Silicon
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REVIEW

Towards Sustainable Lithium-Ion Batteries: A State-of-the-Art Review on Silicon Anodes, Economics, and Recycling

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Abstract

In recent years, natural graphite, widely used as anode material for commercial lithium-ion batteries (LIBs), has been classified by EU as a Strategic Raw Material (SRM) and its partial or total replacement with non-critical or end-of-life (EoL) material is recommended. Silicon is one of the most promising materials to replace natural graphite. In fact, it is known that silicon can form alloys with lithium, with a theoretical energy storage capacity at room temperature of 3579 mA h g^{-1} (when lithiated to $Li_{15}Si_4$), i.e., significantly higher than that of graphite (372 mA h g^{-1}) and comparable with metallic lithium (3860 mA h g^{-1}). The limiting factor of silicon as anodic material for lithium-ion battery systems is represented by its volumetric expansion, which can reach values more than 300% during battery charge–discharge cycles, involving progressive fragmentation and loss of active material and resulting in rapid decrease of the accumulated capacity. Moreover, the recent inclusion of silicon into the EU list of strategic raw materials and the high environmental impact of silicon production from SiO_2, make the recovery and recycling of this material thoroughly recommended, especially from EoL products like photovoltaic (PV) panels. The paper offers a state-of-the-art on challenges and solutions related to the use of silicon as anodic material in LIBs, besides a survey on the Technology Readiness Level (TRL) and the market penetration of silicon anode battery technology.

Keywords Lithium-ion batteries · Silicon anode · Critical raw materials · Solar panel waste · Sustainability

1 Introduction

In recent years, natural graphite has been widely used for the anode of LIBs (see Appendix A). However, graphite has recently been classified by European Community as a SRM (see Appendix B) and, therefore, its partial or total replacement with non-critical or EoL material is recommended [49, 76]. Several studies in literature have reported the possibility of making the anode for LIBs using silicon-based materials, in particular with nanometric grain size, in order to completely or partially replace graphite [37, 54, 55]. It is known that silicon can form alloys with lithium [32], with a theoretical specific capacity of nearly 4200 mA h g^{-1}, which corresponds to a fully lithiated state of $Li_{22}Si_5$ (4.4 Li per Si) [29]. This represents a significant improvement over graphite, which has a specific capacity of only 372 mA h g^{-1} (LiC_6) [29] and even over metallic lithium (3,860 mA h g^{-1}). In fact, alloying-type anodes generally offer much higher specific capacities than carbonaceous anodes. In particular, among the various elements that can form electrochemical alloys with lithium, silicon stands out as the most promising for practical applications due to its exceptionally high gravimetric and volumetric capacities [3, 27]. Silicon also offers benefits due to its low cost, widespread availability, and non-toxic nature [3]. It is largely for this reason that silicon has attracted considerable attention as an anode material in lithium-ion battery research.

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However, the lithium silicide $Li_{22}Si_5$ forms at elevated temperatures (~ 415–450 °C) and it is not typically observed in standard LIB cycling conditions. For instance, in Obrovac and Christensen's work on sputtered Si thin films and Si alloy anodes, $Li_{22}Si_5$ is never reported under practical electrochemical cycling conditions [70]. Electrochemical lithiation of silicon at room temperature predominantly yields an amorphous Li_xSi alloy, as demonstrated by Limthongkul et al. [52], due to electrochemically driven solid-state amorphization. However, at deep lithiation stages (voltages below 50 mV vs. Li/Li^+), the crystalline and metastable $Li_{15}Si_4$ phase appears, as shown in studies by Obrovac and Christensen [70], Sternad et al. [83], Hatchard et al. [29], and Baggetto et al. [5].

The theoretical capacity for $Li_{15}Si_4$ at room temperature is 3579 mA h g^{-1} [70]. This phase indicates deep lithiation and is associated with high capacity, but also with severe volume expansion (~ 280–300%, during full-capacity cycling at room temperature) [3, 76].

The results of research on the use of silicon to fabricate LIB anodes (Fig. 1) have given a total of 9607 patent families [20], the first dating back to 1997 (earliest publication date). This large number of patents demonstrates the scientific and technological interest towards the potential of this material to replace graphite, currently used in commercial batteries.

It is worth mentioning that, despite the cited technological advantages of using silicon as anode material, one of the industrial challenges related to its employ concerns its recent inclusion in the EU list of critical materials [16]. This fact makes the recovery and recycling of this material extremely recommended. A possible solution to the criticality problem could be the recovery of silicon from EoL PV panels (see Appendix C).

Si and Si-based anode materials remain in need of solving various bottlenecks: first, degradation of cycling performance and capacity decay occurs during lithiation/delithiation. In fact, the limiting factor of silicon, as anodic material, is represented by its volumetric expansion, which can reach values up to 300% during charge–discharge cycling [76]. Such a behaviour involves progressive fragmentation and loss of active material (which is electrically isolated from the rest of the electrode) with the progress of charge–discharge cycling, resulting in a rapid decrease of the accumulated capacity. In addition, the formation of Solid Electrolyte Interphase (SEI) onto

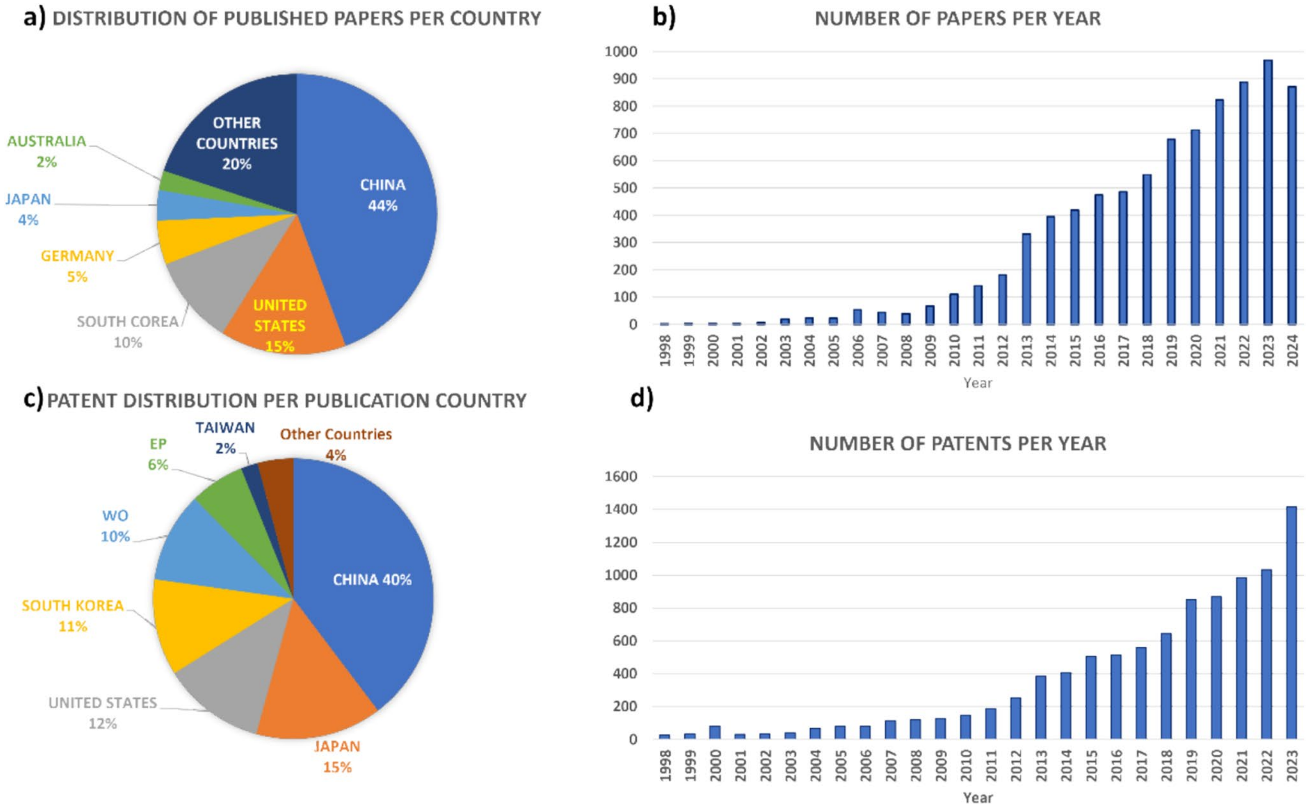


Fig. 1 Number of published papers per country (**a**) and over time (**b**), from 1998 to 2024; Number of patent families per country or organization where the patent application was filed or granted—EP = European Patent Office; WO = World Intellectual Property Organization (WIPO) — (**c**) and over time (earliest publication date) (**d**), from 1998 to 2023

the Si electrode is ascribable to electrolyte reduction i.e., the solid compounds originated from the electrolyte degradation form a passive layer onto the silicon anodes. During the initial charging and discharging cycles, the formation of the SEI film on the surface of Si, repeatedly forming and cracking due to significant volume changes, reduces the conductivity and cycling stability of the anode and also gives poor dynamic stability of the active material surface and inferior cycling performance of the battery. In subsequent cycles, as more electrolyte decomposes and depletes Li^+, new SEI layers form on the surface of the silicon anode, further reducing the Coulombic efficiency (CE) of the anode and results in irreversible capacity loss in the first cycle. Furthermore, the repeated volumetric expansion causes continuous break of the SEI at the electrolyte/electrode interface [47, 51]. Therefore, silicon (not being protected by SEI) comes into direct contact with the electrolyte. This induces a continuous reaction, by the silicon electrode with the electrolytic components, leading to an increase in irreversible capacity accumulated during the battery charge and resulting in a decrease in CE of the battery. In addition, the continuous volumetric expansion promotes electrode fragmentation, leading to active material loss and, therefore, capacity decay. Thus, the continuous formation of the SEI film leads to the loss of both electrolytes and electrode materials, which contributes to the degradation of electrochemical performance in Si-based lithium-ion battery anodes. To overcome the inherent challenges associated with Si and Si-based materials—such as severe volume expansion, poor cycling stability, and limited capacity retention—significant progress has been made through nanoscale engineering and the incorporation of various composite materials. These advancements aim to enhance the structural integrity and overall performance of Si-based anodes [10].

The paper offers a state-of-the-art on challenges and solutions related to the use of silicon as anodic material in LIBs, besides a survey on the TRL and the market penetration of silicon anode battery technology.

