

# Application of Nano Silicon-Carbon Composite Materials in Anode Materials for Lithium-Ion Batteries

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**Abstract.** With the growing global demand for high-energy-density energy storage technologies, silicon (Si) is gradually replacing graphite anodes to become one of the most competitive materials in the lithium-ion battery (LIB) field. The theoretical capacity of silicon is nearly ten times that of graphite. However, silicon's volume expansion and low intrinsic conductivity become its Achilles' heel. This paper reviews strategies for silicon-carbon nanocomposites aimed at solving these problems. The work focuses on analyzing three popular architectures: amorphous carbon encapsulation, reduced graphene oxide (rGO) scaffolding, and carbon nanotube (CNT) reinforcement. The findings show that amorphous carbon creates a shell that buffers volume changes, but it may lead to cracking under stress. rGO modification improves density and conductivity, and smaller rGO sheets are more effective in terms of ion transport than larger ones. CNTs provide strong conductive networks to prevent electrode damage but cause "dead lithium" accumulation and inefficient silicon penetration. So, only by focusing on hybrid designs and advanced manufacturing techniques, it is possible to make silicon anodes more perfect and meet the demands of the global market.

## 1 Introduction

The decreasing amount of fossil fuels and the growing world energy consumption have led the change to sustainable energy storage systems [1]. This has prompted a fast growth in renewable energy sector and the technologies of energy storage. Due to its high capacity and being eco-friendly, the lithium ion batteries (LIBs) have become the star in the new energy vehicle (NEV) market and portable electronic devices [2]. Nonetheless, mass application of NEVs requires a substantial enhancement to high performance batteries of high capacity, charge / discharge rate and reliability [3]. In addition, industries need advanced energy storage systems that provide greater energy and density to meet the needs of electric vehicles and large-scale applications [1, 3] and which ensure the safety of operation by reducing the risks of lithium dendrite formation [1, 2]. At the same time, to achieve good cost-effectiveness people should adopt naturally abundant and environmentally friendly active

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materials [2, 3]. Adherence to these stringent parameters is rather important to the future generation of LIBs that will have to realize a balance between the electrochemical and economic performance at a synergistic level to be able to sustain a sustainable energy vision [1, 3].

Even with the profound developments in the LIB technology, the pressing need to achieve the systems of high energy density with accelerated cycling reliability is still one of the leading issues in the sphere [4]. Conventional commercial LIBs using mainly graphite anodes are less and less capable of meeting the demanding performance standard needed by classical applications [3]. Such a drawback can be mainly explained by the fact that carbonaceous materials are inherently limited by theoretical capacities of energy density and therefore cannot be further enhanced by the gravimetric energy density, which is limited to that of only 372 mAh g<sup>-1</sup> at most [5]. Therefore, the technical bottlenecks are being countered by developing next-generation battery systems with higher storage capacity and a greater cycling stability with massive development [1, 5]. Intrinsic properties and customized structure of electrode materials are the basic determining factor of the performance of a battery; capacity, rate capability, and operational lifespan [1]. Thus, the high-performance electrode materials exploration and the accurate engineering is becoming the hot topic of materials science research at present [5]. In this regard, scientists are turning to other materials that have the potential to liberate the structural and kinetic constraints of the existing systems to realize a synergistic equilibrium between ultra-high specific capacity and structural integrity over a time.

Silicon (Si) has proven to be the most interesting replacement to the traditional graphite anodes because of its theoretical capacity of about 3,579 mAh g<sup>-1</sup>, which is close to 10 times greater than graphite [5]. What's more, silicon is also cost effective and also environmentally benign because silicon will not be used up easily [5]. Yet, there are difficulties with 2 parts. One is that the bulk Si owns poor electrical conductivity. Second, safety concern of Si anode. Because of the low electrical conductivity of Si, conversion to nano-scaled silicon materials has been found to be imperative. The nano-engineered architectures can help to form a significantly increased specific surface area. This expansion essentially reduces the diffusion path of lithium ions and facilitates charge-transfer dynamics, which forms the basis of increased rate performance necessary in the modern EV [2]. Silicon (Si) anodes have a high lithiation coefficient which can be more than 300%, which makes the anode less useful due to high levels of volume enlargement on lithiation. This growth may lead to the pulverisation of the electrode and unstable solid electrolyte interphase (SEI) making its large scale application extremely restrictive [6]. Adding carbonaceous substances into the structural design has emerged as a central research topic in order to overcome these challenges. Carbon modification as a versatile mechanical buffer to collect the Si volume changes, enhance electronic conductivity, and stabilize the anode interface have been proven feasible in numerous studies [7, 8]. In one instance, a fluorine-functionalised core-shell Si@C core was synthesized by high-temperature pyrolysis by Chen et al., this time the carbon shell not only served as an excellent conductor, but as a stable interface that reduced the effects of the volume and provided improved cyclability of the entire battery [9]. Also, Zhang et. al. adopted a polydopamine-mediated approach to the production of SiO<sub>x</sub>/C@C yolk@shell microspheres that proved to have a steady high capacity by efficiently buffering the scaffold swelling via the designed void space [10]. These developments mentioned imply that the strategic design of carbon-modified silicon electrodes is a very promising field of research in developing next-generation energy storage application.

