

Progress and Perspectives on Silicon-Carbon Composite Anodes for Sustainable Energy Storage: A Review

Hasanuzzaman Aoyon¹, Himangshu Bhowmik^{1*}, Mohammad Asaduzzaman Chowdhury¹, Mandira Bhowmik², Shaon Talukdar³, Md Masud Rana¹, Md Mostafizur Rahman¹

¹Dhaka University of Engineering & Technology, Gazipur, Bangladesh

²University of Arizona, Tucson, Arizona, USA

³University of Alabama, Tuscaloosa, AL, USA

Email: *bhowmik@duet.ac.bd

How to cite this paper: Aoyon, H., Bhowmik, H., Chowdhury, M.A., Bhowmik, M., Talukdar, S., Rana, M.M. and Rahman, M.M. (2026) Progress and Perspectives on Silicon-Carbon Composite Anodes for Sustainable Energy Storage: A Review. *Journal of Materials Science and Chemical Engineering*, 14, 1-51.
<https://doi.org/10.4236/msce.2026.1410001>

Received: July 27, 2026

Accepted: September 27, 2026

Published: September 30, 2026

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Abstract

This review uniquely integrates high-performance silicon-carbon (Si-C) anode design with sustainability considerations, encompassing electrochemical performance, material sources, synthesis strategies, and lifecycle aspects. It elucidates key considerations governing electrical conductivity, volume buffering, and solid-electrolyte interphase (SEI) stability, and critically assesses advanced Si-C architectures with perspectives on mitigating silicon volume expansion for durable cycling. This review systematically discusses in detail the recent advances and synthesis methods of various types of carbon-based, silicon-based, metallic and intermetallic, transition-based, phosphorus-based, organic, and multidimensional composite anode technologies used as energy storage materials. A comparative analysis of the structural designs, including void-engineered, yolk-shell, MXene-rich, metal-organic framework, MOF-derived and graphene/carbon nanotubes, CNT-hybrid designs, is provided. In addition, the challenges of industrial-scale production, safety concerns, suitability for various applications and environmental sustainability are highlighted. Research gaps are identified and future directions for improved design and application are also discussed. Finally, this review highlights the importance and potential of Si/C-based composite anodes as enabling materials for durable, high-energy-density and industrially viable lithium-ion batteries, providing valuable guidance for the development of next-generation energy-storage technologies.

Keywords

Composite Anodes, Silicon-Carbon Composite Anodes, Lithium-Ion

1. Introduction

The global energy mix is undergoing rapid transformation, driven by the increasing deployment of renewable energy and international commitments toward carbon neutrality, making efficient and sustainable energy-storage technologies increasingly important. Among available technologies, lithium-ion batteries (LIBs) play a central role in electric vehicles, portable electronics, and renewable-energy storage because of their high energy density, long cycle life, and technological maturity. **Figure 1** illustrates the fundamental operating principle of a LIB during charging and discharging. During charging, an external power source drives lithium ions through the electrolyte from the cathode to the anode, where energy is stored electrochemically. During discharge, lithium ions migrate back to the cathode, while electrons flow through the external circuit to deliver electrical power. Within this broader context, this review specifically focuses on silicon-carbon (Si-C) composite anodes as a promising strategy for achieving high-capacity and durable LIBs. Other anode material classes are discussed selectively as comparative benchmarks, rather than as independent review topics, to contextualize the advantages and limitations of Si-C systems. Particular emphasis is placed on material design, synthesis strategies, structural architectures, electrical conductivity, silicon volume-expansion mitigation, solid-electrolyte interphase stability, electrochemical performance, sustainability, scalability, and practical implementation of Si-C anodes.

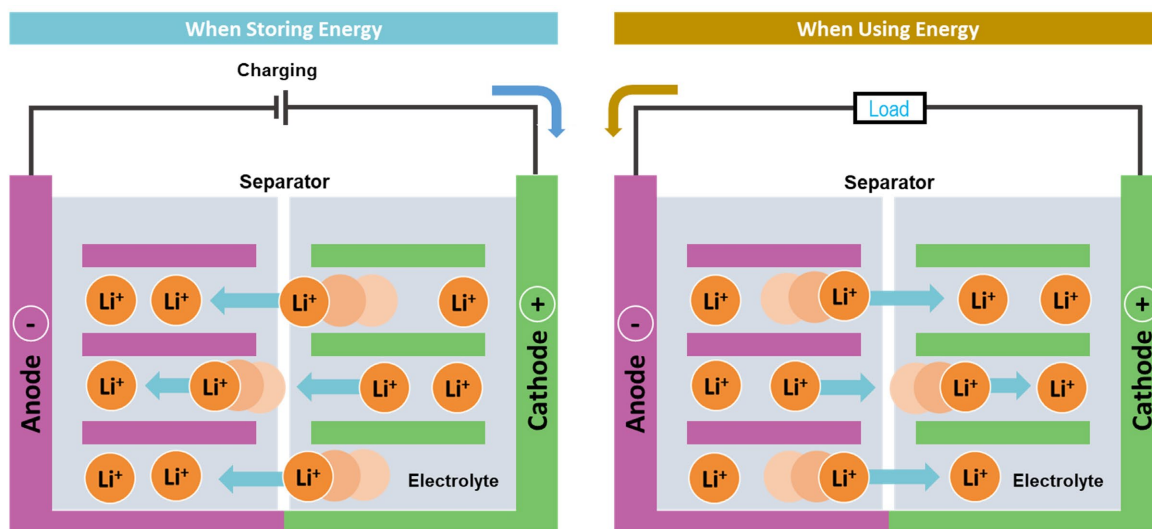


Figure 1. Operating principals of lithium-ion battery during charging and discharging cycles.

