Check for updates

Battery Energy

WILEY

BATTERY ENERGY Open Access

REVIEW OPEN ACCESS

Advancements in Silicon Anodes for Enhanced Lithium-Ion Batteries Performance: Innovations Toward Next-Gen Superbatteries

Norshahirah Mohamad Saidi[1,2] | Muhammad Amirul Aizat Mohd Abdah[1,3] | Muhammad Norhaffis Mustafa[1] | Rashmi Walvekar[4,5] | Mohammad Khalid[6,7,8] | Ajit Khosla[9]

[1]Sunway Centre for Electrochemical Energy and Sustainable Technology (SCEEST), School of Engineering and Technology, Sunway University, Bandar Sunway, Darul Ehsan, Selangor, Malaysia | [2]Department of Physics, Faculty of Science, Universiti Teknologi Malaysia, Skudai, Johor, Malaysia | [3]Department of Chemistry, Faculty of Science, Universiti Teknologi Malaysia, Johor Bahru, Johor, Malaysia | [4]Department of Chemical and Process Engineering, University of Strathclyde, Glasgow, United Kingdom | [5]Chitkara Centre for Research and Development, Chitkara University, Himachal Pradesh, India | [6]Materials and Manufacturing Research Group, James Watt School of Engineering, University of Glasgow, Glasgow, UK | [7]Faculty of Engineering, Manipal University Jaipur, Rajasthan, India | [8]Centre for Research Impact & Outcome, Chitkara University Institute of Engineering and Technology, Chitkara University, Rajpura, Punjab, India | [9]School of Advanced Materials and Nanotechnology, Xidian University, Xi'an, China

Correspondence: Mohammad Khalid (mohammad.khalid@glasgow.ac.uk) | Ajit Khosla (ajitkhosla@xidian.edu.cn)

Received: 17 September 2024 | **Revised:** 2 December 2024 | **Accepted:** 19 December 2024

Funding: The authors would like to thank the financial support from the Sunway University Research Grant (GRTIN-RRO-60-2022) and the International Research Network Grant Scheme (STR-IRNGSSET-GAMRG-01-2022).

Keywords: Lithium-ion battery | Nanostructure | Silicon anode | Superbatteries

ABSTRACT

Silicon (Si)-based materials have emerged as promising alternatives to graphite anodes in lithium-ion (Li-ion) batteries due to their exceptionally high theoretical capacity. However, their practical deployment remains constrained by challenges such as significant volume changes during lithiation, poor electrical conductivity, and the instability of the solid electrolyte interphase (SEI). This review critically examines recent advancements in Si-based nanostructures to enhance stability and electrochemical performance. Distinct from prior studies, it highlights the application of Si anodes in commercial domains, including electric vehicles, consumer electronics, and renewable energy storage systems, where prolonged cycle life and improved power density are crucial. Special emphasis is placed on emerging fabrication techniques, particularly scalable and cost-effective methods such as electrospinning and sol–gel processes, which show promise for industrial adoption. By addressing both the technical innovations and economic considerations surrounding Si anodes, this review provides a comprehensive roadmap for overcoming existing barriers, paving the way for next-generation, high-performance batteries.

1 | Introduction to Si Anodes and the Underlying Science

The depletion of fossil resources has escalated fuel costs, air pollution, and global warming, highlighting the urgent need for renewable, clean energy sources and advanced storage technologies. These are critical to meeting the growing demands of global energy sustainably. Among various energy storage solutions, functional materials are pivotal in determining the performance of electrochemical energy storage (EES) devices such as lithium-ion batteries (LIBs), lithium–sulfur (Li–S) batteries, metal–air batteries, supercapacitors (SCs), and hybrid systems like supercapatteries. Despite significant progress, the development of materials that simultaneously deliver high energy density, stability, efficiency, and safety remains a challenge. Figure 1 highlights key breakthroughs and advancements

This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

© 2025 The Author(s). *Battery Energy* published by Xijing University and John Wiley & Sons Australia, Ltd.

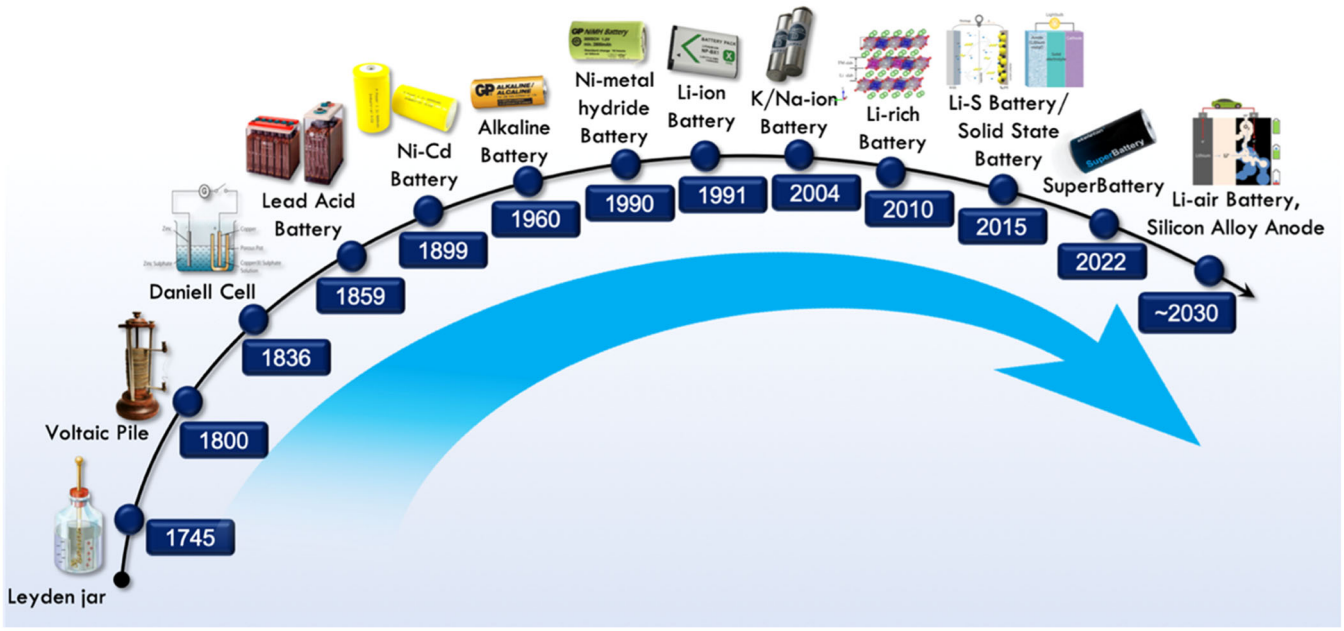

FIGURE 1 | Overview of the advances and innovations in battery technology over time.

in battery technology over time, reflecting the ongoing evolution of EES devices.

Rechargeable batteries have been indispensable since the invention of the lead-acid battery in 1859, particularly in portable applications. Among these, LIBs have emerged as the most successful technology, offering significantly higher energy and power densities than earlier systems like nickel–cadmium (NiCd) and nickel–metal hydride (NiMH) batteries. LIBs are widely regarded as an ideal solution for high-energy, portable electrochemical power sources. Their strengths lie in their high energy density, excellent rate capability, long cycle life, and low self-discharge rates. Despite their advantages, LIBs face challenges that limit their performance and broader application. Safety concerns arise from dendrite formation on lithium metal anodes, which can cause short circuits and reduce cycle life [1, 2]. Additionally, solid-phase diffusion during charging and discharging hinders fast charging, contributing to long charging times and lower power density. Sony addressed some of these issues in 1990 by introducing carbon-based materials for the negative electrode (anode), significantly enhancing performance and safety.

At present, graphite is employed in most commercially accessible anodes because of its distinctive hierarchical structure. The relatively wide gap between two adjacent layers of carbon atoms in graphite provides insertion sites for incorporating Li^+ into graphite, thereby keeping the shape, size, and structure of the anode material constant throughout the charge-discharge cycle. Figure 2a provides a schematic representation of the operational mechanism of LIBs with graphite anodes and Li-based composite cathodes. During the charging process, Li^+ is extracted from the cathode material and intercalated into the anode material, typically graphite. Simultaneously, electrons travel from the cathode to the anode through the external circuit to maintain charge balance. During discharge, the process is reversed, with Li^+ migrating back to the cathode while electrons flow from the anode to the cathode, providing electrical power. This process highlights the intercalation/deintercalation mechanism in LIBs, where graphite's layered structure provides stable insertion sites for Li^+ without significant structural changes. However, the energy density of graphite-based anodes is restricted (120–240 Wh/kg), limiting LIB applications in high-energy-demand scenarios [1].

Supercapacitors (SCs) have gained traction as complementary technologies to LIBs for high-power applications. SCs can achieve higher power densities and longer cycle lives due to the rapid non-Faradaic adsorption and desorption processes occurring on the surfaces of active materials [3, 4]. They are classified into electrochemical double-layer capacitors (EDLCs) and pseudocapacitors. In EDLCs, the surface of the positive electrode absorbs anions, while the surface of the negative electrode absorbs cations, creating a significant potential difference that enables charge storage (Figure 2b). This EDLC energy storage mechanism is an entirely physical process, resulting in excellent stability and long cycle life. In the pseudocapacitive mechanism (Figure 2c), electroactive species such as conducting polymers and transition metal oxides/hydroxides undergo fast and reversible redox reactions on the electrode surface, resulting in higher capacitance and energy density compared with EDLCs [5, 6]. However, in SCs, particularly asymmetric SCs (ASCs), the relatively poor electrochemical performance of most carbon-based materials (~ 200 F/g) poses a significant challenge. To achieve charge balance, the mass of the negative electrode materials often needs to be substantially greater than that of the positive electrode material [7]. This imbalance not only decreases the overall specific capacitance of the ASC but also restricts the effective utilization of high-performance positive electrode materials, thereby limiting their potential in energy storage applications [8, 9].

The selection of electrode materials is critically affecting the performance of both LIBs and SCs [10–16]. Consequently, novel anode materials with high-rate performance, large store capacity, and extended cycles are highly sought after. The Li^+ and electron transport processes within electrode materials strongly affect the

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

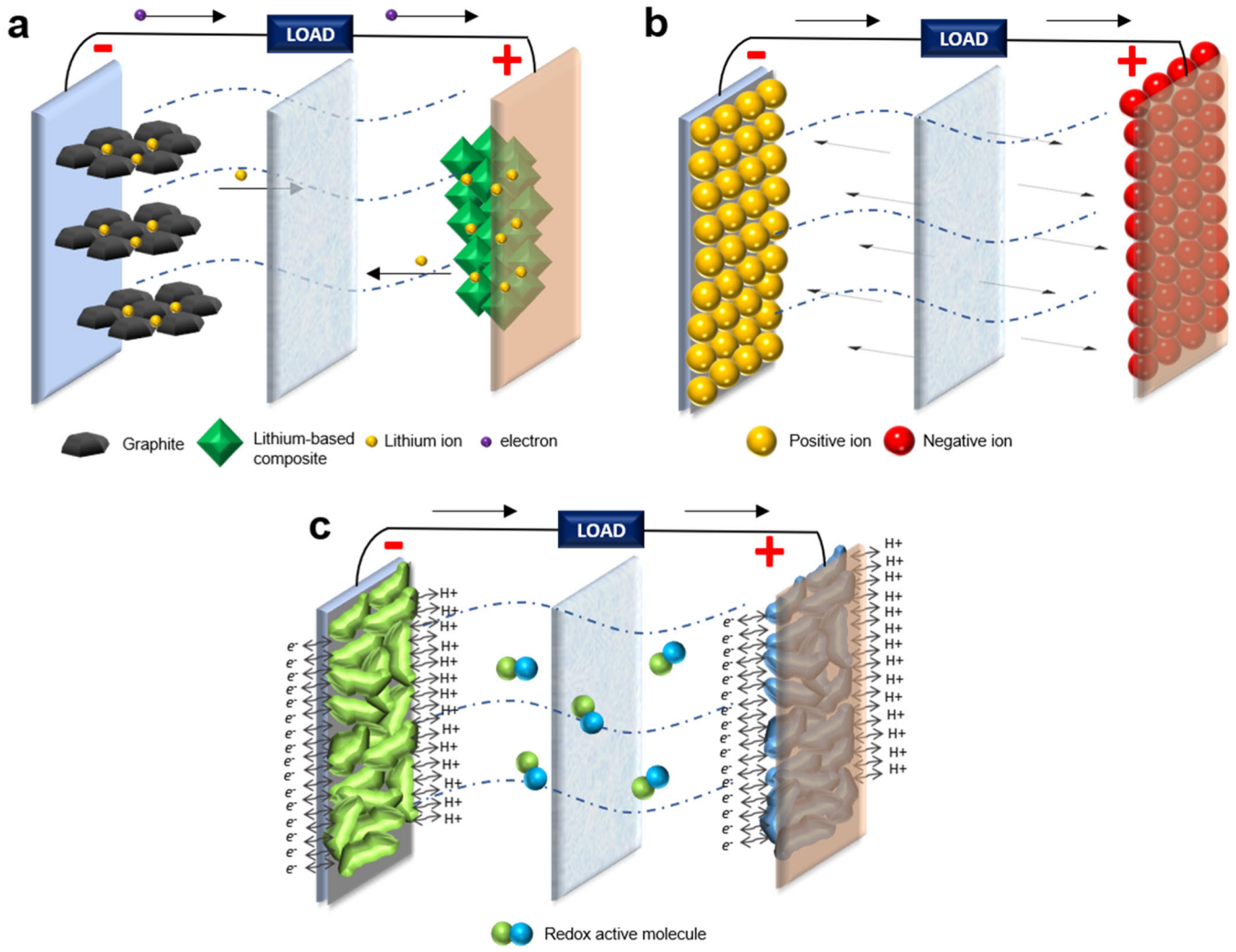

FIGURE 2 | Schematic diagram showing the different energy storage mechanisms of (a) LIBs, (b) EDLCs, and (c) pseudocapacitors.

TABLE 1 | Comparison of different anode materials.

Anode materials	Silicon ($Li_{22}Si_5$)	Carbon (LiC_6)	Lithium (Li)	Magnesium (Li_3Mg)	Aluminum (LiAl)	Tin ($Li_{4.4}Sn$)	Antimony (Li_3Sb)
Theoretical specific capacity (mAh/g)	4200	372	3862	3350	993	994	660
Theoretical volume capacity (mAh/cm^3)	9786	837	2047	4355	2681	7246	4422
Volume expansion (%)	300	12	100	100	96	260	200
Potential versus Li/Li$^+$ (V)	0.37	0.05	0	0.1	0.3	0.6	0.9

rate capability of the devices. New anode materials with significantly higher energy densities are being investigated to replace conventional graphite (or other carbonaceous materials), which cannot fulfill the need for enhanced storage capacity due to its unsatisfactory theoretical specific capacity of 372 mAh/g. Thus, various materials, including amorphous carbon, metal oxides, and alloy complexes, have been extensively studied as potential next-generation anode materials [17–19]. Among all elements used in Li alloy anodes (e.g., Si, Sn, Sb, Al, Au, Pt, Zn, Mg, and C), Si has attracted tremendous attention as an anode material. The performances of different anode materials are compared in Table 1. Si anodes offer a significantly higher theoretical specific capacity (4200 mAh/g), low working potential (0.37 V vs. Li/Li$^+$), and abundant natural silicon resources, making them ideal for next-generation LIBs and SCs. With new advances in anode materials, both LIBs and SCs may be used in a wider range of applications, including the storage of regenerated energy [20], cell phones [21, 22], and electric vehicles [23, 24].

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

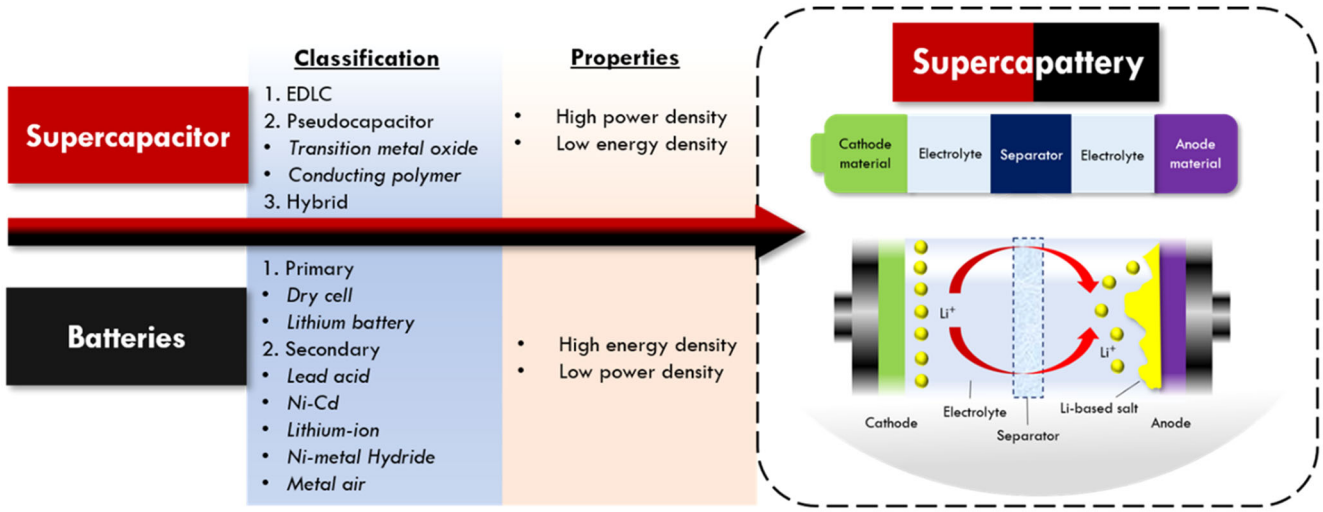

FIGURE 3 | Features of individual supercapacitors, batteries, and their combination in the form of supercapatteries.

