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Recent Progress in the Structural Design of Silicon/Carbon Composite Anodes for Lithium-Ion Batteries

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Abstract. Silicon is considered to be a promising anode material for the next generation of lithium-ion batteries due to its high theoretical specific capacity. However, it will produce significant volume changes during the lithium insertion and delithiation processes, which further cause problems such as particle cracking, repeated rupture and formation of the solid electrolyte interphase (SEI), and loss of electrical contact. Silicon/carbon (Si/C) composite materials provide an effective way to alleviate the above problems by combining the high capacity of silicon with the good electrical conductivity, mechanical support capability and interface stabilisation of carbon materials. This article reviews the structural design of Si/C composite anodes, focusing on three typical structures (carbon-coated, embedded and hollow/porous), and compares their roles and limitations in interface protection, electron transmission, mechanical buffering and volume expansion control. Further analysis revealed that there are trade-offs between different structures in terms of performance, energy density, preparation complexity and scaled application. The future development of Si/C anodes requires further optimisation of the Si/C ratio, interface bonding, pore structure and multiscale structural design, while taking into account high performance, low cost and scalable manufacturing.

Keywords: Silicon/carbon composites, lithium-ion batteries, anode materials, structural design, interfacial engineering

1. Introduction

In the current context of battery material research, the lithium-ion battery is the subject of much attention, because of its high energy density, and is at the forefront of the developmental efforts [1]. The most used anode, the conventional graphite, is characterised by very good stability and is supported by a well-developed production in the commercial frame, but its usefulness is limited by a relatively low specific capacity of 372 mAh g^{-1} [2, 3]. On the other hand, silicon undergoes large volumetric changes when it lithiates, as it forms the Li-Si alloys, with an expansion of up to 300% in the fully lithiated phase. Cyclic swelling and shrinking induce a large mechanical stress in a single silicon particle and in the general electrode structure, which leads to the particle fracture. The exposure of the fresh silicon surface to the electrolyte triggers the reformation of the solid electrolyte interphase (SEI), which leads to the continuous depletion of active lithium ions and electrolyte components.

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At the same time, the electrical connectivity among the particles, and between the particles and the current collector, can also progressively deteriorate. The sum of all the above mechanical degradations and interfacial instabilities results in a fast decrease in the capacity of the silicon (Si) electrode [4]. Even though the so-called silicon anode materials have a very high theoretical capacity, and thus are main candidates for next-generation anodes, pure silicon is far from being a good choice. It has, in fact, several problems during the processes of lithiation and delithiation, like a very important volume expansion, fracture of the particles and development of an unstable solid electrolyte interphase (SEI) [2, 3]. In this context, Si/C composites have become an important strategy in current studies. By taking advantage of the good properties of carbon, like its electrical conductivity, its ability to buffer the mechanical stress and its role in stabilising the interfaces, the Si/C composites avoid many of the drawbacks of the conventional Si anodes. It is to be remarked that the carbon phase in the Si/C composites is not made of one homogeneous material.

Typical carbonaceous materials are graphite, amorphous carbon, porous carbon variants, graphene, carbon nanotubes (CNTs) and carbon nanofibres. Each of these forms plays different roles: for instance, graphite is similar to the substances used in the commercial anode configurations; amorphous or porous carbon may act as a protective coating or a structuring scaffold; while graphene and CNTs allow one to build up very conductive two-dimensional or one-dimensional networks, which enhance both electron transport and mechanical support. Thus, the efficiency of the silicon/carbon composites is not only given by the presence of carbon, but also by the variety of carbon selected and its distribution in the composite [5]. Concerning the structural design, the main goals associated with different Si/C architectures are as follows. Structures with carbon coatings aim to improve the surface protection, the electronic conductivity and the interfacial stability.

A continuous carbon framework is the basis of the embedded architectures, providing at the same time the structural support and the conduction of the electricity. For the hollow or porous structures, the only support is the void inside of the design, which is used to buffer the volumetric expansion with respect to the silicon. It is important to notice that these are not sharply different classes.