High-performance energy storage systems are essential to maintain the viability

of variable renewable sources such as solar and wind power, as their production is not continuous and is dependent on time and natural conditions [1] [2]. At the same time, the rapid growth of electric vehicles (EVs), the transition of industrial and household power systems to smart grids and the proliferation of portable electronic devices have increased the demand for energy storage materials with high energy density, long cycle life and safety [1]. According to the International Energy Agency (IEA), the number of electric vehicles will increase many times over by 2030 compared to the current level, which will further intensify the need for large-scale advanced lithium-ion batteries (LIBs) [3].

Lithium-ion batteries (LIBs) are considered as the foundation of modern energy storage technologies, mainly due to their high energy density, fast charge-discharge capability and relatively long cycling durability. Although significant progress has been made in cathode materials, the anode material poses a significant limitation in determining the overall performance of batteries [4]. Graphite has been used as a commercial LIB anode for decades, due to its high stability, low voltage platform and affordable price. However, the theoretical capacity of graphite is only 372 mAh/g [5] [6], which is a major obstacle to the development of next-generation battery technologies with high energy density. The search for new high-capacity anode materials to overcome this limitation has become a focus of scientific discussion. Silicon (Si) is a very promising alternative due to its extremely high theoretical capacity about 4200 mAh/g [7], abundant availability and environmentally friendly nature. However, the main problem with silicon is the 300% - 400% volume change during the charge-discharge process, which leads to particle cracking, active material loss, formation of unstable SEI layers and rapid performance degradation [8]. As a result, structural maintenance and durability are major challenges in the implementation of silicon-based anode technologies.

To address the above issues, recent review has focused on silicon-carbon (Si/C) composite anodes that attempt to combine the high capacity of silicon with the mechanical and electrical stability of carbon. The carbon matrix absorbs the volume changes of silicon and prevents structural damages, while also enhancing electron conductivity and ensuring stable SEI formation [9]. Different types of carbon such as amorphous carbon, hard carbon, graphene, carbon nanotubes (CNTs) can be integrated with silicon to provide different structural functions. Recent studies have shown that void-structured, yolk-shell and hierarchical pore engineered Si/C designs are particularly effective in controlling volume expansion [10]. Graphene and CNT-rich hybrid structures have been shown to exhibit high electrical conductivity, fast ion transport and improved rate-capability, with capacities exceeding 1500 mAh/g at high currents up to 6C reported [11] [12]. Composite anode technology is not limited to silicon successful composite anodes have been fabricated in various carbon matrices with alloying materials such as Sn, Sb, P and transformation materials such as MnO_x , Fe_2O_3 , Co_3O_4 , MoS_2 , which have opened up new possibilities for high power, stability and rate-capability [13] [14]. However, despite these advances, some challenges remain especially low initial

Coulombic efficiency, complex synthesis methods, expensive materials and large-scale production-scalability issues.

In recent years, more scalable, environmentally friendly and industrially friendly methods such as high-energy ball milling, spray pyrolysis, sol-gel processes, metal-organic framework (MOF)-derived carbonization and MXene-based hybrid composites have gained importance to overcome these limitations [15]. These innovations have opened up new possibilities for high electrical conductivity, improved ion transport, strong structural stability and long cycling life. Overall, the ability to achieve four key aspects of high power, long cycle stability, safety and industrial-friendly manufacturing together has established silicon-carbon-based composite anode technology as one of the most important research areas for future lithium-ion batteries [16]. The contribution of this technology will undoubtedly play an important role in creating sustainable and high-performance energy storage solutions in the renewable energy era.

This review brings together the latest advances and various structural designs of various types of composite anode materials for lithium-ion batteries through a comprehensive study and analysis of existing research. The sub-chapters are discussed in detail in a total of eight main categories including Carbon-Based, Silicon-Based, Alloy and Intermetallic, Conversion-Type, Phosphorus-Based, Organic-Polymeric, Lithium-Metal Supported and Emerging Multifunctional Composite Systems. Each sub-chapter includes a comprehensive review of the design, synthesis methods and electrical performance of composite anodes ranging from graphite to CNTs, graphene, MOF-derived carbon, MXene-based hybrids, silicon nanoparticles, metallic and semi-metallic compounds, transition metal oxides and sulfides, phosphorus-based and organic polymer composites [17]. However, the comprehensive classification and analytical framework has been discussed separately or partially in conventional studies, whereas the main novelty of this review is to present and integrate as a whole in this single review. In particular, the comparative analysis of void-engineered, yolk-shell, MXene-rich, MOF-derived and graphene/CNT-hybrid structures as well as other conventional composite designs and the elucidation of their structure property performance relationships make this review particularly important. Another notable aspect is that this review not only discusses the relevance and challenges from the material perspective, but also from the synthesis process, production scale-up potential, safety concerns and various applications (EV, grid storage, fast charging), which are very important for the industrialization and implementation of such composite anode technologies.