Moving forward in the structure of the document, Sect. 1.1 describes the publication and patent trend relating to the use of Silicon as anodic material in LIBs, while the operating principle and a brief history of rechargeable batteries is illustrated into Appendix A. Section 2 is dedicated to the description of the methods used to overcome the current drawbacks related to the use of silicon as an anode material in LIBs. A survey on the Technology Readiness Level (TRL) of the silicon anode battery technology is offered in Appendix F, while Sect. 3 is dedicated to future perspectives.

1.1 Patents and Academic Research: A State Of The Art

With the rapidly increasing demands of energy markets, the emergent next generation LIBs represent a huge success due to their higher energy and power densities. It is worth noting that the dominance of LIBs in the energy storage industry is connected to their maturity and their in-depth understanding of the complex commercial manufacturing processes involved in the production of their cathode and anode materials. Meanwhile, the huge market demand has fuelled scientific research on LIBs. Since the early 1990s, there has been an extraordinary amount of work on all aspects of lithium-ion chemistry, battery design, manufacture, and application.

Despite the considerable progress of this technology, there is a key issue that need to be addressed: LIBs are required to deliver more energy in shorter times, especially in automotive application. To meet actual and future needs in LIBs market, silicon is being positioned as a promising anode material due to its extremely high theoretical specific capacity. Since the electrochemical performances of LIBs are mainly determined by the properties of its anode and cathode, which can be roughly classified based on intrinsic material properties (e.g., theoretical capacity, ion conductivity, volume change during charge–discharge cycles, and voltage with respect to metallic Li) and extrinsic structure properties (e.g., particle size of the active material, morphology, porosity, and spatial distribution), appropriate synthetic methods are necessary to manipulate the electrode's microstructure. In addition, considering scalability issues and the upcoming massive consumption of LIBs due to the widespread use of electric and hybrid vehicles, industrially viable synthetic methods are urgently needed for mass-production of these new electrode materials. Among different anode materials used for LIBs, silicon has recently attracted increasing attention as one of the most likely alternatives to graphite. Indeed, the highest theoretical specific capacity, ten times higher than graphite, its relatively low lithiation potential (<0.4 V vs Li^+/Li°), makes silicon the most promising anode materials for high energy density LIBs. Therefore, developing strategies to utilize Si resources in energy storage systems has significant value for the advanced electrodes of next-generation LIBs and green sustainable development.

According to the bibliometric analysis of publications in the field of silicon-based electrodes for research on LIBs, China leads global volume of published articles (44%), followed by the United States (15%), South Korea (10%), Germany (5%), Japan (4%), and Australia (2%). At the bottom of the list, there are additional countries (about 70 countries) with less than 150 published articles, accounting for 20% of total publishing (Fig. 1 **(a)**). Figure 1 **(b)** shows the number of publications issued each year, from 1997 to 2024 that are relevant to silicon-based anodes for LIBs. Silicon anodes for LIBs are a booming industry, and the level of intellectual property (IP) activity is growing fast, with numerous newcomers entering the game. To fully comprehend the competitive landscape and technology roadmap, revealing the companies, technical solutions and strategies not identified through standard market analysis, a patent landscape analysis was also performed (Fig. 1 **(c,d)**). This examination revealed that China ranked first also in the patents in the research field of silicon anodes for LIBs, as shown by the number of patents per country and over time, from 1998 to 2023.

Indeed, the silicon anode patent landscape is dominated by Chinese entities representing today 40% of the patent families (with about 7000 patent families and over 9700 documents in this field). In conclusion, China leads in collaborative publications in this field, demonstrating its dominance in terms of the total number of published silicon anodes for LIBs-related articles and patents [26].

2 The Solutions to Technological Challenges

This section will explore methods to overcome the drawbacks associated with the use of silicon as an anode in Li ion batteries. These solutions range from designing nanostructured silicon (Paragraph 2.1), to combining silicon with carbonaceous materials (Paragraph 2.2), including research for multifunctional polymer binders (Paragraph 2.3), the development of artificial SEI layers (Paragraph 2.4), and the opportune choice of the current collectors (Appendix E).

2.1 Silicon Nanostructuration

In typical silicon-containing electrodes used in lithium-ion batteries, lithium insertion causes substantial swelling—up to 300%—due to the formation of lithium silicides. This extreme volume expansion generates significant mechanical stress within the electrode bulk, leading to pronounced macrostructural deformation and disruption of electron-conducting pathways [42]. As previously described, the strain-induced deformation of active particles accelerates electrochemical corrosion processes within the electrode. A major obstacle to the use of silicon as an anode material in lithium-ion batteries is the inevitable formation of an oxide layer on the silicon surface, coupled with the inherently slow kinetics of lithium-silicon alloy formation. As a result, the electrodes show poor retention of reversible capacity and a pronounced accumulation of irreversible capacity. The cycling stability of silicon-containing anodes, however, can be significantly improved through nanostructuring strategies. Nanostructuring effectively mitigates the mechanical degradation of the electrode, preserves robust electrical contact between active particles and the current collector, and enhances lithium-ion mass transport.

Silicon anodes with low-dimensional structures (including 0D Si nanoparticles (Si NPs), 1D Si nanowires (Si NWs), nanofiber and nanotubes, 2D thin films and 3D porous structures/Si with nanoporous structure), compared with bulky silicon anodes, are strongly believed to have several advanced characteristics including larger surface area, fast electron transfer, and shortened lithium diffusion pathway as well as better accommodation with volume changes. Also, the high density of grain boundaries in nanomaterials provides a fast diffusion path for lithium ions and acts as additional Li-storage sites. Nanostructured Si materials (Fig. 2) are potential candidates as anodes for LIBs because they can alleviate the problems of pulverisation and fragmentation of anode and of formation of an unstable SEI on the Si surface.

The preparation techniques for these materials rely on the concept of spatial (structural) stabilization of silicon-containing electrodes to achieve improved cycling stability. This approach, in turn, is based on creating free spaces—such as pores and voids—that can accommodate the swelling Li_xSi alloy formed during charging, thereby mitigating the detrimental effects of its volumetric expansion. Notably, early studies have already demonstrated the excellent cycling stability of thin-film silicon electrodes. In particular, previous research has shown that capacity retention improves with decreasing silicon film thickness and with higher electrode charging currents [42, 69].

Starting in 1994, Jeff Dahn at Simon Fraser University led several studies to place nano-sized silicon in carbon matrices [95]. Although the silicon occupied a very small portion of the material, these studies demonstrated the viability of using nanosized silicon as an anode at ambient temperatures, thus inspiring a new branch of structural engineering. In 1998, Wang from Zhejiang University was the first to make silicon nanoparticles for use in anodes without a carbon matrix through ball milling [86] and in 1999 Li from the Chinese Academy of Science synthesized silicon nanoparticles using a molecular precursor [44]. Both demonstrated reversible reactions with Li at room temperature. From 2000 to 2010, research on silicon took off. More and more reports from research institutions demonstrated promising lifetime through nanosizing, amorphization, restricting expansion, or a combination of these techniques. Some of the inventions, such as silicon nanowires and nanoparticles, led to start-ups like Amprius and Sila Nanotechnologies that are still seeking to commercialize silicon-based anode technologies.

However, one of the main challenges in nano-Si research is the high cost of synthesis and processing. The commercially available Si NPs are sold at unreasonably high price considering that Si is the second abundant element in the earth's crust. The reason is that it occurs in nature mainly in

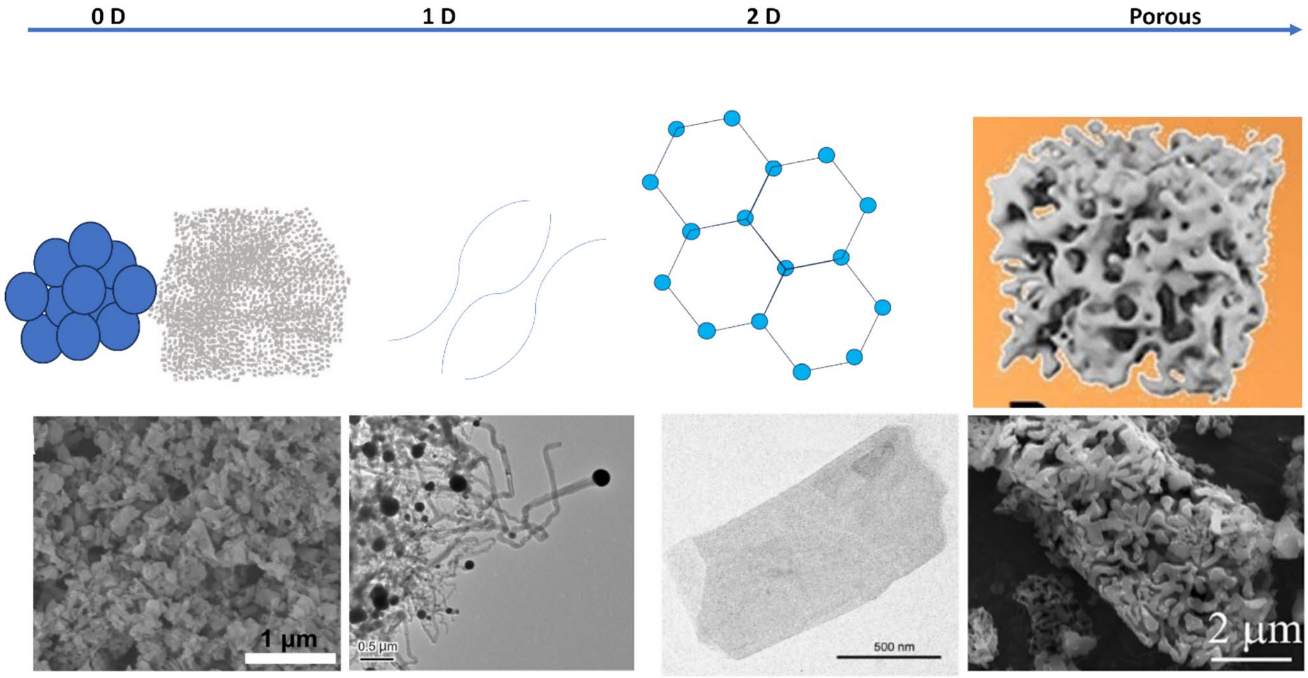


Fig. 2 Silicon nano structuration as a function of number of dimensions. Adapted from references Liu, X. et al. Trash to Treasure: Harmful Fly Ash Derived Silicon Nanoparticles for Enhanced Lithium-Ion Batteries. Silicon 14, 7983–7990 (2022) (0 D); Saana Amiinu, I. et al. A thin Si nanowire network anode for high volumetric capacity and long-life lithium-ion batteries. J. Energy Chem. 81, 20–27 (2023) (1 D); Kanaphan, Y. et al. Multilayer Silicene Nanosheets Derived from a Recycling Process Using End-of-Life Solar Cells Producing a Silicene/Graphite Composite for Anodes in Lithium-Ion Batteries. ACS Sustain. Chem. Eng. 11, 13,545–13,553 (2023) (2 D); Zhang, F. et al. Advances of Synthesis Methods for Porous Silicon-Based Anode Materials. Front. Chem. 10, 889,563 (2022) (Porous)

the form of oxide (silica) and as silicates. Therefore, there is a need for low-cost and scalable preparation methods that can produce nano-Si with desired properties and functionalities to support the development of Si-based anode for LIBs.

