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The Influence of Silicon Anode Morphology on Energy Storage Performance in Lithium-Ion Batteries

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Abstract. The rapid electrification of transportation, together with the continued growth of portable electronics and renewable-energy storage, has increased demand for lithium-ion batteries with higher energy density and longer cycle life. Silicon is widely regarded as a promising alternative to graphite because of its exceptionally high theoretical capacity and low lithiation potential. However, practical application remains limited by severe volume expansion, unstable solid electrolyte interphase (SEI) formation, and poor electrical conductivity. This review examines how silicon-anode morphology affects lithium-storage performance by comparing four representative structures: silicon nanoparticles, nanotube and tube-like architectures, bulk or micron-sized silicon, and thin films. The comparison shows that each morphology offers a different balance among lithium-ion transport, mechanical stability, electrode density, and manufacturability. Nanostructured silicon generally improves stress tolerance and reaction kinetics, whereas larger or denser structures offer advantages in packing density, cost, and industrial compatibility. No single morphology is universally superior. Future progress will therefore depend on combining multiscale structural design with stable interfaces, suitable binders and electrolytes, and scalable manufacturing methods to translate silicon's high theoretical capacity into durable full-cell performance.

Keywords: lithium-ion batteries, silicon anodes, morphology, energy-storage performance, cycling stability

1. Introduction

The rapid growth of portable electronic devices, electric vehicles, and renewable-energy storage systems has created increasing demand for rechargeable batteries with higher energy density, longer cycle life, faster charging capability, and improved safety. Lithium-ion batteries (LIBs) have become the dominant energy-storage technology because of their high energy and power densities, low self-discharge rate, and relatively good cycling stability. However, further increases in the energy density of commercial LIBs are increasingly constrained by conventional graphite anodes. Graphite has a theoretical specific capacity of only 372 mAh g^{-1}, which limits the amount of lithium that can be stored per unit mass of active material [1].

Silicon has therefore attracted considerable attention as a promising alternative anode material. When fully lithiated to form $Li_{15}Si_4$, silicon can deliver a theoretical specific capacity of

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approximately 3579 mAh g^{-1}, nearly ten times that of graphite. It also has a relatively low lithiation potential of approximately 0.4 V versus Li^+/Li, which helps maintain a high operating voltage in a full cell [2]. In addition, silicon is naturally abundant and potentially suitable for large-scale battery production. These characteristics suggest that replacing or supplementing graphite with silicon could substantially increase the energy density of lithium-ion batteries.

Nevertheless, silicon's high theoretical capacity does not automatically translate into stable practical performance. During full lithiation, silicon may expand by approximately 300–400%, generating substantial mechanical stress within the electrode. Repeated expansion and contraction can crack or pulverize particles, disrupt electrical contact, and accelerate capacity loss. The same dimensional changes repeatedly fracture and reform the SEI, consuming active lithium and electrolyte while increasing interfacial resistance. Silicon's relatively low intrinsic electrical conductivity further limits electron transport and contributes to poor rate capability. Large volume changes, unstable interfaces, and low conductivity therefore remain the principal barriers to practical silicon anodes [3].

Morphology engineering is one of the most widely investigated approaches to these problems. Nanoparticles, nanowires, nanotubes, porous structures, and thin films can shorten lithium-ion diffusion pathways and accommodate mechanical deformation more effectively than conventional dense silicon. However, increasing surface area can also promote electrolyte decomposition and excessive SEI formation, reducing initial Coulombic efficiency. Carbon coatings, conductive matrices, advanced binders, electrolyte additives, and prelithiation can provide additional support, but their effectiveness is strongly influenced by the geometry and surface area of the silicon itself [4]. Thus, silicon morphology must be evaluated as a balance among stress accommodation, ion and electron transport, interfacial stability, and electrode density.