Recently, more and more scholars have chosen to use multiscale and hierarchical architectures to combine the different functional parts, such as protective coating, conductive pathways, porous frameworks and interface modification, in a single silicon-carbon (Si/C) composite system [2, 6]. The present review focuses on Si/C composites and focuses on the strategy of structural configurations (i.e. carbon-coated, embedded and hollow morphologies), as well as on recent progress and the future challenges for their practical use [2]. As shown in Figure 1, the architectural designs for Si/C composite anodes are generally categorised into three different categories, i.e. carbon-coated, embedded and hollow structures. For each of these structural variants, we can find how they improve the anode performance: surface coating is used to stabilise the interface, a continuous carbon matrix provides both mechanical strength and electrical conductivity, while the internal voids provide the amount of buffer to reduce the silicon's volumetric expansion. These three structural classifications are the basis for the detailed analysis that we will do in the following sections of this paper.

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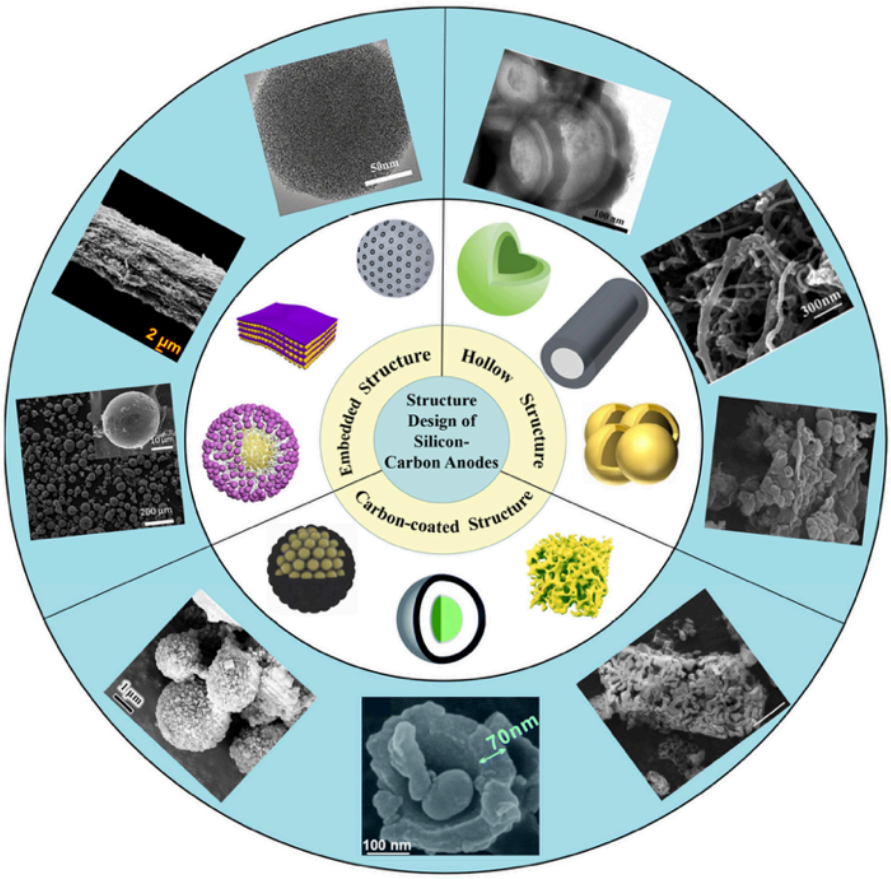


Figure 1. Schematic illustration of the three major structural design strategies for silicon/carbon composite anodes, i.e. carbon-coated, embedded and hollow structures. Figure is reproduced from Mei et al. [2] with CC BY 4.0 licence

2. Carbon-coated structure

For the case of the lithium-ion batteries whose anode material is silicon (Si), the architecture with the carbon coating is simply characterised by a silicon core completely covered by a layer of carbon.

This layer acts both as an electrical passage and as a shield for the inner silicon nucleus. In fact, silicon has intrinsically low electrical conductivity and can be subject to a strong volumetric swelling which may entail a risk of particle fragmentation. Such a mechanical degradation can break electrical connectivity and promote the permanent breaking and reconstruction of the solid electrolyte interphase (SEI) [2, 3]. With a carbon-coated architecture, a stable conductive network is kept during the lithiation and delithiation processes, and it can strengthen the mechanical robustness of the anode material [2, 7]. Moreover, this also strongly reduces the direct interaction between the silicon particles and the electrolyte, which prevents the exposure of fresh silicon surfaces and limits the cyclic failure and repair of the SEI [2, 7]. However, the efficiency of this approach depends on the exact structural configuration, in particular whether the internal silicon core touches the outer shell or is isolated by a void space [2].