Despite their broad applicability, both LIBs and SCs face limitations in individually meeting the growing demands for efficient energy storage systems (EES). To address these challenges, hybrid energy storage solutions are being explored. Integrating the high power density of SCs and the high energy density of LIBs [25], hybrid energy storage systems such as supercapatteries have been developed. Supercapatteries represent a promising solution for next-generation EES, offering high power density, high energy density, and prolonged cycle life, thereby bridging the gap between LIBs and SCs (Figure 3). Thus, supercapatteries offer a balanced approach, delivering both rapid power delivery and substantial energy storage [26–28]. Furthermore, the integration of Si anodes enhances the performance of hybrid systems. Si anodes are notable for their high specific capacity and abundance, making them a critical component in advancing EES technologies. The combination of supercapatteries and Si anodes creates promising pathways for addressing energy storage challenges and expanding the applications of EES devices, including renewable energy storage, mobile devices, and electric vehicles [23, 24]. By leveraging innovative materials and hybrid technologies, the next generation of energy storage systems has the potential to efficiently and sustainably meet the growing global energy demands.

2 | Material Properties of Si as a Promising Anode Material for LiBs

Si has emerged as a promising anode material for LIBs due to its exceptional material properties and abundance. Notably, Si can intercalate with approximately 3.75 Li atoms at ambient temperature and up to 4.4 Li atoms at elevated temperatures (450°C), significantly surpassing the intercalation capacity of carbon, which requires six carbon atoms to produce lithium carbide (Li_6C) [29]. The extraordinarily high theoretical specific capacity of Si, estimated at ~4200 mAh/g for lithium uptake with the $Li_{22}Si_5$ stoichiometry, is over ten times greater than that of graphite, making it an attractive candidate for next-generation LIBs [30, 31].

In addition to its high specific capacity, Si possesses several advantageous characteristics that enhance its suitability for battery applications. These include its natural abundance, with Si comprising approximately 27.7% of the Earth's crust by mass, making it a low-cost material [32–34]. Silicate minerals dominate the Earth's crust and contain Si in various forms, such as oxides (e.g., quartz, sand) and silicates (e.g., feldspar, mica) [35]. Furthermore, Si exhibits a higher Li intercalation potential (0 to 0.370 V vs. Li/Li^+) compared to graphite (0 to 0.25 V vs. Li/Li^+), which helps mitigate the formation of Li dendrites during cycling [36, 37]. This characteristic, combined with its good chemical stability in electrolytes, contributes to enhanced battery safety.

However, the significant volume change associated with the lithiation and delithiation processes can induce mechanical stress and particle fracture, which can adversely affect cycle stability. To mitigate these issues, silicon is often hybridized with conductive materials such as carbon or graphene. This combination helps maintain structural integrity and improves electrical conductivity, facilitating better electron transfer and enhancing rate capability. Additionally, the formation of a stable solid electrolyte interphase (SEI) on the Si surface protects it from degradation while allowing efficient Li^+ transport, further improving cycling stability (Figure 4a) [33]. Additionally, nanostructured Si with a high surface area promotes rapid ion diffusion, leading to improved overall performance and extended cycle life. In comparison, the rigid structure of graphite limits its capacity for Li intercalation, allowing it to store significantly less Li than Si. Consequently, silicon can store Li up to ten times more than an equivalent mass of graphite, leading to a minimum increase in battery energy density of approximately 30% [38, 39].

Furthermore, the high melting temperatures of Li–Si alloys make them advantageous for high-temperature battery operations. To fully appreciate the potential of silicon as an anode material, it is essential to consider the binary phase diagram of Si and Li, which reveals the formation of several stable compounds during lithiation, including $Li_{12}Si_7$, Li_7Si_3, $Li_{13}Si_4$, and $Li_{22}Si_5$. The Li^+ intercalation and deintercalation mechanisms in Si can be summarized using Equations (1)–(3) [40]:

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

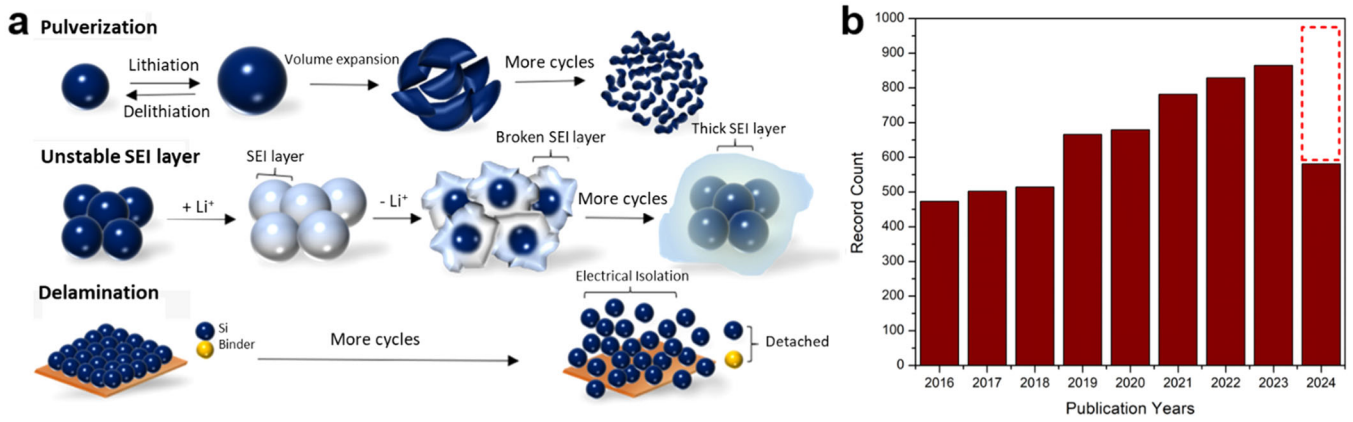

FIGURE 4 | (a) Schematic showing the failure mechanisms of Si anodes upon cycling: pulverization, SEI layer instability, and delamination. (b) Graphical representation of the trend in journal publications on Si anodes for energy storage applications. The corresponding annual publications were retrieved using "silicon," "anode," and "energy" as keywords, and the date range was 2012 to 2023 (Web of Science database, accessed on February 21, 2023).

Discharge process:

$$\mathrm{Si\,(c)} + x\mathrm{Li^+} + xe^- \rightarrow \mathrm{Li}_x\mathrm{Si\,(a)} \quad (1)$$

$$\mathrm{Li}_x\mathrm{Si\,(a)} + (3.75 - x)\,\mathrm{Li^+} + (3.75 - x)\,e^- \rightarrow \mathrm{Li}_{15}\mathrm{Si}_4\,\mathrm{(c)} \quad (2)$$

Charge process:

$$\mathrm{Li}_{15}\mathrm{Si}_4\,\mathrm{(c)} \rightarrow \mathrm{Si(a)} + y\mathrm{Li^+} + ye^- + \mathrm{Li}_{15}\mathrm{Si}_4\,\mathrm{(residual)} \quad (3)$$

2.1 | Challenges Related to Si Anodes

Despite the advantages of Si anodes, several significant challenges hinder their widespread adoption in LIBs. The foremost issue is the substantial volume expansion during the lithiation and delithiation process, reaching approximately 360% for $Li_{22}Si_5$. This dramatic expansion leads to the pulverization of the electrode material, resulting in rapid fracture and a loss of electronic connectivity. Additionally, this volume change contributes to the persistent formation of an SEI, which consumes Li^+ from the electrolyte and further exacerbates capacity loss (Figure 4a).

Another critical challenge is the slow Li^+ diffusion kinetics, estimated between 10^{-14} and $10^{-13}\,cm^2\,s^{-1}$, coupled with poor intrinsic conductivity of Si (approximately $10^{-5}\,S\,m^{-1}$). These factors generate significant local mechanical stress that can detach Si particles from the current collector, thereby reducing electrode capacity during repeated charge–discharge cycles [41]. During lithiation, a concentration gradient of Li^+ develops across the Si particle, leading to hoop compressive stress in the outer layer and hoop tensile stress in the inner part. Conversely, during delithiation, the stress direction reverses, resulting in cracking when the stress exceeds the particle's cohesive strength [42, 43]. Typically, pulverization of crystalline Si occurs during lithiation, while amorphous Si experiences significant degradation during delithiations [29, 44]. This behavior contributes to the poor cycling stability and irreversible capacity decay observed in Si anodes. Strategies such as reducing Si particle size and incorporating conductive additives and binders have been employed to constrain volume changes and enhance cycling performance to mitigate these issues. Consequently, pure Si is seldom used directly as an anode material; instead, Si-based nanostructures and hybrid materials that effectively manage volume expansion while maintaining structural integrity have been developed as alternatives [45].

Moreover, modified Si-based anodes have emerged by synthesizing composites that incorporate carbonaceous materials, metals, alloys, metal-organic frameworks (MOFs), MXenes, polymers, and other substances. Doping carbonaceous coatings with heteroatoms (e.g., N, S, P, and F) can further optimize the physicochemical properties, enhancing electrical conductivity and increasing the number of electrochemically active sites [46]. The adoption of nanoscale Si materials, such as nanowires, nanosheets, and porous nanoparticles, provides unique structural advantages that alleviate the effects of volume expansion while reducing ion and electron transport distances [47]. These advancements have driven extensive research into Si anodes for energy storage, with over 6,300 publications between 2012 and 2023 (Figure 4b). Therefore, it is imperative to discuss recent advancements in Si anode design and their characteristics when applied in LIBs and SCs, as these innovations are crucial for overcoming the inherent challenges associated with Si-based energy storage solutions.

2.2 | Progress in Si Anode Development

2.2.1 | Overview of Recent Advances in Si Anode Technology

Recent developments in Si anode technology have focused on creating "superbatteries," which are characterized by enhanced energy density, enabling greater energy storage in a compact form, and improved power density for faster energy output. These advancements also result in longer cycle lives, allowing for more charge–discharge cycles before capacity degradation, as well as significantly faster charging capabilities.

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/ne2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

TABLE 2 | Overview of recent advances in Si anode technology.

Material	Specific capacity (mAh/g) or (F/g)	Retention (%)	Reference
Polyvinyl alcohol/Si/polyacrylonitrile	900 mAh/g	90% (250 cycles)	[48]
Si@SiO_2-PAA-GL	1192.7 mAh/g	83% (500 cycles)	[49]
SPSC-PANI-MoS_2	637.5 F/g	80%	[50]
Rolled-up amorphous Si nanomembranes	2000 mAh/g	85% (500 cycles)	[51]
Si hollow spheres	2725 mAh/g	60% (500 cycles)	[52]
B-doped porous Si nanowires	2000 mAh/g	80% (200 cycles)	[53]
HN-Si@aTiO_2	2804 mAh/g	42% (100 cycles)	[54]
Silicon microparticles coated with graphene cage	3300 mAh/g	85% (300 cycles)	[55]
C-Si nanocomposite	378 mAh/g	98% (100 cycles)	[56]
Silicon hollow nanoboxes coated with reduced graphene oxide	681 mAh/g	71% (2000 cycles)	[57]
c-silicene	180 mAh/g	100% (3000 cycles)	[58]

Furthermore, enhanced safety features, such as improved thermal stability and reduced risk of overheating or combustion, are critical for developing next-generation superbatteries.

To achieve high energy and power densities, novel anode materials must possess high specific capacities, long lifetimes, and excellent rate capabilities. While cathode materials are nearing their theoretical capacity limits, Si remains a highly attractive candidate for next-generation anodes. Its theoretical specific capacity is approximately ten times greater than that of graphite due to its ability to bind with four Li^+. However, addressing the challenge of Si's substantial volume expansion during cycling is crucial for achieving reliable cycling performance. This challenge is being effectively mitigated through the design of Si-based nanomaterials and their incorporation into hybrid materials. An overview of recent advances in Si anode technology is presented in Table 2.

For instance, Pei et al. [48] developed polyvinyl alcohol/Si/polyacrylonitrile composite fibers through coaxial electrospinning followed by carbonization. The resulting microfiber mats formed an electrically conductive network with a thin carbon skin encasing the Si/carbon nanofiber bundles, which acted as a three-dimensional (3D) current collector, facilitating Li^+ transport and charge transfer during charge/discharge operations. The mesoporous gaps between the short Si nanofibers served as buffers to alleviate stress in the Si nanoparticles (SiNPs) due to volume changes during Li insertion and extraction. This fibrous anode material, with a Si concentration of 40%, achieved a discharge capacity of 900 mAh/g and maintained approximately 90% capacity retention from cycles 50 to 250.

Similarly, Sun et al. [49] enhanced SiNPs by coating them with silicon oxide (Si@SiO_2) using a polyacrylic acid and glycerol polymer binder (PAA-GL) via an in situ dehydration condensation reaction. The Si@SiO_2-PAA-GL electrode exhibited remarkable electrochemical performance due to several factors: (1) the SiO_2 layer mitigated the significant volumetric changes of the SiNPs; (2) strong covalent interactions between Si@SiO_2 and the free carboxyl groups in the PAA-GL binder effectively restricted large-scale particle movement; and (3) the numerous functional groups in the PAA-GL binder provided strong adhesion to the SiO_2 coating, enhancing the structural stability of the electrode. When tested in LIBs, the Si@SiO_2-PAA-GL electrode delivered a high capacity of 1192.7 mAh/g with a minimal capacity decay rate of 0.0337% per cycle after 500 cycles at a current density of 2.0 C.

For SC applications, Wang et al. [50] utilized a partial etching technique to synthesize SiO_2 peristaltic supporting carbon (SPSC), which was then combined with polyaniline (PANI). Molybdenum disulfide (MoS_2) nanosheets were deposited onto the surface of the unfixed PANI through a redox reaction, resulting in the creation of the SPSC-PANI-MoS_2 nanocomposite. This composite demonstrated remarkable supercapacitive performance, achieving a specific capacitance of 637.5 F/g at a current density of 0.5 A/g. Additionally, the assembled SPSC-PANI-MoS_2//AC configuration exhibited a specific energy of 22.6 Wh/kg and a specific power of 550.0 W/kg, along with exceptional cycle stability at a specific capacitance of 80.4%. These studies highlight the versatility of Si anodes and the potential of advanced synthesis techniques to overcome intrinsic material challenges. A detailed exploration of these innovative approaches is provided in the following sections.

2.3 | Strategies for Overcoming the Challenges of Using Si Anodes

2.3.1 | Si Composites With Carbonaceous Materials

Si composites with carbonaceous materials are commonly employed to address the significant volume expansion and enhance the electrical conductivity of Si-based electrodes. This approach improves conductivity from 10^{-5} to 10^{-3} S/cm and mitigates the issues associated with Si's inherent volume changes during cycling [59, 60]. Various Si/carbon composite structures have been developed to optimize these properties. For instance, Shi et al. [61] synthesized porous Si/carbon composites (C@void/Si-G) via ball milling with sodium chloride (NaCl) template, followed by heating and cleaning. Figure 5a shows the detailed synthesis processes of the C@void/Si-G and C@Si-G composites.