Accordingly, this review first summarizes the lithium-storage mechanism and the main morphology-related challenges of silicon anodes. It then compares four representative forms - nanoparticles, nanotube and tube-like structures, bulk or micron-sized silicon, and thin films - in terms of preparation, advantages, limitations, and practical relevance. The final section discusses technical innovation, scalable manufacturing, and development trends. The purpose is to clarify how morphology affects electrochemical performance and to identify design principles that can support higher-energy and longer-lasting lithium-ion batteries.

2. Progress of silicon anode materials and lithium-storage mechanism

Silicon stores lithium through an alloying reaction rather than the intercalation mechanism used by graphite. During the initial lithiation of crystalline silicon, lithium first forms amorphous Li_xSi phases; at sufficiently deep lithiation and a low cutoff voltage, crystalline $Li_{15}Si_4$ may subsequently form. During delithiation, the material does not simply return to its original crystalline state but generally evolves through amorphous and partially crystalline silicon-rich phases. These repeated phase transformations break and reform Si-Si bonds, generating large internal stresses that progressively damage the electrode. Morphology is therefore not a secondary structural detail: it directly influences stress relaxation, lithium transport, electrical continuity, and interfacial stability during repeated charge–discharge cycles [5].

Recent research has shifted from maximizing half-cell capacity toward identifying structures that can operate under more practical conditions. Greater attention is now given to cycle life, initial Coulombic efficiency, mechanical durability, areal loading, electrode density, and manufacturability. This shift highlights a central trade-off. Making silicon smaller, more porous, or more mechanically compliant can suppress fracture and shorten diffusion pathways, but excessive nanostructuring

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increases surface area, parasitic reactions, and inactive-material demand while reducing tap density and volumetric energy density. The most effective morphology is therefore not necessarily the smallest or most complex; it is the one that best balances transport, mechanical stability, interfacial chemistry, electrode density, and scalable fabrication.

3. Different morphologies of silicon anodes

3.1. Silicon nanoparticles

Silicon nanoparticles are among the most widely studied silicon morphologies because nanosizing provides a direct means of reducing fracture. They are commonly produced by magnesiothermic reduction, high-energy ball milling, vapor-phase synthesis, spray drying, or solution-based synthesis followed by thermal treatment. These methods differ in cost, yield, particle-size control, purity, and suitability for scale-up. The principal advantage of nanoparticles is that their small dimensions distribute strain more evenly and shorten lithium-ion diffusion pathways. Below a critical particle size, fracture becomes significantly less likely, making nanoparticles useful for studying the effects of size, surface chemistry, and coating design on electrochemical performance [6].

Nanoparticles are also highly compatible with surface and interface engineering. Carbon, metal-oxide, or artificial-interphase coatings can be applied relatively uniformly, which is why nanosilicon is frequently used in core-shell, multilayer, yolk-shell, and hybrid composite structures. A 2024 study of double-layer-coated silicon nanoparticles reported that combined carbon and TiO_2 protection improved structural and interfacial stability [7]. However, the same high surface area that enables effective coating also accelerates electrolyte decomposition and repeated SEI formation. This consumes active lithium, lowers initial Coulombic efficiency, and can increase the need for binders, conductive additives, electrolyte, or prelithiation. Agglomeration and low tap density may further reduce volumetric energy density [8].

For practical batteries, silicon nanoparticles are therefore more attractive as components of engineered composite anodes than as unsupported active material. They are especially useful when high specific capacity and rapid lithium transport are prioritized. In electric-vehicle cells, a more realistic route is to incorporate nanosilicon at a controlled fraction in silicon-graphite blends or to assemble it into denser secondary particles. This preserves some of the mechanical benefits of nanosizing while reducing surface reactivity and maintaining compatibility with existing slurry-coating processes.

3.2. Silicon nanotubes and CNT-supported tube-like architectures

Silicon nanotubes and related tube-like architectures address mechanical degradation primarily through geometry rather than size reduction alone. Their hollow interior provides space into which the silicon wall can expand during lithiation, limiting outward deformation and reducing the risk of catastrophic fracture. The thin wall also shortens radial lithium-ion diffusion distances, while a connected one-dimensional framework can provide continuous pathways for electron transport. Common preparation routes include template-assisted deposition, chemical vapor deposition, reduction of silica nanotubes, and the formation of silicon within carbon-nanotube frameworks [9].