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2.1. Core-shell structure

In the architecture of a core-shell design, the outer carbon layer is always in immediate physical contact with the inner Si part. The carbonaceous coating plays two roles: it coats the encapsulated Si core as a protection and, at the same time, it establishes an electrically conductive skeleton [4].

Nevertheless, such immediate physical interaction brings some mechanical limitations to the silicon component. Although the core-shell architecture is characterised by its simplicity and has considerable commercial viability, which gives us an uncomplicated strategy for improving the silicon-based anode, it is not without considerable disadvantages [2]. Specifically, with such a direct interface, the volumetric expansion of the silicon anode during the lithiation process can apply a direct force to the carbon coating. This means that the undue stress will accumulate and finally lead to the structural fracture of the shell [2, 7]. For example, in 2024, Fang and colleagues reported the production of the silicon nanowire-carbon core-shell anode by a single-step atmospheric-pressure chemical vapour deposition technique. In this configuration, the silicon nanowires are the inner active species, while the external carbon layer gives both protective benefits and the channels of the electronic transport. It should be noted that this research did not need the extra metal catalysts which are usually needed for nanowire synthesis, which makes it a comparatively simple manufacturing procedure with the scale-up prospect. The resulting electrode showed a reversible specific capacity of about 745.8 mAh g^{-1} and maintained approximately 91.3% of its initial capacity following 100 cycles at a current density of 0.5 A g^{-1}, which shows that an appropriately engineered carbon coating could improve at the same time the electrical conductivity and the cycling durability [8]. In order to reduce the mechanical stress induced by the direct contact during the expansion of the volume, new structural designs have been proposed to better bear the cracking problems.

2.2. Yolk-shell structure

The yolk-shell architecture [9, 10] is able to avoid the shell fracture induced by the volumetric expansion of the silicon nucleus by introducing an empty space between the silicon nucleus and the carbonaceous coating. It can relieve mechanical strain on the outer layer, keep the structure of the carbon shell and improve the stability of the external solid electrolyte interphase (SEI) [9, 10]. An example of recent progress is the F-Si@Void@C yolk-shell composite reported by Liu et al. (2024). In this structure, there is an internal void between the silicon core and the carbon shell, which provides enough room for the silicon to expand during the lithiation, and thus decreases the mechanical stress.

At the same time, the external carbon coating also plays a role in reducing the direct interaction between silicon and the electrolyte, while allowing the electron conduction. In this research, the fluorine-ion interface engineering was also used to increase the concentration of LiF in the formed solid electrolyte interphase (SEI) to enhance the stability at the interface. Thus, the composite still has a reversible specific capacity of about 1166 mAh g^{-1} after 900 charge-discharge cycles at a current density of 0.5 A g^{-1} [11]. The evolution of the internal void structure is better shown in Figure 2. For instance, as shown in Figure 2(a), Zhou and co-workers used the polystyrene (PS) to form the sacrificial template around the silicon particles, and then a polyaniline (PANI) layer was deposited on them.

After the carbonisation step, the PS part is removed, and the PANI is transformed into the nitrogen-doped carbon network, and then the Si@void@NCN architecture is obtained [12]. In the TEM micrograph given in Figure 2(b), we can clearly see this void space between the middle Si core and the outside carbon shell. This inner cavity is a buffer zone to accommodate the big volume

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expansion of the silicon in the lithiation process. However, this structure design has some disadvantages. Firstly, the fabrication procedure is very complicated and usually uses the sacrificial template with the selective etching technique [2, 9]; that is to say, it increases the total manufacturing complexity. Secondly, if the dimension of the void is not regulated very accurately, the volumetric energy density may be adversely affected [6]. Summing up, these difficulties are obstacles to the scalable production of such materials [2, 3].