This is a comprehensive review of nowadays which talks about current design strategies of Si-carbon composite anodes in lithium-ion battery, including a research on the tradeoff between anode structure and its electrochemical performance, and gives advice for future research directions.

## **2 Case description**

### **2.1 Amorphous carbon modified silicon electrodes definition**

An amorphous carbon (a-C) modified silicon electrode is defined as a composite architecture where silicon active species are surrounded by a carbon shell or embedded in a disordered carbon network. The basic reason for such change is isotropic mechanical characteristics of amorphous carbon. The physical buffer offered by these properties can easily handle extreme volumetric changes which can often be greater than 300% in silicon and these changes are inevitable in lithiation and delithiation processes. In addition, the a-C layer will also act as a conformal protective shield that does not allow silicon to directly contact with the organic electrolyte. This isolation can be effectively used in preventing the irreversible and unstopped break down of the electrolyte to form a thin and kinetically stable solid electrolyte interphase (SEI). Therefore, such a change reduces the loss of active material and improves the initial Coulombic performance and cyclic stability of the anode system in general.

### **2.2 rGO modified silicon electrode definition**

A reduced graphene oxide (rGO) modified silicon electrode is described as a heterogeneous film in which atom-thick, two-dimensional sheets of graphene are used as a flexible conductive skeleton and mechanical support to silicon nanoparticles or nanowires. These films have high aspect ratios and good carrier mobility, and they are always designed to wrap, sandwich or interweave across the silicon phase to create a strong and stable network of conductive structure in three-dimension. The rGO modification is important because it possesses a high mechanical toughness which enables it to keep electrical percolation when the electrode particles are subjected to the process of pulverization or fracturing of the underlying silicon particles. In addition, functional groups and structural defects left on the rGO surface improve interfacial compatibility and electrolyte wettability so that fast lithium ion diffusion at the electrode-electrolyte interface and efficient electron transport can be guaranteed. The architecture especially works well in dealing with the low intrinsic conductivity of silicon and owns an ability to maintain structural integrity of the material during dynamic loading.

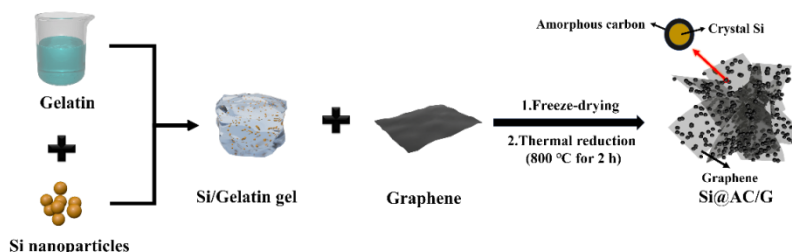
### **2.3 CNTs modified silicon electrodes definition**

Carbon nanotube (CNT) modified silicon electrode is a refined composite system in which one-dimensionally arranged carbon nanotubes are embedded in the silicon structure to create a long and woven electron transmission network. This change, which takes advantage of CNTs being a quasi-one-dimensional structure and their ballistic transport properties, is an efficiency bridge between isolated silicon grains, and the charge transfer resistance of the bulk electrode can be greatly reduced. Mechanically, carbon nanotubes are used to reinforce structures, like steel reinforcement in buildings. They enhance tensile strength while preventing the expansion of electrode films. CNT modification effectively prevents active material delamination in current collectors by providing permeable conductive pathways to keep consistent stability even under repeated volume changes. This structural design ensures low polarization effects and maintains high capacity even under high-load working conditions.