Recent studies often focus on silicon-carbon nanotube composites rather than isolated, free-standing silicon nanotubes because a conductive tubular framework can provide both mechanical support and electrical continuity. In a 2024 study, interfacial Si-C bonding in a Si/CNT composite produced an initial reversible capacity of approximately 1100.2 mAh g^{-1} at 0.2 A g^{-1}, retained 922.4

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mAh g^{-1} after 200 cycles, and delivered 612.3 mAh g^{-1} after 1000 cycles. Other work has confined silicon particles inside carbon nanotubes, where the tubular framework buffered large volume changes and improved reversibility and cycling stability [10]. These results show that the value of tube-like design lies not only in the hollow shape itself but also in its ability to preserve mechanically adaptive and electrically continuous pathways.

The major limitations of nanotube architectures are fabrication complexity and low packing density. Template methods, multistep deposition, reduction, or etching can increase cost and complicate large-scale manufacturing, while internal void space may lower volumetric capacity even when cycle life improves. Nanotube-based silicon is therefore most suitable at present for high-performance research electrodes, flexible devices, and systems that benefit from directional transport or self-supporting conductive frameworks. Broader commercial use will require simpler synthesis and denser electrode assembly without eliminating the internal space needed for expansion.

3.3. Micron-sized silicon

Although nanostructures dominate much academic research, bulk and micron-sized silicon remain highly relevant because they are closer to practical industrial materials. Here, bulk silicon refers mainly to dense silicon particles or domains with micrometer-scale dimensions rather than a single monolithic block. Such materials can be produced by crushing, milling, and classifying commercial silicon or by recovering silicon-rich waste from semiconductor and photovoltaic manufacturing. Compared with nanoparticles, micron-sized silicon generally provides higher tap density, lower specific surface area, lower processing cost, and better compatibility with conventional slurry mixing, coating, and calendaring. Its lower surface area can also reduce parasitic reactions and improve initial Coulombic efficiency, making it attractive for high-areal-loading electrodes [11].

The main weakness of micron-sized silicon is severe mechanical fracture during cycling. Larger particles develop stronger lithium-concentration and stress gradients, so cracks can propagate during lithiation. Once fracture occurs, fresh silicon surfaces are exposed, leading to repeated SEI formation, loss of active lithium, and electrical isolation of fragments. Recent work therefore seeks to retain the density and cost advantages of micron silicon while controlling mechanical and interfacial failure through protective coatings, artificial interphases, conductive networks, internal porosity, optimized electrolytes, and mechanical confinement. A 2024 study reported that volume confinement improved SEI uniformity and enabled a silicon-based anode to retain a specific capacity of 937 mAh g^{-1} after 200 cycles [12].

Micron silicon represents a different design philosophy from extreme nanosizing: rather than avoiding stress primarily through very small dimensions, it attempts to manage stress through confinement, interface regulation, and electrode engineering. Its density and compatibility with established manufacturing make it particularly relevant to silicon-graphite composites and potentially to future silicon-dominant anodes. However, successful implementation still requires robust conductive support, stable interfaces, and carefully designed binders or electrolytes.

3.4. Thin-film silicon

Silicon thin-film anodes consist of continuous or patterned silicon layers deposited directly onto a current collector or substrate rather than dispersed as particles in a conventional composite electrode. A variety of deposition techniques have been investigated, including chemical vapor deposition (CVD), physical vapor deposition (PVD), pulsed-laser deposition (PLD), magnetron

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sputtering, vacuum evaporation, electron-beam evaporation, and electrodeposition. These methods allow the thickness, composition, density, and surface morphology of silicon films to be controlled with relatively high precision. The choice of deposition method also influences film uniformity, adhesion to the substrate, production cost, and electrochemical performance. Thin films are therefore particularly useful for studying the relationship between silicon structure, mechanical stress, and solid electrolyte interphase formation.

