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Composite Design of Silicon-Based Anode Materials for High-Performance Lithium-Ion Batteries: A Systematic Review

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Abstract. Pure silicon anodes are regarded as promising candidates for high-energy lithium-ion batteries, yet their practical use is still limited by severe mechanical degradation and sluggish charge-transfer behavior. Rather than treating silicon as an isolated active phase, recent studies increasingly design it as part of integrated composite systems in which each component performs a specific function. This review reorganizes the progress of silicon-based composites from four perspectives: carbon-supported structures that provide conductive and deformable frameworks, metal-containing hybrids that reinforce the electrode and reduce impedance, silicon oxide-related heterostructures that generate interfacial buffer phases, and polymer/inorganic architectures that combine adhesion, elasticity, and interphase regulation. Emphasis is placed on the cooperative roles of different phases in relieving volume strain, preserving electrical contact, stabilizing the SEI, and promoting ion/electron transport. The discussion aims to clarify the design logic of advanced silicon-based anodes and to provide guidance for constructing practical composite electrodes for next-generation high-energy lithium-ion batteries.

Keywords: silicon-based composite anodes, lithium-ion batteries, volume variation, structural regulation, synergistic mechanism

1. Introduction

Lithium-ion batteries (LIBs) have become one of the most widely used electrochemical energy-storage systems because they can support portable electronics, electric vehicles, and other clean-energy applications [1]. Nevertheless, the energy density of current commercial cells is increasingly restricted by graphite anodes, whose theoretical capacity is only 372 mAh g^{-1} [2]. To meet the demand for batteries with higher energy output, many alternative anode chemistries have been investigated. Among them, silicon (Si) is especially attractive because its theoretical capacity can approach 4200 mAh g^{-1} when forming $Li_{4.4}Si$, far exceeding that of graphite [3, 4]. In addition, silicon operates at a relatively low lithiation potential and benefits from abundant reserves and environmental compatibility, which gives it clear potential for upgrading present LIB technology.

However, the advantages of silicon are accompanied by pronounced structural and interfacial instability [5]. During repeated lithiation and delithiation, silicon experiences volume changes that can exceed 300%, producing large internal stresses and causing cracking, pulverization, and loss of electrical connection with the conductive network or current collector [4, 6]. The same dimensional fluctuation also repeatedly damages the solid electrolyte interphase (SEI). Continuous rupture and rebuilding of the SEI consume active lithium and electrolyte, lower the initial Coulombic efficiency, and increase cell resistance during long-term cycling [4, 7]. For this reason, silicon anode research has gradually shifted from simply using pure Si particles to constructing silicon-based composites [8]. In such composites, silicon acts as the high-capacity component, while the surrounding host phase is designed to provide mechanical constraint, conductive pathways, ion-transport channels, or interfacial protection. The performance improvement therefore comes from the cooperative action of multiple components rather than from silicon alone.

According to the function and composition of the auxiliary phase, the following sections discuss four representative families of silicon-based composites. Silicon/carbon systems take advantage of the structural diversity and high conductivity of carbon frameworks. Silicon/metal composites employ metallic or metal-compound phases to enhance mechanical support and reduce charge-transfer resistance. Silicon oxide-based materials exploit conversion-derived products and heterointerfaces to buffer expansion and regulate the SEI. Silicon/polymer composites introduce flexible, adhesive, or chemically functional networks that can maintain electrode integrity and promote interphase stability. Across these systems, an effective composite design should simultaneously accommodate volume strain, retain continuous ion/electron transport, and guide the formation of a stable and reversible interfacial layer [9].

2. Silicon/carbon composite anodes

Combining silicon with carbon is one of the most established approaches for mitigating the major weaknesses of Si anodes. Carbon can improve electronic conductivity, dilute local stress, and protect active particles from direct exposure to the electrolyte. Recent work has moved beyond simple carbon coating and has focused on chemical bonding at the interface, engineered pores or voids, dense three-dimensional assemblies, and controlled particle size. These strategies allow silicon/carbon anodes to better balance high capacity, structural durability, and cycling stability.