### 3 Analysis and challenge

#### 3.1 Amorphous carbon modified silicon electrode

Overall, the amorphous carbon modified silicon electrode preparation technique employed mainly involved gelatin-assisted pyrolysis technique. For instance, one synthesis of the 10-micron sheets of graphene with integrated Si@Amorphous Carbon (Si@AC) composite was reported to be as follows [11]: 1 g of gelatin was dissolved in 100 ml of deionized water at 60 °C, whereas 1 g of Si nanoparticles (30-150 nm) was dispersed in 50 ml of water by using 30 minutes of ultrasonic. Si dispersion was then added dropwise into the gelatin solution which was stirred under constant temperature of 60 °C till a hydrogel was formed. The mixture was then supplemented with 10 µm graphene that was optimized in terms of its size to produce a aerogel after 48 hours of freeze-drying. The aerogel was also carbonized at 800 °C under nitrogen protection and maintained at 2 hours to form a dual-network structure with the Si nanoparticles which are coated by nitrogen filled amorphous carbon and pegged on the graphene substrates. Fig. 1 presented a schematic view of the synthesis process of composite materials of Si@AC/G [11].



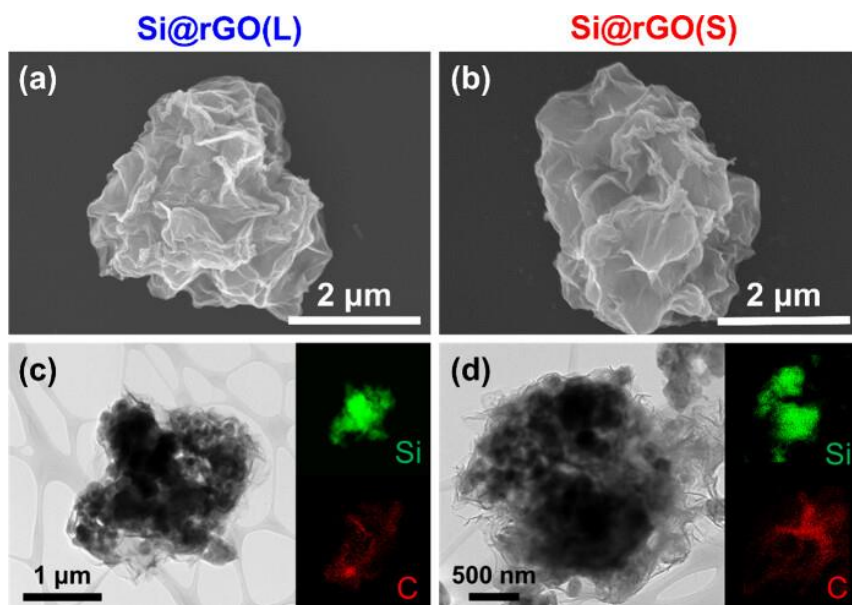
**Fig. 1.** The synthesis process of the Si@AC/G composites is schematically presented [11].

The bulk Silicon Amorphous Carbon composite prepared had excellent performance improvement over the normal silicon based anodes. The Si nanoparticles exhibited steady core-shell with the 10-nm-thick carbon shell coating. As a result of N-doped amorphous carbon and graphene scaffolding being used synergetically, the Si@Amorphous Carbon composite attained a reversible capacity of 1,192 mAh<sup>-1</sup> g<sup>-1</sup> in 300 cycles and owned a high capacity retention of 92.4%. This was absolutely better than the conventional Si-carbon composites which was 432 mAh g<sup>-1</sup> and the bare Si anodes which was about 6 mAh g<sup>-1</sup>. In addition, the large nitrogen content, the C-N to C-C ratio was 1.59, was useful in increasing the electrical conductivity and having more Li-ion storage sites, which were the disordered defects. Good rate performance was also shown when reaching a superior rate ability of 812 mAh g<sup>-1</sup> at 2C [11].

Nevertheless, there are also many inherent challenges and technical flaws. Extreme volume fluctuations during lithiation/delithiation in silicon materials can do harm to the integrity of the amorphous carbon filler encapsulation. This leads to carbon shell cracking so the capacity will decrease. While nitrogen-enriched amorphous carbon enhances conductivity, if the disordered carbon structure undergoes side reactions with the electrolyte, it may cause continuous growth of the solid electrolyte interphase (SEI) and result in low initial coulombic efficiency. In addition, the mechanical elasticity of the amorphous carbon framework may not withstand stress accumulation during repeated charge-discharge cycles. It is believed that only by increasing the thickness and structural stability of the amorphous carbon coating can these shortcomings be solved [11].