This review focuses primarily on silicon-carbon (Si-C) composite anodes as a promising platform for next-generation high-performance lithium-ion batteries, while other anode material classes are discussed selectively as comparative benchmarks to contextualize the advantages and limitations of Si-C systems. The review uniquely integrates high-performance material design with sustainability considerations, emphasizing electrochemical performance alongside material sources,

synthesis strategies, scalability, and lifecycle aspects. Particular attention is given to electrical conductivity, silicon volume-change buffering, and solid-electrolyte interphase (SEI) stability, combining experimental findings with theoretical insights to clarify structure-property-performance relationships. Advanced Si-C architectures, including yolk-shell, core-shell, porous, and hierarchical structures, are critically evaluated in terms of their ability to accommodate silicon's large volume expansion, preserve conductive networks, stabilize the SEI, and enhance cycling performance. Persistent challenges associated with structural durability, scalable synthesis, cost, material utilization, and practical electrode implementation are also identified. Future research directions emphasize rational structural engineering, sustainable carbon sources, environmentally responsible synthesis, scalable manufacturing, and lifecycle-conscious material design. By positioning non-Si-C materials as comparative benchmarks, this review provides a focused framework linking electrochemical performance, structural innovation, sustainability, and practical implementation to guide the development of next-generation Si-C anode technologies.

2. Advancements in Composite Anode Materials

The development of advanced anode materials is critical for achieving high-energy-density and long-life lithium-ion batteries (LIBs). Although cathode materials have received considerable research attention, anodes play an equally important role in determining key cell-performance parameters, including cycle life, power density, rate capability, and energy density [18]. Optimization of anode properties, including electrical conductivity, electrochemical activity, chemical and mechanical stability, structural integrity, and morphology, is therefore essential for improving overall cell performance [19]. Among emerging anode materials, silicon is particularly attractive because of its exceptionally high theoretical capacity. However, its practical application is constrained by severe volume expansion during lithiation/delithiation, unstable solid-electrolyte interphase (SEI) formation, loss of electrical contact, and rapid capacity degradation. Recent advances in lithium-sulfur batteries have focused on suppressing polysulfide migration, accelerating sulfur redox kinetics, and stabilizing electrode-electrolyte interfaces. Fan *et al.* demonstrated that coordinated thermodynamic and kinetic regulation can improve sulfur utilization and enable high-performance operation at low temperatures [20]. Zhou *et al.* developed TiO₂-TiN heterostructures that combine effective polysulfide adsorption with rapid diffusion and catalytic conversion, thereby supporting prolonged cycling stability [21]. Wu *et al.* showed that systematic regulation of the active-material distribution and ion-diffusion pathway can reduce degradation and enable ultralong cycle life [22]. Furthermore, Qu *et al.* introduced a multifunctional sandwich-structured electrolyte that restricts polysulfide shuttling, promotes uniform lithium-ion transport, and improves the electrochemical performance of lithium-sulfur batteries [23]. These representative approaches are schematically summarized in **Figure 2**.