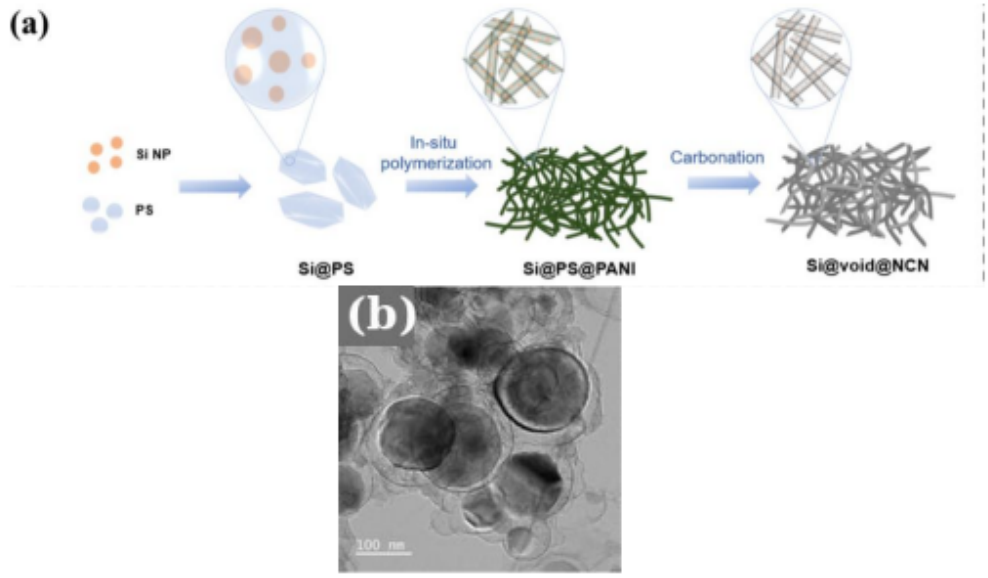


Figure 2. Representative yolk-shell Si/C architecture. (a) Schematic diagram of the synthesis of Si@void@NCN using a sacrificial polystyrene template followed by in situ polymerisation and carbonisation; (b) Transmission electron microscope image showing the internal cavity between the silicon core and the surrounding carbon network. Reprinted from Zhou et al. [12] with permission from Elsevier, under the CC BY 4.0 licence agreement

2.3. Comparison

In general, the core-shell configurations are engineered to maintain the electrical conductivity and to protect the surface of the core material. However, the problems related to the volume expansion and mechanical stress are still unavoidable from the point of view both of theory and practice [2, 7]. Although the yolk-shell architectures could be a way to solve these problems, they need more complicated design rules and more expensive fabrication. Also, good control of the void sizes is needed to make sure that the energy density is not lost [5, 6].

3. Embedded structure

The "embedded architecture" looks like a very attractive strategy within the world of silicon/carbon composite engineering. In different configurations, it comes as the insertion of silicon in amorphous carbon, in porous carbon scaffolds, in carbon nanotube (CNT) frameworks or in three-dimensional networks; in all cases, one puts the active material in a carbonaceous host [2, 13, 14]. If we think about the bad points of the silicon, that is to say, the important volumetric expansion, the particle fracture, the disconnection from the electrical way and the poor conductivity, the underlying idea of this design is to protect the silicon from having to bear on its own the mechanical stresses. If one puts the silicon in an unbroken carbon matrix or network, the host is able to provide a sustained

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structural reinforcement. This is the structural feature which distinguishes the embedded structure from the usual carbon-coated structure. The working of the embedded structure is complex and multi-layered. About the conductive network, the unbroken electron transport channel given by the carbon matrix assures that the electrical connectivity remains, and the structural stability is kept even in the case of fracture of silicon [13, 14].

From the mechanical point of view, the carbon matrix acts as a supporting scaffold which takes the stress induced by the volumetric expansion of the silicon, and thus keeps the whole electrode intact [13, 14]. Moreover, this archetypal architecture also ensures a sustained electrical contact; when the silicon particles are embedded into the carbon network, their displacement is limited, which means that this reduces the risks of particle detachment and of electrical isolation [13, 14]. These basic mechanisms give the silicon-carbon composites interesting benefits for the application in lithium-ion batteries. In 2024, Zhou and co-workers embedded the nano-silicon in a three-dimensional honeycomb-structured carbon. The channels of the honeycomb structure accommodate the volume expansion of the silicon, and thus protect the structural coherence of the carbon skeleton; at the same time, they provide shorter paths for the diffusion of the lithium ions.