2.1.1 0D Silicon Structures

Several studies [19, 56, 76, 81] have explored the possibility of preparing 0D Si NPs from industrial waste or recycled silicon. These methods are environmentally friendly and economical, as they utilize abundant and renewable resources. Liu et al. have described one possible solution by using fly ash, a by-product of coal combustion, as a raw material for synthesizing spherical shape Si NPs with an average of 50 nm [56]. Fly ash is considered as a hazardous waste due to its environmental and health impacts. However, fly ash contains a significant amount of silicon oxide, which can be converted into crystalline nano-Si by acid leaching and magnesio-thermic reduction. The resulting nanoparticles have a high purity ($> 99\%$), a porous structure and a specific surface area of 220 $m^2\ g^{-1}$. The Si NPs obtained by this method demonstrate good lithium storage performances, achieving a high specific capacity of around 1030 $mA\ h\ g^{-1}$ after 500 cycles at a current density of 3.6 $A\ g^{-1}$.

Another potential source of recycled silicon is the high-quality silicon wafers from EoL-PV panels. Eshraghi et al. have showed an efficient way to recover nano-Si from PV waste and used it to make high performance LIBs [19]. They have used a combination of alkaline and acidic solutions to wash away the main contaminants (Pb, Ag, and Al) followed by ball milling to obtain ultrahigh purity nanosized Si powder. The electrochemical performance of the samples depends on whether they are leached or not. Leaching is necessary to remove Al and Ag residues that hinder the effective reduction of Si particle size during ball milling. The effects of milling speed and duration on the structural and morphological properties of Si powders and their electrochemical behaviour have been investigated. The ball-milled Si wafer fragments show a wide range of particle sizes from nanometres to micrometres, while maintaining the crystalline structure. When used as anode material for LIBs, the high purity nano-Si displayed a capacity of around 1285 $mA\ h\ g^{-1}$ after 50 cycles.

Rahman et al. have developed a novel technique to harvest high quality nano-Si from PV waste panels [76]. The process involves a KOH-based purification step and a ball milling treatment to produce nano powder having a mean particle diameter of 51 nm. Then, the nano-Si grains are mixed with graphite and tested as anode electrode in LIBs, showing a charge capacity of 426 mA h g^{-1} after 600 cycles, a capacity retention of 70%, a rate capability of 215 mA h g^{-1} at 5 C, and an average Coulombic efficiency of 99.4%.

Likewise, Sim et al. have designed a recovery process that involves only one step: dissolving expired Si cell in hot diluted phosphoric acid [81]. This step leaches out both the Al back contact and the anti-reflecting coating of silicon nitride (SiN_x) whilst detaches the Ag electrodes. Using this approach, the researchers have achieved a 98.9% recovery rate with 99.2% Si purity. Recovered Si powders are treated by ball milling to reduce the size to around 100 nm and then tested as anode in LIBs. After 500 cycles at a charging rate of 1 C, a specific capacity of 1086.6 mA h g^{-1} is obtained while maintaining a Coulombic efficiency of $>99\%$.

Other 0D Si contenders for lithium-ion anodes also look promising. Liu et al. of the Bijing National Laboratory for Molecular Sciences in China, have developed a NP anode material by reducing silica at the nanoscale [53]. They have studied an innovative method for the preparation of colloidal Si NPs from silica by means of magnesio-thermal reduction. Using silica NPs instead of bulk silica as the starting material, the authors have achieved the reduction of silica by magnesium in a much shorter time and at a much lower temperature. The reaction is completed in 10 min by mechanical milling at room temperature, while normally it requires high temperatures to proceed. The resulting Si NPs, which have a highly reactive surface, are then functionalized with 1-pentanol to form a hydrophobic colloid having an average diameter of 40 nm and later are carbon-coated by a single carbonization step. The carbon-coated Si NP anode exhibits a reversible capacity of 1756 mA h g_{-1} after 500 cycles at a specific current of 2.1 A g^{-1}.

Among the various methods for preparing 0D silicon nanoparticles, ball milling stands out for its scalability and versatility, particularly in utilizing low-cost, widely available raw materials such as photovoltaic waste and low-grade silicon sources such as metallurgical silicon and ferrosilicon. By mechanically pulverizing bulk silicon using high-energy collisions within a milling chamber, ball milling not only reduces particle size to below 150 nm but also helps alleviate the critical issue of silicon's volumetric expansion during cycling. However, excessive milling time can cause undesirable particle agglomeration, resulting in larger aggregates that impair cycling performance. To mitigate these issues and improve electrochemical behaviour, ball milling-produced Si nanoparticles are commonly combined with carbon sources to form Si/C composites. This strategy effectively trades off a modest reduction in capacity for significantly enhanced stability. Moreover, extended milling tends to induce amorphization of the silicon, which facilitates lithium-ion diffusion but may require additional treatments—such as chemical etching or ultrasonic dispersion—to improve uniformity and performance [45, 50].

Despite these trade-offs, ball milling remains among the most industrially promising routes for producing Si-based anodes, thanks to its simplicity, environmental compatibility, and readiness for mass production. It serves not only as a scalable synthesis method but also as a crucial link between laboratory innovation and commercial deployment.

2.1.2 1D Silicon Structures

Among 1 D structure, Silicon NWs are seen as a promising advancement in LIB technology [45, 50, 109], potentially leading to batteries that charge faster and provide longer range. NWs are grown directly on the metallic current collector substrate. This configuration offers several benefits and enhances the rate performance of metal oxide cathode materials. One advantage is that the nanowires have a small diameter that enables them to withstand the large volume changes without cracking, unlike bulk or micron-sized materials. Second, Si NW is in electrical contact with the metallic current collector, ensuring that all the NWs contribute to the capacity. Third, Si NWs provide direct 1D electronic pathways for efficient charge transport. In contrast, in particle-based electrode structures, electronic charge carriers must move through small interparticle contact areas. Moreover, as every NW is connected to the current-carrying electrode, there is no need for binders or conducting additives, which add extra weight.

Lu et al. have proposed a recycling strategy that can turn PV waste-Si into high-performance and stable SiNWs@reduced graphene oxide electrodes for LIBs [59]. The process is carried out in two steps. At the beginning, waste-Si particles and graphene oxide are assembled to create a structure where waste-Si particles are located between the graphene-oxide sheets. Then, by applying an electro-thermal shock, the waste-Si particles can be heated and cooled (≈2100 K for 10 ms) rapidly, inducing the formation of SiNWs having a diameter of 50 nm within the gaps between graphene oxide layers. The confined space provided by the reduced-graphene oxide sheets and the oxide layer on the surface of waste-Si particles are the key factors for the growth of SiNWs. When applied as a binder-free anode for LIBs, the SiNWs@reduced graphene oxide electrode exhibits a Coulombic efficiency of 89.5% and robust cycle stability (2381.7 mA h g^{-1} at 1 A g^{-1} for more than 500 cycles) at Si content of 76%.

Molten salt electrolysis presents a cost-effective and environmentally friendly pathway for synthesizing 1D Si-NWs. This technique involves the electrochemical reduction of silicon dioxide in a molten salt—typically calcium chloride—at elevated temperatures. When voltage is applied, the Si^{4+} ions are reduced to elemental silicon, forming nanowire structures directly on a conductive substrate.

The appeal of molten salt electrolysis lies in its simplicity, scalability, and use of abundant, low-cost precursors without relying on toxic gaseous reagents like silane. Unlike more conventional vapor-phase methods, molten salt electrolysis offers a safer and potentially industrially viable route for fabricating 1D nanostructures. However, challenges such as achieving uniform nanowire morphology, optimizing crystallinity, and managing high-temperature operations still limit its maturity [45, 50].

Despite being in its early development stages, molten salt electrolysis shows strong potential as a green and scalable alternative for producing high-performance Si NWs in future lithium-ion battery technologies.

2.1.3 2D Silicon Structures

Two-dimensional Si materials, owing to their nanoscale thickness and microscopic lateral dimensions, are other promising nanostructures for use in LIBs. Well-designed 2D silicon anodes provide a short path for lithium-ion diffusion, a quick charge transfer at the interface and a large contact area with the electrolyte, which leads to improved electrochemical performance [45, 50]. Among them, silicene, a 2D Si allotrope with single-layer thickness, has been attracting increasing attention in recent years. However, silicene is also highly unstable and reactive in ambient conditions, which poses significant challenges for its synthesis and application. Recent research has explored an efficient method to exfoliate a milled Si powder derived EoL PV panels into a multilayer silicene with thicknesses between 1.73 to 3 nm [36]. The silicene/graphite anode shows a Coulombic efficiency above 97% and a stable reversible capacity of around 290 mA h g^{-1} at a current density of 1 C after 500 cycles.

In addition to silicene, 2D Si nanosheets (NSs) are also considered as promising anode material candidate in LIBs and are usually prepared by means of exfoliation and reduction methods. For example, Chen et al. have proposed a novel technique for making a sheet-stacked silicon-carbon (Si/C) porous composite via magnesio-reduction combined to carbon coating with CO_2 [8, 9]. They have used Si waste as raw material, that is waste generated during the diamond wire cutting process in the PV fabrication. As a result, after 300 cycles at 1.0 A g^{-1}, the Si/C anode delivers a remaining capacity of 693 mA h g^{-1}. Park et al. [74] have synthetized uniform carbon layer-coated Si NSs (C-Si NSs) in three simple steps: 1) synthesis of Si NSs directly on graphite foil via chemical vapor deposition (CVD) methods, 2) deposition of parylene C on SiNSs; 3) thermal carbonization to obtain a uniform carbon layer with thickness of around 5, 10 and 25 nm on SiNSs 13 nm thick. The electrochemical tests depended on the carbon-coat thickness, suggesting that a suitable carbon coat on Si NSs increases the conductivity of electrons and ions and acts as buffer against volumetric changes. Among the thicknesses examined, the 10 nm layer showed the best performance in terms of charge–discharge efficiency (~ 100%) and the cycle performance (~ 2100 mA h g^{-1} after 300 cycles).