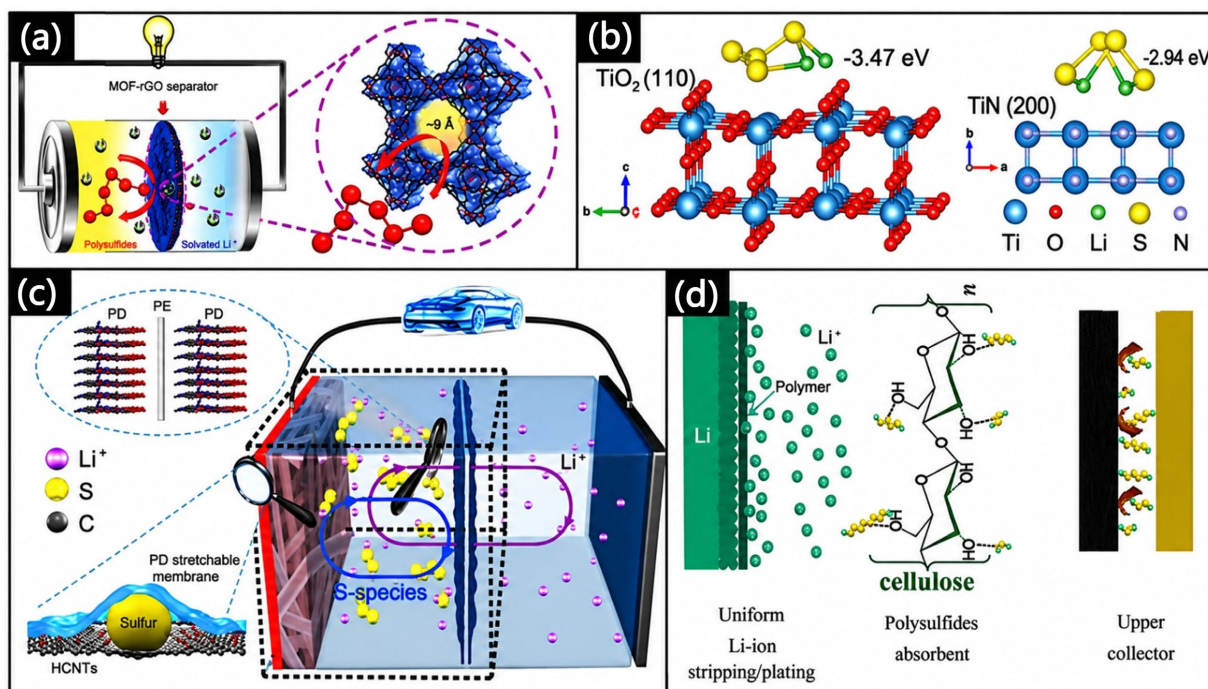


Figure 2. Illustrations of (a) MOF/GO-coated separator with a MOF window [20], (b) PS interactions with TiO_2 and TiN electronegativity [21], (c) S-HCNTs cathode for PS inhibition [22], (d) Polymer electrolyte for PS inhibition [23].

Silicon-carbon (Si-C) composite anodes provide an effective strategy for addressing these limitations by combining the high lithium-storage capacity of silicon with the electrical conductivity, structural flexibility, and mechanical stability of carbon. **Table 1** provides a comparative summary of the electrochemical performance, advantages, and limitations of representative silicon-based composite anodes. Carbon-coated and carbon-integrated silicon composites have demonstrated significant potential for buffering silicon volume changes, maintaining conductive pathways, and stabilizing the SEI during repeated cycling [24] [25]. For example, Si nanoparticle composites incorporating biomass-derived carbon or graphite can provide improved conductivity and mechanical buffering, with reported capacities of approximately 880 - 883 mAh/g and favorable capacity retention over prolonged cycling. Advanced carbon-shell architectures can provide additional structural confinement and interfacial protection, with some systems achieving capacities as high as 3207 mAh/g while maintaining improved cycling stability [26].

Further improvements can be achieved through robust binders, controlled Si-C interfaces, porous structures, and optimized particle architectures that preserve electrode integrity during repeated volume changes. Moreover, sustainable carbon sources and scalable synthesis routes offer promising pathways toward environmentally responsible and commercially viable Si-C electrodes. Nevertheless, no single Si-C architecture simultaneously maximizes capacity, cycling stability, rate capability, sustainability, cost-effectiveness, and scalability. Consequently, rational integration of silicon with appropriately engineered carbon structures re-

mains essential for developing practical high-performance anodes for electric vehicles and stationary energy-storage applications [27].

Table 1. Electrochemical performance of Si-based composite anodes for LIBs.

Composite	Electrochemical performance	Advantages	Limitations
Si/C/G (Si-amorphous C-Gr) [28]	2469 mAh/g (50 cycles, 0.2 A/g), >1500 mAh/g at 2 A/g for 300 cycles; 471 mAh/g at 32 A/g	Gr-C coating provides good conductivity, strong structural buffering of Si expansion, high-rate capability	Gr/C ratio optimization required; C decreases gravimetric energy density
Si@Fe ₃ C/Fe ₃ O ₄ -C foam (Si embedded in Fe ₃ O ₄ /C matrix) [29]	1116 mAh/g (250 cycles, 0.1 A/g), 858 mAh/g (500 cycles, 1 A/g)	Porous carbon foam + Fe ₃ C/Fe ₃ O ₄ provide strong structural support, conductivity, and volume-change buffering	Multi-phase composite increases synthesis complexity, metal interfaces degrade over long cycling
Si/CNT (Si nanoparticles + C nanotubes) [30]	1100 mAh/g initial; 922 mAh/g after 200 cycles 612 mAh/g after 1000 cycles (high-rate)	CNTs give strong mechanical strength + high conductivity, good Si-C bonding, excellent long-cycle stability	Interfacial uniformity difficult, CNT networks costly, scale-up issues
Si/rGrO/C (Si + reduced GrO + sucrose-derived C) [31]	1209 mAh/g initial; 92% retention after 100 cycles; stable up to 6C	Carbon anchor maintains electron pathways, rGO provides flexibility, low volume-change	Moderate ICE, carbon increases inactive mass
Si/NG (Si nanoparticles + nanographite) [32]	1569 mAh/g initial; 1138 mAh/g after 100 cycles	Nanographite improves electrical conductivity and buffers volume expansion, simple synthesis	Noticeable capacity fading, SEI growth still significant
SiO _x /3D-Gr with Si-N-C bonding [33]	Reported as durable with superior interface stability	Laser-induced Si-N-C bonding gives chemically reinforced interface, very stable at long-cycle	Fabrication costly, precise processing needed, scaling difficult
Si/G/C (Si+GO + C coating) [34]	70% retention after 267 cycles (1 A/g)	Carbon coating + GO improves Si dispersion and conductivity, stable SEI formation	Long-term capacity fade possible, carbon layer thickness must be optimized