2.1. Interfacial bonding and covalent anchoring

Stable contact between silicon and the carbon phase is essential for preventing active-material isolation during cycling. Guo et al. obtained a Si/SiC/CNT three-phase heterostructure through one-step magnesiothermic reduction [10], as presented in Figure 1a. In this architecture, an interfacial SiC layer generated by Si–C bonding serves as a chemical anchor that fixes Si nanoparticles on carbon nanotubes. This strong interface helps suppress particle detachment and enables the electrode to operate for 1000 cycles at 5 A g^{-1} (Figure 1b). Tian et al. used the alkaline environment and steric effect associated with polydopamine to form ultrafine SiO_2 quantum dots on a carbon framework [11]. After magnesiothermic reduction, NC@Si nanospheres were produced (Figure 1c). The small Si domains reduce expansion-induced damage, while the in-situ-formed thin Si_3N_4 layer provides mechanical reinforcement and stable multipoint electrical contact with the N-doped carbon matrix. As a result, the electrode delivered a capacity retention of 75.4% after 100 cycles at 0.1 A g^{-1}.

2.2. Void buffering and spatial architecture

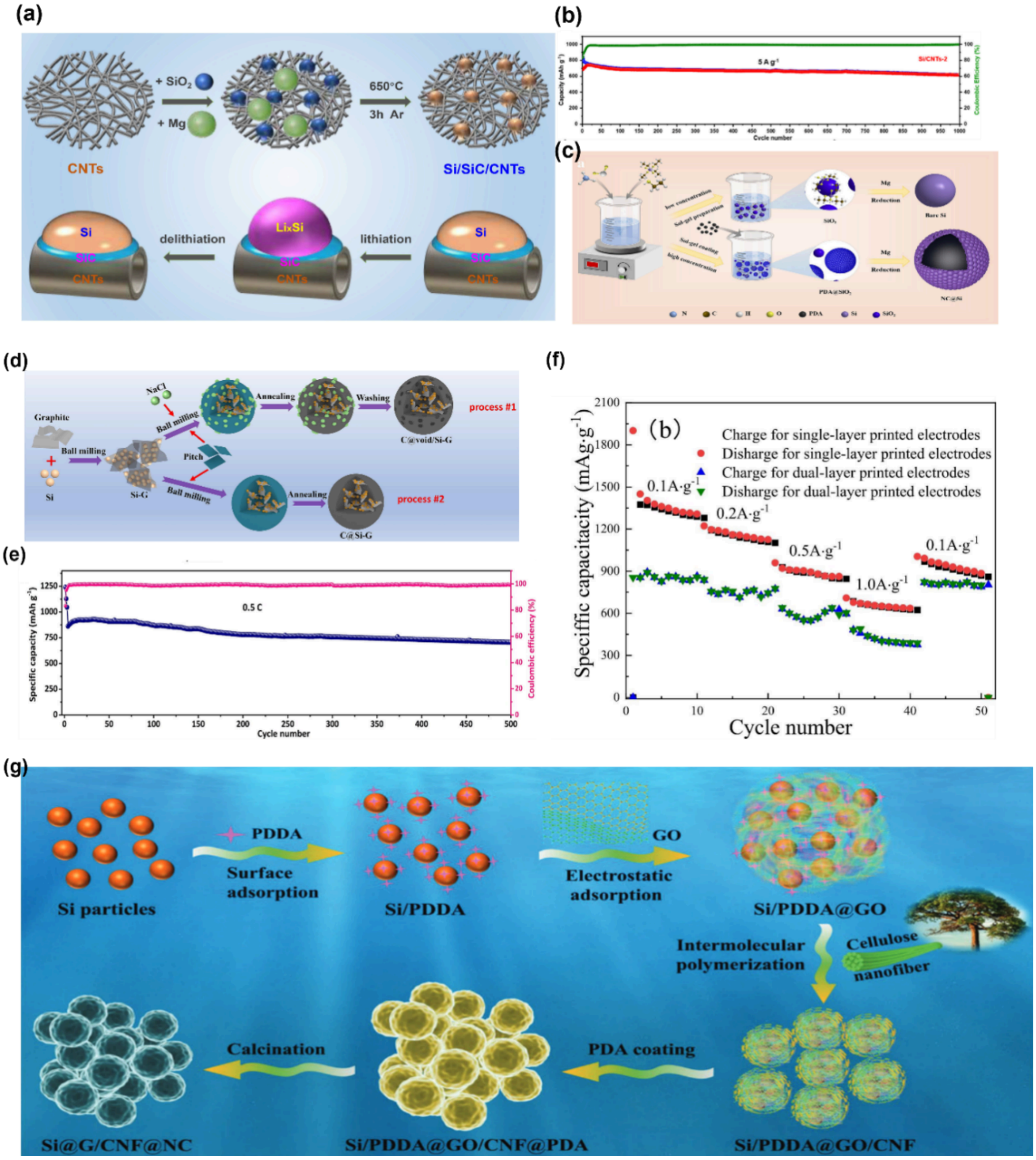

Figure 1. Representative silicon/carbon composite designs: (a) synthesis and lithium-storage mechanism of Si/SiC/CNTs [10]; (b) cycling behavior of Si/SiC/CNTs [10]; (c) preparation route for NC@Si [11]; (d) formation of the C@void/Si-G composite [12]; (e) cycling stability of C@void/Si-G [12]; (f) rate capability of the bilayer free-standing Si/C anode [13]; and (g) structural design of Si@G/CNF@NC [14]

Another important design principle is to reserve internal space that can absorb the expansion of silicon. Shi et al. fabricated a porous C@void/Si-G composite with NaCl as a removable template [12] (Figure 1d). In this material, graphite contributes electronic conduction, pyrolytic carbon reduces side reactions, and the pores generated after salt removal provide room for Si expansion.

The electrode retained 81.9% of its capacity after 500 cycles at 0.5 C (Figure 1e), while the overall volume increase of the electrode was limited to about 41%. Zhang et al. further reported a bilayer free-standing Si/C anode prepared by microelectronic printing [13]. The electrode contains layered slit-like spaces narrower than 50 nm, which act as elastic reservoirs for volume change. Together with an amorphous carbon coating, this structure maintains the integrity of the electrode and gives excellent rate recovery, with 98.67% capacity retention when the current density returned from 1.0 A g^{-1} to 0.1 A g^{-1} (Figure 1f).

2.3. Three-dimensional networks for high-loading electrodes