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

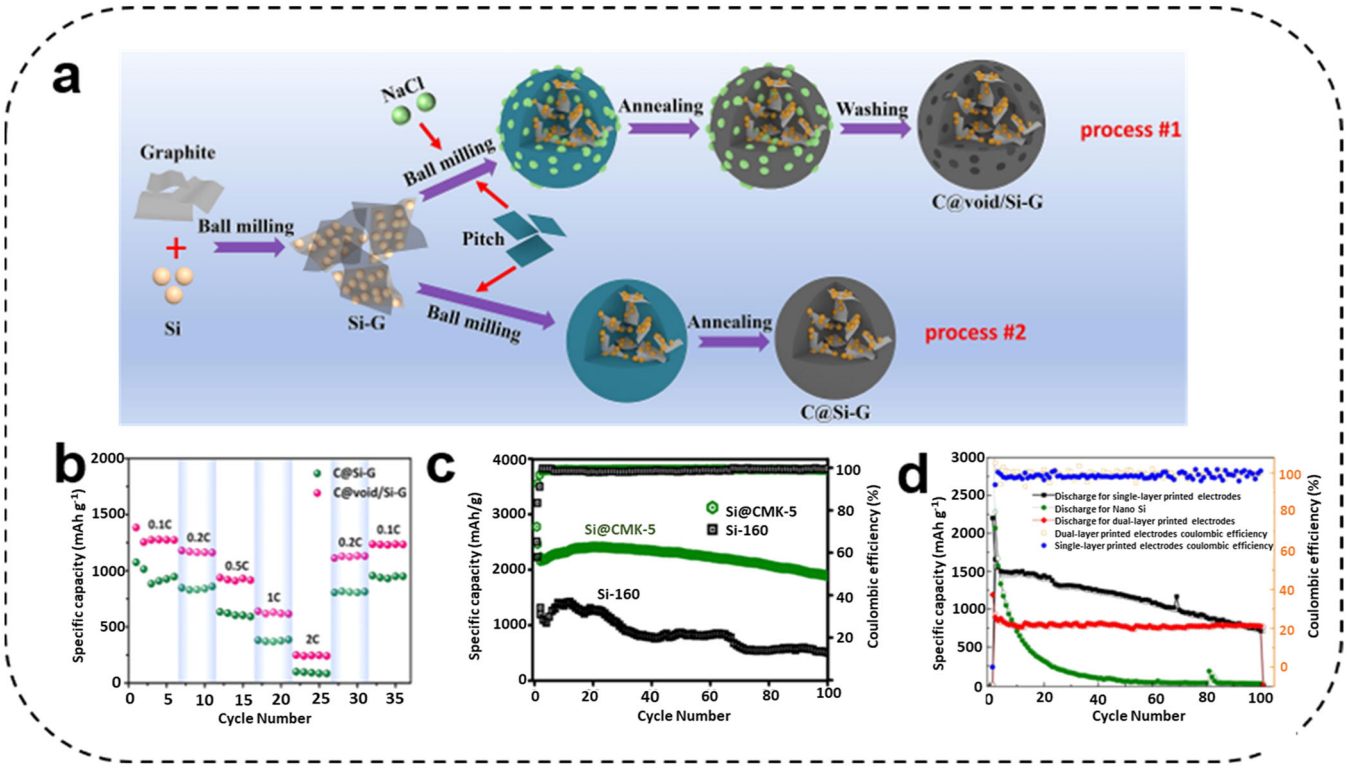

FIGURE 5 | (a) Schematic diagram showing the preparation of C@Si-G and C@void/Si-G composites. (b) Rate capability of the as-prepared C@Si-G and C@void/Si-G electrodes at various current densities [12]. (c) Cycle stability and Coulombic efficiency of Si-160 and the Si@CMK-5 composite at a rate of 0.5 C [62]. (d) Cycle stability and Coulombic efficiency of microelectronically printed electrodes at a current density of 0.1 A/g [63].

This structure offers multiple advantages: (1) the pyrolytic carbon layer around Si/graphite composites forms a stable SEI that reduces electrolyte consumption and accommodates volume expansion, (2) SiNPs anchored to graphite improve electrical conductivity, and (3) NaCl removal creates a porous region that shortens the Li^+ diffusion path and buffers volumetric changes. The C@void/Si-G electrode exhibited excellent rate capabilities, delivering specific capacities of 1373.3, 1168.9, 937.8, 639.3, and 248.9 mAh/g at 0.1, 0.2, 0.5, 1.0, and 2.0 C, respectively. After cycling, the capacity recovered to approximately 1128.9 and 1236.0 mAh/g at 0.2 and 0.1 C, respectively, demonstrating exceptional structural stability (Figure 5b).

In another study, Ha et al. [62] developed a mesoporous carbon-supported Si composite (Si@CMK-5) by calcination using SBA-15 nanorods as a template. The mesoporous carbon framework increased the composite's stability and conductivity, leading to impressive electrochemical performance with an initial capacity of 3555.4 mAh/g. After 100 cycles at 0.5 C, the Si@CMK-5 composite retained a discharge capacity of 1900.8 mAh/g, nearly four times higher than the Si-160 electrode (Figure 5c). This enhanced capacity resulted from the synergistic effect of CMK-5 and SBA-15, which improved the surface area and mesoporous structure, facilitating Li^+ loading, reducing volume expansion, and enhancing conductivity. Additionally, oxygen defects formed during magnesiothermic reduction helped mitigate Si's volume expansion and shorten the Li^+ diffusion path.

Zhang et al. [63] addressed the significant volumetric changes in Si by fabricating a flexible Si/carbon composite with a layered slit structure, achieved through microelectronic printing and colloidal curing. This design allows for structural adaptation during Li^+ intercalation and delithiation, preserving the anode's integrity. A thin amorphous carbon layer covering the monolayer Si anode significantly enhanced its electrochemical performance. As shown in Figure 5d, the printed double-layer anode exhibited an initial reversible capacity of 941.1 mAh/g and retained 769.8 mAh/g after 100 cycles. Scanning electron microscopy confirmed minimal layering and peeling on the electrode surface, indicating that the structural integrity and cycling stability were well-maintained until the amorphous carbon layer was fully depleted.

2.3.2 | Si Composites With Other Materials

Recent studies have demonstrated that combining SiNPs with various materials, such as 2D materials, conducting polymers, and MOFs, can significantly improve the performance of LIBs and SCs. Among 2D materials, MXenes ($M_{n+1}X_nT_x$, where M represents an early transition metal, X denotes carbon or nitrogen, T_x indicates surface terminations, and $n = 1–4$) are particularly noteworthy due to their excellent electrical conductivity and layered structures, which can accommodate the volumetric changes of Si during cycling, thereby prolonging the cycle life of Si anodes. For example, Zhang et al. [64] developed a self-standing SiNP structure anchored on nitrogen-doped carbon (NC) foam and covered with a MXene layer (MXene@SiNPs@NC foam) (Figure 6a). The MXene layer and NC foam framework provided efficient electron transport and

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/hte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

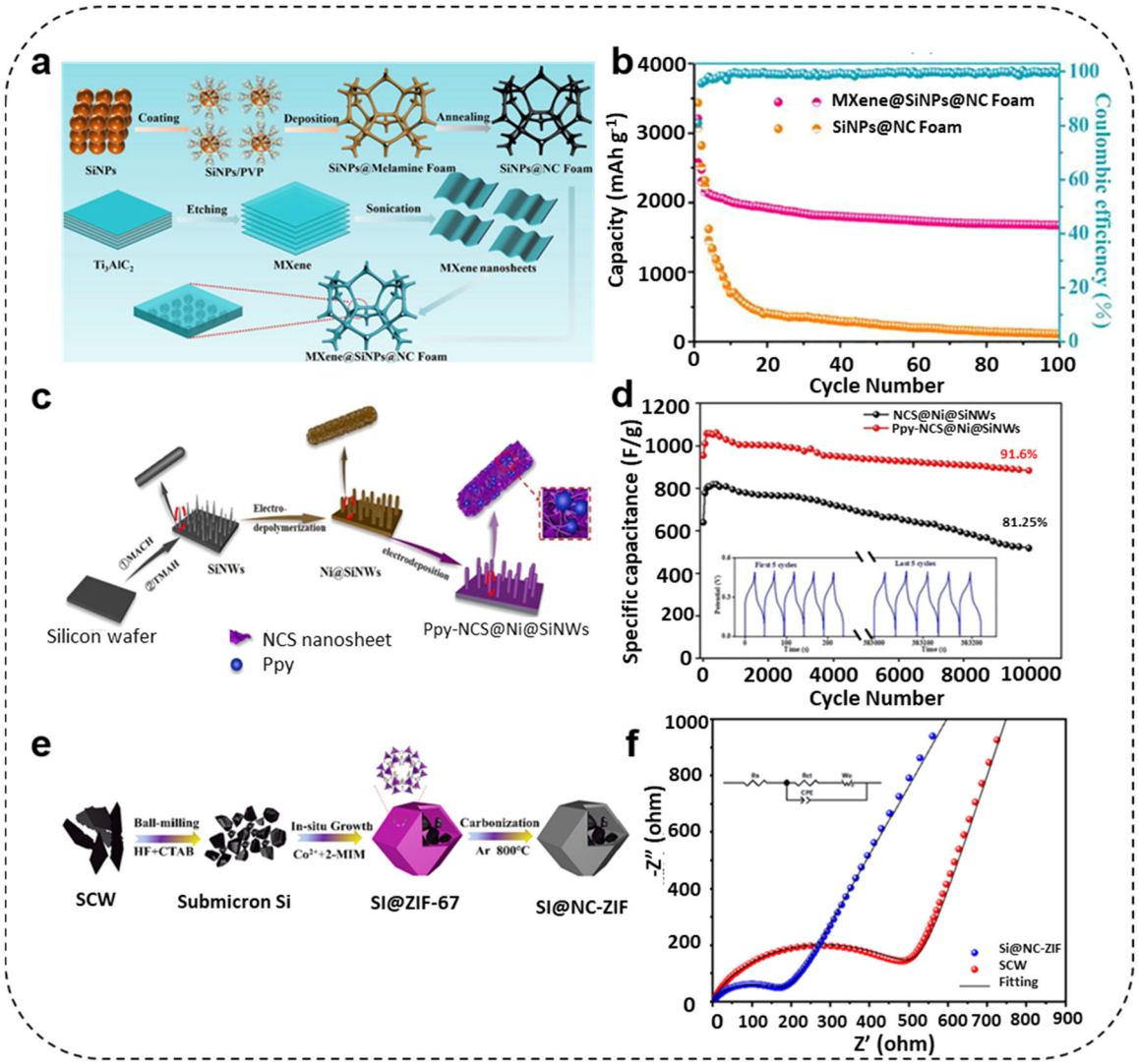

FIGURE 6 | (a) Schematic diagram showing the preparation of MXene@SiNPs@NC foam. (b) Long-term stability of MXene@SiNPs@NC and SiNPs@NC foams at a rate of 0.1 C [64]. (c) Schematic diagram showing the preparation of PPy-NCS@Ni@SiNWs. (d) Long-term stability of NCS@Ni@SiNWs and PPy-NCS@Ni@SiNWs electrodes at 20 A/g. The inset shows the galvanostatic charge/discharge curves for the first and last five cycles of the PPy-NCS@Ni@SiNWs electrode [65]. (e) Schematic diagram showing the preparation of Si@NC-ZIF composite. (f) Nyquist plots of the SCW and Si@NC-ZIF composites in the frequency range of 100 kHz to 0.01 Hz at room temperature [66].

ion diffusion pathways, while the void spaces between the MXene layer and the NC foam skeleton buffered the large volumetric changes during lithiation/delithiation. As shown in Figure 6b, the MXene@SiNPs@NC foam anode exhibited impressive cycling stability, maintaining a specific capacity of 857 mAh/g after 500 cycles at a current density of 0.5 C and 1658 mAh/g after 100 cycles at 0.1 C. In full-cell configurations, this anode achieved a gravimetric energy density of 433 Wh/kg when paired with an NCM111 cathode.

Additionally, Shen et al. [65] fabricated Si nanowire (SiNW) arrays passivated by a multi-shelled polypyrrole-nickel-cobalt sulfide@-nickel (PPy-NCS@Ni@SiNWs) composite via electrodeposition and subsequent solution processing (Figure 6c). The combination of the high surface area of SiNWs and the high capacitance of the PPy-NCS layer enabled the PPy-NCS@Ni@SiNWs electrode to deliver a specific capacitance of 1602 F/g at 1 A/g, excellent rate capability (1348 F/g at 30 A/g), and an impressive capacitance retention of 91.6% after 10,000 cycles (Figure 6d).

MOFs have gained significant attention for their unique properties, including high porosity, large surface area, crystalline structures, and tunable pore sizes. Boorboorajdari et al. [66] reported the fabrication of Si@NC-ZIF composites, where ZIF-67 was formed in situ on the surface of silicon cutting waste (SCW) and subsequently carbonized to create Si-based composites with submicron-scale pores and cage-like structures (Figure 6e). The carbon matrix formed from ZIF-67 incorporated various gaps that effectively buffered the volumetric changes of Si during cycling. The Si@NC-ZIF composite demonstrated outstanding rate performance and long-term stability, maintaining a capacity of 1623.05 mAh/g with an 86.29% retention rate after 100 cycles at 200 mA/g and a capacity of 1034.19 mAh/g after 200 cycles at 500 mA/g. As illustrated in Figure 6f, the charge transfer resistance (R_{ct}) values for SCW and the Si@NC-ZIF composite were 542 Ω and 189 Ω, respectively, indicating that the ZIF-derived carbon coating significantly enhanced the electrical conductivity, electrochemical contact, and electron transport of the Si anodes, thereby improving their rate capability.

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

2.3.3 | Si Composites With Heteroatomic Doping

Heteroatomic doping, particularly with nitrogen (N), sulfur (S), phosphorus (P), and fluorine (F), has emerged as an effective strategy to enhance the electrochemical performance of Si anodes by modifying their chemical reactivity, as well as their electronic and physical properties [46, 67, 68]. Doping with N has been shown to improve the electrical conductivity of Si, accelerate charge transfer, and increase the availability of active sites, which are beneficial for Li^+ diffusion and storage during cycling [69]. Similarly, the addition of sulfur can enhance the electrochemical performance of Si by improving surface wettability and reducing R_{ct} [70].

Pan et al. [71] used a simple alkali etching and hydrothermal method to co-dope Si with N and S atoms, resulting in an amorphous, ball-flower-like NiO-loaded silicon oxycarbide-derived carbon composite (NSCDC/NiO; Figure 7a). The NSCDC/NiO composite exhibited a capacitance of 804 F/g at a current density of 1 A/g, nearly four times that of the pure NSCDC electrode (201 F/g; Figure 7b). Furthermore, an asymmetric supercapacitor (ASC) based on NSCDC/NiO//NSCDC demonstrated excellent cycling stability, retaining 84% of its capacitance after 5000 cycles. These superior performances can be attributed to the amorphous structure of NSCDC/NiO, which promotes effective charge transport, provides a high surface area, and mitigates stress during redox reactions.

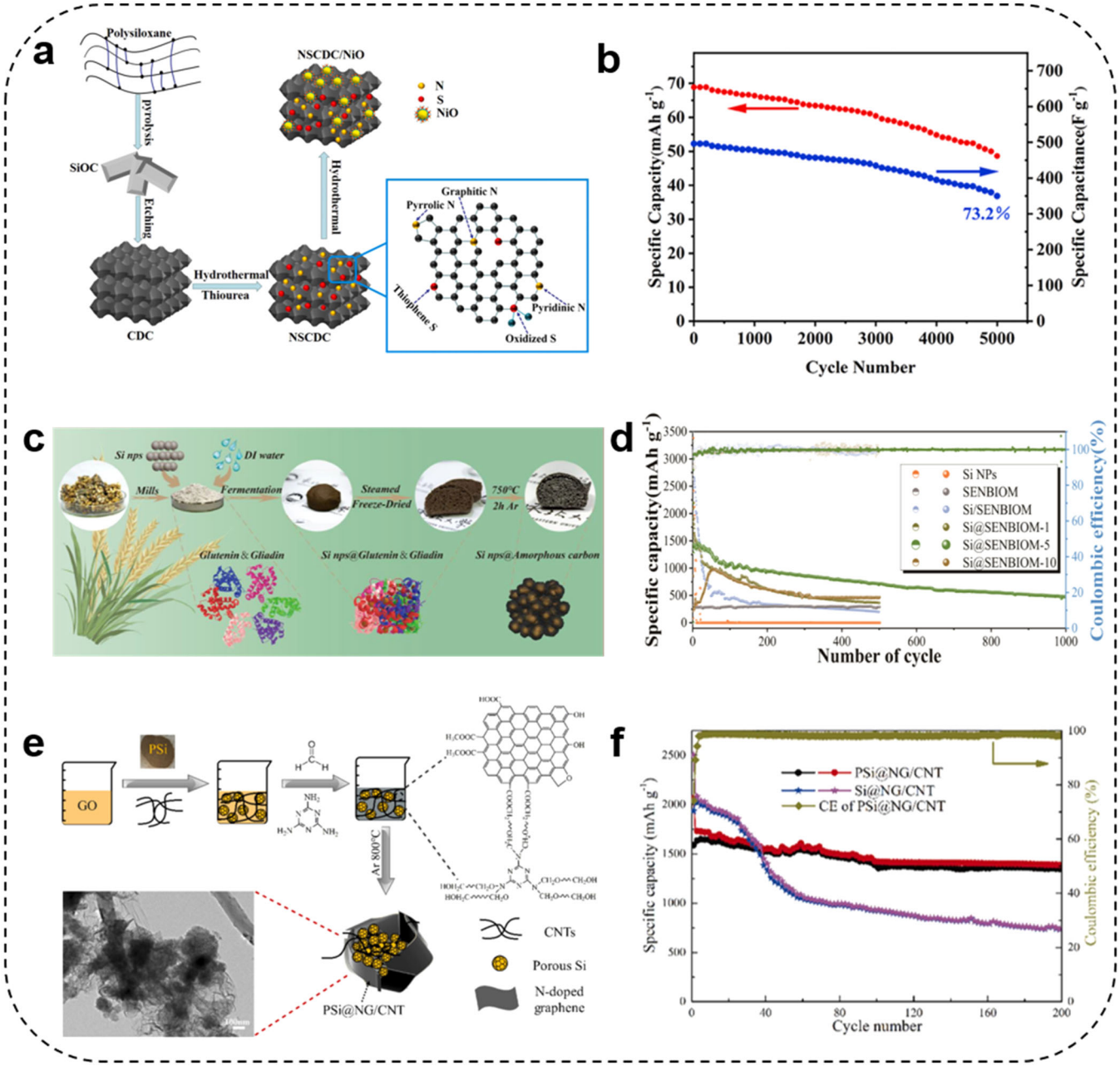

FIGURE 7 | (a) Schematic diagram showing the preparation of NSCDC/NiO composites. (b) Long-term cycling performance of the NSCDC/NiO-10 composite at 5 A/g [71]. (c) Schematic diagram showing the preparation of Si@SENBIOM-5 electrode. (d) Long-term cycling performances of SiNPs, SENBIOM, Si/SENBIOM-5, Si@SENBIOM-1, Si@SENBIOM-5, and Si@SENBIOM-10 at 1 A/g [72]. (e) Schematic diagram showing the preparation of PSi@NG/CNT composites. (f) Long-term cycling performance of PSi@NG/CNT- and Si@NG/CNT-based electrodes at 0.2 A/g [73].

