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Challenges in Achieving High Energy Density in Lithium-Ion Batteries: Anode Materials and Silicon Modification Strategies

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Abstract. Due to the development of electric cars, portable electronic devices, and energy storage, lithium-ion batteries (LIBs) have experienced an increase in energy density demand. However, restrictions associated with the design of classical electrodes hinder the improvement of batteries' characteristics. The present review article examines three types of anode materials, namely, graphite, graphene, and silicon, as well as their distinct features. First, despite low cost and reliable cycling ability, the capacity of graphite is limited by 372 mAh g^{-1}. Second, high conductivity and lithium storage capacities are provided by graphene, but there is a possibility of aggregation of its sheets that can cause instability. Last but not least, silicon attracts special attention owing to its potential capacity of 3579 mAh g^{-1}. But the constant expansion of silicon makes it harmful for the structure of the electrode and solid electrolyte interphase. In order to resolve these issues, three modification approaches, i.e., structural engineering, composite with carbon materials, and modification of the interface, are proposed. Experimental data support positive outcomes related to the use of such modifications and improvement of the electrode capacity and performance. At last, the drawbacks of silicon-containing anodes, namely production difficulties, electrode swelling, and gap between the experiment and industrial practice, are discussed.

Keywords: Lithium-ion batteries, silicon anodes, energy density, silicon–carbon composites, interface engineering

1. Introduction

Lithium-ion battery (LIB) cells find application in portable devices, vehicles, and stationary electricity storage due to their high energy density and good cycle life. Pursuit of improved performance at longer charge intervals led to making energy density a key topic of study. High energy density, however, may be achieved only by improving electrode materials and ensuring the cycle life, safety and economic feasibility.

An anode plays an important role in the performance characteristics of a lithium-ion cell. Graphite is one of the most popular anode materials for LIBs due to its inexpensive nature, structural stability and capability of reversible lithium intercalation. Graphite anode material has the theoretical upper capacity limit equal to 372 mAh g^{-1} [1]. Various carbon-based materials have been

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proposed for LIB anodes, including graphene. Such characteristics as high conductivity, high mechanical stability and large surface area may help to improve the electron transfer in electrodes as well as increase the amount of active sites for storing lithium atoms. However, graphene sheets tend to stack together while forming an electrode, decreasing the accessibility of active centers and limiting practical electrochemical performance of the electrode [2]. Different methods of increasing porosity of graphene and graphene composites have been developed for improving lithium storage properties [3].

Silicon is known to have the theoretical specific capacity value much higher than graphite anode: it equals 3579 mAh g^{-1}. However, during lithiation process silicon shows great volume expansion that might lead to particles breaking and damaging the protective layer [4]. Therefore, the increase in energy density of batteries is not only associated with choosing a material with a greater theoretical capacity but also with ensuring structural stability and electrochemical reversibility of the chosen material. This review will provide a comparison of the properties and drawbacks of graphite, graphene, and silicon anodes. Then it will discuss three approaches to modification of silicon-based anodes – structural modification, designing of silicon-carbon composites and interface engineering. The last section will address the open issues and future trends.

2. Conventional lithium-ion battery anode materials

2.1. Graphite

Graphite remains one of the most commonly used anode materials. Being cheap, toxicologically safe, abundant, and having a rather small electrochemical potential, it becomes applicable in all types of equipment, from personal computers to electric vehicles and battery storage systems. Upon charging, lithium ions intercalate between layers of graphite, thereby creating lithium–graphite intercalation compounds. The mechanism of intercalation is more or less reversible, thus providing good electrochemical performance of graphite electrodes [1].

A key limitation of graphite is its theoretical specific capacity of only 372 mAh g^{-1}. Although graphite is stable and inexpensive, its limited capacity restricts further gains in LIB energy density. Therefore, enhancing the capacity of the anode has become one of the key research areas for next-generation batteries. Recent research on graphite has also focused on interpreting its electrochemical and electronic behavior during lithiation, which may help improve the design of future anode materials [1]. In addition, graphite undergoes a comparatively small volume change during lithiation, which helps explain its relatively stable cycling behavior compared with silicon [5].