### 3.2 rGO modified silicon electrodes

Generally, Si@rGO (Si@reduced Graphene Oxide) can be prepared through the combination of spray drying and thermal reduction. In order to study how the size of the graphene affects it, two kinds of graphene oxide (GO) with various lateral dimensions, namely, large-sized GO (GO(L)) and small-sized GO (GO(S)). One of the synthesis processes was as follows [12]: The preparation process involves mechanically stirring and ultrasonically dispersing 2 g of graphene oxide dispersion (solid content), 8 g of silicon powder, 0.05 g of single-walled carbon nanotubes (SWCNTs), and 0.1 g of polyvinyl alcohol (PVA) in deionized water. Intensive mechanical stirring was used to prepare a homogeneous slurry and ultrasonic treatment was used to transfer the mixture. The result of this solution was then subjected to a spray dryer at a temperature of 110 °C to better convert the droplets of the slurry into solid secondary composite particles. The spray-dried powder was then thermally reduced at 950 °C in argon atmosphere for an hour at a ramp rate of 10 °C min<sup>-1</sup>, which led to the formation of core-shell structured Si@rGO(L) and Si@rGO(S) composite. In Fig. 2, the products of the synthesis of the composite particles were revealed [12].



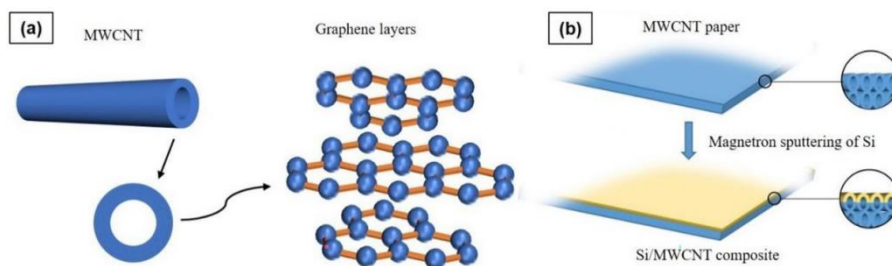
**Fig. 2.** SEM images of (a) Si recombinant composite materials on rGO(L) and (b) Si recombinant composite materials on rGO(S). TEM image and elemental mapping of (c) Si@rGO(L) and (d) Si@rGO(S) composite materials [12].

The resulting prepared Si@rGO composite anodes were of great benefit in terms of the high tap density and high electrochemical capacity. In detail, the tap density of the composite of Si@rGO(S) had 1.02 g<sup>-1</sup> cm<sup>3</sup>, or an increased amount of 175 percent of 0.37 g cm<sup>-3</sup> of submicron silicon. The measured surface area was 20.3 m<sup>2</sup> g<sup>-1</sup> which was quite higher than the 7.01 m<sup>2</sup> g<sup>-1</sup> of pure silicon. When it comes to electrochemical performance, the Si@rGO(S) anode showed outstanding specific capacity of 2586 mAh g<sup>-1</sup> at 0.1C, and the Si@rGO(L) anode recorded 2181 mAh g<sup>-1</sup>. Besides, the initial Coulombic efficiency of the 2 composites was about 93% with cycling efficiency of over 99.5% following the formation cycles. The capacity retention of the Si@rGO(S) anode (150 cycles) was 78% (1,549 mAh g<sup>-1</sup>), much larger than 16% retention of the pure silicon anode. These findings affirmed that the small-sized rGO coating was useful in the stability of the interface and offered ample diffusion routes of the Li-ion [12].

There were also inherent problems in the Si@rGO anodes, including the mechanical strain due to the volume expansion and the size of sheets of rGO on ion transport kinetics. Lithiation caused a volumetric growth of silicon core of more than 300 % which may cause the active particles to crush and the solid electrolyte interface (SEI) to become unsteady. Large-sized Si@rGO(L) also formed more tortuous pathways in analysis which greatly hindered the diffusion of lithium ions in comparison to Si@rGO(S) type. Moreover, the sheet-restacking defects on the rGO surface may influence the structural stability under repeated mechanical strain. In addition, the composite materials should be highly stable to the environment so that the silicon core will not be oxidized. For the next generation of lithium-ion batteries, it is necessary to reduce the size of rGO films in order to optimize both the mechanical buffering and efficient ion transport [12].