Table 1 summaries the various silicon-carbon composites have demonstrated improved electrochemical performance by enhancing conductivity and buffering silicon volume expansion. The Si/amorphous-carbon/graphite composite delivered 2469 mAh·g⁻¹ after 50 cycles at 0.2 A·g⁻¹, over 1500 mAh·g⁻¹ for 300 cycles at 2 A·g⁻¹, and 471 mAh·g⁻¹ at 32 A·g⁻¹ [28]. However, its graphite-to-carbon ratio requires optimization to minimize inactive mass. The porous Si@Fe₃C/Fe₃O₄-carbon foam achieved 1116 mAh·g⁻¹ after 250 cycles and 858 mAh·g⁻¹ after 500 cycles, although its multiphase structure complicates synthesis [29]. The mechanically robust and conductive Si/CNT network retained 922 mAh·g⁻¹ after 200 cycles and 612 mAh·g⁻¹ after 1000 high-rate cycles, but CNT cost and interfacial uniformity remain concerns [30]. The Si/rGO/C composite provided 1209 mAh·g⁻¹ initially, 92% retention after 100 cycles, and stable performance up to 6C [31]. Similarly, Si/nanographite delivered 1569 mAh·g⁻¹ initially and 1138 mAh·g⁻¹ after 100 cycles [32]. Chemically reinforced Si-N-C bonding improved the interfacial durability of SiO_x/3D-graphene, although laser processing limits scalability

[33]. Finally, carbon-coated Si/graphene oxide retained 70% capacity after 267 cycles at $1 \text{ A}\cdot\text{g}^{-1}$, but coating thickness requires careful optimization [34].

2.1. Carbon-Based Composite Anodes

Composite anode materials have emerged as a critical area of research in advancing lithium-ion battery technology, addressing key limitations of traditional graphite anodes, primarily their limited specific capacity and poor cycling stability under fast charge/discharge conditions [5] [35]. Among the diverse classes of composites, carbon-based composites remain foundational due to their excellent electrical conductivity, mechanical flexibility and chemical stability. Recent studies have focused on hybrid structures such as graphene combined with carbon nanotubes or amorphous carbon coatings to improve electrode integrity and electron transport pathways. For example, recent work demonstrated a graphene-CNT hybrid network that maintained over 90% capacity retention after 1000 cycles at high current densities, overcoming the common issue of capacity fading in conventional carbons [36]. Despite these advancements, challenges remain in optimizing the balance between porosity, mechanical strength and areal capacity to ensure practical energy densities suitable for commercial applications.

Carbon materials with diverse morphologies and hierarchical structures are widely regarded as viable anode components in lithium-ion batteries due to their accessibility, mechanical and thermal robustness, electrochemical stability, low cost and ability to support reversible lithium intercalation/de-intercalation [37] [38]. However, despite these favorable properties, carbon-based anodes are not chemically inert. At elevated temperatures, lithiated or delithiated carbonaceous electrodes can undergo vigorous reactions with non-aqueous electrolytes, while even under ambient conditions, parasitic side reactions and continuous SEI growth may occur, negatively impacting long-term cycling stability and safety [39]. These issues underscore the need for careful material selection and interface engineering in composite anode design.

Table 2 summarizes the advanced multilayered and three-dimensional Si-carbon architectures provide continuous conductive pathways and internal space to accommodate silicon expansion. The hydrothermally synthesized Si/C/graphite double-layer composite, consisting of a silicon core, inner carbon coating, and outer graphite layer, delivered $2469 \text{ mAh}\cdot\text{g}^{-1}$ after 50 cycles at $0.2 \text{ A}\cdot\text{g}^{-1}$. It retained $1500 \text{ mAh}\cdot\text{g}^{-1}$ after 300 cycles at $2 \text{ A}\cdot\text{g}^{-1}$ and achieved $471 \text{ mAh}\cdot\text{g}^{-1}$ at $32 \text{ A}\cdot\text{g}^{-1}$ [28]. The flexible Si/graphite/carbon nanofiber electrode incorporated silicon into a three-dimensional conductive network, providing a capacity exceeding $2000 \text{ mAh}\cdot\text{g}^{-1}$, structural stability, and favorable rate performance [40]. A carbon-coated Si nanoparticle/hollow graphite-fiber composite achieved $1498 \text{ mAh}\cdot\text{g}^{-1}$ at $200 \text{ mA}\cdot\text{g}^{-1}$ and $462 \text{ mAh}\cdot\text{g}^{-1}$ at $10 \text{ A}\cdot\text{g}^{-1}$, while reportedly exhibiting no capacity decay over 2000 cycles at $10 \text{ A}\cdot\text{g}^{-1}$ [41]. The milled, spray-dried, and pyrolyzed SiO/C/graphite composite delivered $963 \text{ mAh}\cdot\text{g}^{-1}$ at $100 \text{ mA}\cdot\text{g}^{-1}$, retained $950 \text{ mAh}\cdot\text{g}^{-1}$ after 100 cycles, and provided $670 \text{ mAh}\cdot\text{g}^{-1}$ at $1 \text{ A}\cdot\text{g}^{-1}$ [42]. Finally, the

pitch-derived hard-carbon@Si-P composite, comprising a hard-carbon core and nanosilicon shell, exhibited a low capacity-decay rate of 0.16% per cycle over 200 cycles at 1 A·g⁻¹ [43].

Table 2. Performance comparison of C-based composite anodes.