2.2. Graphene

There are certain features of the graphene that make it an attractive candidate for an anode material: it possesses a high value of electrical conductivity, great mechanical strength, and large surface area. Thus, due to the specific structure of graphene, the material allows the formation of fast electronic pathways and effective ions intercalation. Therefore, one may say that the utilization of graphene allows achieving advantages in comparison with the conventional graphite structure from the point of view of electrical conductivity and rate performance [2].

According to their theoretical lithiation composition, one may state that the theoretical specific capacity of graphene reaches 744 mAh g^{-1} for Li_2C_6 lithiation which is almost twice as high as that of the graphite 372 mAh g^{-1} for LiC_6 [6]. Nevertheless, there are some obstacles in replacing graphite by graphene. Graphene sheets are likely to agglomerate during electrode synthesis that

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leads to a decrease in surface area available for lithium ions incorporation. In addition, closely-packed sheets allow formation of relatively long pathways of lithium-ion transport which negatively affects the electrochemical performance. In one comparative study, graphene sheets delivered reversible capacities of 1130 mAh g^{-1} at 0.2 mA cm^{-2} and 636 mAh g^{-1} at 1 mA cm^{-2}, although capacity fading and lower first-cycle Coulombic efficiency were also observed [7].

For this reason, researchers pay attention to the creation of the porous graphene, graphene with heteroatom doping, or graphene composites. These approaches help to provide the increase in accessible surface area, creation of new active sites, and improvement of ion transport. Hence, graphene is valuable for creating the composite structures as the conductive material rather than the replacement of graphite anode material. According to figure 1 below, one may note the following theoretical specific capacities for graphite, graphene, and silicon: 372, 744, 3579 mAh g^{-1}, correspondingly.

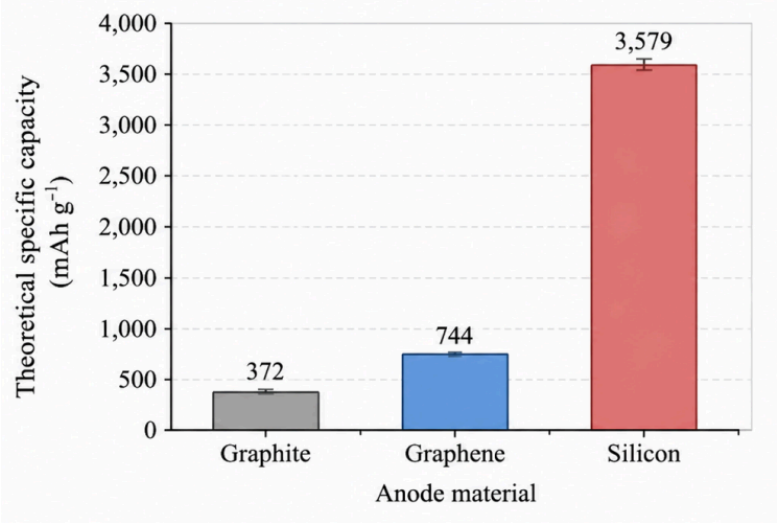


Figure 1. Comparison of the theoretical specific capacities of graphite, graphene and silicon as anode materials for lithium-ion batteries

2.3. Silicon

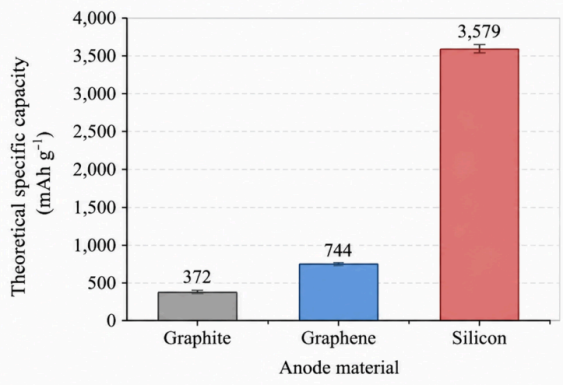


Figure 1. Theoretical specific capacities of graphite, graphene and silicon anode materials.