To further improve the electrochemical performances of 2D Si NSs as anodes in LIBs, Tang and his group developed porous Si NSs 30 nm thick via a soft template and a subsequent magnesiothermic reduction [85]. The method mainly includes the production of mesoporous SiO_2 NSs and its magnesiothermic reduction with the assistance of NaCl as the heat scavenger. High resolution transmission electron microscopy (HRTEM) observation indicates that SiNSs are composed of interconnected silicon nanocrystals with size of ~ 10 nm. When used as anode material for LIBs, the SiNSs exhibited a reversible capacity of 800 mA h g^{-1} after 900 cycles at a rate as high as 8400 mA g^{-1}.

Recently, Saana Amiinu et al. have developed a method to produce dense networks of indium-seeded Si NWs on a 3D copper-silicide (CS) current collector as binder-free, high-capacity anode (Si NWs@3D-CS) [77]. In this method, 3D nanostructured copper-silicide network is first prepared by functionalizing the copper-foil surface with silylbenzene, then loaded with catalyst seeds which act as nucleation sites and finally employed to nanowire growth by thermal decomposition of Si-precursors in liquid medium. Due to its nano branched structure (the average branch-size is of 140 nm), 3D network presents high surface area. This allows the indium seeds to be evenly and uniformly distributed within the entire structure, promoting therefore the nucleation and growth of a well-connected network of Si NWs with small dimension (average size around 70 nm). The resulting Si NWs@3D-CS electrode shows a Coulombic efficiency of > 99.6%, stable performance for more than 900 cycles with 88.7% capacity retention, and it delivers a volumetric capacity of ~ 1086.1 mA h cm^{-3} at 5C.

2.1.4 3D Silicon Structures

Going further from 2 to 3D structures, it is worth mentioning that porous Si nanostructures can improve the performance of Si anodes by creating space for silicon to expand when it intercalates lithium ions. In this way, the whole anode does not change its volume much and avoids breaking. It also helps to maintain a stable SEI layer on the anode surface that can ensure good electrical conductivity [45, 50, 107].

A novel method of synthesizing highly porous SiO_x/nanoSi@C composites has been developed by using gas-phase magnesium to thermally reduce low-cost fumed silica at 650 °C [31]. This technique allows the magnesium vapor to penetrate the SiO_2 and control both rate and location of the magnesium thermal reduction reaction, which otherwise would be too fast and depthless. This approach also regulates the distribution and the relative composition of SiO_x and nanoSi in the active material network. The resulting composites had a porous structure of Si NPs embedded in silica matrix. The resulting SiO_x/nanoSi@C anodes showed outstanding electrochemical performance, which maintained a reversible capacity of 1067.9 mA h g^{-1} after 400 cycles at a current density of 1 A g^{-1}. Here, the nano-Si component enhanced the reversible capacity of the composite, the porous SiO_x matrix mitigated the volume expansion of crystalline silicon by forming Li_2O and lithium silicate layers on the surface, and the carbon coatings further reduced volume expansion, improving the contact between the active material and the electrolyte.

The preparation of porous silicon structures through the thermal reduction of silica using magnesium has been extensively studied [45, 50]. During the process, magnesium serves as a strong reducing agent, transforming silica into silicon while simultaneously creating a porous structure. This method is cost-effective and straightforward, as it relies on readily available raw materials and offers relatively easy scalability for industrial production.

However, there are some notable challenges: a) byproduct removal: the reaction generates magnesium oxide, which must be thoroughly removed to prevent degradation of the material's performance; b) reaction kinetics: achieving uniform porosity and consistent product quality requires precise control and careful optimization of the reaction conditions.

To produce porous Si from EoL PV panels, C. Zhang et al. have studied a combined ball-milling and alloying/dealloying in molten salt process [106]. In this method, Si wafers are first treated with alkaline and acidic solutions to wash away the main contaminants (Ag, Al and SiN_x) and, after milling, meso-porous Si powder (m-Si) is obtained. Then, an alloying-dealloying approach in molten LiCl–KCl is dressed to remove the surface oxide layer of m-Si powders and create nanostructures. This is achieved through an electrochemical reaction: Li^+ is first reduced to form a Li-Si alloy with m-Si at the cathode, and then, the Li-Si alloy is used as the anode in the same molten salt to release Li^+ back into the salt, producing porous-Si (p-Si) by exploiting the volume change effect. The resulting p-Si has a higher specific surface area than the m-Si (19.4 m^2 g^{-1} and 6.1 m^2 g^{-1} respectively) with pore size ranging from 2 to 30 nm. Electrode expansion analysis confirms that alloying/dealloying process can produce p-Si structures that can act as buffers when acting as the anode in LIBs. After 200 cycles, the m-Si electrode expanded by about 236% in thickness. The p-Si electrode, which had an initial thickness of 12.4 µm, also grew to 23.6 µm, resulting in a thickness increase of 90.3%. Furthermore, p-Si anode shows a capacity of 2427.7 mA h g^{-1} at 1 A g^{-1} after 200 cycles with a capacity retention rate of 91.5% (1383.3 mA h g^{-1} after 500 cycles).

Table 1 summarizes the results presented in Sect. 2.1, organized by Si nanostructure dimensionality, with studies employing recovered Si highlighted in green.

2.2 Si/C Composite Anode for LIBs

This paragraph will explore the effort in overcoming the drawbacks related to the use of silicon as anode material trough the development and structural design of Si/C composite, in which carbon may form conductive networks, promoting electron and ions transport and preventing Si expansion [17, 21, 65]. Si/C composites exhibit excellent cyclic stability, lithiation capability, and electrical conductivity, resulting in high specific capacity and greatly increased anode conductivity. Moreover, carbon materials can operate as a buffer for the silicon anode, preventing direct contact between electrolyte and silicon, generating a stable SEI, and reducing volume variations during the Li^+ insertion/deinsertion processes. As a result, the electrode's structural integrity is preserved.

An overview of several structural Si/C composite anodes, depending on the dimensionality of carbon material's sources (Fig. 3), is provided in Appendix D of the Supporting Information, while the main performances reported in literature for different Si/C composite nanostructure are given in Table 2.

2.3 Polymeric Binders

Although the content of binder in each component of the electrode is very small (less than 10 wt.%), it has a vital impact on the full performance of the battery. For the anode manufacture, the binder play an important role in binding the silicon active material with the conductive agent and, therefore, the binder structure can significantly affect the mechanical durability, adhesion, ionic/electronic conductivities, and SEI stability of the silicon anodes [18, 110].

The traditional polyvinylidene fluoride (PVdF) binder has low tensile strength and weak Van Der Waals binding force, so it cannot provide enough adhesion to resist the severe volume change of Si after a long cycle [41]. The cycling stability of Si-based anodes is strongly dependent on the type of polymer binders employed. Rational design of polymer binders with strong adhesive strength could efficiently alleviate the volume expansion of Si particles, reducing the cracking or pulverization of active materials and improving the cycling stability of Si-based anodes. Depending on the polymer structure, polymer binders for Si-based anodes are simply divided into four categories, namely linear, branched, crosslinked, and conjugated conductive polymer binders [18, 108] (Fig. 4).

Linear binders (e.g., Carboxymethyl cellulose -CMC-, PVDF, and alginate—Alg) have shown great potential in

Table 1 Performance comparison of technological solutions used to improve Si behaviour as anodic material

		Anode material	**Specific Capacity (mA hg^{-1})**	**Current Density (A g^{-1})**	**Rate**	**Cycles**	**Reference**
Silicon Nano structuration	0D	Si NPs prepared from industrial waste fly ash via magnesiothermic reaction	1030	3.6		500	X. Liu et al., 2022
			3173		0.1 C	3	
		Si NPs obtained by ball milling process on leached PV cells	1285			50	Eshraghi et al., 2020
		Hybrid recovered PV nano-Si/graphite anodes	426			600	Rahman et al., 2021
		Recovered PV nano-Si	1087		1 C	500	Sim et al., 2023
		C coated Si NPs produced using Mg reduction	1756	2.1		500	Z. Liu et al., 2017)
	1D	Nanobranched networks of indium-seed Si NWs on copper-silicide collectors silicon NW	1783		0.1 C	900	Saana Amiinu et al., 2023
		Si NWs grown on reduced graphene oxide	2382	1		500	Lu et al., 2021
	2D	Silicene/graphite composites	290		1 C	500	Kanaphan et al., 2023
		Sheet-stacked Si/C composites	693	1		300	M. Chen et al., 2022
		C coated Si Nanosheets	2100		0.15 C	300	S. W. Park et al., 2021
		Porous Si nanosheets	800	8.4		900	Tang et al., 2019
	3D	Porous composites of Si NPs distributed into SiO$_x$ matrices	1068	1		400	Hong et al., 2023
		Porous Si from EoL PV panels by a combined ball-milling and alloying/dealloying process	2428	1		200	C. Zhang et al., 2021

Unless otherwise indicated, the specific capacity refers to the value calculated after the number of cycles and at the current density listed in the designated columns. References utilizing recovered Si are highlighted in green