Composite	Synthesis/Structure	First-cycle capacity	Electrochemical performance	Rate capability
Si/C/Gr double-layer [28]	Hydrothermal + coating, Si core, C inner, Gr outer	2469 mAh/g (50 cycles @ 0.2 A/g)	Maintained 1500 mAh/g after 300 cycles at 2 A/g	471 mAh/g at 32 A/g
3D Flexible Si/Gr/C Nanofiber (FSiGCNF) [40]	3D architecture with atomic-scale expansion control, Si embedded in Gr/C nanofibers	2000+ mAh/g (high capacity, see design)	High structural stability over many cycles (due to free space design)	Good rate performance due to conductive network
Si NP/Hollow G Fiber/C coating [41]	CVD + nanofiber/C coating, sandwich-Si nanoparticles tied-up on Gr/CNT aerogel, hollow graphite fiber	1498 mAh/g (at 200 mA/g)	Very stable cycling: e.g., 2000 cycles at 10 A/g with no capacity decay	462 mAh/g at 10 A/g
SiO/C/Graphite [42]	Milling + spray drying + pyrolysis, SiO plates, amorphous C coating, graphite sheets	963 mAh/g (at 100 Ma/g)	950 mAh/g retained after 100 cycles	670 mAh/g at 1 A/g
Hard-C@Si-P (HC@Si-P) [43]	Pitch-derived carbon coating, hard C core, nano-silicon shell	—	Very stable cycling, 0.16% capacity decay/cycle over 200 cycles at 1 A/g	—

2.1.1. Graphite-Based Anode

Graphite has remained the dominant commercial anode material since its introduction in the first lithium-ion battery (LIB) in 1994, owing to its low cost, wide availability and stable operation at a low lithiation potential (0.1 V vs. Li/Li⁺). During lithiation, lithium ions intercalate between graphene layers, forming staged lithium-graphite intercalation compounds with a theoretical gravimetric capacity of 372 mAh/g and a volumetric capacity of approximately 850 mAh/cm³ [44] [45]. The ordered staging mechanism characterized by alternating lithium and graphene layers confers excellent structural reversibility under standard operating conditions.

Despite these advantages, graphite exhibits severe electrochemical limitations under low-temperature and high-rate operating conditions. At reduced temperatures, increased bulk electrode resistance, charge-transfer resistance (R_{ct}), and solid-electrolyte interphase (SEI) impedance hinder lithium-ion diffusion and intercalation kinetics [46]. The resulting polarization lowers the anode potential toward that of metallic lithium, thereby increasing the risk of lithium plating. Experimental results show that graphite retains only 12% of its room-temperature capacity at -20°C, compared with 92% under ambient conditions [47]. GITT and EIS analyses further reveal that, at -30°C, SEI resistance increases approximately 27-fold and electrolyte resistance rises by nearly one order of magnitude relative to 25°C, with R_{ct} becoming the primary contrib-

utor to polarization [48]. These kinetic limitations restrict the use of graphite in fast-charging and low-temperature applications. Nevertheless, structural engineering strategies, including particle-size control, porous architectures, interface optimization, and composite design can improve ion transport, reaction kinetics, and structural stability under low-temperature conditions [49].

2.1.2. Carbon Nanotube and Nanofiber Composites

Carbon nanotube and nanofiber composites have emerged as highly effective anode materials due to their ability to store Li-ions through intercalation rather than alloying, offering minimal volume expansion and superior structural stability for high-rate, long-life lithium-ion batteries [12] [45]. As graphitic allotropes, CNTs exhibit exceptional tensile strength, high electrical conductivity and resistance to chemical degradation, enabling enhanced capacity and reduced pulverization compared with conventional graphite anodes [13] [36] [50]. Their electronic properties are chirality-dependent, with metallic CNTs delivering lithium insertion capacities up to 400% higher than semiconducting CNTs [51]. Recent work demonstrated that integrating CNTs into $\text{Bi}_5\text{Nb}_3\text{O}_{15}$ via a simple solid-state CNT-modulation approach forms a conductive 3D network that improves electron/ion transport, buffers volume change, and significantly enhances rate performance [52]. **Figure 3** shows these findings highlight carbon nanotube (CNT)-based structural modulation as a highly effective strategy for advancing next-generation intercalation-type anode materials by improving electron transport, accommodating volume changes, and enhancing electrochemical stability. Mechanochemical engineering further expands these opportunities. The high-energy ball milling exfoliates graphite into graphene while simultaneously enabling heteroatom doping and the assembly of graphene-based composite electrodes. This scalable

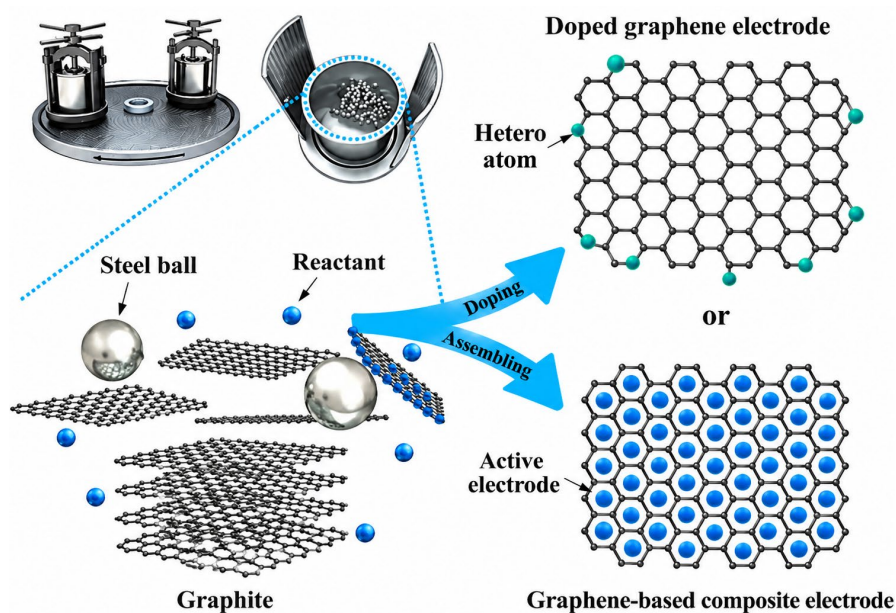


Figure 3. Graphite and graphene functional structure [53].