Figure 2. Schematic illustration of the volume change of graphite and silicon during lithiation.

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Silicon has been studied as an alternative to graphite because it can store substantially more lithium. At 3579 mAh g^{-1}, the theoretical specific capacity of silicon is about 9.6 times that of graphite (372 mAh g^{-1}) [4, 5]. Silicon is also abundant and operates at a low lithiation potential [4]. Despite this potential, silicon has an important limitation: its volume changes substantially during lithiation and delithiation. Repeated expansion and contraction can generate stress that fractures particles and interrupts electrical connections within the electrode. Some silicon may therefore no longer contribute to electrochemical storage. Silicon can experience around 280% volume expansion with lithiation compared to graphite, which shows less volume expansion [5]. As depicted in Figure 2, silicon suffers more volume expansion during lithiation than graphite.

3. Modification strategies for silicon anodes

3.1. Structural modification

Structural engineering seeks to manage silicon expansion and contraction by introducing internal space within mechanically stable designs. Researchers have explored several designs, including silicon nanoparticles, porous structures, nanowires, core-shell particles and yolk-shell particles. Out of all of these structures, a yolk-shell architecture is distinguished by the separation of the core of silicon and outer conductive shell using an internal void. This gap allows the silicon core to expand, while the outer shell helps preserve electrical contact and reduces exposure of fresh silicon surfaces to the electrolyte [8].

Experimental studies from the year 2023 back up this idea. The specific capacity of the Si@HNC composite material having 54.4 wt% silicon was 865 mAh g^{-1} after 300 cycles of usage at 1.0 A g^{-1} current density, keeping 98% of its capacity. However, when the loading of material per unit area was increased to 3.2 mg cm^{-2}, the specific energy storage capacity of the Si@HNC composite was 1013 mAh g^{-1} after 400 cycles of usage at 0.5 A g^{-1} current density [8].

While there are many benefits associated with nanostructure of silicon, nanostructure does not make it fit for the market automatically. Nano structuring may cause increased irreversibility because of high surface area, and complex synthesis may lower the tap density.

3.2. Silicon–carbon composite modification

Combining silicon with graphite or other carbon materials is a practical option because the materials offer different advantages. While silicon is used for increasing lithium storage capability, graphite and carbon coatings are used for their conductive and mechanical qualities and increased packing density [4, 9].

Yan et al. developed a silicon–graphite composite containing 31.6 wt% silicon. Its reversible capacity is 1080 mAh g^{-1} with the initial Coulombic efficiency of 86.3% and retention of 94.3% of the capacity even after 100 cycles. Moreover, it has been used in pouch-type 1.5 Ah full cells with an electrode density of 1.6 g cm^{-3} and areal capacity of 3.46 mAh cm^{-2} [9].

Silicon content presents a trade-off: adding more silicon may raise capacity, but may also worsen swelling, lithium loss and electrode damage. Therefore, the objective of Si/C composite design is not simply to maximise silicon content. A useful composite must balance capacity, initial Coulombic efficiency, cycling stability, electrode density and manufacturability.

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3.3. Interface and SEI modification

Controlling the SEI is another important aspect of improving silicon anodes. In conventional silicon electrodes, large changes in particle size can repeatedly crack the SEI and expose fresh silicon to the electrolyte. The newly exposed surface can react with the electrolyte again, consuming lithium and electrolyte and contributing to resistance growth and capacity loss. Thus, interface engineering can be used for formation of a thin and uniform SEI [4, 10].

In a 2024 study, a fluorinated binder was applied to form an SEI layer that contains lithium fluoride ion at the surface of silicon anode in lean additive electrolyte. The capacity retention of this modified anode is up to 93.7% after 400 cycles at the rate of 0.9 A g^{-1}. The full cell has shown the areal capacity of 4.23 mAh cm^{-2} with 93.4% capacity retention in 200 cycles [10].