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

Sun et al. [72] developed Si@Self-encapsulated N-doped biochar derived from moldy wheat flour to improve the Li storage performance of the Si anodes (Figure 7c). The process involved mixing SiNPs with water, resulting in effective self-coating, followed by annealing, which led to the formation of a loose, homogeneous cladding layer containing nitrogen due to the stretchable and viscous properties of the gluten network. This approach successfully created a stable SEI that prevented direct contact between SiNPs and the electrolyte, thereby increasing conductivity. The Si@Self-encapsulated N-doped biochar (Si@SENBIOM-5) electrode exhibited high conductivity and excellent charge/discharge cycling performance, with an initial Coulombic efficiency of 88.6%. The reversible capacity of this electrode was maintained at 712.6 mAh/g after 500 cycles and 478 mAh/g after 1000 cycles at current densities of up to 1 A/g. Even at 2 A/g, the electrode maintained a capacity of 348.28 mAh/g after 1000 cycles, indicating good long-term cycle stability.

Tang et al. [73] incorporated porous Si (PSi) into an N-doped graphene and carbon nanotube (CNT) framework (PSi@NG/CNT) using a one-pot carbonization process with scalable magnesiothermic reduction (Figure 7e). The synergistic interactions between the N-doped graphene, CNTs, and PSi contributed to the enhanced electrochemical performance of the PSi@NG/CNT composite. The flexible N-doped graphene served as an active matrix to improve electrical conductivity and partially buffer volume fluctuations during the Li-Si alloying and dealloying processes. The CNTs acted as "welding points" that activated more Si and connected the graphene sheets into a stable network, preventing aggregation. Additionally, the porous structure facilitated the movement of Li^+ ions and the penetration of the electrolyte, ensuring good contact between the electrolyte and the electrode while mitigating the pulverization of PSi caused by volume expansion during cycling. When used as an anode for LIBs, the PSi@NG/CNT composite demonstrated superior rate performance compared to SiNP composites, achieving a specific discharge capacity of 1333 mAh/g at 0.2 A/g after 200 cycles, with a high capacity retention of 79.7%.

2.3.4 | Si-Based Nanoscale Materials

The limitations of Si anodes can be mitigated by developing advanced negative electrode materials. Recent progress has seen the successful fabrication of Si-based nanostructures, including SiNPs [74], SiNWs [75], and Si nanotubes [76], which have been successfully synthesized to enhance mechanical tolerance, buffer volume expansion, and improve electron and ion transport, thus mitigating the low initial coulombic efficiency commonly associated with Si anodes. For instance, Zhou et al. [77] developed a core-shell nanostructured SiNW/poly(3,4-ethylenedioxythiophene)/polypyrrole (SiNWs/PEDOT@PPy) composite electrode using a two-step process that involved in situ photopolymerization followed by chemical oxidative polymerization (Figure 8a). The electrochemical properties of this composite were influenced by the length of the SiNWs, which was adjustable by varying the duration of metal-assisted chemical etching. The incorporation of PEDOT as a surface modifier significantly enhanced the supercapacitive performance of the composite. At a current density of 0.5 mA/cm^2, the SiNWs/PEDOT@PPy electrode achieved an area-specific capacitance of 106 mF/cm^2 and maintained its core-shell structure with a capacitance retention of 80.2% after 5000 cycles (Figure 8b).

In another approach, Bencheikh et al. [78] demonstrated the synthesis of SiNWs decorated with ruthenium nanoparticles (RuNPs) for Si-based micro-supercapacitors (micro-SCs). The SiNWs were fabricated using the vapor–liquid–solid growth mechanism, and RuNPs were deposited via electroless plating. The RuNPs' pseudocapacitive behavior and the large surface area of the SiNWs significantly enhanced the energy storage capacity. After 25,000 charge–discharge cycles at 1 mA/cm^2, the RuNPs/SiNWs electrode maintained a specific capacitance of 36.25 mF/cm^2 with 87% retention (Figure 8c). Additionally, Zhang et al. [79] developed a 3D honeycomb-structured composite consisting of reduced graphene oxide (rGO), acetylene black (ACET), and SiNPs. This composite was fabricated through mechanical ball milling of ACET and Si, followed by encapsulation in a graphene oxide suspension. The final Si/C/rGO composite was obtained by reducing graphene oxide using hydrazine hydrate ($N_2H_4H_2O$). As illustrated in Figure 8d, the Si/C/rGO composite exhibited excellent cycling performance as a LIB anode, achieving an initial coulombic efficiency (ICE) of 85%. Specific capacities were notably high, with 502 mAh/g at 2 A/g and 1004 mAh/g at 1 A/g after 270 cycles and a capacity retention of 374 mAh/g at 3 A/g after 220 cycles, demonstrating superior performance compared to the theoretical capacity of graphite. Furthermore, an excellent rate performance of 460 mAh/g was achieved at 5 A/g. The performance characteristics of these various nanostructures are summarized in Table 3.

3 | Characteristics of Si Anodes

3.1 | Structure and Properties of Si Anodes

A rational design of Si-based nanostructures is among the most promising strategies to mitigate electrode deformation and crack formation during volume expansion [80]. Several structural designs, such as nanosized, porous, and hollow materials, along with core-shell and yolk-shell architectures, are being explored for Si anodes to improve their performance. Si nanomaterials, with dimensions ranging from 1 to 100 nm, can be classified into four types: nanoparticles, nanotubes, nanosheets, and nanowires.

Nanoparticles, which are zero-dimensional (0D) nanomaterials with spherical shapes, benefit from short ion diffusion paths that enhance the rate capability of batteries. One-dimensional (1D) nanotubes and nanowires possess longitudinal axes that allow for free expansion during lithiation, providing effective pathways for charge carrier transport and improving energy density [81, 82]. Nanosheets, 2D structures, exhibit dimensions perpendicular to their plane, which helps reduce volume changes during lithiation and delithiation while offering a high surface area with numerous exposed redox-active sites, thereby enhancing electrochemical performance [83].

The incorporation of porous structures in Si anodes enhances structural stability by accommodating volume expansion

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

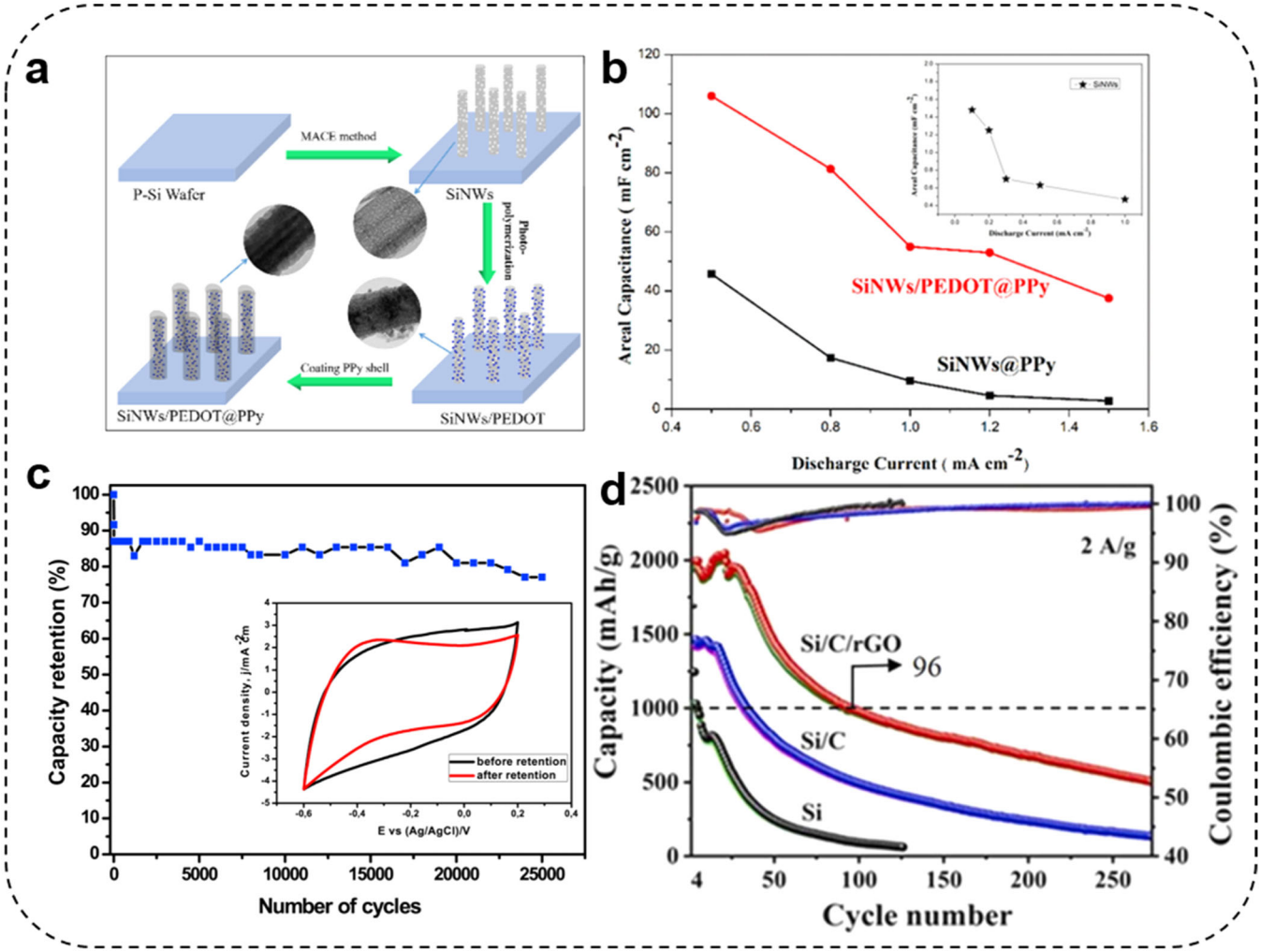

FIGURE 8 | (a) Schematic diagram showing the preparation of SiNWs/PEDOT@PPy electrodes. (b) Plots of areal capacitance as a function of current density for SiNWs@PPy and SiNWs/PEDOT@PPy electrodes [77]. (c) Cycle stability of Ru/Si NWs electrode over 25,000 cycles. The inset shows the cyclic voltammetry curves before and after the cycling test at a scan rate of 100 V/s [78]. (d) Long-term cycle stability of Si, Si/C, and Si/C/rGO electrodes at a current density of 2 A/g [79].

TABLE 3 | Characteristic performance of various nanostructures.

Silicon Nanostructure	Specific capacity (mAh/g)	Cycle stability (cycles)	Volume expansion control	Remarks
Si Nanoparticles (SiNPs)	~3500–4200	100–200	Limited without structural support	High theoretical capacity; prone to pulverization.
Si Nanowires (SiNWs)	~3000–3800	300–500	Good tolerance to volume changes	Better cycle stability due to high surface area and 1D structure.
Yolk-Shell Si/C Structures	~2000–3500	500–1000	Very effective	Void space buffers expansion; stable SEI formation.
Si/Carbon Nanotube (CNT) Composites	~1500–3000	400–800	Moderate	Enhanced conductivity; CNTs provide structural flexibility.
Si/Graphene Composites	~2000–3500	600–1000	Moderate	High conductivity; graphene sheets buffer expansion.
Porous Si Structures	~2000–3000	500–700	Effective	Porous design improves Li diffusion; reduces internal stress.
Doped Si (e.g., N, P doped)	~2500–3800	400–600	Limited	Doping enhances conductivity and stabilizes SEI layer.

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/hne2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

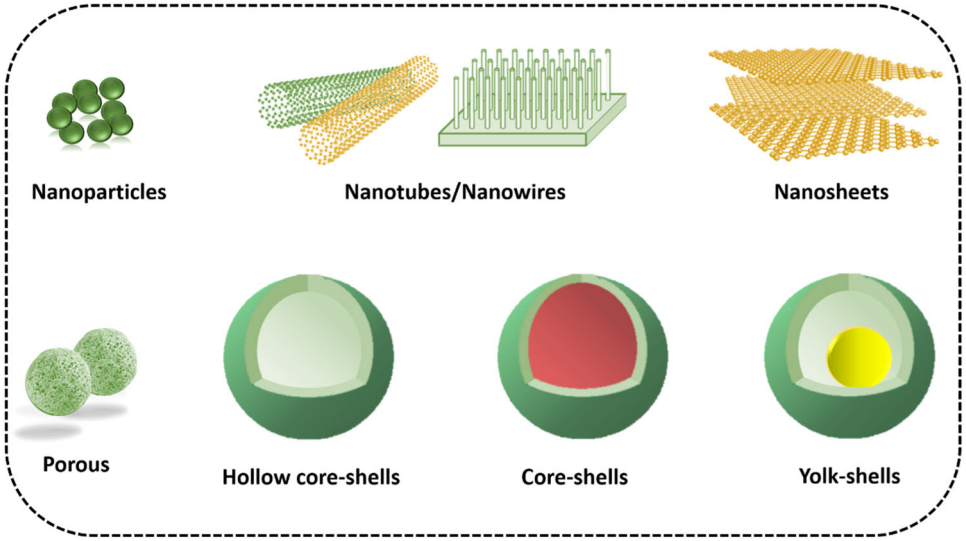

FIGURE 9 | Various Si structures designed to overcome the shortcomings of Si anodes.

through voids, which also facilitates Li^+ mobility and ion diffusion. Hollow 3D core-shell structures similarly buffer the volume expansion of SiNPs. The outer shell promotes direct contact between the SiNPs and the electrolyte, enhancing SEI stability [84]. The 3D open-channel architecture exposes the inner layer to the electrolyte, facilitating continuous SEI growth on the interconnected surface and reducing the initial coulombic efficiency (ICE) of the electrode. Core-shell structures, commonly proposed for Si/C anodes, consist of an inner Si core coated with a carbon-based outer shell, which protects the inner Si framework and minimizes direct electrolyte contact [85, 86]. This configuration stabilizes the SEI, improving cycle stability and coulombic efficiency.

Yolk-shell structures, distinct from core-shell designs, feature an inner Si yolk and an outer carbon shell separated by a void space, which confines the volume expansion of the Si yolk without compromising the outer shell. This design accommodates the Si yolk's volume changes while maintaining the electrode's microstructure, enhancing battery safety. An ideal yolk-shell structure should incorporate a conductive and rigid outer shell to improve electrical conductivity and maintain the electrode's volume during cycling [41, 87]. Various structural designs for Si anodes are illustrated in Figure 9, each addressing the limitations of pure Si anodes. The advantageous properties of Si anodes for battery applications are summarized in Table 4, along with recommendations for further enhancements.

Recent studies have focused on utilizing micron-sized Si particles or embedding them within carbon frameworks to improve structural stability and maintain conductive pathways during repeated cycling. For example, An et al. [90] developed a porous Si/C composite microsphere (P-Si/C@C) using a cost-effective spray-drying and chemical vapor deposition method. This composite structure, featuring Si nanoparticles within a micron-sized carbon matrix, effectively mitigated the volume expansion of Si while enhancing structural stability through an outer carbon shell. The P-Si/C@C anode demonstrated a high ICE of 89.8% and a reversible capacity of 1269.6 mAh/g, retaining 87.1% capacity over 820 cycles. In practical tests within a 3.2 Ah cylindrical cell, the composite retained 81.4% capacity after 1200 cycles, highlighting its potential for long-life applications in electric vehicles.

Another study by An et al. [91] produced a novel "ant-nest-like" porous Si structure via a top-down approach using magnesium-silicon alloy as the starting material. This structure featured interconnected nanoligaments and bicontinuous nanopores that effectively accommodated volume expansion and prevented structural disintegration. With a carbon coating, this porous Si anode achieved a high capacity of 1271 mAh/g, retaining 90% of its capacity over 1000 cycles with minimal electrode swelling. When paired with a $Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2$ cathode, the anode delivered an energy density of 502 Wh/kg and 84% capacity retention after 400 cycles, marking it as a viable option for high-energy-density applications.

Xu et al. [92] developed a scalable methodology for producing kilogram-scale graphite-Si-C/TiO_2 composites (GSCT), embedding Si nanoparticles within a micron-scale graphite framework coated with a C/TiO_2 layer [92]. This composite achieved a high ICE of 80%–83% and a tap density of 0.82 g/cm^3, while maintaining a high reversible capacity of 919.8 mAh/g over 900 cycles, demonstrating its stability for long-term cycling. The incorporation of a thin TiO_2 layer and carbon coating improved structural integrity and interfacial compatibility, validated through extensive electrochemical testing. When assembled into a full cell, the GSCT anode delivered a high energy density of 288.4 Wh/kg, indicating substantial commercial potential for Si-C anodes. Collectively, these studies highlight significant advancements in the design and scalability of Si-C anodes, providing promising pathways for the development of high-capacity, stable, and scalable lithium-ion battery technologies.