Different deposition methods offer distinct advantages and limitations. Physical and vapor-phase techniques such as magnetron sputtering, PVD, and CVD can produce highly uniform and controllable silicon layers, but they often require specialized equipment, relatively high energy consumption, or slow deposition rates, which can limit large-scale production. Electrodeposition offers a potentially simpler and lower-cost alternative because silicon can be deposited directly onto a conductive substrate under comparatively mild conditions. For example, Feng et al. prepared amorphous silicon thin films on copper foil by pulsed electrodeposition and reported a reversible capacity of approximately 1200 mAh g^{-1} with no obvious capacity fading after 100 cycles at a current density of 200 mA g^{-1} [13]. This result illustrates how both deposition method and film structure can influence the cycling stability of silicon anodes.

The main advantages of thin-film silicon are its structural uniformity, well-defined geometry, short lithium-ion diffusion pathways, and direct electrical contact with the current collector. These characteristics reduce some of the contact losses and structural heterogeneity found in conventional powder-based electrodes. However, a fundamental trade-off exists between mechanical stability and practical loading. Very thin films can better accommodate lithiation-induced stress but contain insufficient active material to provide high areal capacity. Increasing film thickness raises the amount of active silicon, but thicker films experience greater mechanical stress during repeated expansion and contraction and may eventually crack or delaminate from the current collector. In addition, many high-quality deposition techniques remain relatively expensive and difficult to scale.

Overall, silicon thin films are particularly valuable for mechanistic studies because their thickness and interface with the current collector can be precisely controlled. They may also be suitable for microbatteries, integrated electronics, flexible devices, and other applications where precise structural control is more important than very high active-material loading. For conventional large-format lithium-ion batteries, however, limited areal capacity, mechanical instability at greater film thickness, and manufacturing cost remain major barriers. Future development will therefore require deposition methods that combine good structural control and strong interfacial adhesion with higher throughput and lower production cost.

3.5. Comparative assessment of morphology and performance

A useful comparison of silicon morphologies must consider more than specific capacity alone. Nanostructures often perform well because short diffusion distances and improved stress accommodation protect the active material, but the same features can increase surface reactivity and reduce packing density. Denser micron-scale materials move in the opposite direction: they are easier to process and can support higher electrode density, yet their larger dimensions make stress management more difficult. Thin films provide unusually clear control over thickness and interfacial contact, whereas tubular structures emphasize internal expansion space and continuous transport pathways. These differences show that morphology changes not only electrochemical behavior but also the practical way in which an electrode can be manufactured, loaded, and cycled.

For this reason, the most suitable morphology depends on the intended application and the performance metric being prioritized. Laboratory studies may favor nanoparticles, nanotubes, or thin

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films because their geometry can isolate specific mechanisms and produce strong rate or cycling behavior. Commercial electrodes, however, must also meet requirements for areal loading, electrode density, initial efficiency, scalable processing, and cost. The comparison therefore supports a broader design principle: morphology should be selected as part of an electrode system rather than judged as an isolated material property. This perspective helps explain why current research increasingly combines nanoscale features with micron-scale particles, conductive supports, protective interfaces, and compatible processing methods.

4. Suggestions and future prospects for silicon anodes

The comparison above shows that silicon-anode development is moving away from optimizing a single structural feature in isolation. The central problem is no longer simply how to reduce particle size or create additional free volume, but how to preserve the advantages of structural engineering under realistic electrode conditions. Mechanical stability and interfacial control must therefore be considered together with electrode density, loading, cost, and manufacturability. Future designs are likely to integrate features from several morphology classes rather than rely on one idealized structure.

4.1. Technical innovation

Future technical development should focus on multiscale and hierarchical structures rather than simple size reduction. Micron-sized secondary particles containing nanoscale silicon domains, for example, could combine high packing density and industrially convenient handling with short diffusion pathways and improved stress tolerance. Porous cores, yolk-shell structures, gradient coatings, and interconnected conductive networks may provide controlled expansion space, but pore volume must be optimized: insufficient void space cannot relieve stress, whereas excessive porosity increases electrolyte consumption and lowers electrode density.