For practical application, Si anodes must also overcome the problems of low tap density and limited areal capacity. Zhang et al. developed a densely packed Si@G/CNF@NC composite through electrostatic assembly followed by polydopamine-derived carbon encapsulation [14] (Figure 1g). The integrated three-dimensional conductive framework increased the tap density to 0.78 g cm^{-3} and constrained the volume expansion to below 17.6%. Even at a high active-material loading of 10.8 mg cm^{-2}, the electrode achieved a volumetric capacity of 2197.7 mAh cm^{-3} and retained 91.1% of its capacity after 100 cycles, indicating that dense conductive networks are valuable for moving Si/C composites toward high-energy practical electrodes.

3. Silicon/metal and metal-compound composites

In addition to carbon, metal-related components can introduce functions that are difficult to achieve with soft carbon matrices alone. Metals, metal oxides, sulfides, and silicides may serve as mechanically rigid supports, highly conductive networks, or electrochemically active buffers. Their inclusion can improve charge transfer, strengthen the electrode skeleton, and help stabilize the Si host during repeated volume variation.

3.1. Intermetallics and alloys as conductive scaffolds

Silicides and alloy phases can build internal conductive frameworks that protect structural continuity during cycling. Wang et al. synthesized Si/$TiSi_2$ nanocomposites by molten-salt electrolysis [15] (Figure 2a). The $TiSi_2$ phase has high electrical conductivity and helps both electron transport and stress buffering, allowing the composite to retain 1133.8 mAh g^{-1} after 200 cycles at 200 mA g^{-1} with an initial Coulombic efficiency of 83.99% (Figure 2b,c). Li et al. prepared a phosphorus-doped silicon-copper alloy composite (P-doped Si-Cu) [16] (Figure 2d). In this system, the relatively inert Cu_3Si phase mitigates expansion, while phosphorus doping produces n-type Si and improves conductivity. The optimized electrode maintained 1048 mAh g^{-1} after 60 cycles at 100 mA g^{-1} (Figure 2e), and the charge-transfer resistance decreased markedly to 73.65 Ω.

3.2. Metal oxides and sulfides for interfacial stabilization and potential regulation

Metal oxides and sulfides are often introduced as protective layers or active matrix phases because they can stabilize the SEI and buffer volume change in a staged manner. Vats et al. combined graphite with Si@TiO_2 core-shell nanoparticles [17] (Figure 2f). The TiO_2 shell alleviated Si expansion, and the composite containing 10% core-shell particles retained 75% of its capacity after 100 cycles, outperforming the pure silicon electrode. Liu et al. designed a ZnS and nitrogen-doped carbon co-modified SiO composite (ZnS&NC/SiO) [18] (Figure 2g). Because ZnS and SiO react with lithium at different potentials, their volume changes can partly compensate each other during cycling, while the N-doped carbon coating enhances electronic conduction. This cooperative design enabled the electrode to retain 748 mAh g^{-1} after 1000 cycles at 1.0 A g^{-1} (Figure 2h).