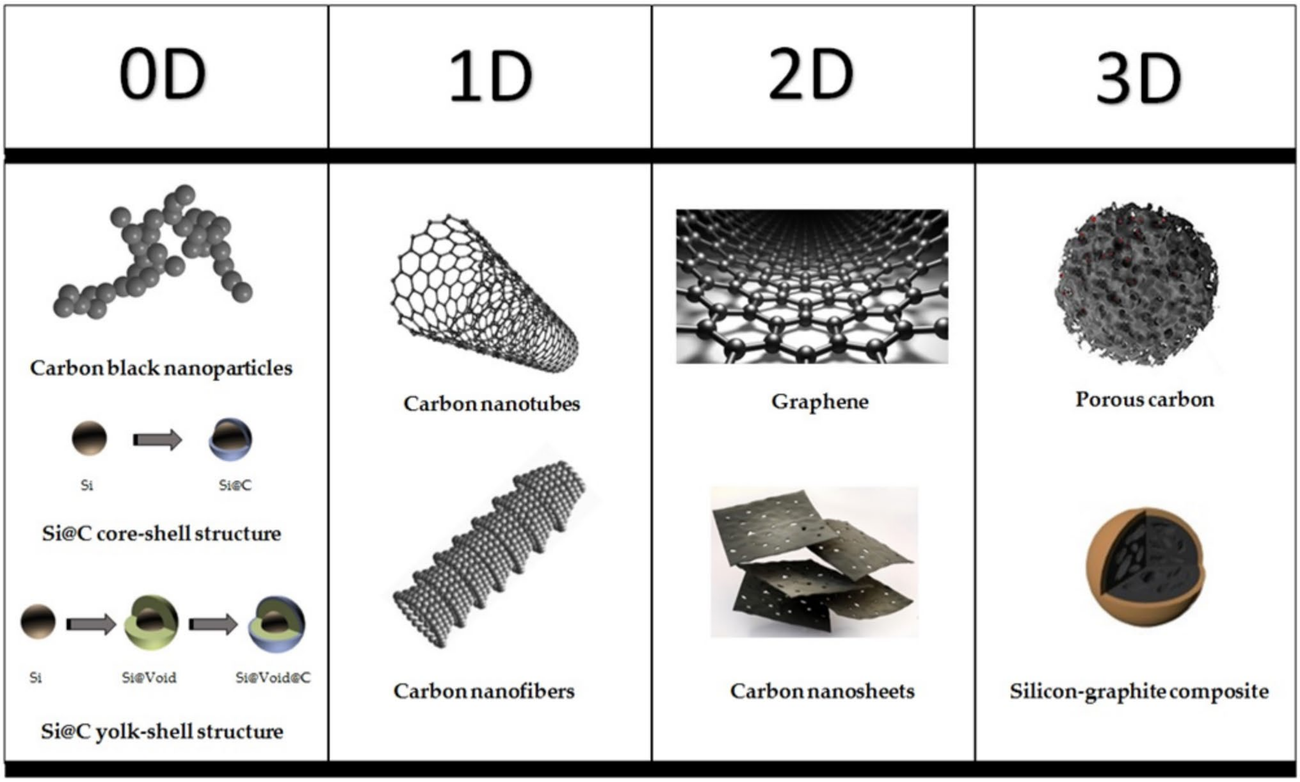


Fig. 3 Dimensional classification of carbon material sources

improving the electrochemical performance of Si-based anodes and are still widely used. Branched-chain binders derived from linear binders (i.e., Poly(acrylic acid sodium salt)-grafted-Carboxymethyl Cellulose -NaPAA-g-CMC- or Poly(vinyl alcohol)-graft-poly(acrylic acid), PVA-g-PAA) also show good performance. To further alleviate a series of problems during cycling of Si-based anodes, cross-linked binders with 3D structures have become a research hotspot. Cross-linking polymerization is divided into two types: covalent cross-linking and dynamic cross-linking. Covalent cross-linking mainly enhances the mechanical strength of polymers by forming chemical bonds, while dynamic cross-linking realizes the self-healing function of the adhesive through hydrogen bonding and electrostatic interaction. Conductive binders can act as both binders and conductive additives and can provide excellent electron or ion transport channels [57]. Compared with the one-dimensional contacts by linear polymer binders, branched polymer binders tend to form multidimensional contacts with Si particles, thus distributing or dissipating the mechanical stresses generated by Si volume change to multiple branched chains or points. Therefore, branched polymer binders are expected to exert stronger adhesive strength and more effective suppression of volume expansion of Si-based anodes than the linear polymer binders [108]. Among these, self-healing polymers based on three-dimensionally interconnected network are regarded as a highly promising means to accommodate the large volume changes of Si-based anodes [12]. The adaptability of the binder to different particle sizes should also attract more attention.

At present, there are many studies on the binder, but there is no comparison between the particle size of silicon and the adaptability of the binder. For silicon particles with larger particle size, the binder is required to have self-healing ability, which can be achieved through structural design [43], although innovative strategies are still needed to foster their application in practical use and their commercialization [99].

For example, a three-dimensional binder with “fishing nets” structure using widely sourced raw materials CMC and β-CD (β-cyclodextrins) was designed. As a green binder, CMC is directly soluble in water and has excellent compatibility with electrolytes. On the other hand, CMC has high brittleness, and its low elongation could not suffer the volume change of Si anode. Cross-linked cyclodextrins have the advantage of dispersing the stress at the anchor point and moderating the significant volume changes of the Si anode. The hydrogen bonding present inside β-CD-CMC can self-repair the cracks generated in the electrode during cycling to a certain extent. The Si/β-CD-CMC electrode maintains a reversible capacity of 1702 mA h g^{-1} even after 200 cycles, resulting to be significantly more performant than the Si/β -CD and Si/CMC electrodes. Thus β-CD-CMC

Table 2 Performance reported in literature for different Si/C composite structures with different carbon material dimensionalities

		Anode Material	**Specific Capacity (mA hg^{-1})**	**Current Density (A g^{-1})**	**Cycles**	**Reference**
Si/C composites	**0D C sources**	Si/C core-shell particles	1200	0.2	300	J. S. Kim et al., 2015
		Carbon-coated porous silicon	1271	2.1	1000	An et al., 2019
		Yolk-shell carbon-coated rigid SiO_2 confining Si NPs and CNTs	1500	0.5	450	L. Zhang et al., 2019
		Si/C encapsulated nanocomposite	1576	0.4	100	Chen et al., 2018
	1D C sources	Vertically aligned CNTs containing nanoscale amorphous/nanocrystalline Si droplets	2050	0.1	25	W. Wang & Kumta, 2010
		CNTs framework anchored with double carbon coated Si NPs	943	0.84	1000	H. Zhang et al., 2019
		Gel polymer electrolyte infiltration into silicon-coated vertically aligned carbon nanofibers	1070	2.6	100	Pandey et al., 2015
	2D C sources	Si/C nanofiber/graphene composite	965	0.1	100	Cong et al., 2021
		Nitrogen-doped carbon matrix-encapsulated Si NPs/carbon nanofibers	1371	0.1	100	Cong et al., 2022
		Porous silicon NWs in graphene nanoribbon papers	1500	1	300	Salvatierra et al., 2016
	3D C-sources	MOF-derived carbon shell/silicon composite	820	5	1000	R. Gao et al., 2020
		Silicon NPs/mesoporous carbon forming pomegranate-structure microspheres	581	0.2	100	Shen et al., 2017
		Carbon-coated silicon NWs on carbon fabric substrate	2061	1	500	Wang et al., 2017
		Si/C/graphite micro/nanocomposite	938	0.1	300	J. Wang et al., 2016

Table 2 (continued)

		Al_2O_3 coating layer on mesoporous Si nanospheres	1002	2	500	N. Li et al., 2019
		Sandwich heterostructures of MoS_2 2D nanosheets with Si NPs	1399	0.5	500	Marriam et al., 2023
		C coated $Si@SiO_x$NPs	1333	0.1	100	X. Li et al., 2022
		$Si/s\text{-}C@TiO_2$ composite: Si NPs embedded in spherical carbon layer and titania shell on the outer surface	780	0.2	100	X. Chen et al., 2022
		Network of core/shell Si NPs/TiO_2 structures with Ag NWs	First discharge capacity of 3524	0.4	100	J. Li et al., 2023

is an effective green binder for Si-based electrodes and offers a new option for future industrial development [34].

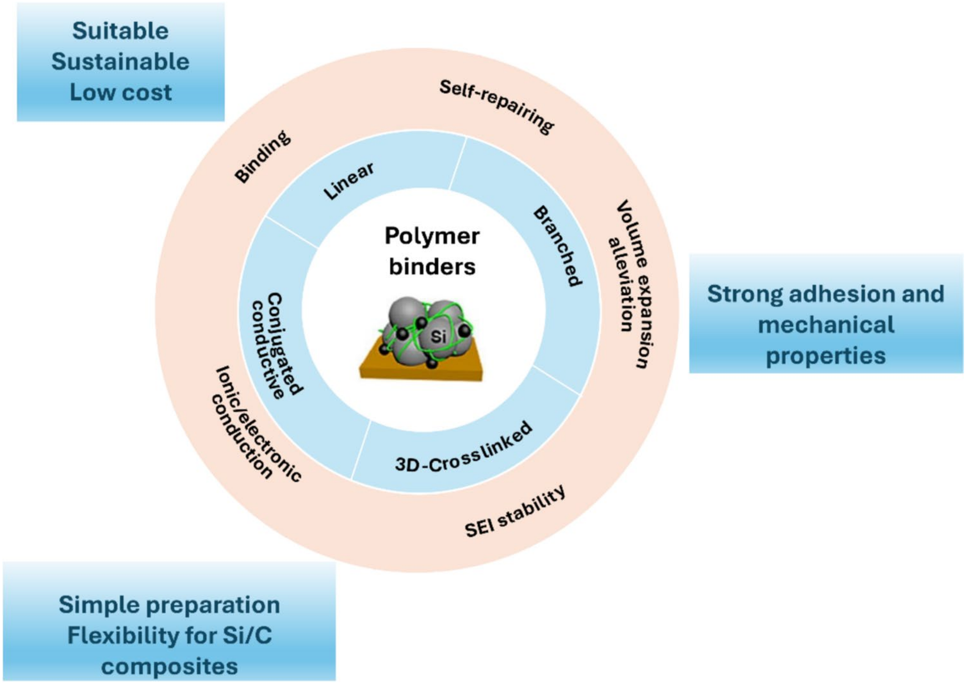
Natural adhesives have the advantages of being sustainable, low cost, of high molecular weight, and with great bonding groups such as carboxyl, hydroxyl, and amino, making them ideal in applications for Si electrodes. The Si@CMGG electrode prepared by a carboxymethylation method (CM) of the natural vegetable guar gum (GG) maintains a reversible capacity of 1865 mA h g^{-1} after 200 cycles and has a capacity decay rate of 0.12% per cycle. The self – healing adhesive can continuously repair the cracks caused by the volume expansion of the Si anode [41, 92, 93].

The development of a binder should be more inclined to have multiple functions. The electrode binder has more functions than bonding, which can not only reduce the inactive material content of the anode electrode and improve the mass loading but also save the manufacturing cost. The SEI is very important for the electrode. If the stability of the SEI can be improved by a binder, the cyclic performance of the silicon electrode will be greatly improved. Electronic conductivity and ionic conductivity are very important for the silicon anode. When the binder has a certain conductivity, it can improve the CE and capacity retention of the anode [43].