At the same time, the unbroken three-dimensional carbon framework ensures reliable electrical connectivity. After 1400 charge-discharge cycles at a current density of 2 A g^{-1}, the refined electrode kept a specific capacity of about 1294.3 mAh g^{-1}, which is a retention of about 85.5%. These results are in line with the general idea of the embedded architectures, where the carbon host plays several very important roles: it provides a mechanical reinforcement, it helps efficient ion diffusion and it guarantees electrical percolation [15]. In particular, the unbroken conductive network increases the electronic conductivity, the mechanical cushioning effect improves the structure, and the confinement of the active particles limits their displacement, so it is possible to have a more general matrix engineering. However, this configuration has some drawbacks. If the proportion of carbon is too high, it will decrease the total specific capacity [2, 6]. Moreover, such an excess can deteriorate the capability of the matrix to withstand the very large volume expansion of the silicon. Moreover, even if the structure is constrained, one cannot be sure that during long cycling the silicon particles remain adhered to the matrix.

The heterogeneity of the dispersion of silicon in the matrix is another factor which influences the stress concentration. It is well known that the embedded configurations are highly effective in guaranteeing both the mechanical integrity and the electrical continuity. However, their final efficiency depends on several parameters, such as the level of silicon loading, the nature of the Si-C bonds and the homogeneity of the dispersion of silicon [2, 6]. It should be noticed that, in real applications, the strategies of embedding, carbon coating and the construction of the conductive networks are never done separately. For instance, in the contemporary multidimensional carbon framework design, one can find in general several approaches integrated: a carbon matrix is used to anchor the silicon particles in a uniform way, a carbon nanotube (CNT) network allows efficient long-distance electron transport, and an outer carbon layer controls the interface between the silicon and the electrolyte. These integrated approaches show that the future track of Si/C structural engineering will probably be more the combination of various functional structures than the reliance on any single architectural design [16]. Figure 3 is more intuitive.

In this section, we explain the idea of multi-structural synergistic design. First, silicon is embedded into a carbon matrix; then, an outer carbon layer is applied and a kind of conductive network composed of carbon nanotubes (CNTs) is added, and finally these parts are merged. The function of the outside carbon shell is to improve the interface between the silicon and electrolyte. And the CNT framework is able to conduct the electrons over long distances and provide more

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mechanical stability. So, the strategies of carbon coating and embedded or networked structures are not conflicting, but they work in a hierarchical configuration to complement each other.

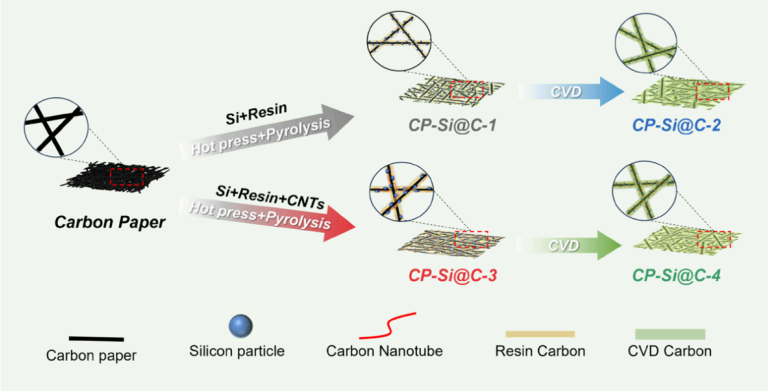


Figure 3. Schematic illustration of multidimensional carbon-framework designs combining a carbon matrix, CNT conductive network and conformal carbon coating for Si-containing composite anodes. Reprinted from Guo et al. [16] with permission from CC BY 4.0 licence

4. Hollow and porous Si/C structures

An often-used way to improve anode material of the lithium-ion battery is the engineering of the silicon/carbon (Si/C) composites with the hollow and porous architectures. Contrary to the configurations in which the carbon only coats or embeds the silicon, in this methodology, the Si/C mixture is considered as a whole, and one introduces either big internal voids or a dispersion of finer pores. In this sense, the former represents a single large cavity, whereas the latter is a network of small channels. These structural features are designed to buffer the volumetric swelling that is characteristic of the silicon [14, 17]. In the face of problems like severe volume expansion, the stress build-up and the particle fracture, the idea is that it is more efficient to give enough room for the expansion than to try to strictly restrict it. Therefore, the capacity to accommodate the volume changes is a key mechanism within this design.