3.3. Metallic networks and nanoscale conductive phases

Highly conductive metallic nanophases can also improve interface kinetics while reinforcing the electrode structure. Kang et al. constructed a three-dimensional branched polyimide/silver interfacial layer (Si@PMTI-Ag) on silicon particles [19]. In this design, ultrafine Ag nanoclusters with a size of approximately 2.5 nm are embedded within the polyimide network (Figure 2i), giving the coating both mechanical robustness and dual ion/electron transport capability. Owing to this integrated interface, the electrode retained a reversible capacity of 1103.9 mAh g^{-1} after 350 cycles at 1 A g^{-1} (Figure 2j).

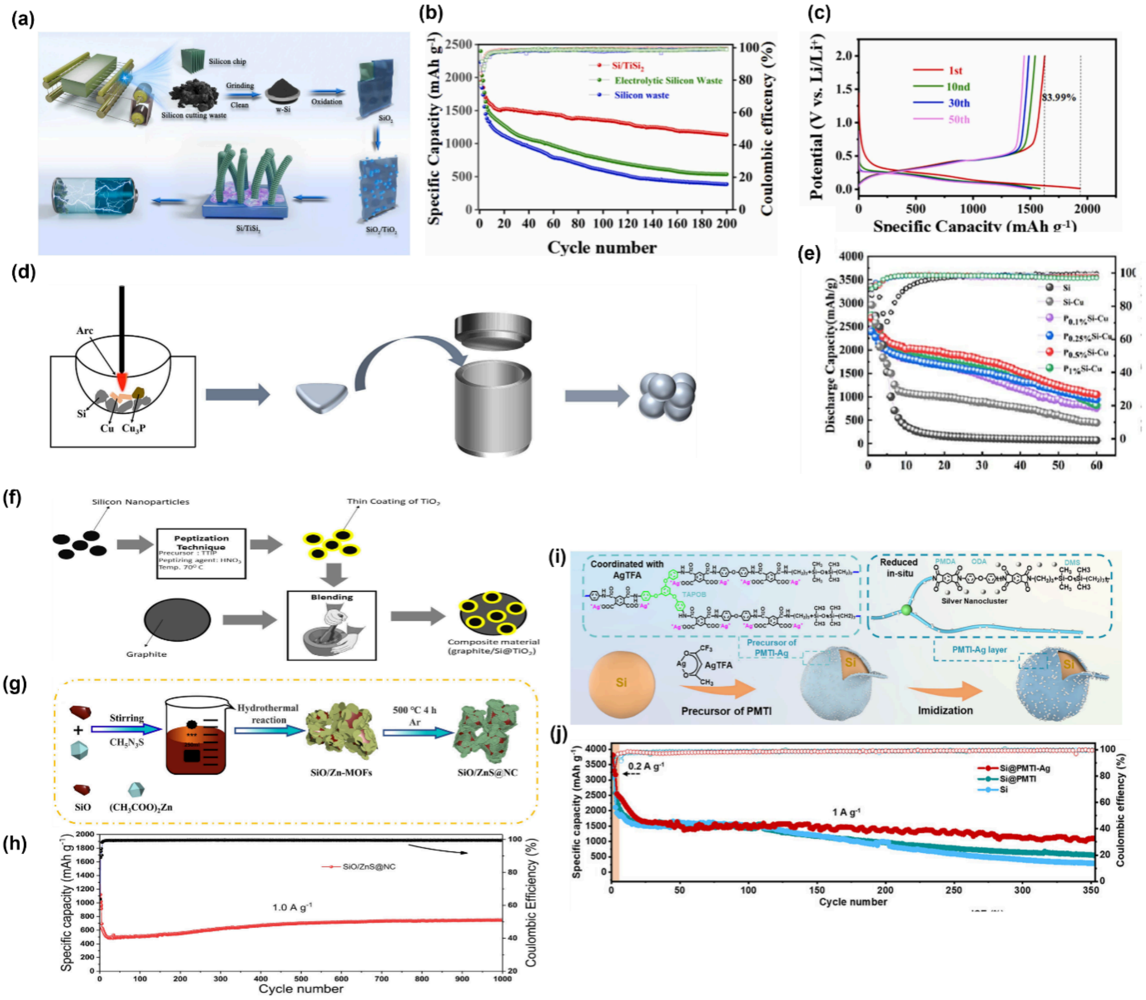

Figure 2. Examples of silicon/metal and metal-compound composites: (a) synthesis of Si/TiSi₂ [15]; (b) cycling performance and (c) charge-discharge curves of Si/TiSi₂ [15]; (d) preparation of P-doped Si-Cu [16]; (e) cycling stability of P-doped Si-Cu [16]; (f) graphite/Si@TiO₂ composite design [17]; (g) synthesis of SiO/ZnS@NC [18]; (h) cycling performance of SiO/ZnS@NC [18]; (i) construction of Si@PMTI-Ag; and (j) cycling behavior of Si@PMTI-Ag [19]

4. Silicon oxide-based anodes

Silicon oxides (SiO_x) generally exhibit lower volume variation than crystalline silicon while still offering high lithium-storage capacity. Their application, however, is limited by poor intrinsic conductivity and low initial Coulombic efficiency. Current studies therefore focus on converting the oxide component from a passive phase into a functional buffer through electrochemical activation, coating regulation, and stress-management design.

4.1. Electrochemical activation and heteroatom doping

For SiO_x electrodes, one effective route is to activate the oxide component during the initial discharge so that it contributes to a stable composite structure. Dong et al. prepared carbon-coated SiO_2 ($C@SiO_2$) through dopamine polymerization followed by carbonization and then applied a 2 mV potentiostatic activation process [20] (Figure 3a). The carbon layer improves electronic conductivity, whereas the