mobile phones, thereby promoting the development of the electronic technology.

2.2. Silicon/carbon nanotubes Composite material

Silicon/carbon nanotubes composites are formed by combining carbon nanotubes—characterized by excellent electrical conductivity and toughness—with silicon materials. They not only retain the high theoretical specific capacity of silicon materials but also address the common problem of silicon-based anodes through combination with carbon materials.

Carbon nanotubes form an interwoven network structure in the composite, providing a buffer for silicon materials that undergo significant volume changes during charge-discharge cycles. This prevents the integrity of the overall anode from being affected by the volume variation of silicon nanoparticle within the material. The excellent electrical conductivity of carbon nanotubes can also compensate for the low electronic conductivity of silicon-based anodes; thus, the incorporation of this carbon material rapidly enhances the overall performance of single silicon anodes. Such composite anodes exhibit characteristics such as high specific capacity and long cycle life. Chen Dingqiong and his research team tested lithium-ion batteries using silicon/carbon nanotubes/amorphous carbon composite anodes (amorphous carbon serves to anchor carbon nanotubes) prepared with polyvinyl alcohol gel as the carbon source under 0.1C condition. They found that the initial specific capacity was approximately 800 $mAh \cdot g^{-1}$, and 97% of the capacity was still retained after 100 cycles [6]. However, this type of anode also faces some issues—for instance, carbon nanotubes are very expensive, which economically restricts the large-scale production and commercial application of silicon/carbon nanotubes composite anodes. Mechanical ball milling and chemical vapor deposition can also be employed to fabricate such composite anodes.

In summary, silicon/carbon nanotubes composites possess advantages including high performance, high initial specific capacity, strong reversibility, long cycle life, and high energy density. These outstanding features endow them with broad application prospects in high-tech industries in the future, such as aerospace. Nevertheless, the high cost of carbon nanotubes limits the more widespread application of this anode.

2.3. Silicon/Graphene composite material

Silicon/graphene composites are formed by combining graphene with silicon or silicon oxides. As an advanced material, graphene possesses excellent electrical conductivity, a high specific surface area, and superior mechanical toughness. When compounded with silicon anode materials, the resulting composite anode can effectively address the common issues faced by single silicon-based anodes.

Owing to its single-layer carbon atom structure, graphene adheres to the surface of silicon materials in the composite anode to form a film-like structure. This structure isolates the electrolyte, inhibits the formation and thickening of the solid electrolyte interphase (SEI) layer, and thereby improves the utilization efficiency of active lithium atoms in the battery. Additionally, this film-like structure can promote the alloying reaction between lithium ions and silicon materials, leading to a significant enhancement in the performance of batteries based on silicon/graphene anodes [7]. Cho et al. [8] prepared a silicon/graphene composite anode via the thermal decomposition of silane and assembled it into a battery for testing. The results showed that the initial Coulombic efficiency reached 92.5%, and the specific capacity remained at 1103 $mAh \cdot g^{-1}$ after 1000 cycles at a current density of 14 $A \cdot g^{-1}$. Moreover, during the experiment, the composite anode of the battery spontaneously contracted after cycling, solving the problem of large volume change of single silicon anodes upon cycling [9]. It is evident that this composite material exhibits outstanding specific capacity and cycle life. However, certain factors limit the application of this anode. For example, graphene, as an advanced material, cannot be produced on a large scale, and both its cost and production difficulty are relatively high. Therefore, similar to the batteries based on silicon/carbon nanotubes anodes mentioned earlier, this type of battery may find wider applications in high-tech fields. The preparation of this anode typically involves methods such as CVD (Chemical Vapor Deposition) and thermal decomposition, which are associated with high costs, harsh reaction conditions, and low yields, making them unsuitable for large-scale production [10].

3. Outlook of Silicon-carbon composites for lithium-ion Battery anodes

As a decisive structure, determining the performance of lithium-ion batteries, silicon-carbon anode materials still have some drawbacks. Therefore, the development of lithium-ion battery anode materials should remain a focus in the future.

In future research, emphasis should be placed on technological innovations. For example, new carbon-based materials with higher electrical conductivity, better mechanical toughness, and more suitable structures should be compounded with silicon materials to continuously reduce the impact of silicon's common problem on the overall anode performance, and to more effectively utilize the theoretical specific capacity of silicon to enhance the overall battery performance. Similar to silicon/graphene composite anodes, research can be continued on composite anode materials with a certain degree of self-compaction, rather than passively absorbing the stress generated by silicon materials through physical structures (passive physical stress absorption may ultimately still damage the integrity of the anode). It is necessary to develop new synthesis methods for anode materials to reduce the required reaction conditions, costs, and to improve the safety of materials used. This will enable high-performance silicon-carbon composite anodes to be easily popularized among the public.

In the future, after overcoming the aforementioned issues of cost and synthesis technology, lithium-ion batteries based on silicon-carbon anodes are likely to be widely adopted, replacing traditional lithium batteries. They will find extensive applications in fields such as automotive, mobile communications, and even aerospace. The trend of becoming smaller in volume while storing more energy represents the future development direction of lithium-ion batteries with silicon-carbon composite anodes.

4. Conclusion

Through the research on lithium-ion battery anode materials, this paper finds that silicon/graphite anode materials are easy to produce on a large scale, but their electrochemical performance is not particularly outstanding compared with other silicon-carbon composite materials. In contrast, silicon/carbon nanotubes and silicon/graphene anode materials exhibit excellent electrochemical properties, which are generally superior to those of silicon/graphite anode materials; however, there is still lots of space for improvement in their fabrication processes and production costs. Under such circumstances, researchers cannot easily develop silicon-carbon composite anode materials that simultaneously possess excellent electrochemical properties, low cost, and facile preparation. Therefore, retaining and enhancing the performance of silicon-carbon anodes while significantly reducing their material cost and preparation difficulty is a crucial goal for enabling the large-scale production and application of silicon-carbon composite anodes at present.

The main research purpose of this paper is to summarize the characteristics and potential application prospects of several currently feasible silicon-carbon composite anode materials. This work is expected to provide a theoretical basis for the future development of anode materials and alleviate the pressure of future researches.

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