Morphology must also be designed together with the binder, electrolyte, and SEI. Elastic or self-healing binders, artificial interphases, conductive coatings, and optimized electrolyte additives are most effective when matched to the geometry and surface area of the silicon. Future studies should therefore place greater emphasis on realistic full-cell conditions and report areal capacity, electrode density, initial Coulombic efficiency, electrolyte quantity, and electrode swelling. Operando microscopy, spectroscopy, tomography, and stress measurements can further connect structural evolution with electrochemical failure and guide morphology design under practical conditions.

4.2. Cost reduction and scalable manufacturing

Cost reduction will depend less on creating increasingly complicated nanostructures than on simplifying production while maintaining structural reliability. Silane-based synthesis, high-vacuum deposition, and sacrificial templates can produce highly controlled structures, but their equipment, energy, and precursor requirements may be unsuitable for large-scale electric-vehicle battery production. More scalable routes include mechanical milling, solution processing, reduction of inexpensive silica sources, and recovery of silicon from photovoltaic or semiconductor waste. Water-based binders and low-toxicity processing can further reduce environmental and manufacturing costs.

Industrial assessment must also consider more than the price of silicon powder. Energy use, purification, solvent recovery, production yield, inactive-material content, safety, and expected cycle

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life all affect the true cost of an electrode. A slightly lower-capacity morphology that can be manufactured reproducibly at high yield may be more valuable than a record-setting nanostructure that requires multiple templates, coating steps, and washing procedures. Dense micron-scale secondary particles containing nanoscale internal features may therefore offer a practical compromise between structural control and scalable handling.

4.3. Development trends

In the near term, commercial lithium-ion batteries are likely to increase silicon content gradually through silicon-graphite blended anodes. Graphite can help limit electrode swelling and preserve compatibility with established manufacturing lines, while silicon increases capacity. As binders, electrolytes, prelithiation, coatings, and particle structures improve, silicon-dominant anodes may become more realistic. The broader trend is therefore toward engineered particles that combine micron-scale density with nanoscale or porous features where they provide the greatest mechanical and interfacial benefit.

Another important direction is the integration of silicon with solid-state batteries. Solid electrolytes may improve safety and energy density, but silicon expansion can create voids and high-resistance gaps at solid-solid interfaces. Elastic electrolytes, pressure-free cell designs, and three-dimensional interfacial networks are therefore important research directions. More generally, future work should evaluate morphology under realistic cell constraints rather than focusing only on the highest half-cell capacity. Useful full-cell performance will require practical loading, limited electrolyte, controlled swelling, high initial efficiency, consistent testing, and acceptable manufacturing cost.

5. Conclusion

This review shows that silicon-anode morphology governs a fundamental trade-off among lithium-ion transport, mechanical stability, interfacial stability, electrode density, and manufacturability. Nanoscale structures improve reaction kinetics and stress tolerance, whereas denser micron-scale structures offer advantages in volumetric energy density and industrial processing. Therefore, no single morphology is optimal for every application. Hierarchical designs that combine micron-scale packing density with nanoscale stress-relief features appear to provide the most promising balance between electrochemical performance and practical manufacturability.

Future materials should be assessed using practical indicators such as initial Coulombic efficiency, areal capacity, volumetric energy density, electrode swelling, cycle life, production cost, and compatibility with full-cell manufacturing. Future research should also integrate morphology engineering with conductive networks, elastic binders, artificial interphases, optimized electrolytes, and prelithiation strategies. Greater attention should be given to scalable and low-cost preparation methods, standardized testing procedures, realistic electrode loadings, and limited-electrolyte conditions. By coordinating structural design, interface control, and manufacturing requirements, future studies may translate silicon's high theoretical capacity into stable full-cell performance. This progress could support the development of lithium-ion batteries with higher energy density and longer service life for electric vehicles, portable electronics, and renewable-energy storage systems.

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