activation reaction between SiO_2 and lithium generates Li_4SiO_4 buffer domains and electrochemically active Si phases. Through this combined effect, the $C@SiO_2$ electrode delivered 735.43 mAh g^{-1} after 100 cycles at 100 mA g^{-1} and 414 mAh g^{-1} after 400 cycles at 2.0 A g^{-1} (Figure 3b,c), showing good cycling durability. You et al. synthesized fluorine-doped carbon-coated $Si/SiO_x@F\text{-}C$ by pyrolyzing PVDF [21]. Fluorine doping enhanced conductivity and promoted the formation of a LiF-rich SEI with favorable mechanical strength and chemical stability. Accordingly, the electrode retained 82.12% of its capacity after 2400 cycles at 3000 mA g^{-1} (Figure 3d).

4.2. Controlled etching and porosity regulation

Regulating the morphology of the SiO_x shell is another way to provide expansion space while protecting the Si/electrolyte interface. Jing et al. used a one-step etching-oxidation method in LiOH solution to tailor the $Si@SiO_x$ coating [22] (Figure 3e). By changing the water content in the ethanol-water medium, the thickness and morphology of the oxide shell could be adjusted. The optimized SLH-50 sample possessed an island-like porous SiO_x layer, which not only offered buffering space for Si expansion but also helped form a stable SEI enriched with LiF and Si-F species. This electrode maintained 841.0 mAh g^{-1} after 500 cycles at 1 A g^{-1}, and the electrode-level volume expansion decreased from 358% for bare Si to 248% (Figure 3f).

4.3. Stress management and yolk-shell structures

The mechanical characteristics of SiO_x coatings, including Young's modulus, strongly influence the macroscopic deformation of Si electrodes. Wei et al. investigated the stress-buffering effect of SiO_2 layers in silicon anodes [23] (Figure 3g). They designed a yolk-shell $Si@SiO_2@void@NC$ composite and demonstrated that a SiO_2 interlayer with an appropriate modulus can distribute cycling stress and suppress the expansion of the whole electrode. Benefiting from this stress-management architecture, the composite delivered 577 mAh g^{-1} after 1000 cycles at 1 A g^{-1} (Figure 3h).