The presence of voids or porous structures gives us the space to accommodate the volume expansion in the process of the lithiation [14, 17]. In this way, it can alleviate the direct mechanical strain in the adjacent carbon matrix and it also prevents the stress from accumulating in some regions. The other mechanism is the redistribution of the stress, i.e. it spreads the forces that come from the expansion over a wider domain of the structure. In this way, the risk of cracking in some local regions and the pulverisation of the particle are reduced [14, 17]. Finally, the transport of the ions is higher.

The presence of porous channels is meant to limit the diffusion path length for the lithium ion (Li^+) and thus allows the electrolyte to go deeper and speeds up the reaction kinetics [14, 17]. The advantages of such a kind of architecture are obvious: it reduces the volume fluctuations that are typical of the silicon, it does not favour the formation of fracture and it strengthens the total structure. Moreover, the reduction of the distance to transport the ions allows an enhancement of the rate capability and of the cycling durability. Yu et al. (2025) have performed a direct study of the relation between the pore evolution and the mechanical robustness in porous Si/C composites. They have shown that to maximise the porosity is not always good: while a moderately developed pore network is able to accommodate well the silicon expansion and to keep the electrode stable, if the degree of porosity is too high, the mechanical strength of the carbon matrix is severely damaged and

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the electrode disintegrates, and the capacity is lost very fast. After 500 cycles at 1 C, the material shows a capacity retention of 74.4%.

These results show that the crucial point of developing hollow or porous architectures is not simply to have the biggest porosity, but to have an optimum balance between available free volume and mechanical robustness [18]. However, such designs have several difficulties. If the porosity level is too high, the tap density will be diminished and the total volumetric energy density will be reduced [10, 17]. Besides, the expanded surface allows a greater direct interaction with the electrolyte, which favours the formation of thick SEI layers and induces several parasitic side reactions. It is also clear that too many pores are going to break the mechanical integrity of the carbon skeleton. Moreover, the structural engineering is complicated; as for embedded configurations, the dependence on templates, etching procedures and precise pore regulation makes the fabrication complex [2, 17]. In all, this category of structure mainly compensates for the volume expansion by creating internal free space, which also improves the ion transport capability.

However, in order to ascertain whether the ultimate result fulfils the necessary requirements, one needs to find an optimal equilibrium between porosity, structural rigidity, specific surface area and volumetric energy density [6, 17].

5. Conclusion

As we have described in this overview, even though the silicon (Si) has quite a lot of promise as an anode material for lithium-ion batteries, its practical use has been, and still is, hindered by some crucial limitations: large volumetric expansion, structural fracture and pulverisation, the formation of an unstable solid electrolyte interphase (SEI), disconnection from the electrical network and intrinsically low conductivity. In order to compensate for these deficiencies, carbon is often used because of its good electrical conductivity, the possibility to give a mechanical reinforcement and the possibility to stabilise the interfaces. In this paper, we stress three particular structural configurations, each of which is dedicated to facing some particular problems. The carbon-coated architectures concern mostly the improvement of the surface protection and of the electrical conduction; moreover, in case of the direct coating, one could generate a stress in the shell layer. The embedded structures are designed to improve the conductive paths and to give a mechanical support, but they need a precise balance among different parameters, such as the silicon loading level, the nature of the Si-C bonding and the distribution of the material.

Hollow or porous configurations are well suited for the volume expansion and the stress relief. However, too many pores can be detrimental to the material density and its practical use. In this review, altogether, it seems that no structural design is able to solve all these problems at once. Therefore, future progress is probably expected to be made on improving the silicon-to-carbon ratio, on improving the interfacial adhesion between the silicon and carbon, and on better control of the coating thickness or pore dimensions. However, future Si/C architectures should consider not only gravimetric capacity but also the initial Coulombic efficiency (ICE), the areal capacity, the tap and the volumetric densities, the electrode swelling behaviour, the cycling durability and the possibility of manufacturing at scale. Finally, nowadays the composite engineering is going from basic structures to complex multiscale and multifunctional structures. The aim is to realise a design that offers better performance, is manufacturable and not too expensive, and can be used in large-scale commercial applications.

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