### 3.3 CNTs modified silicon electrodes

The modification of carbon nanotube tissues with silicon coatings is a primary strategy to enhance the electrochemical performance of flexible anodes for lithium-ion batteries. Silicon-coated multi-walled carbon nanotube (MWCNT) tissue can be fabricated as follows [13]: Keep the 73- $\mu\text{m}$  thick MWCNT tissue whose areal density is 2 mg cm<sup>-2</sup> under vacuum for over 10 hours to get rid of absorbed air moisture. Put the Si-coated MWCNT (Si/MWCNT) tissue into the magnetron sputtering machine and use the n-type 4-in silicon wafers as the sputtering target for 5, 10, and 20 minutes respectively under argon atmosphere at a constant current of 80 mA to make sure that the oxide layer get removed. This led to Si weight loadings of 0.5, 1, and 2mg cm<sup>-2</sup>. Fig. 3 shows how to make Si-coated MWCNT composite. Later, those produced Si/MWCNT sheets were cut into 0.6 cm $\times$ 0.6 cm squares as the anodes in the electrochemical measurements [13].



**Fig. 3.** Schematic illustrations of:(a) a pristine multi-walled carbon nanotube (MWCNT) structure; and(b) the manufacturing of a silicon (Si)/MWCNT composite from a pristine MWCNT tissue via magnetron sputtering of silicon [13].

The Si/MWCNT composite excelled in capacity and kinetic properties. The sputtered silicon formed a conformal and uniform coating on the tissue surface with an average thickness of approximately 427 nm. This electrode composite improved gravimetric capacity from 109 to 290 mAh g<sup>-1</sup> and achieved 94% of coulombic efficiency after went through the initial ten formation cycles. These improvements largely thank to Si coating that can protect the graphene layers from peeling and directly being exposed to electrolyte. Furthermore, the Si/MWCNT anode possessed a lower charge transfer resistance ( $R_{ct}$ ) of less than 0.1  $\Omega$  compared to the pure MWCNT whose resistance is 1577  $\Omega$ , which enabled faster lithium ion diffusion kinetics [13].

The Si/MWCNT system also presents several challenges and inherent defects that limit its industrial application. The formation of a solid electrolyte interphase (SEI) on the vast surface area of the MWCNTs will cause irreversible capacity loss. Most silicon is on the top surface which is up to 27.7 wt.% and fails to penetrate the inner layers which according to

the reference only 0.1 wt% of Si can penetrate its depth. The bulk of the MWCNT is not utilized adequately. In addition, “dead lithium” is accumulated, which is about 0.54 mg consumed after 100 cycles. To overcome these limitations, it is believed the essential to combine this sputtering method with advanced strategies such as pre-lithiation or artificial SEI layer fabrication. Furthermore, optimizing sputtering parameters to penetrate deeper into the tissue can significantly enhance overall energy density and cycling stability [13].

## 4 Summary & suggestion

Table 1 showed the progress and challenges of different Si-carbon anodes. The amorphous carbon modification, prepared by the pyrolysis in the presence of gelatin, provides a robust dual-network structure and shows much higher cycling stability. The main progress of such material is its high capacity retention and it can be easily doped with nitrogen which can give extra sites of Li-ion storage and improve charge-transfer kinetics. These systems also face the problems of mechanical integrity of carbon shells which could shred them under the pressure of repeated lithiation. Additionally, the natural property incompatibility of silicon and graphene often causes the Si agglomeration and makes ion hard to transport at the basal planes of the graphene. It is proposed that researchers can optimize the size of graphene to promote mechanical durability and diffusion kinetics and researchers should aim to develop a more homogeneous distribution of silicon nanoparticles as well.

**Table 1.** Comparison the advantage and disadvantage for various Si-carbon anodes.

Method	Progress	Challenges
Amorphous Carbon Modified Silicon Electrode	<ul style="list-style-type: none"> <li>- Good capacity retention</li> <li>- Easy for nitrogen doping</li> <li>- Synergistic dual-network architecture</li> </ul>	<ul style="list-style-type: none"> <li>- Gradual pulverization of carbon shells</li> <li>- Phase separation and Si agglomeration</li> <li>- Kinetic bottlenecks</li> </ul>
rGO Modified Silicon Electrodes	<ul style="list-style-type: none"> <li>- High tap density and specific capacity</li> <li>- High initial Coulombic efficiency</li> <li>- Stable interface and diffusion pathways</li> </ul>	<ul style="list-style-type: none"> <li>- Huge volume expansion stress</li> <li>- Tortuous pathways in large sheets</li> <li>- Sheet-restacking derived defects</li> </ul>
CNTs Modified Silicon Electrodes	<ul style="list-style-type: none"> <li>- Ultra-low charge transfer resistance</li> <li>- Protection from peeling and being exposed to electrolyte</li> </ul>	<ul style="list-style-type: none"> <li>- High irreversible capacity loss</li> <li>- Limited Si penetration</li> <li>- Accumulation of "dead lithium"</li> </ul>