3.2 | Si Anode Fabrication Techniques and Scalability

For practical applications, the desired structure and properties of Si anodes must be imparted through a simple, cost-effective,

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

TABLE 4 | Merits and demerits of Si anodes.

Properties of Si anodes	Significant findings related to battery applications	Description
Merits		
Gravimetric capacity	Si anodes exhibit a high theoretical specific capacity of 4200 mAh/g.	The high specific capacity of Si anodes means that they can store more energy per unit of volume or weight.
Volumetric capacity	Si can deliver a volumetric capacity of 2357 mAh/cm^3 at room temperature [88].	A higher gravimetric or volumetric energy density means the ability to store energy in a lighter or smaller package.
Structural properties	Structural flexibility.	Si can be engineered into different structures such as NPs, nanotubes, and nanosheets to optimize its performance in battery anodes.
Production cost	The production of Si anodes is cost-effective.	Si is the second most abundant element in the Earth's crust and is widely available, making it a low-cost material for use in battery anodes. The high energy density and long lifespan of Si anodes make them cost-effective in electric vehicle applications.
Demerits		
Volume expansion	Large volume expansion (up to nearly 300%) during cycling.	This disadvantage can cause mechanical stress and lead to electrode degradation and failure. Improvement: 1) Design a nanostructured Si material. 2) Combination Si with another material (i.e., carbon or metal oxide).
Electrical conductivity	Low electrical conductivity of 6.7×10^{-4} S/cm [89].	The low electrical conductivity can limit the power output, especially at high current rates. Improvement: 1) Develop Si-based composite materials such as Si–carbon nanofibers and Si–graphene hybrids. 2) Modify the electrode morphology, adjust the particle size distribution, or select an appropriate electrolyte solvent or salt.
Cycling performance	Si anodes have poor cycling performance.	The large volume change and low electrical conductivity of Si anodes lead to poor cycle stability and rate capability. Improvement: 1) The volume change can be reduced by reducing the particle size to the nanoscale (e.g., NPs and NWs). 2) The electric conductivity of Si anodes can be improved by coating the Si particles with carbon, metal oxides, or polymers.

and easily scalable fabrication technique. Si anode materials can be synthesized via either bottom-up or top-down methods. Among bottom-up methods, chemical vapor deposition (CVD) is the most commonly used for producing stable Si anode materials with high purity and performance [93]. The CVD process involves depositing a solid material from a gaseous phase onto a heated substrate through chemical reactions. In battery applications, CVD is the most popular and simple method to deposit various forms of carbon onto the surfaces of Si particles [94]. Amorphous carbon nanotubes (ACNTs) consisting of carbon clusters have been deposited onto the surfaces of Si particles [95]. The fabrication process of the Si/ACNT composites via impregnation and in situ CVD is illustrated in Figure 10a. Compared to Si, carbon has a higher electrical conductivity. Ideally, the deposition of a carbon material onto the surface of Si via CVD will improve the electrical conductivity of the anode. The morphological evaluation of the Si/ACNT composite (Figure 10b) indicated that an interwoven ACNT network wrapped the Si particles. The ACNTs provide a large amount of void space (Figure 10b(iii) and (iv)), which is beneficial for accommodating the volume expansion of the SiNPs and for electrolyte penetration. These voids enhance the transport of Li^+ within the device. Similarly, CNTs grown on the hard carbon (HC) encapsulating Si were shown to improve the conductivity of the anode [96]. For example, Kim et al. synthesized Si@HC@CNT via CVD under acetylene gas [96], as shown in Figure 10c. The deposition of CNTs enhanced the conductivity of the anode by providing an electrical network between Si and carbon in the composite. Meanwhile, HC derived from natural polysaccharides provided an effective conductive matrix for Si particles.

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/tne2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

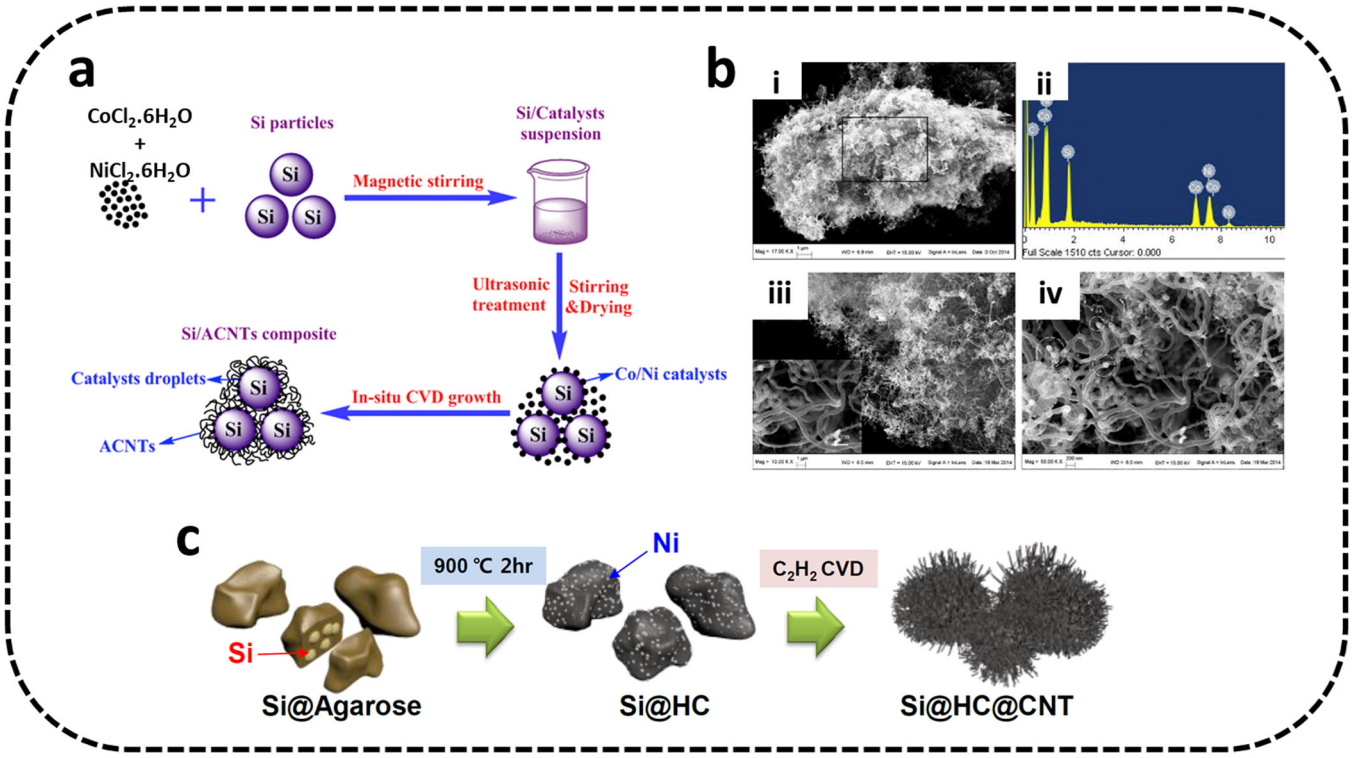

FIGURE 10 | (a) Schematic illustration showing the fabrication of Si/ACNT composite via impregnation and in situ CVD to grow coiled ACNTs on Si particles [95]. (b) Field-emission scanning electron microscopy images and energy-dispersive X-ray spectrum of the synthesized Si/ANCT composite [95]. (c) Schematic illustration of the facile CVD-based synthesis of Si@HC@CNT [96].

The sol–gel method is a widely used bottom-up synthesis technique for fabricating nanomaterials with high purity, uniformity, and controlled chemical composition under mild conditions of low temperature and pressure. This method is particularly suited for large-scale production due to its simplicity and versatility [97]. For instance, the Si@TiO_2@rGO composite has been synthesized by combining sol–gel processing with graphene encapsulation [98, 99]. In this approach, the sol–gel method is employed to coat SiNPs with a TiO_2 shell. TiO_2 serves as an effective anode material due to its minimal volume expansion (< 4%) during lithiation and its porous structure, which enhances Li^+ ion diffusion [100]. The subsequent encapsulation with flexible graphene sheets introduces additional benefits, such as improved electrical conductivity and the creation of microscopic pores that facilitate Li^+ transport, thereby enhancing the charge–discharge performance of lithium-ion batteries [101]. Two fabrication strategies for Si@TiO_2@rGO are depicted in Figure 11a,b. The approach in Figure 11b incorporates an electrostatic interaction-assisted spray-drying step, yielding pomegranate-shaped Si@TiO_2@rGO microspheres with unique structural properties. Although sol–gel synthesis has also been utilized for producing Si/C nanofibers and Si-based composites, challenges remain in achieving consistent quality and scalability for mass production. Similarly, while CVD offers advantages for depositing carbon materials onto Si surfaces to enhance conductivity and stability, its high capital and operational costs, driven by the need for specialized equipment and stringent environmental controls, limit its economic feasibility for large-scale applications. Continued advancements in these methods are critical to addressing these limitations and enabling broader commercial adoption.

A critical factor influencing the scalability of energy storage materials is the ability to produce consistent, high-quality products at industrial volumes while minimizing production costs. Future research should prioritize simplifying synthesis protocols, reducing raw material costs, and integrating renewable resources. Techniques that enable room-temperature synthesis, such as certain top-down mechanical milling approaches [102], present a promising direction by lowering energy consumption and the associated costs of high-temperature methods.

Mechanical milling can be classified into two categories: wet milling, which uses a liquid medium, and dry milling, which does not [103]. In wet milling, slight modifications to milling time can result in significant changes in particle size, with longer milling times yielding smaller particles. However, wet milling is susceptible to product contamination, prompting many mechanical milling processes to be conducted in a dry environment. During mechanical milling, both mechanical action and chemical reactions contribute to particle size reduction [104]. Initially, mechanical force brings the material to a nonequilibrium state, resulting in high-energy collisions between milling balls that fracture the particles. This repeated fracturing is responsible for reducing particle size.

Recently, Zhang et al. reported a high-energy mechanical milling (HEMM) process to produce silicon/carbon (Si/C) microspheres as anode materials [105]. They incorporated ethanol during the HEMM process to prevent thermally induced oxidation of silicon and to facilitate particle size reduction. Subsequently, sucrose was coated onto the SiNPs through a hydrothermal process followed by carbonization. This coating

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

encapsulates the SiNPs in carbon microspheres, minimizing detrimental interactions between the SiNPs and the electrolyte.

Electrospinning is another simple and effective top-down method to produce Si/C composite nanofibers containing SiNPs that are uniformly embedded in the carbon fibers [106]. During nanofiber production, polymers such as polyacrylonitrile, polyvinyl pyrrolidone, and polyamic acid are carbonized and serve as the main carbon source. Kim et al. [107] used polyacrylonitrile as a carbon source to electrospin Si/C nanofibers, resulting in an extensive contact area between Si and the carbon phase. This type of Si nanofiber is expected to provide abundant conduction pathways for electron transfer while also helping to minimize the volume expansion and electrochemical isolation during cycling. Wang et al. [108] reported a coaxial electrospinning method to produce Si/CNF composites from polyamic acid as a carbon precursor. The prepared Si/CNF composites showed good mechanical strength, high surface area, and excellent electronic conductivity. This coaxial electrospinning method was performed at low temperatures without sophisticated equipment. As illustrated in Figure 12b, pumpkin-like Si/C nanocomposites were sheathed in N-doped porous carbon nanofibers, and Si/C-ZIF-8/CNFs with uniform diameters were produced by electrospinning followed by heat treatment [109]. The Si/ZIF-8 nanofibers were produced during electrospinning from a polyacrylonitrile and dimethylformamide solvent mixture. After heat treatment under nitrogen atmosphere at 950°C, ZIF-8 and polyacrylonitrile were decomposed and transformed into N-doped porous carbon layers. This work demonstrates that a conductive network formed by N-doped porous carbon nanofibers can improve the electrical conductivity of Si, which is beneficial. SiNPs–CNTs composites have also been produced using a combination of mechanical milling and electrospinning [110], as illustrated in Figure 12c. In this process, mechanical milling is first used to downsize the Si particles to the nanoscale. Subsequent electrospinning using polyacrylonitrile as a carbon precursor then forms the SiNPs–CNTs composite. This combination of techniques offers smaller nanoparticles with higher surface areas, which helps improve the rate performance by enhancing the contact between the anode material and the electrolyte.

4 | Applications of Si Anodes in Superbatteries

4.1 | Potential for Si Anodes to Revolutionize the Battery Industry

4.1.1 | Consumer Electronics and Mobile Devices

Si anodes hold the potential to significantly transform the battery industry, particularly in consumer electronics and mobile devices. Traditionally, most of these devices rely on LIBs that use graphite as the anode material. However, Si anodes offer several advantages, including higher energy density, faster charging times, and longer lifespans [111]. The integration of Si anodes in electronic and mobile devices could greatly enhance their performance and energy storage capabilities, benefiting both consumers and manufacturers.

Leading companies such as Tesla, Samsung, and LG are actively exploring the incorporation of Si anodes into their battery designs. In 2015, Samsung introduced an innovative battery design featuring Si anodes layered with graphene, claiming a potential battery life of up to 21 h, significantly exceeding that of conventional lithium-ion batteries. Six years later, Samsung SDI launched its PRiMX battery brand, which combines a high-nickel cathode with Si anodes to enhance runtime, lifespan, and charging speed. LG has also pursued Si anode technology to improve battery capacity and reduce charging times. In collaboration with researchers at the University of California, San Diego, LG Energy Solution developed a battery that employs a solid-state electrolyte in conjunction with all-Si anodes, eliminating the need for carbon and binders. This innovative approach replaced the conventional liquid electrolyte with a sulfide-based solid electrolyte, resulting in a remarkably stable battery.

Overall, batteries utilizing Si anodes demonstrate strong potential for applications in wearable technologies and other electronic devices, such as drones and remote sensors, due to their superior energy density, faster charging capabilities, and enhanced stability compared to traditional LIBs. Despite these promising advancements, it may still take several years before Si anode technology becomes widely accessible in the market.

4.1.2 | Electric Vehicles and Renewable Energy Storage

The global shift toward sustainability necessitates reducing fossil fuel consumption, as their combustion contributes to harmful emissions such as nitrogen oxides, worsening air pollution and accelerating climate change. Transitioning to renewable energy storage is essential to mitigate these challenges. Since vehicles are significant sources of nitrogen oxide emissions, adopting electric vehicles (EVs) powered by Li metal anodes is a key strategy. However, pure Li metal anodes face limitations, prompting significant interest in Si-based anode materials. Si alloys, particularly $Li_{22}Si_5$, offer a superior volumetric capacity (2190 mAh/cm^3 in the fully charged state) compared to pure Li. Si anodes are promising for EV applications due to their high theoretical specific capacity, enabling fast charging at room temperature without compromising driving range [112]. Tesla, for example, uses batteries with 5% Si in the anodes of Panasonic cells in the Model X [113], and in 2020, Tesla announced a new “tabless” battery technology incorporating Si anodes, claiming a 16% increase in driving range. However, the timeline for the widespread availability of this technology remains unclear. Other companies, including LG and Samsung, are also advancing Si anode battery technologies to enhance EV performance and reduce costs. In 2023, LG Chem introduced a pure Si development prototype for EVs, and BMW has plans to integrate Si anodes in future EV battery models. Sila Nanotechnologies aims to commercialize Si anodes by 2025. Despite the promising potential of Si anode batteries for electric vehicles, several challenges remain to be addressed. Notably, the expansion and contraction of Si during charge–discharge cycles can lead to battery degradation over time. Researchers are actively investigating solutions to these challenges, and further advancements in this area are anticipated in the coming years.

4.1.3 | Military and Aerospace Applications

Experimental use of Si anodes in military technologies, particularly in unmanned aerial vehicles (UAVs), has gained traction.