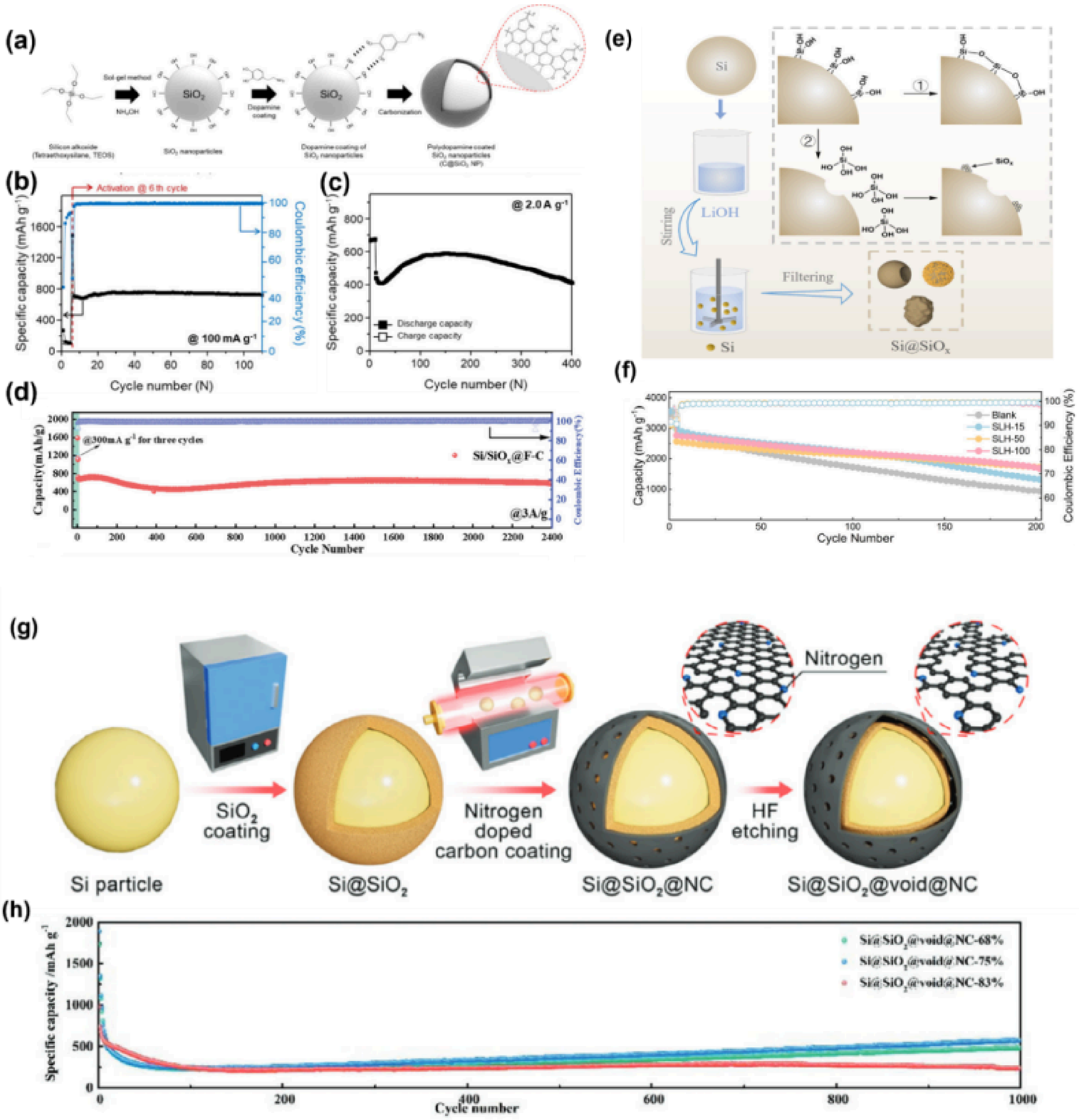

Figure 3. Silicon oxide-related anode designs: (a) preparation of C@SiO$_2$ nanoparticles [20]; (b) cycling performance of C@SiO$_2$ at 100 mA g^{-1} [20]; (c) long-term cycling of C@SiO$_2$ at 2.0 A g^{-1} [20]; (d) cycling behavior of Si/SiO$_x$@F-C [21]; (e) synthesis of Si@SiO$_x$ [22]; (f) cycling performance of Si@SiO$_x$ [22]; (g) formation of Si@SiO$_2$@void@NC [23]; and (h) cycling stability of Si@SiO$_2$@void@NC [23]

5. Silicon/polymer composite anodes

Compared with rigid inorganic layers, polymer-based designs provide a more deformable route for stabilizing silicon electrodes. Polymers can function as binders that hold the electrode together, as artificial coatings that isolate Si from the electrolyte, and as chemical regulators that influence SEI composition. Through these roles, polymer-containing composites can improve mechanical resilience, interfacial stability, and reaction kinetics.