For a large-scale application of LIBs, safety has become a more and more concerning issue. A multifunctional three-dimensional PAA-FREP polymer binder was prepared by cross-linking polyacrylic acid (PAA) with a flame-retardant epoxy resin (FREP) containing phosphorus and nitrogen elements. The cross-linking between FREP and PAA and the introduction of the epoxy functional group greatly improve the mechanical property and cycle stability of the Si electrode. Also, most importantly, the FREP component in the binder leads to admirable flame retardance. In these ways, the multifunctional binder exhibits excellent mechanical, electrochemical, and safety performances [54, 55].

It is believed that the development trends of new binders should include the following aspects: first, most binder test data are confined to the half-cells, which widen the gap to the real-world applications of Si anodic LIBs. Compared

Fig. 4 An overview of categories, functions and properties of polymer binders for Si-based anodes. Adapted from reference [108]



with half cells, full cells do not have excess lithium ions and have a higher potential window. Therefore, comprehensive data on testing binders in full cells are required. Secondly, the current synthesis process of most binders is still very complicated and not suitable for large-scale production. Third, compared with binders soluble in organic solvents, water-soluble binders are cost-effective and environmentally friendly. Finally, new binders should be able to adapt to the traditional graphite negative electrode. Moreover, it is worth noting that at present, composite materials made of graphite and Si are used as the active material. Therefore, the binder should not only adapt to the Si anodes but also be suitable for the graphite ones or for anodes constituted by composite based onto Silicon and Carbon materials [18, 57].

2.4 Surface/Interface Engineering

In recent years, several strategies of surface/interface engineering of Si anodes have been developed to overcome the several technical issues related to the fast reactivity at the interface of Si anode and the electrolyte, which limit the electrochemical performance of Si-based LIBs because bring to low Initial Coulombic Efficiency (ICE) and poor cycling stability, by seriously restricting their application. The most used surface/interface engineering methods include surface coatings, surface functionalization and the growth of an artificial SEI (ASEI).

The surface coating is an effective way to prevent the direct contact between Si anode and the electrolyte, suppress the repeated broken and generation processes of SEI, provide rapid transport channels for Li ions and electrons, and to stabilize the volume expansion [66]. A variety of materials have been used as coating, including carbons [68, 84, 97], metals [105], metal oxides [48, 60, 61, 64], conductive polymers [100] and solid electrolytes [28], mainly used to coat Si nanostructures.

The application of a stable carbon material coating to the Si surface is a globally used technique to extend the Si anode's cycle life. This is because a carbon coating layer serves multiple purposes: it prevents the aggregation of Si NPs, enhances conductivity, and alleviates mechanical stress. A recent work on stable core shell Si@SiO$_x$/C anode [45, 50] produced via spray and pyrolysis method, reported a high specific capacity of 1333 mA h g^{-1} after 100 cycles at current density of 100 mA g^{-1}. The authors attributed the improved electrochemical performance to the core shell structure, which prevented the aggregation of Si nanoparticles and alleviated the volume expansion during lithiation. The spray and pyrolysis method were also simple, scalable, and cost-effective for mass production of the anode material.

Carbons such as graphene [65, 71], graphene oxide [22] and pyrolytic carbons [91] have been the most widely used as a coating for Si nanostructures because of their ability to improve remarkably the electrical conductivity of anodes and to accommodate the huge Si volume changes during lithium alloying/dealloying process. Moreover, the carbon-based coatings efficiently prevent electrolyte infiltration into the Si particles via interparticle cracks by building a more stable SEI layer and the carbon shell avoids the agglomeration of the NPs. However, low cycling CE is still a remaining issue for the Si/C composite anode.

Coating Si-based electrodes with metal oxides is another effective approach to improve their electrochemical performance by alleviating the volume expansion of Si particles upon lithiation/de-lithiation proceed. Moreover, the metallic oxides protective layer impedes the corrosion of electrolyte to internal Si particles, thus retaining the structural integrity of Si/MO composites and improving cycle stability. This is notably true for composites showing a core- shell structure, where Si NP is the nucleus and the transition metal oxide the buffer layer which protect the core from direct contact with the electrolyte. The common metallic oxides coating materials include TiO_2 [98], Al_2O_3 [39, 58] and zeolites [82]. Specifically, TiO_2 is a transition metal oxide that attracts a lot of attention for its outstanding properties, such as high electrical conductivity, low volume expansion ($<4\%$), great mechanical strength, thermal stability and cost effectiveness. For example, Chen et al. have prepared Si NP/s-C using the hydrothermal method, which embeds commercial Si NPs in spherical carbon and coat an amorphous TiO_2 shell on the outer surface [8, 9]. Li et al. have explored a dual strategy to enhance the cycling stability of the silicon-based anode [47, 51]. First, they have grown Si NPs/TiO_2 core–shell structure and then have doped it with Ag NWs to form a conductive network, improving the materials conductivity.

Coating Si particles with various metals has proven to be a useful strategy to improve electrical conductivity, decrease polarization, and buffer volume changes. An effective metal coating must be inert to the electrolyte and allow for Li-ion insertion into Si. Noble metal particles, such as Ag [6], Au, etc., and other transition metal particles, such as Cu [67], Fe, Ge [60, 61], etc., are often used to modify Si-based materials.

The rational design of MoS_2 nanosheets with Si NPs can create a MoS_2@Si sandwich nanoarchitecture that can synergize the advantages of both materials [63]. Here, the layered MoS_2 nanosheets can buffer the volume expansion of Si NPs and facilitate the lithium-ion diffusion during the electrochemical cycles. The volume expansion of Si NPs is reduced to 68% after 500 cycles, compared to 430% for pure Si NPs. The composite anode also shows excellent rate capability of 2331 mA h g^{-1} at 100 mAg^{-1} with the Coulombic efficiency of above 98%.

Polymer coating (especially conductive polymers) is also feasible in relieving fracture of Si particles. Polymers have the advantages of flexibility and elasticity and act as the flexible medium to mitigate the volume changes during cycling, endowing Si with the ability to self-healing. The most used conductive polymers for high performance LIB anodes are PANI [96], PPy [73] and PEDOT [30].

Another way to reduce the fast reactivity (side reactions) between Si anodes and electrolytes is the anode surface functionalization. As a result, a stable and robust protective layer can be constructed on the surface of Si anode, particularly on the Si nanoparticles. A variety of interfacial engineering strategies have emerged as solutions to these challenges. For example, Si nanoparticles were surface-functionalized with epoxy group treated by hydrogen peroxide followed by a silanization reaction [33]. Among these, there is also the prelithiation, i.e., introduction of additional Li in the anode Si materials via a controlled manner [23, 35]. This abundance of Li can not only provide excessive depletion ions for the stabilization of the SEI but also can effectively improve initial CE and increase the average cell potential. Prelithiation can be accomplished through two different methods. In one method, electrochemical prelithiation can be realized by charging/discharging a Si-based cathode with a temporary Li anode. The other method involves the physical blending of Si-based active materials with Li metal foil or powder in an inert atmosphere. Both methods have been shown to stabilize the interface between the active material and the electrolyte.

In addition to the surface coating and functionalization, the intentional formation of an ASEI with superior ion conductivity, mechanical strength and good flexibility is another promising approach to protect the Si-based anode from reactions with chemicals in the organic solvent and thus significantly improve the cycling stability by increasing the initial capacity loss, the initial CE and stabilize SEI growth [90]. Artificial SEI layers can be created with electrolyte additives, novel binders, surface coatings, etc. Desirable artificial SEI layers should exhibit superior chemical inertness and mechanical strength to lessen the dissolution of SEI components in the electrolyte and avoid cracking of the artificial SEI. For example, numerous electrolyte additives for the formation of an artificial SEI layer were discussed by Zhang et al. [102–104]. The use of blended polymer binders, such as PAA and CMC, has been found to result in more mechanically robust SEI layers, which can suppress initial capacity loss [75]. Coatings of single-component inorganic oxides or polymers have also been used to facilitate the formation of an artificial SEI [102–104]. Wang et al. proposed the formation of in situ ASEI on the surface of the FeSi/C anode by a nucleophilic reaction of polysulphides with VC (vinylene carbonate) and FEC (fluoroethylene carbonate) molecules [92, 93]. The as-formed ASEI layer is mechanically dense and ionically conducting and therefore can effectively prevent the electrolyte infiltration and decomposition while allowing Li^+ transport across, thus stabilizing the interface. As a result, the reversible capacity reached 1409.4 mA h g^{-1}, the cycling stability was over 650 cycles and the CE was 99,8%.

Finally, suitable ionic liquid-based electrolyte formulations were founded to show very good compatibility towards

Si electrodes. In particular, bis(fluorosulfonyl)imide (FSI) ionic liquids displayed excellent cycling behaviour in silicon nanowire anodes, i.e., a capacity close to 1000 mA h g^{-1} were recorded after more than 2000 continuous cycles carried out at 1C. This is ascribable to the good film-forming ability of the FSI anion in combination with its fast ion transport properties [62, 79].

2.5 Challenges in Scaling Laboratory Findings to Commercial Applications

Anodes containing silicon (Si or SiO_x) and their carbon composites have already been commercialized in the Tesla Model X (2015) and Model 3 (2017) through Panasonic [101]. These innovations extended driving ranges to over 330 km for the Model X and more than 490 km for the Model 3. Incorporating small amounts of silicon into predominantly carbon-based anodes offers a practical compromise: it boosts energy density while minimizing volumetric fluctuations and limiting capacity loss from electrical disconnection. In these composites, porous voids within the graphite matrix accommodate ultrasmall silicon particles, whose volume expansion is mitigated by their nanoscale size and carbon encapsulation, reducing mechanical stress. This synergy between silicon and graphite further stabilizes the active material by creating an interfacial barrier with the electrolyte, thereby improving durability.

The optimal silicon ratio varies depending on the technology employed. Typically, the mass fraction of Si or SiO_x is kept below 5 wt% [40, 101], since higher amounts make it difficult to maintain strong bonding among the electrode components and the solid electrolyte interphase (SEI) layer. Kim et al. [40] reported the evolution of specific capacity as a function of Si/SiO_x content across various commercial 18,650 cells, including Samsung 30Q, Samsung 25R, LG MJ1, Samsung 35E, Samsung 48G, Sony VTC6, Sony VTC5A, and LG HG2. In SiO_x–graphite composites, SiO_x contents above 10 wt% can deliver higher energy densities than graphite-only cells, but beyond 20 wt% the performance drops due to swelling and porosity challenges. At SiO_x contents exceeding 70%, energy densities can even fall below those of commercial graphite cells.