mechanochemical process tailors the electronic structure, increases the density of electrochemically active sites, and constructs conductive networks that uniformly disperse active materials. Consequently, the resulting graphene-based electrodes exhibit enhanced structural integrity, accelerated lithium-ion diffusion, improved electrical conductivity, superior cycling stability, and enhanced overall electrochemical performance, making mechanochemical synthesis a promising route for the development of high-performance lithium-ion battery anodes [53].

2.1.3. Graphene and Graphene Oxide-Based Composites

Graphene and graphene oxide (GO) based composites remain among the most promising anode materials for lithium-ion batteries (LIBs) due to their high surface area, excellent electrical conductivity and theoretical Li-storage capacity 744 mAh/g [54]. A primary challenge is the intrinsic tendency of graphene sheets to restack via van der Waals forces, which restricts ion-accessible surfaces and suppresses rate performance. Heteroatom functionalization including B, N, S, and halogen doping effectively alleviates these limitations by enlarging interlayer spacing, introducing active defects and improving electronic structure, with N-doping showing the most consistent enhancements in conductivity and Li-ion adsorption [55]. Hybrid graphene-based architectures further demonstrate significant electrochemical benefits. For example, $\text{Li}_4\text{Ti}_5\text{O}_{12}$ /graphene nanocomposites prepared via atomic layer deposition or hydrothermal lithiation exhibit ultrafast ion diffusion and strong cycling durability, maintaining 120.8 mAh/g at 100 C and 90% capacity retention over 2500 cycles [56]. Carbon nanofiber (CNF) integrated systems, such as Fe/ Fe_3C -CNF hybrids, also achieve high reversible capacities 270 - 380 mAh/g by reducing charge-transfer resistance and improving low-temperature performance [57]. Temperature-dependent studies further reveal that graphene-based anodes approach theoretical capacities at 10°C but suffer sharp capacity decline at lower temperatures. Additionally, carbon-coated $\text{Li}_5\text{Cr}_7\text{Ti}_6\text{O}_{25}$ nanofibers produced via electrospinning show superior structural integrity and maintain 84% of their capacity after 500 cycles at 1 A/g [58]. These results underscore the importance of heteroatom engineering, hierarchical structuring and carbon-nanostructure integration in enabling high-rate, high-stability graphene-based anodes for next-generation LIBs.

2.1.4. Carbon-Carbon Hybrids and 3D Porous Carbon Frameworks

Three-dimensional (3D) carbon-carbon hybrids, combining carbon allotropes such as graphene and carbon nanotubes (CNTs) and 3D porous carbon frameworks have emerged as highly promising anode scaffolds for next-generation lithium-ion batteries. By integrating two-dimensional graphene sheets with one-dimensional CNTs in a 3D interpenetrating network, these hybrids prevent graphene restacking and maximize the complementary advantages of each allotrope graphene provides exceptional electronic conductivity, while CNTs ensure robust structural support and pore connectivity [59]. Similarly, purely carbon-based 3D frameworks such as carbonized wood-derived carbon current collectors or non-

woven carbon nano-fiber (CNF) matrices offer lightweight, highly conductive structures with interconnected porosity that facilitate rapid electron and ion transport, enable very thick electrodes with high mass loading and buffer mechanical stresses during cycling [60]. These architectures support enhanced electrochemical kinetics, improved rate capability and better cycle stability compared with conventional carbon anodes. In template 3D porous carbons ordered mesoporous carbon, reported specific capacities reach up to 900 - 1100 mAh/g under moderate current densities, thanks to their enormous surface area ($>1000 \text{ m}^2/\text{g}$) and high pore volume [61]. Despite challenges such as first-cycle irreversible capacity loss largely due to solid electrolyte interphase (SEI) formation on the extensive surface area and the intrinsic limitation of pure carbon frameworks in storing lithium compared with alloy or conversion-type materials, 3D carbon-carbon hybrids remain highly attractive as conductive, mechanically robust scaffolds for hosting high-capacity silicon, Sn, or metal-oxide anodes, addressing key issues of conductivity, volume change and cycle life [62].