Si@rGO composites are produced by spray drying and thermal reduction and they are characterized by much greater tap density and fine specific capacities. RGO of small size offers a stable interface and diffusion pathways of lithium-ions. Nevertheless, the volume expansion of silicon of more than 300 % continues to be an issue to the system itself that causes electrode pulverization and solid electrolyte interphase (SEI) instability. On the other hand, large rGO surfaces can form tortuous pathways that slow down the ion-flux, and sheet-restacking dislocations can destroy the structural integrity of the composites over time. Dimensional restructure of rGO sheets should be developed so that mechanical buffering can be stabilized and fast ion transport can be realized as well. This may also enhance environmental stability to prevent silicon core from being oxidized.

The modification of carbon nanotubes (CNT) indicates an advanced method of fabricating long-range electronic transmission frameworks, mostly achieved by the magnetron sputtering methods. It is such an effective technique that it can minimize the charge transfer resistance and protect the inner graphene layers from peeling and reacting with electrolyte. The sputtering, however, can contribute to a large irreversible loss of capacity during the initial formation of SEI. To overcome the above flaws, it is necessary to add in improved techniques like pre-lithiation, or developing artificial SEI layers. The optimization of sputtering parameters for getting deeper access into the porous tissue might help produce reliable and high-energy density anodes.

To help make the next-generation silicon-carbon anodes more perfect, it is essential to look for a balance between electrochemical performance and industrial viability. Future research should focus on hybrid architectures that combine the high conductivity of carbon nanotubes (CNTs) with the conformal protection of amorphous carbon shells. The research should also spare no effort to avoid the risks of "dead lithium" accumulation and enhance the environmental stability of silicon cores. Only by optimizing these nanostructures and enhancing the stability of the interface between active materials and electrolytes can high-performance energy storage systems be developed to meet the demands of the global electric vehicle market.

## 5 Conclusion

This review has reviewed the importance of nano silicon-carbon composite architectures in the development of anode materials of the future lithium-ion batteries. This study was aimed at tackling the theoretical constraints of conventional graphite anodes. The work also covered the shortcomings of silicon especially its massive growth and low electrical conduction. Evaluations of carbon modification strategies including amorphous carbon encapsulation, reduced graphene oxide (rGO) scaffolding and carbon nanotube (CNT) reinforcement provided three aspects of findings in respect of their structural and electrochemical trade-offs.

Firstly, the Si@AC composite is the basic solution to the volume change buffering. The study shows that the nitrogen-doped amorphous carbon layers are efficient in separating silicon to the electrolyte hence stabilizing solid electrolyte interphase (SEI). But the mechanical elasticity of the carbon shell is a limitation, which may cause the structure to crack and reduce capacity during repeated charge-discharge cycles. This requires an accuracy of shell thickness and homogeneity.

Secondly, the Si@rGO shows an efficient method to increase tap density and specific capacity. A key result of this review is that there is a difference between the large reduced graphene oxide (rGO(L)) sheets and the small ones(rGO(S)). Although reduced graphene oxide is an ideal mechanical buffer, it is necessary to adjust the size of the sheet. It was found that smaller sheets were identified to be the most efficient in the diffusion process. At the same time, when the sheets were larger, they would form tortuous pathways that had negative efforts to the system.

Thirdly, the Si@MWCNT forms a better long-range conductive structure. CNTs serve to stop the expansion of electrode at a microscopic level and help charge to transfer more easily. But this is limited by the low silicon diffusion through the tissue layers at the interior and also the massive loss of irreversible capacity when it undergoes the first formation cycles.

In conclusion, although each of the carbon modification strategies has their unique benefits, none of them can address all the electrochemical bottlenecks of silicon anodes at the moment. To achieve high-performance silicon anodes, it should explore multi-dimensional hybrid designs to combine the conformal protection of amorphous carbon and the high conductivity and mechanical toughness of CNTs or graphene. In addition, the adoption of the latest production methods, including pre-lithiation, and artificial SEI

engineering should be used to avoid the build up of the dead lithium and gain the industrial viability required of the electric vehicle market.

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