Modification of the interface does not eliminate entirely the inherent volumetric increase that silicon undergoes. This is why SEI modification is most useful when used in conjunction with structural and silicon-carbon composite engineering. These approaches address different parts of the same problem: structural designs accommodate expansion, carbon supports conduction, and interface changes reduce repeated electrolyte reactions.

4. Outlook and future perspectives

Despite progress in silicon-anode research, several challenges continue to restrict their use in high-energy-density lithium-ion batteries. This is why future work must not only target high specific capacity but also long-term stability and manufacturability.

One continuing problem is how to control silicon volume changes during charging and discharging. Porous and yolk-shell designs provide empty space that silicon can occupy as it expands during cycling. However, such materials can be difficult to synthesize and possess low packing density. Hence, high specific capacity of the material does not guarantee higher energy density of the whole battery system.

Also significant is the optimisation of Si-C based composites where carbon-based compounds may help in providing better conductivity properties as well as maintaining the structural stability of the electrode, and adding silicon is expected to provide extra lithium ions capacity. At the same time, increasing the content of silicon leads to an additional swelling effect and higher losses of lithium. Thus, the further development is expected to include the finding of the optimal proportion of silicon concentration versus its capacity and density [9].

The stability of the interface is one more problem that should be addressed. Repeated silicon expansion can damage the SEI layer, allowing further electrolyte reactions and lithium loss. Although the preliminary results obtained from the implementation of the modified binders and interface design were positive, further work needs to be done on the effectiveness of these approaches.

Ultimately, future investigations ought to focus on how these strategies can be used in combination, that is, structural modification, silicon-carbon composites and interface engineering. Since each of these techniques deals with a particular problem related to silicon degradation, their effectiveness can be enhanced through using them in combination. Nevertheless, the efficiency of the combination of these techniques has to be assessed in view of the issues of cost-effectiveness, scalability, safety and energy density.

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5. Conclusion

Increasing the energy density of lithium-ion batteries is a key goal for future energy storage technology design. This review focuses on not only traditional but novel anode materials like graphite, graphene, and silicon, as well as different approaches to increasing the electrochemical characteristics of silicon-based anodes.

Graphite is currently used commercially not only because of its comparatively low price and ability to enable reversible lithium intercalation, but due to its limitations that set a barrier to further increase the battery energy density: the theory specifies its maximum specific capacity as 372 mAh g^{-1}. The conductive and tough material with high surface area like graphene is able to improve electronic conductivity and offer extra storage sites. On the other hand, its aggregation and restacking can reduce its usefulness, which is why graphene works particularly well as a part of composites.Among these materials, silicon has the highest theoretical specific capacity, approximately 3579 mAh g^{-1}, making it a promising option for next-generation high-energy-density batteries. However, silicon expansion during lithiation can damage the electrode, break electronic connections and cause repeated SEI formation. Therefore, it hampers its cycling stability and prevents the utilization of theoretical capacity of silicon in practice.

Three approaches have been investigated to address these problems. First of all, structural modification can ensure a place for volume change and mechanical damage minimization. Secondly, silicon-carbon composite combines high lithium storage capacity of silicon and conductivity of carbon-based materials. Finally, interface engineering tries to stabilize SEI and minimize electrolyte decomposition. Experimental results of the studies presented in this review confirm their effectiveness.

Nevertheless, the improvement of the material properties alone will not be enough for ensuring high energy density in batteries. It should also take into account other factors, such as electrode loading, manufacturing problems, loss of lithium irreversibly, and cycling stability. Therefore, the best solution would be the implementation of all above-mentioned methods together.

Overall, silicon anodes have clear potential to raise Li-ion battery energy density, but practical designs must balance capacity with stability, cost and ease of production. Future work should examine how these trade-offs affect full-cell performance.

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