5.1. Multifunctional binders and self-healing networks

Binder design has evolved from simple adhesion toward multifunctional networks capable of accommodating the large strain of silicon. Wang et al. developed a flexible polymer-skeleton binder based on poly(acrylic acid)-citric acid-polyethylenimine (PAA-CA-PEI, PCI) [24] (Figure 4a). The binder uses rigid amide linkages to create a tough framework and multiple hydrogen bonds to enable self-healing. It shows strong adhesion, with a peel force of 3.16 N, as well as good toughness. A silicon electrode using the PCI100 binder retained 2017.4 mAh g^{-1} after 200 cycles at 0.2 C, corresponding to 80.7% capacity retention, and still delivered 1745.3 mAh g^{-1} after 500 cycles at 0.3 C (Figure 4b). Thickness measurements further indicated that PCI100 suppressed electrode swelling, with only about 41% thickness increase after cycling, while XPS results suggested that it favored the formation of a more stable SEI.

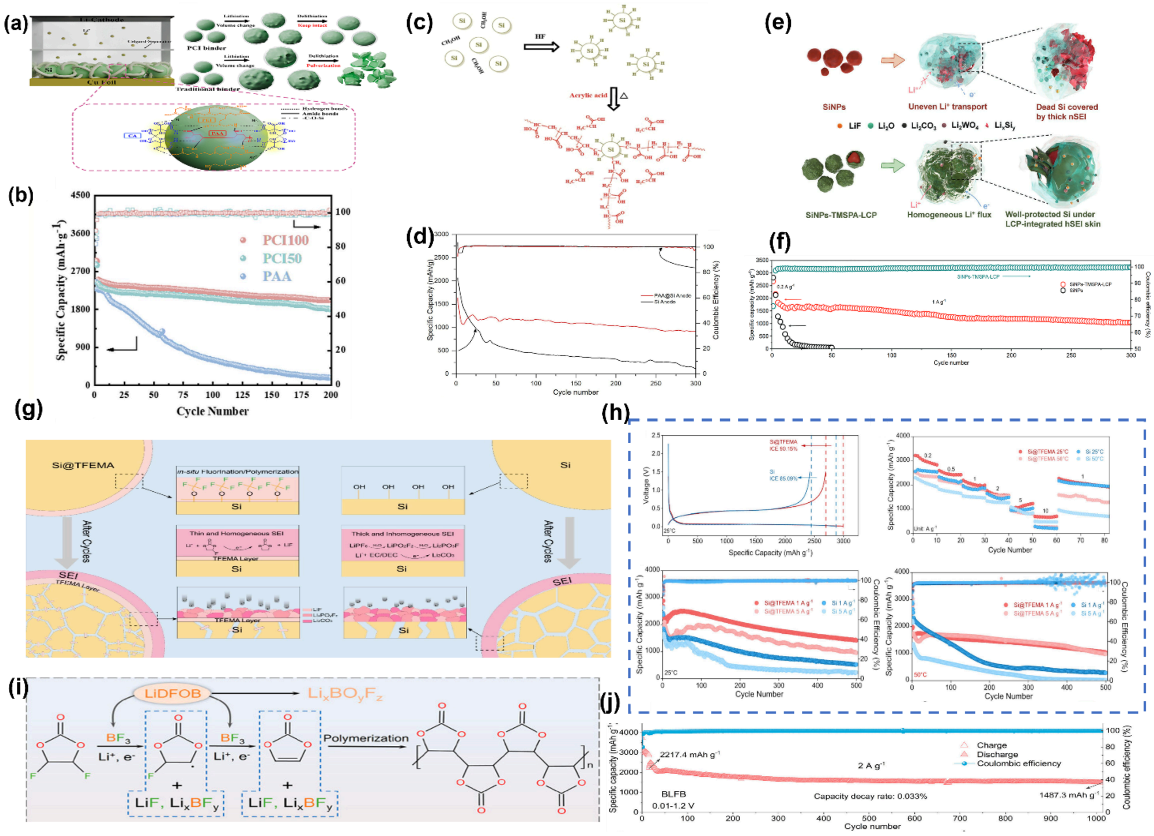

Figure 4. Polymer-based strategies for silicon anodes: (a) working mechanism of the PCI binder [24]; (b) cycling comparison of Si electrodes using PCI and PAA binders [24]; (c) preparation of PAA@Si [25]; (d) cycling performance of PAA@Si [25]; (e) structural evolution and Li^+ transport in SiNPs and SiNPs-TMSPA-LCP [26]; (f) cycling stability of SiNPs-TMSPA-LCP [26]; (g) SEI formation on Si@TFEMA and bare Si [27]; (h) electrochemical performance of Si@TFEMA and bare Si [27]; (i) reaction mechanism related to LiDFOB [28]; and (j) cycling performance of BLFB [28]