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

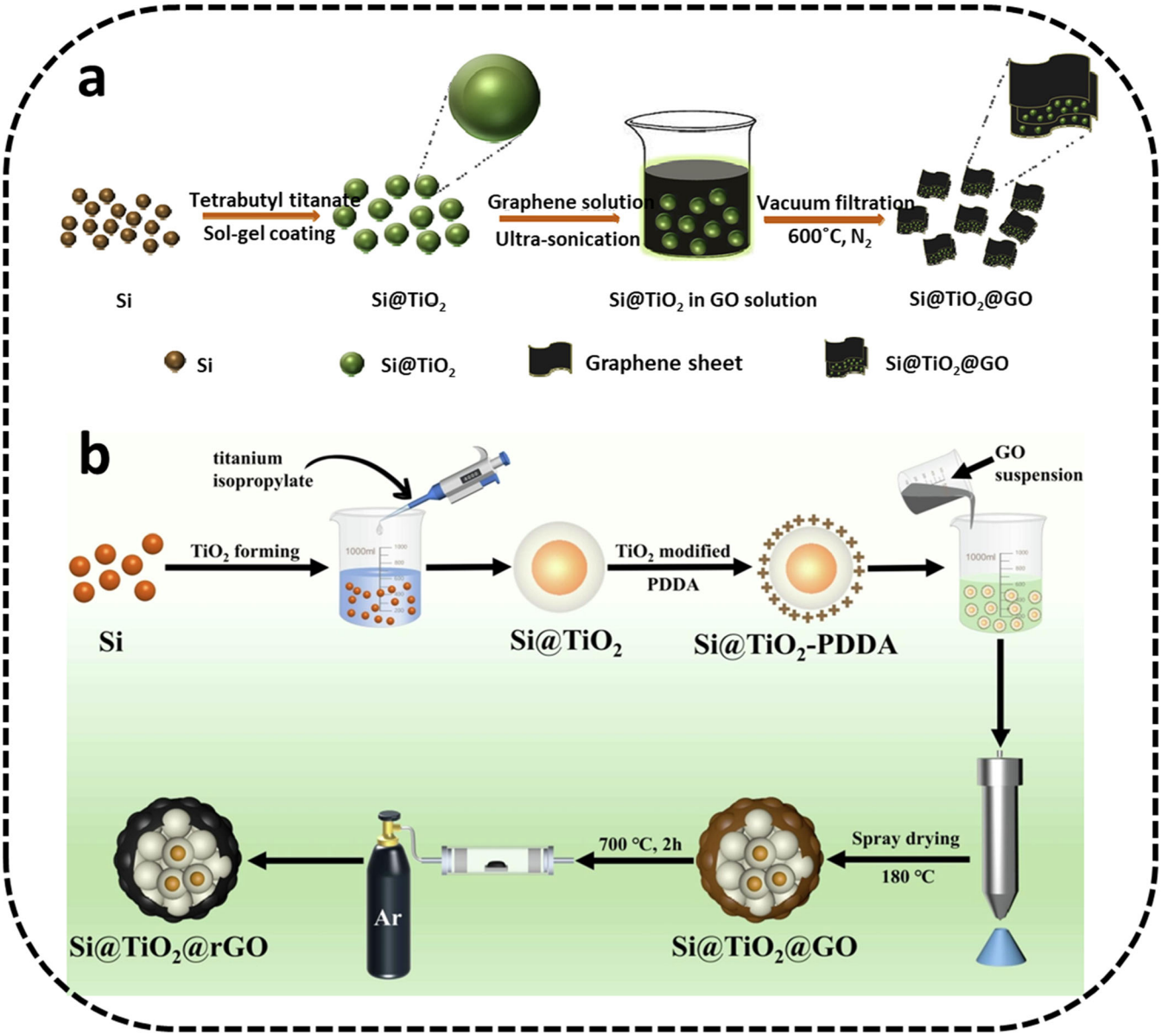

FIGURE 11 | (a) Schematic illustration of the preparation of Si@TiO_2 [98]. (b) Schematic illustration of the preparation of pomegranate-shaped Si@TiO_2@rGO microspheres [99].

Recently, US Army's Rapid Capabilities and Critical Technologies Office began work with Amprius Technologies, the first company to use 100% Si anodes in LIBs. These batteries, specifically designed for small UAVs, demonstrated significantly longer flight times and enhanced power density compared to conventional LIBs. However, integrating Si anodes into military applications presents unique challenges, including stringent durability, reliability, and security requirements that exceed those in commercial sectors. Concurrently, ongoing research within the aerospace industry focuses on optimizing battery performance and energy density. Si anodes, known for their potential to deliver higher energy density and extended lifespan relative to traditional graphite anodes, are particularly advantageous for space missions where weight and longevity are critical.

In a collaborative effort with the University of Maryland, the Army Research Laboratory has developed innovative solutions to enhance battery performance. Their research indicates that incorporating a self-healing protective layer can significantly mitigate the degradation of both the electrolyte and Si anodes, resulting in battery designs that achieve over 100 cycles with minimal degradation, a significant improvement compared to previous iterations, which typically lasted only tens of cycles [114]. While Si anodes are not yet utilized in aerospace applications, ongoing research suggests that they may enhance the performance of batteries used in space exploration in the future.

4.1.4 | Potential for New Applications and Innovations in Energy Storage

Si and its derived materials are central to the development of advanced anode materials essential for driving the fourth industrial revolution (4IR). Characterized by the integration of digital technologies such as the Internet of Things (IoT),

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

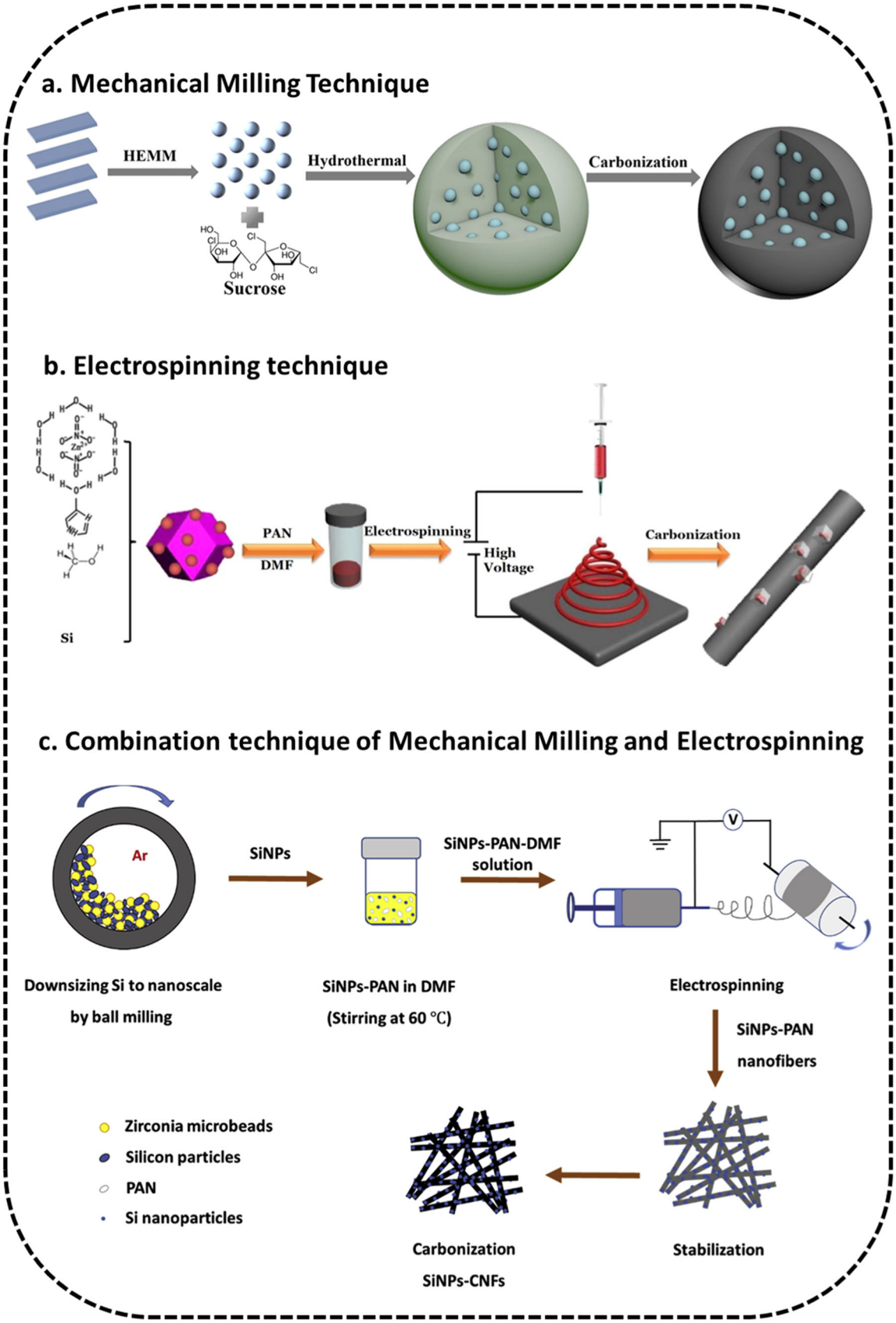

FIGURE 12 | (a) Schematic illustration of the synthesis of Si@C microspheres via mechanical milling [98]. (b) Schematic illustration of the preparation of Si/C-ZIF-8/CNFs via electrospinning [109]. (c) Schematic of the synthesis of Si/C via mechanical milling followed by electrospinning [110].

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/he2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

artificial intelligence (AI), and robotics, the 4IR is driving a significant increase in energy demand alongside the need for efficient, reliable, and sustainable energy storage solutions. Si anodes offer the potential for higher energy density, longer battery life, and faster charging, which are essential for meeting the growing energy storage requirements associated with this technological transformation. Moreover, Si anodes facilitate the development of innovative energy storage applications, including grid-scale storage solutions for renewable energy sources like wind and solar power. Their implementation can enhance energy storage capacity and discharge efficiency, contributing to grid stability and reducing dependence on fossil fuel-based power generation. These advancements are pivotal for achieving a more sustainable and decentralized energy infrastructure, aligning with the overarching goals of the 4IR.

Ongoing research is focused on developing stable Si composite materials, optimizing the formation of solid electrolyte interphases, and assessing performance across thousands of charge/discharge cycles. As these efforts progress, Si anodes are poised to enable new applications in energy storage across various sectors, marking them as a key innovation in the transition toward a sustainable energy future.

4.1.5 | Enhancing Energy Density and Cycle Life of Superbatteries With Si Anodes

Superbatteries have a high energy density of 10 kW/kg and are safer to handle than common batteries. Si anodes can potentially improve the energy density and life cycle of superbatteries because Si anodes can store more ions than graphite anodes commonly used in commercial batteries. This is a result of the high theoretical capacity of Si for ion storage, which is approximately 10 times higher than that of graphite. However, the practical use of Si as an anode material has been limited due to its tendency to expand and contract during charging and discharging cycles, leading to mechanical stress and, ultimately, electrode degradation and failure. To address this issue, researchers have developed various approaches, including nanostructuring, coating, and alloying, to improve the stability and durability of Si anodes. For example, nanostructuring can create porous Si structures that accommodate the volume change of Si without causing excessive stress, while coating can form a protective layer to prevent direct contact between Si and the electrolyte. Coating SiNWs with carbon improves the reversible capacity up to 1500 mAh/g [115]. Alloying Si with other materials, such as carbon or tin, can enhance its stability and conductivity. Based on these strategies, Si anodes have demonstrated improved performance in superbatteries in terms of cycle stability and rate capability, leading to higher energy density and longer battery life.

5 | Challenges and Limitations

5.1 | Current Limitations of Si Anode Technology

The primary limitation of Si anode technology is the significant volume expansion and contraction that occurs during lithiation and delithiation. This cyclical volume change reduces the electrical contact between the current collector and the active material, leading to the pulverization of Si due to hoop compressive and tensile stresses. When the stress induced by lithiation exceeds the cohesive strength of the Si surface, cracking occurs. Over time, as the number of charge–discharge cycles increases, the electrode may begin to peel, resulting in capacity degradation due to the fluctuating volume of the entire electrode [40]. Another significant challenge is the formation of the SEI during the lithiation of Si at low potentials (below 0.5 V vs. Li/Li^+). This process involves side reactions between the electrolyte and the Si surface, resulting in the generation of SEI layers. However, the substantial volume changes associated with Si lead to the rupture of the SEI film, exposing fresh Si to the electrolyte. This exposure consumes active Li^+ to form additional SEI layers, leading to the accumulation of "dead lithium," which is lithium trapped within the SEI and unavailable for electrochemical reactions. As the amount of dead lithium increases, both the Coulombic efficiency and ionic conductivity decrease, ultimately contributing to the failure of batteries utilizing Si anodes [29].

5.2 | Challenges of Si Anodes in Solid-State Electrolytes

The performance and stability of Si-based anodes in LIBs and solid-state batteries (SSBs) are significantly influenced by electrolyte optimization, particularly the SEI. The large volume changes Si undergoes during lithiation and delithiation challenge SEI stability and mechanical integrity, limiting cycling performance. Solid-state electrolytes (SSEs) offer distinct advantages over traditional liquid electrolytes, notably in enhancing safety and improving SEI durability. SSEs have been shown to optimize SEI formation and improve the mechanical stability of Si anodes, making them a promising solution for SSBs.

Han et al. [116] suggested replacing conventional carbonate-based electrolytes with SSEs to improve LIB safety. This demonstrates that SSEs reduce electrolyte leakage and side reactions due to their lower reactivity and reduced permeability. These SSEs also promote the formation of a stable SEI on the Si surface, which is crucial for maintaining anode integrity during cycling. Si's high specific capacity makes it an attractive option for next-generation SSBs; however, its substantial volume changes lead to poor mechanical stability and SEI degradation. Han et al. address these issues with a Si/MXene architecture engineered with nitrogen doping, which enhances both electrochemical and mechanical stability by improving interfacial bonding and forming an amorphous carbon shell. This design helps maintain the SEI integrity and supports stable cycling and high-rate performance, with nitrogen-doped MXene layers facilitating lithium-ion transport.

Gu et al.'s [117] work focused on Si anodes with a shell-coated design ($Si@SiO_2@LPO@C$), which includes an outer solid-state electrolyte layer that effectively shields the Si from the liquid electrolyte. This layered structure not only reduces the energy barrier for lithium-ion transport but also promotes a more uniform SEI. The SSE, particularly one derived from poly (ethylene oxide) (PEO)/LiTFSI, forms a LiF-rich SEI with

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

favorable electrochemical characteristics. This SEI is highly stable, only forming on the outer surface of the Si anode, which reduces side reactions and limits SEI growth within the anode structure itself. Gu et al. show that this design enables a high capacity and stable cycle performance in both liquid and solid-state LIBs. The introduction of the Li_3PO_4 and carbon shell enhances mechanical integrity, allowing the Si to endure the stresses associated with volume change without compromising the SEI. This enables Si anodes to reach high capacities, maintaining 1012.4 mAh/g over 200 cycles. Similarly, Han et al. developed a porous Si anode decorated with silver nanoparticles (PS-Ag-C) to address mechanical challenges associated with Si volume expansion [118]. The silver nanoparticles and a thin carbon layer improve interfacial contact, facilitating continuous charge transfer and stabilizing the SEI. Coupling the PS-Ag-C anode with an SSE such as PVDF-HFP/$Li_{1.3}Al_{0.3}Ti_{1.7}(PO_4)_3$ (LATP) results in a LiF-rich SEI that enhances interfacial stability. This design achieves high areal capacities and maintains cycling stability due to favorable SEI formation and stress relief from the porous Si structure.

5.3 | Technical and Economic Challenges

The implementation of Si anodes in advanced batteries faces significant technical and economic challenges, particularly related to cost and scalability. The high manufacturing cost of Si anodes arises from complex fabrication processes and the need to convert bulk Si into nanoscale structures. Consequently, transitioning from conventional graphite anodes to Si anodes will require the development of innovative, cost-effective production methods. For instance, Gao et al. [119] explored cost-effective methods for producing porous Si anodes using low-temperature aluminothermic reduction of two natural raw materials: attapulgite, a magnesium-aluminum silicate clay with a needle-like morphology, and mica, a mineral group characterized by layered structures of silica tetrahedra (SiO_4). The resulting porous Si composites demonstrated impressive specific capacities of 799 and 697 mAh/g, respectively. Similarly, in another study, pure SiNPs for high-performance LIB anodes were produced from rice husks, an agricultural byproduct. The resulting anode showed an extraordinarily high specific capacity of 2790 mAh/g, seven times higher than a graphite anode. This high specific capacity can be attributed to the porosity and small sizes of the SiNPs recovered from rice husks, which result in a large surface area of the Si anode. The simplicity, cost efficiency, and scalability of the method reported to produce rice husk-derived SiNPs make it attractive for producing next-generation LIBs. In the future, to realize the transition from graphite anodes to Si anodes, new techniques are needed to overcome the above technical and economic challenges.

5.4 | Advances and Future Directions in Si Anode Technology

Si anodes are poised for significant advancements in LIBs, particularly for EV applications. Current LIB technology typically achieves an energy density of approximately 150 Wh/kg, but EVs require higher energy densities and enhanced operational safety. The integration of high-voltage cathodes and the development of high-capacity anode and cathode materials are essential for increasing the overall energy density of Li batteries. Crucially, the anode capacity must exceed that of the cathode to maintain a positive capacity ratio; otherwise, during deep charging, excess Li^+ can accumulate, leading to Li metal layer formation on the anode surface. This poses significant safety risks, including the formation of lithium oxides and unwanted reactions with the electrolyte.

The introduction of Si into anodes can improve energy density and decrease recharging times, ultimately reducing costs. Researchers are actively working to enhance composite materials and fabrication techniques for Si anodes, leveraging Si's unique properties to boost the performance of graphite-based LIBs. However, both Si and graphite must be optimized to achieve a synergistic effect for optimal battery performance.

Si also shows promise in SCs, where its high theoretical specific capacity and abundance make it an attractive electrode material. Studies indicate that Si-based composites exhibit excellent performance in SC applications, attributed to their superior capacity and conductivity [65, 66, 120]. Furthermore, Si-based composites have potential applications in other areas, including supercapatteries [121], solar cells [122], and fuel cells [123].