In summary, moderate additions of silicon can enhance energy density but do not guarantee cost-effectiveness, since controlling silicon's volume changes increases manufacturing costs. Higher silicon contents require advanced binders to maintain electrode cohesion, along with improved electrolytes capable of forming a mechanically robust SEI on the silicon surface.

Fluorinated ethylene carbonate (FEC) has proven to be an effective additive to ethylene carbonate (EC)-based electrolytes, promoting a more stable SEI and becoming essential in most silicon-based systems [101]. Nonetheless, significant challenges remain to achieve the high energy density, long cycle life, and reliability needed for large-scale electric vehicle adoption. Mechanical degradation, interfacial instability, and process variability must be addressed to fully exploit silicon-containing anodes. A major obstacle is translating promising laboratory-scale results into full-cell configurations compatible with industrial production. Although half-cell tests often report reversible capacities exceeding 2000 mA h g^{-1} for silicon (as shown in Tables 1 and 2), they frequently ignore critical factors like areal capacity, electrode density, and lithium losses, which are essential for practical applications. Kim et al. [40] highlighted the measurement gap between laboratory and industrial results. While lab tests typically report energy density based on active material alone, industry evaluates the entire cell, considering inactive materials, safety, cycle life, swelling, and cost. Normalizing specific capacity to silicon mass in lab settings often leads to overestimated expectations for commercial feasibility.

Silicon anodes undergo significant volume changes during cycling, requiring extra space within the cell to accommodate expansion, which ultimately lowers their achievable energy density despite their high theoretical capacity. Increasing electrode porosity can help manage this swelling but results in thicker, less dense electrodes. Li et al. [46] and Kusenko [42] showed that high capacities are often achieved using ultrathin or highly porous electrodes with extremely low active material loadings—sometimes just fractions of mg cm^{-2}—resulting in very low areal capacities similar to those of early thin-film electrodes. Although these structures may deliver good cycling stability, their volumetric capacity is far below commercial graphite (about 500 mA h cm^{-3} at room temperature), making them unsuitable for practical deployment [11, 46].

To be commercially competitive, silicon-based anodes must simultaneously achieve high areal capacities (over 3 mA h cm^{-2}), elevated electrode densities (above 1.3 g cm^{-3}), and low binder contents (ideally below 3 wt%) [46]. Cheng et al. [11] emphasized that these specifications are critical to improving volumetric capacity and matching current graphite standards. However, achieving these targets requires thicker electrodes, which exacerbate silicon's volumetric expansion—up to 300% during lithiation—leading to fracture, polarization, uneven lithiation, and progressive capacity loss, even beyond the first cycles.

A particularly critical degradation mechanism is the instability of the SEI layer on silicon, which tends to crack and reform repeatedly, consuming electrolyte and irreversibly trapping lithium. In half-cells with lithium metal counter-electrodes, this lithium loss is artificially compensated, giving an overly optimistic picture of long-term stability. In full cells, where the lithium supply is limited, even a small drop in coulombic efficiency—from 99.9% to 99.0%—can cause severe capacity fade. A cell operating at 99.0% efficiency retains only 37% of its capacity after 100 cycles, while improving it to 99.9% raises capacity retention by 28%. Therefore, commercial cells demand coulombic efficiencies exceeding 99.8%, and to support this, electrode volume change should be contained below 70% [46].

Recent research has made progress in boosting silicon-based anodes' volumetric capacity. Cheng et al. [11] summarized strategies such as raising gravimetric capacity, optimizing the active material mass ratio, enhancing initial electrode density, and reducing both electrode swelling and the negative-to-positive capacity ratio (N/P ratio). Yet, to replace graphite in commercial LIBs, silicon-based anodes must demonstrate comparable performance under industrial conditions, including areal loadings above 3 mA h cm^{-2}, electrode densities beyond 1.40 g cm^{-3}, swelling ratios below 17%, and N/P ratios near 1.1.

Despite these advances, the volumetric energy density of full or pouch cells with silicon anodes still falls short, usually not exceeding 1000 Wh L^{-1} [11]. For instance, patent US 2024/0297337 A1 describes a lithium-ion cell with a silicon–carbon anode containing 10–60 wt% silicon (particle size D50 of 3–8 μm), binder content of 5–10 wt%, and a coating density between 0.8–1.75 g/cm^3. The cathode combines a primary cathode material (e.g., LCO, NCM, NCA, or LFP) with a supplementary high-lithium-density material, showing first-cycle capacities of 350–750 mA h/g. The mass ratio between primary and supplementary cathode is typically 15:1 to 50:1, with supplementary cathodes including lithium-rich layered oxides or fluorinated compounds. The full cell achieves coulombic efficiencies above 95% after 20 cycles and over 98% after 40 cycles, demonstrating volumetric energy density improvements of 20–30% compared to conventional cells based on graphite anodes and standard transition metal oxides, which usually reach around 650 Wh L^{-1} [46].

An MGS (macropore-coordinated graphite–Si) electrode achieved an impressive swelling ratio of only 19% after 100 cycles, comparable to graphite's 17%, while delivering a volumetric capacity of 493.9 mA h cm^{-3} and an energy density of 1825.7 Wh L^{-1} after 100 cycles. Generally, highly porous architectures beneficial in lab-scale studies tend to collapse during calendering, making them incompatible with industrial manufacturing [46]. Moreover, many synthesis methods developed in academia—such as chemical vapor deposition or ball milling—remain too expensive or unscalable. Failure mechanisms in high-loading full-cell configurations are still not fully understood, posing another barrier to commercialization.

Overcoming these obstacles will require developing new silicon architectures that balance mechanical stress, porosity, and volume to enable thick, high-capacity electrodes compatible with industrial calendering processes. Additionally, scalable, cost-effective, and reproducible synthesis methods must be prioritized [11]. Advanced in situ characterization techniques are also essential to track SEI evolution, phase transitions, and electrode swelling during cycling. Besides maximizing volumetric capacity, commercial designs must address initial coulombic efficiency, calendar life, and safety.

Several studies have modeled full cells pairing Si/C anodes with various cathode chemistries. For example, a Si/C anode with 600 mA h g^{-1} specific capacity and a compacted density of 1.10 g cm^{-3} showed a theoretical gravimetric energy density improvement of 50% over graphite, with a practical gain of about 20% [46]. Greater gains can be achieved by combining such anodes with high-capacity cathodes like NCA or lithium-rich layered oxides, surpassing traditional LCO or NCM111. Such configurations have reached energy densities up to 374 Wh kg^{-1}. Enhancing compacted electrode density and using prelithiation further increases energy density while reducing cathode material demand.

Despite these achievements, full-cell systems with silicon-based anodes still face challenges meeting the cycle life and energy density targets required for commercial viability. Li et al. [46] showed through advanced spectroscopy and microscopy that SEI instability, parasitic reactions, and irreversible lithium losses remain key failure mechanisms. Unlike half-cells, full cells lack excess lithium, making cyclable lithium management a critical challenge. Accumulated degradation products and detachment of active particles can rapidly lead to cell collapse.

In conclusion, to make silicon a viable next-generation lithium-ion battery material, dense and stable anodes must be developed alongside high-capacity cathodes, effective irreversible loss management, and a deeper understanding of full-cell failure mechanisms. As research and technology evolve, silicon anodes remain a promising path to higher energy densities. Armand et al. [3] suggest key performance indicators—energy density, power density, cycle life, cost per kWh, safety, and sustainability metrics—to steer future research. They also highlight the urgent need for efficient recycling processes to recover strategic materials like lithium, nickel, cobalt, and manganese. Recycling should be integrated from the design stage, with better disassembly, separation, and refining technologies to reduce costs, emissions, and resource consumption. Finally, non-invasive analysis techniques can support predictive maintenance, improving battery safety and reliability [2, 94].

3 Summary and Future Perspectives

In summary, while significant advancements have been made in silicon (Si) anodes for lithium-ion batteries (LIBs), substantial challenges remain for widespread adoption and industrial application. Silicon's high theoretical capacity and energy density make it an attractive alternative to traditional graphite anodes, especially in high-demand sectors like electric vehicles and portable electronics. However, Si's inherent limitations—such as its tendency to undergo large volume expansion during charging cycles—require innovative solutions to improve cycle stability, CE and overall lifespan.

This review explores a variety of approaches aimed at enhancing silicon's performance, including nanostructuring, silicon-carbon (Si/C) composites, and surface/interface engineering. Each method addresses specific issues like mechanical stability, conductivity, and sustainability. Nanostructuring, for instance, offers ways to counteract silicon's volume changes, while Si/C composites leverage carbon's conductive properties to enhance stability and cycling performance. Additionally, recovery of silicon from photovoltaic (PV) waste and the development of eco-friendly, water-soluble binders provide sustainable paths forward, with significant environmental benefits. Tab. 1 presents a comparative analysis of the technological solutions aimed at enhancing silicon's performance as an anode material in lithium-ion batteries through different silicon nanostructures (0D, 1D, 2D, and 3D), each designed to address the challenges posed by silicon's high volume expansion during cycling. For example, 0D silicon nanoparticles produced through magnesiothermic reactions from fly ash exhibit good performance with a specific capacity of 1030 mA h g^{-1} after 500 cycles [56], demonstrating the potential of recycled industrial waste materials as a sustainable resource for battery anodes. Also, the nano-powder derived from photovoltaic panels has shown excellent results (1087 mA h g^{-1} after 500 cycles, [81]. Similarly, Tab. 1 underscores the benefits of complex composites, such as silicon nanowires grown on reduced graphene oxide [59] and silicene/graphite composites [36], which show high capacities (2382 and 290 mA h g^{-1}, respectively) after 500 cycles, while leveraging structural stability and conductivity. These innovations reflect a trend toward more sustainable anode materials that integrate recycled or recovered silicon sources from end-of-life photovoltaic cells, demonstrating high capacities and competitive cycle stability. Appendix C offers more details concerning the criticality of silicon as raw material for Europe and a brief state-of-the art of extraction methods from EoL PVs.