5.2. Artificial interfacial layers and conductive encapsulation

Directly coating silicon particles with an elastic interfacial layer can create a local buffer and reduce undesirable contact between silicon and the electrolyte. Yuca et al. grafted poly(acrylic acid) onto Si nanoparticles through hydrofluoric-acid etching and in-situ polymerization to obtain PAA@Si [25] (Figure 4c). Although the initial capacity decreased to some extent, the modified electrode retained 56% of its capacity after 300 cycles at C/3 (Figure 4d), much higher than that of unmodified Si. EIS analysis indicated that PAA modification increased the lithium-ion diffusion coefficient from 3.97×10^{-11} to 2.36×10^{-6} cm² s⁻¹, and SEM observations confirmed that the flexible layer helped preserve electrode morphology. Pan et al. proposed an integrated hybrid SEI by introducing a layered conductive polyaniline coating on Si nanoparticles [26] (Figure 4e). The layered coating reserved confined space for electrolyte storage and in-situ SEI generation, which promoted a more uniform LiF distribution. The resulting electrode exhibited 942 mAh g^{-1} at 5 A g^{-1} and maintained more than 1000 mAh g^{-1} after 300 cycles at 1 A g^{-1}, while limiting volume expansion to 49% (Figure 4f).

5.3. Chemically functionalized coatings and SEI regulation

Recent polymer strategies increasingly use specific functional groups to guide the formation of inorganic-rich and mechanically stable SEI layers. Ma et al. encapsulated recycled micro-sized silicon scrap with an elastic polymer layer through in-situ cross-linking of a silane coupling agent and trifluoroethyl methacrylate, producing Si@TFEMA [27] (Figure 4g). The trifluoromethyl groups in the coating promoted the formation of a LiF-rich SEI. Consequently, the Si@TFEMA electrode showed a high initial Coulombic efficiency of 90.15%, a capacity of about 1000 mAh g^{-1} even at 10 A g^{-1}, and 70% capacity retention after 360 cycles at 1 A g^{-1} (Figure 4h). Yang et al. controlled SEI composition using trans-difluoroethylene carbonate together with lithium difluoro(oxalato)borate as dual additives [28] (Figure 4i). This electrolyte strategy generated a composite interphase consisting of a LiF-dominated inner layer and a LiF-polyVC interwoven outer layer, enabling the Si electrode to retain 1487.3 mAh g^{-1} after 1000 cycles at 2 A g^{-1} (Figure 4j).

6. Conclusion and outlook

The development of silicon anodes is now closely linked to composite engineering. The studies summarized above show that the key problems of silicon cannot be solved by capacity enhancement alone. Carbon frameworks mainly provide flexibility and conductivity; metal-containing phases contribute reinforcement and fast charge transfer; oxide components act as conversion-derived buffers; and polymers offer adhesion, elasticity, and interfacial regulation [29].

For silicon composites to move from laboratory demonstrations toward commercial batteries, evaluation should focus on the behavior of the entire composite electrode rather than on individual components. This requires attention to how interfacial chemistry, such as Si-C or Si-metal bonding, affects stress distribution and reaction kinetics [30]. Precise control of composite architecture is equally important, because excessive porosity can sacrifice volumetric energy density, whereas insufficient buffer space accelerates mechanical failure [31]. Therefore, dense hierarchical structures with efficient void utilization should be prioritized. In addition, practical validation should be carried out in full-cell configurations, under high-loading conditions above 3.0 mg cm^{-2}, and in electrolyte systems that are close to real battery operation.

Overall, silicon-based composites represent an engineering platform in which organic and inorganic components work together to overcome the intrinsic limitations of silicon. Continued optimization of phase distribution, interfacial chemistry, mechanical tolerance, and full-cell compatibility will be essential for unlocking the high-energy potential of Si-based lithium-ion batteries.

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