Future advancements in Si anode technology are expected to broaden its applications beyond conventional energy storage, facilitating a new generation of energy harvesting devices. Key areas of focus include enhancing the durability and stability of Si anodes to support continuous energy harvesting in applications such as environmental sensors and wearable health devices, where long-term operation is critical. Innovations in Si nanostructuring, including porous and core-shell designs, are being explored to manage volume expansion, extend cycle life, and reduce degradation. Moreover, integrating Si anodes with hybrid energy systems, such as supercapatteries or hybrid capacitors, could enhance energy conversion and storage efficiency, making them ideal for applications with variable energy inputs, such as IoT devices. The development of heteroatom-doped Si anodes may further improve electrical conductivity and solid-electrolyte interphase stability, enabling these anodes to withstand high-frequency cycling demands in IoT applications. As Si anode technology continues to evolve, it increasingly aligns with the requirements of low-power energy harvesting electronics, advancing the potential for autonomous, self-powered systems across various applications.

6 | Conclusion

Si anodes hold significant promise for next-generation LIBs due to their high theoretical specific capacity, which far exceeds that of conventional graphite anodes. Recent advancements in Si nanostructuring and composite designs have effectively addressed key challenges, including volume expansion, low conductivity, and instability of the SEI, leading to improvements in cycle life, power density, and overall stability. These innovations position Si anodes as a compelling choice for applications demanding high energy density and long cycle life, such as electric vehicles, consumer electronics, and renewable energy grid storage. However, challenges to commercializing Si anodes persist, including the need for scalable fabrication techniques

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

and strategies to mitigate electrode degradation. Emerging methods, such as mechanical milling, electrospinning, and the application of carbon or polymer coatings, show strong potential to overcome these barriers. Collaborative efforts across industry, academia, and government will be critical in accelerating development and optimizing cost-effective, sustainable production processes. With continued technological advancements, Si anodes are expected to drive the commercialization of LIBs, meeting the energy demands of the 4IR. Ultimately, Si anodes will play a pivotal role in advancing energy storage solutions, contributing to a more sustainable and efficient global energy infrastructure.

Acknowledgments

The authors would like to thank the financial support from the Sunway University Research Grant (GRTIN-RRO-60-2022) and the International Research Network Grant Scheme (STR-IRNGSSET-GAMRG-01-2022). We also thank LetPub (www.letpub.com) for linguistic assistance and pre-submission expert review.

Conflicts of Interest

The authors declare no conflicts of interest.

References

1. Y. Li, Q. Li, and Z. Tan, "A Review of Electrospun Nanofiber-Based Separators for Rechargeable Lithium-Ion Batteries," *Journal of Power Sources* 443 (2019): 227262.

2. W. Zhang, S. Zhang, L. Fan, et al., "Tuning the LUMO Energy of an Organic Interphase to Stabilize Lithium Metal Batteries," *ACS Energy Letters* 4 (2019): 644–650.

3. S. Ould Amrouche, D. Rekioua, T. Rekioua, and S. Bacha, "Overview of Energy Storage in Renewable Energy Systems," *International Journal of Hydrogen Energy* 41 (2016): 20914–20927.

4. V. Aravindan, J. Gnanaraj, Y.-S. Lee, and S. Madhavi, "Insertion-Type Electrodes for Nonaqueous Li-Ion Capacitors," *Chemical Reviews* 114 (2014): 11619–11635.

5. H. Ji, C. Liu, T. Wang, et al., "Porous Hybrid Composites of Few-Layer MoS_2 Nanosheets Embedded in a Carbon Matrix With an Excellent Supercapacitor Electrode Performance," *Small* 11 (2015): 6480–6490.

6. L. Li, H. Yang, J. Yang, et al., "Hierarchical Carbon@Ni_3S_2@MoS_2 Double Core–Shell Nanorods for High-Performance Supercapacitors," *Journal of Materials Chemistry A* 4 (2016): 1319–1325.

7. Y. Wang, Y. Song, and Y. Xia, "Electrochemical Capacitors: Mechanism, Materials, Systems, Characterization and Applications," *Chemical Society Reviews* 45 (2016): 5925–5950.

8. J. Lu, Y. Zhang, X. Gong, et al., "High-Yield Synthesis of Ultrathin Silicon Nanosheets by Physical Grinding Enables Robust Lithium-Ion Storage," *Chemical Engineering Journal* 446 (2022): 137022.

9. O. Gerard, A. Numan, S. Krishnan, M. Khalid, R. Subramaniam, and R. Kasi, "A Review on the Recent Advances in Binder-Free Electrodes for Electrochemical Energy Storage Application," *Journal of Energy Storage* 50 (2022): 104283.

10. Q. Zhou, M. Bao, and X. Ni, "A Novel Surface Modification of Silicon Nanowires by Polydopamine to Prepare SiNWs/NC@NiO Electrode for High-Performance Supercapacitor," *Surface and Coatings Technology* 406 (2021): 126660.

11. J. Ou, B. Li, H. Deng, K. Li, and H. Wang, "A Carbon-Covered Silicon Material Modified by Phytic Acid With 3D Conductive Network as Anode for Lithium-Ion Batteries," *Advanced Powder Technology* 34 (2023): 103891.

12. O. Gerard, A. Numan, M. A. A. M. Abdah, M. Khalid, S. Ramesh, and K. Ramesh, "Rapid Synthesis of Nickel-Copper Phosphate Electrode by Microwave-Assisted Hydrothermal Reaction for Supercapattery," *Journal of Energy Storage* 61 (2023): 106813.

13. Y. Ren, X. Zhou, J. Tang, et al., "Boron-Doped Spherical Hollow-Porous Silicon Local Lattice Expansion Toward a High-Performance Lithium-Ion-Battery Anode," *Inorganic Chemistry* 58 (2019): 4592–4599.

14. S. Zhu, J. Zhou, Y. Guan, et al., "Hierarchical Graphene-Scaffolded Silicon/Graphite Composites as High Performance Anodes for Lithium-Ion Batteries," *Small* 14 (2018): 1802457.

15. L. Sun, Y. Liu, R. Shao, J. Wu, R. Jiang, and Z. Jin, "Recent Progress and Future Perspective on Practical Silicon Anode-Based Lithium Ion Batteries," *Energy Storage Materials* 46 (2022): 482–502.

16. A. Numan, Y. Zhan, M. Khalid, and M. Hatamvand, "Chapter Three—Introduction to Supercapattery," *Advances in Supercapacitor and Supercapattery* (2021): 45–61, https://doi.org/10.1016/B978-0-12-819897-1.00008-2.

17. Y. Tang, Y. Zhang, W. Li, B. Ma, and X. Chen, "Rational Material Design for Ultrafast Rechargeable Lithium-Ion Batteries," *Chemical Society Reviews* 44 (2015): 5926–5940.

18. M. Osiak, H. Geaney, E. Armstrong, and C. O'Dwyer, "Structuring Materials for Lithium-Ion Batteries: Advancements in Nanomaterial Structure, Composition, and Defined Assembly on Cell Performance," *Journal of Materials Chemistry A* 2 (2014): 9433–9460.

19. A. Gopalakrishnan and S. Badhulika, "Binder-Free Polyaniline Sheathed Crumpled Cobalt Diselenide Nanoparticles as an Advanced Electrode for High Specific Energy Asymmetric Supercapacitor," *Journal of Energy Storage* 41 (2021): 102853.

20. L. Kin, Z. Liu, O. Astakhov, et al., "Efficient Area Matched Converter Aided Solar Charging of Lithium Ion Batteries Using High Voltage Perovskite Solar Cells," *ACS Applied Energy Materials* 3 (2020): 431–439.

21. H. Wan, L. Cai, F. Han, J. P. Mwizerwa, C. Wang, and X. Yao, "Construction of 3D Electronic/Ionic Conduction Networks for All-Solid-State Lithium Batteries," *Small* 15 (2019): 1905849.

22. M. B. Askari, S. M. Rozati, P. Salarizadeh, and S. Azizi, "Reduced Graphene Oxide Supported Co_3O_4–Ni_3S_4 Ternary Nanohybrid for Electrochemical Energy Storage," *Ceramics International* 48 (2022): 16123–16130.

23. Q. Wei, Y. Wu, S. Li, R. Chen, J. Ding, and C. Zhang, "Spent Lithium Ion Battery (LIB) Recycle From Electric Vehicles: A Mini-Review," *Science of the Total Environment* 866 (2023): 161380.

24. D. Zhang, Y. Zhang, H. Liu, Y. Xu, J. Wu, and P. Li, "Effect of Pyrolysis Temperature on Carbon Materials Derived From Reed Residue Waste Biomass for Use in Supercapacitor Electrodes," *Journal of Physics and Chemistry of Solids* 178 (2023): 111318.

25. T. P. Mofokeng, A. K. Ipadeola, Z. N. Tetana, and K. I. Ozoemena, "Defect-Engineered Nanostructured Ni/MOF-Derived Carbons for an Efficient Aqueous Battery-Type Energy Storage Device," *ACS Omega* 5 (2020): 20461–20472.

26. M. Jayachandran, A. Rose, T. Maiyalagan, N. Poongodi, and T. Vijayakumar, "Effect of Various Aqueous Electrolytes on the Electrochemical Performance of A-MnO_2 Nanorods as Electrode Materials for Supercapacitor Application," *Electrochimica Acta* 366 (2021): 137412.

27. H. Zhang, Y. Bai, H. Chen, et al., "Oxygen-Defect-Rich 3D Porous Cobalt-Gallium Layered Double Hydroxide for High-Performance Supercapacitor Application," *Journal of Colloid and Interface Science* 608 (2022): 1837–1845.

28. L. Li, J. Meng, M. Zhang, T. Liu, and C. Zhang, "Recent Advances in Conductive Polymer Hydrogel Composites and Nanocomposites for

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

Flexible Electrochemical Supercapacitors," *Chemical Communications* 58 (2022): 185–207.

29. H. Wu and Y. Cui, "Designing Nanostructured Si Anodes for High Energy Lithium Ion Batteries," *Nano Today* 7 (2012): 414–429.

30. B. Fuchsbichler, C. Stangl, H. Kren, F. Uhlig, and S. Koller, "High Capacity Graphite–Silicon Composite Anode Material for Lithium-Ion Batteries," *Journal of Power Sources* 196 (2011): 2889–2892.

31. Y. Son, J. Ma, N. Kim, et al., "Quantification of Pseudocapacitive Contribution in Nanocage-Shaped Silicon–Carbon Composite Anode," *Advanced Energy Materials* 9 (2019): 1803480.

32. Y. Lai, H. Li, Q. Yang, et al., "Revisit the Progress of Binders for a Silicon-Based Anode From the Perspective of Designed Binder Structure and Special Sized Silicon Nanoparticles," *Industrial & Engineering Chemistry Research* 61 (2022): 6246–6268.

33. H. Liu, Q. Sun, H. Zhang, et al., "The Application Road of Silicon-Based Anode in Lithium-Ion Batteries: From Liquid Electrolyte to Solid-State Electrolyte," *Energy Storage Materials* 55 (2023): 244–263.

34. N. N. Greenwood and A. Earnshaw, eds., *Chemistry of the Elements* (second edition) (Oxford: Butterworth-Heinemann, 1997), 328–366.

35. D. L. Sparks, B. Singh, and M. G. Siebecker, "Soil Minerals," *Environmental Soil Chemistry (Third Edition)* (2023): 39–104, https://doi.org/10.1016/B978-0-443-14034-1.00002-2.

36. M. Raić, L. Mikac, I. Marić, et al., "Nanostructured Silicon as Potential Anode Material for Li-Ion Batteries," *Molecules* 25 (2020): 891.

37. S. J. An, J. Li, C. Daniel, D. Mohanty, S. Nagpure, and D. L. Wood, "The State of Understanding of the Lithium-Ion-Battery Graphite Solid Electrolyte Interphase (SEI) and its Relationship to Formation Cycling," *Carbon* 105 (2016): 52–76.

38. R. N. Seefurth and R. A. Sharma, "Investigation of Lithium Utilization From a Lithium-Silicon Electrode," *Journal of the Electrochemical Society* 124 (1977): 1207–1214.

39. R. A. Sharma and R. N. Seefurth, "Thermodynamic Properties of the Lithium-Silicon System," *Journal of the Electrochemical Society* 123 (1976): 1763–1768.

40. C.-M. Park, J.-H. Kim, H. Kim, and H.-J. Sohn, "Li-Alloy Based Anode Materials for Li Secondary Batteries," *Chemical Society Reviews* 39 (2010): 3115–3141.

41. L. Zhang, C. Wang, Y. Dou, et al., "A Yolk–Shell Structured Silicon Anode With Superior Conductivity and High Tap Density for Full Lithium-Ion Batteries," *Angewandte Chemie International Edition* 58 (2019): 8824–8828.

42. M. Ashuri, Q. He, and L. L. Shaw, "Silicon as a Potential Anode Material for Li-Ion Batteries: Where Size, Geometry and Structure Matter," *Nanoscale* 8 (2016): 74–103.

43. J.-Y. Li, Q. Xu, G. Li, Y.-X. Yin, L.-J. Wan, and Y.-G. Guo, "Research Progress Regarding Si-Based Anode Materials Towards Practical Application in High Energy Density Li-Ion Batteries," *Materials Chemistry Frontiers* 1 (2017): 1691–1708.

44. M. Ge, X. Fang, J. Rong, and C. Zhou, "Review of Porous Silicon Preparation and Its Application for Lithium-Ion Battery Anodes," *Nanotechnology* 24 (2013): 422001.

45. L. Li, Y. Deng, K. Hu, et al., "Nanostructure Designing and Hybridizing of High-Capacity Silicon-Based Anode for Lithium-Ion Batteries," *Progress in Natural Science: Materials International* 33 (2023): 16–36.

46. Z. Zhang, F. Xi, X. Chen, et al., "Improved Lithium-Ion Batteries With Coral-Like Anodes Made of Recycled Spherical Porous Silicon Coated With Nitrogen-Doped Carbon," *Environmental Chemistry Letters* 20 (2022): 3377–3385.

47. K. Feng, M. Li, W. Liu, et al., "Silicon-Based Anodes for Lithium-Ion Batteries: From Fundamentals to Practical Applications," *Small* 14 (2018): 1702737.

48. Y. Pei, Y. Wang, A.-Y. Chang, et al., "Nanofiber-in-Microfiber Carbon/Silicon Composite Anode With High Silicon Content for Lithium-Ion Batteries," *Carbon* 203 (2023): 436–444.

49. S. Sun, D. He, P. Li, et al., "Improved Adhesion of Cross-Linked Binder and SiO_2-Coating Enhances Structural and Cyclic Stability of Silicon Electrodes for Lithium-Ion Batteries," *Journal of Power Sources* 454 (2020): 227907.

50. H. Wang, "Silicon Dioxide Peristaltic Supporting Carbon and MoS_2 Nanosheets Co-Anchored Polyaniline for High-Performance Supercapacitors," *Synthetic Metals* 277 (2021): 116772.

51. S. Huang, L. Liu, Y. Zheng, et al., "Efficient Sodium Storage in Rolled-Up Amorphous Si Nanomembranes," *Advanced Materials* 30 (2018): 1706637.

52. Y. Yao, M. T. McDowell, I. Ryu, et al., "Interconnected Silicon Hollow Nanospheres for Lithium-Ion Battery Anodes With Long Cycle Life," *Nano Letters* 11 (2011): 2949–2954.

53. H. Dong, H. Xie, Y. Xu, Q. Song, Z. Ning, and H. Yin, "High-Rate Performance Boron-Doped Silicon Flakes Anode Using a Molten Salts Method for Lithium-Ion Batteries," *Journal of Alloys and Compounds* 911 (2022): 164965.

54. X.-W. Jiao, Y.-H. Tian, and X.-J. Zhang, "Hollow Si Nanospheres With Amorphous TiO_2 Layer Used as Anode for High-Performance Li-Ion Battery," *Applied Surface Science* 566 (2021): 150682.

55. C. Li, T. Shi, D. Li, H. Yoshitake, and H. Wang, "Effect of Surface Modification on Electrochemical Performance of Nano-Sized Si as an Anode Material for Li-Ion Batteries," *RSC Advances* 6 (2016): 34715–34723.

56. S. Sekar, A. T. Aqueel Ahmed, D. Y. Kim, and S. Lee, "One-Pot Synthesized Biomass C-Si Nanocomposites as an Anodic Material for High-Performance Sodium-Ion Battery," *Nanomaterials* 10 (2020): 1728.

57. Y. Zhang, Y.-C. Tang, X.-T. Li, et al., "Porous Amorphous Silicon Hollow Nanoboxes Coated With Reduced Graphene Oxide as Stable Anodes for Sodium-Ion Batteries," *ACS Omega* 7 (2022): 30208–30214.

58. Y. Sun, Y. Zhao, Z. Liang, et al., "Reversible Potassium-Ion Alloying Storage in Crystalline Silicene," *Chemical Engineering Journal* 435 (2022): 134961.

59. X. Shen, Z. Tian, R. Fan, et al., "Research Progress on Silicon/Carbon Composite Anode Materials for Lithium-Ion Battery," *Journal of Energy Chemistry* 27 (2018): 1067–1090.

60. R. Fang, W. Xiao, C. Miao, et al., "Improved Lithium Storage Performance of Pomegranate-Like Si@NC/rGO Composite Anodes by Facile In-Situ Nitrogen Doped Carbon Coating and Freeze Drying Processes," *Journal of Alloys and Compounds* 834 (2020): 155230.