Tab. 2, summarizing literature results more deeply discussed in Appendix D, provides an in-depth comparison of silicon-carbon (Si/C) composite structures, organized by the dimensionality of the carbon sources (0D, 1D, 2D, and 3D). These Si/C composites demonstrate diverse approaches to addressing silicon's inherent volume expansion issue and improving conductivity, two critical factors in the performance of lithium-ion battery anodes. The table shows that 0D Si/C composites, like core–shell particles and carbon-coated porous silicon, provide stable cycling performance, with specific capacities reaching up to 1271 mA h g^{-1} over 1000 cycles [1]. 1D and 2D Si/C composites offer additional advantages, particularly through vertically aligned carbon nanotubes (CNTs) and graphene, which provide robust electron pathways and support higher energy storage. For instance, the CNTs framework with double carbon-coated silicon nanoparticles achieves high cycle stability (943 mA h g^{-1} over 1000 cycles), highlighting the effectiveness of carbon nanotubes in maintaining structural integrity during repeated charging cycles [102–104]. The 3D composites in the table illustrate advanced architectures, such as MOF (Metal–Organic Frameworks) -derived carbon shells and pomegranate-like microspheres, which balance high capacity with cycle life. These structures encapsulate silicon in porous carbon frameworks, effectively buffering against volume changes while ensuring efficient electron and ion transport. Notably, the sandwich structures, like MoS_2 with Si NPs, illustrate the multifunctional benefits of integrating additional materials to enhance both structural stability and ion diffusion.

Future research must continue to bridge academic insights with industry demands, particularly in achieving scalable, cost-effective manufacturing processes. Moreover, environmental and geopolitical criticality of raw materials for LIBs such as Lithium, graphite (see Appendix B for more details) and PVdF polymers must be considered. Emphasis on sustainable raw material sourcing and optimized binder design can support the transition of silicon anode technology from laboratory settings to practical, commercial use. This transition will be crucial to meeting the growing demands for high-performance, sustainable energy storage solutions in a variety of applications, from consumer electronics to renewable energy systems. The market for silicon-based batteries is set for rapid expansion. From an estimated USD 55 million in 2023, the industry is projected to reach USD 414 million by 2028, with an impressive annual growth rate of nearly 50%. This growth underscores a rising global interest in replacing or complementing graphite with silicon to achieve smaller, more powerful batteries.

Several leading companies are driving this innovation. Among the top Chinese companies leading the development and production of silicon-based anode materials for LIBs are BTR (Shenzhen BTR New Energy Materials Inc.), Shanshan Technology (Ningbo Shanshan Co., Ltd), Putailai (Jiangxi Zichen Technology), Zeto Energy (Jiangxi Zhengtuo New Energy Technology), Shenzhen XFH Technology and Hunan Zhongke Xingcheng Graphite (Hunan Zhongke Electric).

Table 3 Companies working on silicon as anode material

Company	State	Anode technology
Amprius Technologies https://www.amprius.com/	U.S	In-situ aligned silicon nanowires' template
Enovix Corporation https://www.enovix.com/	U.S	Pure silicon 3D cell architecture
Enevate Corporation https://www.enevate.com/	U.S	Enevate's XFC-Energy® Technology, pure silicon anode
NanoGraf Corporation https://nanograf.com/	U.S	Patented doped silicon alloy material architecture The specific blend of silicon-based alloys and a protective inorganic and organic coating assists in the stabilization of the active material during charge and discharge processes
Sila Technologies https://www.silanano.com/	U.S	Titan Silicon™
Group14 Technologies, Inc.https://www.group14.technology/	U.S	SCC55™, a silicon-carbon composite powder which employs a unique hard carbon-based scaffolding to retain silicon in its amorphous, nano-sized and carbon-encased form, which consists of integrated intra-particle void space, with low surface area, and is made with readily available ultra-high purity materials
BTR New Material Group Co.https://www.btrchina.com/en/	China	Large-scale production of silicon-carbon (Si–C) composite anodes
Shanshan Technology https://shanshantech.com/	China	Silicon-oxygen and silicon-carbon composite materials
Putailai (Jiangxi Zichen Technology) https://www.putailai.com/	China	Silicon-oxygen and silicon-carbon composite materials
Zeto Energy (Jiangxi Zhengtuo New Energy Technology) http://www.jxzeto.com/	China	Silicon-carbon and artificial/natural graphite composite materials
Shenzhen XFH Technology http://www.xiangfenghua.com/	China	Nano-silicon and silicon oxides as additives for commercial anode blends
Hunan Zhongke Xingcheng Graphite (Hunan Zhongke Electric) https://en.shinzoom.com/cpzx_xq/3.html?utm_source=chatgpt.com	China	Nano-silicon and Si–C
E-magy https://e-magy.com/	Netherlands	Nano-porous silicon obtained from metallurgical grade silicon

These companies have made significant advancements in scaling up the manufacturing of silicon-graphite composite anodes, which combine the high energy density of silicon with the stability of graphite to enhance battery performance. BTR, in particular, is a global leader and pioneer in this field, having successfully industrialized silicon-carbon composite anodes and supplying major battery manufacturers such as CATL and BYD. At the same time, companies such as Putailai and Shanshan are scaling up production and intensifying research and development efforts to enhance the durability and cost-effectiveness of their silicon anode materials, further strengthening China's leadership in the global lithium-ion battery materials market [24, 47, 51]. Shenzhen XFH Technology develops nano-silicon and silicon oxides as additives for commercial anode blends, while Hunan Zhongke Xingcheng Graphite is active in the production of nano-silicon and Si–C in collaboration with Chinese research institutes.

In addition to the major Chinese players, numerous startups and emerging companies worldwide are making significant contributions to silicon-based anode development. A notable example is Group14 Technologies, a U.S.-based startup specializing in silicon–carbon composite anodes through its proprietary SCC55™ technology. Group14 has attracted major investments from industry leaders such as Porsche, SK Inc., and BASF, and is building commercial-scale production facilities in both the United States and South Korea. Other firms, including Enevate, Sila Nanotechnologies, and OneD Battery Sciences, are also advancing innovative silicon anode concepts, ranging from silicon nanowire architectures to silane-based composites. Although these startups do not yet match the production volumes of established Chinese manufacturers, they play a crucial role in driving innovation and offering high-performance anode alternatives for next-generation batteries, especially for electric vehicles and consumer electronics.

Table 3 outlines the geographic and technological landscape of companies working on silicon-based anode materials (with further details in Appendix F); however, it should be noted that this is not an exhaustive list. For example, U.S. firms such as Amprius Technologies and Enovix Corporation are making progress in developing high-energy, safer silicon-based batteries. Similarly, Enevate Corporation and NanoGraf Corporation focus on enhancing both the energy density and lifespan of batteries for consumer electronics. Sila Technologies is not only advancing silicon battery technology but also scaling production to meet the expected surge in demand. In Europe, E-magy stands out as a promising technology company supplying nanoporous silicon for high-energy LIBs, using cost-effective upgraded metallurgical-grade silicon that is abundantly available and environmentally friendly.

4 Conclusions

While the future of silicon anode materials in lithium-ion batteries is promising due to their potential for significantly higher energy density, several challenges must be addressed before widespread adoption can occur. These challenges range from technical hurdles – such as managing silicon's substantial volume expansion during charge–discharge cycles and ensuring long-term cycle stability—to broader issues like manufacturing scalability, cost-effectiveness, and supply chain complexities. Overcoming these obstacles will be crucial for silicon anodes to realize their potential in next-generation battery technologies, including electric vehicles and portable electronics. Additionally, a price premium for silicon materials is expected to persist in the medium term, reflecting current production costs and technical challenges. However, the strong long-term outlook for electric vehicles, ongoing advancements in silicon anode technologies, and increasing production capacities underscore the significant growth potential of this market. As efficiency improves and economies of scale are realized, silicon anodes are positioned to become a key enabler of next-generation lithium-ion batteries.

Beyond consumer electronics, the reach of silicon anode technology is broadening, with increasing adoption in electric vehicles, medical devices, and even aerospace applications. This trend is driven by the unique advantages silicon brings in terms of energy storage and longevity, making it a promising solution for industries requiring compact, high-capacity, and durable energy sources. The review thus paints a picture of a transformative shift in battery technology, powered by the move toward silicon, with the potential to redefine energy storage across multiple sectors. Besides exploring methods to overcome the drawbacks associated with the use of silicon as anode material in LIBs, the review wants also to join the academic research with the industrial development for the next generation of high-energy density lithium-ion batteries. A particular attention is posed onto the sustainability aspects of the electrode design, that see for example the recovering of silicon material from PV waste but also the use of environmentally friendly, water-soluble binders as some of the proposed future not only academic but above all industrial answers.

Abbreviations Alg: Alginate; ASEI: Artificial Solid Electrolyte Interphase; CaF_2: Calcium fluoride; CNTs: Carbon nanotubes; CM: Carboxymethylation method; CNFs: Carbon nanofibers; CMC: Carboxymethyl cellulose; CEI: Cathode electrolyte interphase; β-CD: β-Cyclodextrins; CVD: Chemical vapor deposition; CE: Coulombic efficiency; CCs: Current collectors; 2DS: 2 Degrees scenario following the Paris Agreements; DEC: Diethyl carbonate; DMC: Dimethyl carbonate; EoL: End of life; EC: Ethylene carbonate; EVA: Ethylene-vinyl acetate; EMC: Ethyl methyl carbonate; EPIA: European Photovoltaic Industry Association; FREP: Flame-retardant epoxy resin; FEC: Fluoroethylene carbonate; GG: Guar Gum; HRTEM: High resolution transmission electron microscopy; ICE: Initial Coulombic efficiency; IP: Intellectual property; l-c: Life Cycles; LCO: Lithium cobalt oxide; LIBs: Lithium-ion batteries; LMO: Lithium-ion manganese oxide; MOFs: Metal-organic frameworks; MO: Metallic oxide; NCA: Lithium-ion battery with nickel, cobalt, aluminum oxide cathode; NMCO: Lithium-ion battery with nickel, manganese, cobalt oxide cathode; MOF: Metal-Organic Frameworks; n.cV: Nominal cell voltage; PV: Photovoltaic; PAA: Polyacrylic acid; NaPAA-g-CMC: Poly (acrylic acid sodium)-grafted-carboxymethyl cellulose; PANI: Polyaniline; PPy: Polypyrrole; PEDOT: Poly(3,4-ethylenedioxythiophene); PET: Polyethylene Terephthalate; PVA-g-PAA: Polyvinyl alcohol -grafted- Polyacrylic acid; PVDF: Polyvinylidene fluoride; Si/C: Silicon-carbon; SiN_x: Silicon nitride; Si NPs: Si nanoparticles; Si NWs: Si nanowires; Si NSs: Si nanosheets; SEI: Solid electrolyte interphase; SE: Specific Energy; SHE: Standard hydrogen electrode; SRM: Strategic Raw Materials; TRL: Technology Readiness Level; VC: Vinylene carbonate; WEEE: Waste Electrical and Electronic Equipment

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Declarations

Competing interest The authors declare no competing interests.

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