61. H. Shi, W. Zhang, D. Wang, et al., "Facile Preparation of Silicon/Carbon Composite With Porous Architecture for Advanced Lithium-Ion Battery Anode," *Journal of Electroanalytical Chemistry* 937 (2023): 117427.

62. T.-H. Ha, B. S. Reddy, H.-R. Ryu, et al., "A Study on the Electrochemical Properties of Silicon/Carbon Composite for Lithium-Ion Battery," *Journal of Energy Storage* 63 (2023): 107045.

63. M. Zhang, J. Li, C. Sun, Z. Wang, Y. Li, and D. Zhang, "Durable Flexible Dual-Layer and Free-Standing Silicon/Carbon Composite Anode for Lithium-Ion Batteries," *Journal of Alloys and Compounds* 932 (2023): 167687.

64. W. Zhang, H. Shi, D. Wang, et al., "Three-Dimensional Ti_3C_2 MXene@Silicon@Nitrogen-Doped Carbon Foam for High Performance Self-Standing Lithium-Ion Battery Anodes," *Journal of Electroanalytical Chemistry* 921 (2022): 116664.

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/ne2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

65. X. Shen, X. Wei, T. Wang, S. Li, and H. Li, "Polypyrrole Embedded in Nickel-Cobalt Sulfide Nanosheets Grown on Nickel Particles Passivated Silicon Nanowire Arrays for High-Performance Supercapacitors," *Chemical Engineering Journal* 461 (2023): 141745.

66. F. BoorboorAjdari, M. Izadpanah Ostad, M. Niknam Shahrak, et al., "Investigating MCM-41/Metal-Organic Framework Nanocomposites as Silicon-Containing Electrodes for Supercapacitor," *Surfaces and Interfaces* 29 (2022): 101796.

67. J. Chen, J. Wang, S. Zhang, et al., "Si Nanoparticles Confined in N, P- Doped Double Carbon as Efficient Anode Materials for Lithium Ion Batteries," *Journal of Alloys and Compounds* 935 (2023): 167850.

68. Y. Li, X. Liu, J. Zhang, H. Yu, and J. Zhang, "Carbon-Coated Si/N-Doped Porous Carbon Nanofibre Derived From Metal–Organic Frameworks for Li-Ion Battery Anodes," *Journal of Alloys and Compounds* 902 (2022): 163635.

69. L. Shi, W. Wang, A. Wang, K. Yuan, and Y. Yang, "Facile Synthesis of Scalable Pore-Containing Silicon/Nitrogen-Rich Carbon Composites From Waste Contact Mass of Organosilane Industry as Anode Materials for Lithium-Ion Batteries," *Journal of Materials Chemistry A: Materials for Energy and Sustainability* 2 (2014): 20213–20220.

70. X. Wang, G. Li, M. H. Seo, F. M. Hassan, M. A. Hoque, and Z. Chen, "Sulfur Atoms Bridging Few-Layered MoS_2 With S-doped Graphene Enable Highly Robust Anode for Lithium-Ion Batteries," *Advanced Energy Materials* 5 (2015): 1501106.

71. J. Pan, W. Zhong, Z. Gao, et al., "N, S-Doped Silicon Oxycarbide-Drived Carbon/Amorphous Ball-Flower-Like NiO as High Performance Electrode in Asymmetric Supercapacitors," *Ceramics International* 47 (2021): 27833–27842.

72. C. Sun, A. Du, G. Deng, et al., "Naturally Nitrogen-Doped Self-Encapsulated Biochar Materials Based on Mouldy Wheat Flour for Silicon Anode in Lithium-Ion Batteries," *Electrochimica Acta* 450 (2023): 142269.

73. X. Tang, D. Zeng, D. Chen, W. Guo, J. Fu, and L. Zou, "Porous Silicon Particles Embedded in N-Doped Graphene and Carbon Nanotube Framework for High-Performance Lithium-Ion Batteries," *Journal of Alloys and Compounds* 927 (2022): 167055.

74. A. Nazir, H. T. T. Le, A. Kasbe, and C.-J. Park, "Si Nanoparticles Confined Within a Conductive 2D Porous Cu-Based Metal–Organic Framework $(Cu_3(HITP)_2)$ as Potential Anodes for High-Capacity Li-Ion Batteries," *Chemical Engineering Journal* 405 (2021): 126963.

75. I. Saana Amiinu, S. Imtiaz, H. Geaney, et al., "A Thin Si Nanowire Network Anode for High Volumetric Capacity and Long-Life Lithium-Ion Batteries," *Journal of Energy Chemistry* 81 (2023): 20–27.

76. Y. Zhuo, H. Sun, M. H. Uddin, et al., "An Additive-Free Silicon Anode in Nanotube Morphology as a Model Lithium Ion Battery Material," *Electrochimica Acta* 388 (2021): 138522.

77. Q. Zhou, Y. Zhou, M. Bao, and X. Ni, "Modified Silicon Nanowires@Polypyrrole Core-Shell Nanostructures by Poly(3,4-ethylenedioxythiophene) for High Performance On-Chip Micro-Supercapacitors," *Applied Surface Science* 487 (2019): 236–243.

78. Y. Bencheikh, M. Harnois, R. Jijie, et al., "High Performance Silicon Nanowires/Ruthenium Nanoparticles Micro-Supercapacitors," *Electrochimica Acta* 311 (2019): 150–159.

79. Q. Zhang, Y. Yang, D. Wang, et al., "A Silicon/Carbon/Reduced-Graphene Composite of Honeycomb Structure for High-Performance Lithium-Ion Batteries," *Journal of Alloys and Compounds* 944 (2023): 169185.

80. S. Chen, X. Wu, Z. Liu, et al., "Mg_2Si Promoted Magnesio-Mechanical Reduction of Silica Into Silicon Nanoparticles for High-Performance Li-Ion Batteries," *Journal of Solid State Chemistry* 302 (2021): 122408.

81. A. Krause, U. Langklotz, D. Pohl, et al., "Surface Related Differences Between Uncoated Versus Carbon-Coated Silicon Nanowire Electrodes on Performance in Lithium Ion Batteries," *Journal of Energy Storage* 27 (2020): 101052.

82. J. T. Warner, "Lithium-Ion Battery Operation," *Lithium-Ion Battery Chemistries* (2019): 43–77, https://doi.org/10.1016/B978-0-12-814778-8.00003-X.

83. N. Li, X. Xu, B. Sun, K. Xie, W. Huang, and T. Yu, "Recent Nanosheet-Based Materials for Monovalent and Multivalent Ions Storage," *Energy Storage Materials* 25 (2020): 382–403.

84. Y. Mei, Y. He, H. Zhu, et al., "Recent Advances in the Structural Design of Silicon/Carbon Anodes for Lithium Ion Batteries: A Review," *Coatings* 13 (2023): 436.

85. Z. Shang, X. Liu, J. Liu, et al., "Double Core-Shell Structure Stabilized Porous Si@Graphene@TiO_2 Microsphere Anode With Excellent Cyclability and High Coulombic Efficiency," *Electrochimica Acta* 407 (2022): 139893.

86. C. Li, C. Yuan, J. Zhu, et al., "Fabrication of Silicon Nanoparticles/Porous Carbon@Porous Carbon Nanofibers Core-Shell Structured Composites as High-Performance Anodes for Lithium-Ion Batteries," *Colloids and Surfaces, A: Physicochemical and Engineering Aspects* 655 (2022): 129721.

87. W. Zhang, J. Li, P. Guan, et al., "One-Pot Sol–Gel Synthesis of Si/C Yolk-Shell Anodes for High Performance Lithium-Ion Batteries," *Journal of Alloys and Compounds* 835 (2020): 155135.

88. J. Han, H. Li, D. Kong, et al., "Realizing High Volumetric Lithium Storage by Compact and Mechanically Stable Anode Designs," *ACS Energy Letters* 5 (2020): 1986–1995.

89. S. Suh, H. Choi, K. Eom, and H.-J. Kim, "Enhancing the Electrochemical Properties of a Si Anode by Introducing Cobalt Metal as a Conductive Buffer for Lithium-Ion Batteries," *Journal of Alloys and Compounds* 827 (2020): 154102.

90. W. An, P. He, Z. Che, et al., "Scalable Synthesis of Pore-Rich Si/C@C Core–Shell-Structured Microspheres for Practical Long-Life Lithium-Ion Battery Anodes," *ACS Applied Materials & Interfaces* 14 (2022): 10308–10318.

91. W. An, B. Gao, S. Mei, et al., "Scalable Synthesis of Ant-Nest-Like Bulk Porous Silicon for High-Performance Lithium-Ion Battery Anodes," *Nature Communications* 10 (2019): 1447.

92. C. Xu, L. Shen, W. Zhang, et al., "Efficient Implementation of Kilogram-Scale, High-Capacity and Long-Life Si-C/TiO_2 Anodes," *Energy Storage Materials* 56 (2023): 319–330.

93. H. Jin, Q. Sun, J. Wang, C. Ma, L. Ling, and W. Qiao, "Preparation and Electrochemical Properties of Novel Silicon-Carbon Composite Anode Materials With a Core-Shell Structure," *New Carbon Materials* 36 (2021): 390–400.

94. X. Li, H. Su, C. Ma, et al., "Optimizations of Graphitic Carbon/Silicon Hybrids for Scalable Preparation With High-Performance Lithium-Ion Storage," *ACS Sustainable Chemistry & Engineering* 10 (2022): 5590–5598.

95. T. Zhao, S. She, X. Ji, et al., "In-Situ Growth Amorphous Carbon Nanotube on Silicon Particles as Lithium-Ion Battery Anode Materials," *Journal of Alloys and Compounds* 708 (2017): 500–507.

96. C. Kim, S. Choi, S. Yoo, et al., "A Facile Route for Growth of CNTs on Si@Hard Carbon for Conductive Agent Incorporating Anodes for Lithium-Ion Batteries," *Nanoscale* 7 (2015): 11286–11290.

97. D. Bokov, A. Turki Jalil, S. Chupradit, et al., "Nanomaterial by Sol-Gel Method: Synthesis and Application," *Advances in Materials Science and Engineering* 2021 (2021): 5102014.

98. R. Fang, C. Miao, H. Mou, and W. Xiao, "Facile Synthesis of Si@-TiO_2@rGO Composite With Sandwich-Like Nanostructure as Superior Performance Anodes for Lithium Ion Batteries," *Journal of Alloys and Compounds* 818 (2020): 152884.

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/bte2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

99. Z. Wang, Z. Xu, Y. Yuan, et al., "Microspheres Comprise Si Nanoparticles Modified With TiO_2 and Wrapped by Graphene as High-Performance Anode for Lithium-Ion Batteries," *Applied Surface Science* 598 (2022): 153790.

100. M. Ge, Y. Tang, O. I. Malyi, et al., "Mechanically Reinforced Localized Structure Design to Stabilize Solid–Electrolyte Interface of the Composited Electrode of Si Nanoparticles and TiO_2 Nanotubes," *Small* 16 (2020): 2002094.

101. A. Jamaluddin, B. Umesh, F. Chen, J.-K. Chang, and C.-Y. Su, "Facile Synthesis of Core–Shell Structured Si@Graphene Balls as a High-Performance Anode for Lithium-Ion Batteries," *Nanoscale* 12 (2020): 9616–9627.

102. S. S. Bishoyi and S. K. Behera, "Synthesis and Structural Characterization of Nanocrystalline Silicon by High Energy Mechanical Milling Using Al_2O_3 Media," *Advanced Powder Technology* 33 (2022): 103639.

103. J. Nzabahimana, Z. Liu, S. Guo, L. Wang, and X. Hu, "Top-Down Synthesis of Silicon/Carbon Composite Anode Materials for Lithium-Ion Batteries: Mechanical Milling and Etching," *Chemsuschem* 13 (2020): 1923–1946.

104. P. Y. E. Koraag, A. M. Firdaus, N. H. Hawari, et al., "Covalently Bonded Ball-Milled Silicon/CNT Nanocomposite as Lithium-Ion Battery Anode Material," *Batteries* 8 (2022): 165.

105. W. Zhang, D. Wang, H. Shi, et al., "Industrial Waste Micron-Sized Silicon Use for Si@C Microspheres Anodes in Low-Cost Lithium-Ion Batteries," *Sustainable Materials and Technologies* 33 (2022): e00454.

106. U. Zubair, J. Amici, S. M. Crespiera, C. Francia, and S. Bodoardo, "Rational Design of Porous Carbon Matrices to Enable Efficient Lithiated Silicon Sulfur Full Cell," *Carbon* 145 (2019): 100–111.

107. K.-B. Kim, N. A. Dunlap, S. S. Han, et al., "Nanostructured Si/C Fibers as a Highly Reversible Anode Material for All-Solid-State Lithium-Ion Batteries," *Journal of the Electrochemical Society* 165 (2018): A1903–A1908.

108. F. Wang, S. Zhang, J. Zhang, M. Han, G. Pan, and M. Chen, "Rational Synthesis of Silicon Into Polyimide-Derived Hollow Electrospun Carbon Nanofibers for Enhanced Lithium Storage," *e-Polymers* 20 (2020): 491–499.

109. Y. Zeng, Y. Huang, N. Liu, et al., "N-Doped Porous Carbon Nanofibers Sheathed Pumpkin-Like Si/C Composites as Free-Standing Anodes for Lithium-Ion Batteries," *Journal of Energy Chemistry* 54 (2021): 727–735.

110. V. Ghanooni Ahmadabadi, K. Shirvanimoghaddam, R. Kerr, N. Showkath, and M. Naebe, "Structure-Rate Performance Relationship in Si Nanoparticles-Carbon Nanofiber Composite as Flexible Anode for Lithium-Ion Batteries," *Electrochimica Acta* 330 (2020): 135232.

111. S. Chae, S.-H. Choi, N. Kim, J. Sung, and J. Cho, "Integration of Graphite and Silicon Anodes for the Commercialization of High-Energy Lithium-Ion Batteries," *Angewandte Chemie International Edition* 59 (2020): 110–135.

112. M. Fichtner, "Recent Research and Progress in Batteries for Electric Vehicles," *Batteries & Supercaps* 5 (2022): e202100224.

113. M. Lavigne Philippot, D. Costa, G. Cardellini, et al., "Life Cycle Assessment of a Lithium-Ion Battery With a Silicon Anode for Electric Vehicles," *Journal of Energy Storage* 60 (2023): 106635.

114. J. Chen, X. Fan, Q. Li, et al., "Electrolyte Design for LiF-Rich Solid–Electrolyte Interfaces to Enable High-Performance Microsized Alloy Anodes for Batteries," *Nature Energy* 5 (2020): 386–397.

115. J. Lu, Z. Chen, Z. Ma, F. Pan, L. A. Curtiss, and K. Amine, "The Role of Nanotechnology in the Development of Battery Materials for Electric Vehicles," *Nature Nanotechnology* 11 (2016): 1031–1038.

116. X. Han, L. Gu, Z. Sun, et al., "Manipulating Charge-Transfer Kinetics and a Flow-Domain Lif-Rich Interphase to Enable High-Performance Microsized Silicon–Silver–Carbon Composite Anodes for Solid-State Batteries," *Energy & Environmental Science* 16 (2023): 5395–5408.

117. L. Gu, J. Han, M. Chen, et al., "Enabling Robust Structural and Interfacial Stability of Micron-Si Anode Toward High-Performance Liquid and Solid-State Lithium-Ion Batteries," *Energy Storage Materials* 52 (2022): 547–561.

118. X. Han, W. Zhou, M. Chen, et al., "Interfacial Nitrogen Engineering of Robust Silicon/Mxene Anode Toward High Energy Solid-State Lithium-Ion Batteries," *Journal of Energy Chemistry* 67 (2022): 727–735.

119. S. Gao, D. Yang, Y. Pan, et al., "From Natural Material to High-Performance Silicon Based Anode: Towards Cost-Efficient Silicon Based Electrodes in High-Performance Li-Ion Batteries," *Electrochimica Acta* 327 (2019): 135058.

120. I.-N. Bratosin, C. Romanitan, G. Craciun, et al., "Graphitized Porous Silicon Decorated With Cobalt Hexacyanoferrate Nanocubes as Hybrid Electrode for High-Performance Supercapacitors," *Electrochimica Acta* 424 (2022): 140632.

121. M. Diantoro, I. Istiqomah, O. Puji Dwi Lestari, et al., "A Comprehensive Study of Binder Polymer for Supercapattery Electrode Based on Activated Carbon and Nickel-Silicon Composite," *Materials Science for Energy Technologies* 6 (2023): 368–381.

122. L. Jiao, P. Liu, C. Yan, L. Qu, Q. Wang, and H. Li, "Hologram Quantitative Structure-Activity Relationship Guided in Silicon Molecular Design of Phenothiazine Dye Sensitizers for Dye-Sensitized Solar Cells," *Solar Energy* 250 (2023): 51–58.

123. V. Jaswal and Y. Kadapakkam Nandabalan, "Rice Husk-Derived Silicon Nanostructured Anode to Enhance Power Generation in Microbial Fuel Cell Treating Distillery Wastewater," *Journal of Environmental Management* 328 (2023): 116912.

27681696, 2025, 5, Downloaded from https://onlinelibrary.wiley.com/doi/10.1002/ne2.20240048 by University Of Strathclyde, Wiley Online Library on [17/09/2025]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License