Table 3 summarizes the performance of various Silicon-based and carbon-based composite anodes. high capacity with improved structural and interfacial stability. The (Si/G/Gr)@C composite, prepared through spray processing and pyrolysis, delivered a first-discharge capacity of $860.4 \text{ mAh}\cdot\text{g}^{-1}$ with an initial Coulombic efficiency of 90.9% and retained 74.5% of its capacity after 200 cycles [63]. A sustainable Si/reed-catkin-derived carbon composite, fabricated using magnesiothermic reduction and porous biomass carbon, achieved $318.4 \text{ mAh}\cdot\text{g}^{-1}$ after 100 cycles at $200 \text{ mA}\cdot\text{g}^{-1}$ and retained $229.3 \text{ mAh}\cdot\text{g}^{-1}$ after 1000 cycles at $1 \text{ A}\cdot\text{g}^{-1}$ [64]. Although its capacity was relatively moderate, the porous biomass-derived framework supported prolonged cycling. A scalable Si-graphene nanocomposite exhibited $1307 \text{ mAh}\cdot\text{g}^{-1}$ at 0.1C and an initial Coulombic efficiency of 84.7%. It retained 97% and 89% of its capacity after 25 and 50 cycles, respectively, demonstrating the potential of scalable processing for practical electrode production [65]. Similarly, a Si/G@graphite composite produced through spray drying and heat treatment delivered approximately $850 \text{ mAh}\cdot\text{g}^{-1}$ during the initial cycles and maintained $500 \text{ mAh}\cdot\text{g}^{-1}$ after 50 cycles at $100 \text{ mA}\cdot\text{g}^{-1}$ [66]. These results highlight the roles of conductive carbon networks, protective coatings, and scalable synthesis in improving silicon-anode performance.

Table 3. Summary of Si-C composite performance metrics.

Composite	Synthesis/Structure	First-cycle capacity	ICE (%)	Cycle performance
(Si/G/Gr)@C [63]	Nano-Si + G + Gr, C coating (spray + pyrolysis)	860.4 mAh/g (1st discharge)	90.9%	74.5% capacity retention after 200 cycles
Si/Reed-Catkin biomass C (Si/RC) [64]	Magnesium thermal reduction, biomass-derived porous C	318.4 mAh/g (at 200 mA/g) after 100 cycles	—	229.3 mAh/g after 1000 cycles @ 1 A/g
Si-Gr (Scalable) [65]	Scalable graphene nanocomposite (MDPI)	1307 mAh/g at 0.1 C	84.7% (first cycle)	97% retention at 25th cycle; 89% at 50th cycle
Si/G@Gr [66]	Spray drying + heat treatment	850 mAh/g (in earlier cycles)	—	500 mAh/g maintained at 50 cycles @ 100 mA/g

2.2. Silicon-Based Composite Anodes

Silicon (Si) is widely recognized as a leading candidate for next-generation lithium-ion battery (LIB) anodes due to its exceptionally high theoretical gravimetric capacity 4200 mAh/g and volumetric capacity of 2357 mAh/cm³, far surpassing commercial graphite [16] [40] [67].

Despite its natural abundance, environmental benignity, and low operating potential, the practical application of silicon is limited by its substantial volume change of approximately 300% - 400% during lithiation and delithiation. This repeated expansion and contraction causes mechanical fracture, unstable solid-electrolyte interphase (SEI) formation, and low Coulombic efficiency [68] [69]. To address these limitations, various structural and compositional strategies have been developed. **Figure 4** summarizes recently reported silicon-based anode architectures for lithium-ion batteries, classified by dimensionality—0D, 1D, 2D, and 3D—and their integration with different carbon-based materials [69]. To mitigate these limitations, recent advances have focused on silicon-based composite architectures, particularly Si-carbon hybrids, graphene-reinforced matrices and engineered yolk-shell structures that buffer volumetric strain while improving electronic conductivity [70]. Yolk-shell Si@C frameworks have shown notable durability, with recent studies reporting capacity retentions exceeding 80% after hundreds of cycles at high current densities [71]. Amorphous columnar Si films have further demonstrated superior low-temperature stability, maintaining capacities around 600 mAh/g at 20 °C, outperforming graphite-based controls [72]. Additionally, prototype Si-rich pouch cells integrating Si composite anodes with high-energy NCA cathodes have achieved energy-dense operation with >80% capacity retention over extended cycling under both ambient and elevated temperature conditions [73]. It was illustrated the dimensional classification of carbon-based materials and their structural roles in silicon-based energy storage applications. The circular framework categorizes carbon nanostructures into 0D, 1D, 2D, and 3D architectures around a central silicon (Si) reference, highlighting their contributions to Si-C composite anodes. 0D carbon materials (fullerenes, carbon quantum dots, and nanoparticles) provide abundant active sites and short ion-diffusion pathways, enhancing interfacial reactions. 1D structures (nanotubes, nanofibers, and nanowires) establish conductive networks and accommodate silicon-induced mechanical stress. 2D carbon materials (graphene and layered carbon sheets) offer high conductivity, large surface area, and flexible frameworks for rapid charge transfer and deformation buffering. 3D carbon architectures (porous carbon, aerogels, and interconnected networks) provide hierarchical structures for silicon confinement and electrolyte transport.

This dimensional diversity enables tailored optimization of conductivity, mechanical stability, ion diffusion, and interfacial compatibility in Si-C anodes. By integrating silicon with carbon frameworks of different dimensionalities, limitations associated with silicon's poor conductivity and large volume expansion can be effectively mitigated, resulting in enhanced cycling stability, rate capability, and

electrochemical performance for next-generation lithium-ion batteries. Dimensional classification of carbon-based materials (0D - 3D) surrounding silicon (Si), highlighting the structural versatility of carbon architectures for advanced Si-based energy storage applications.

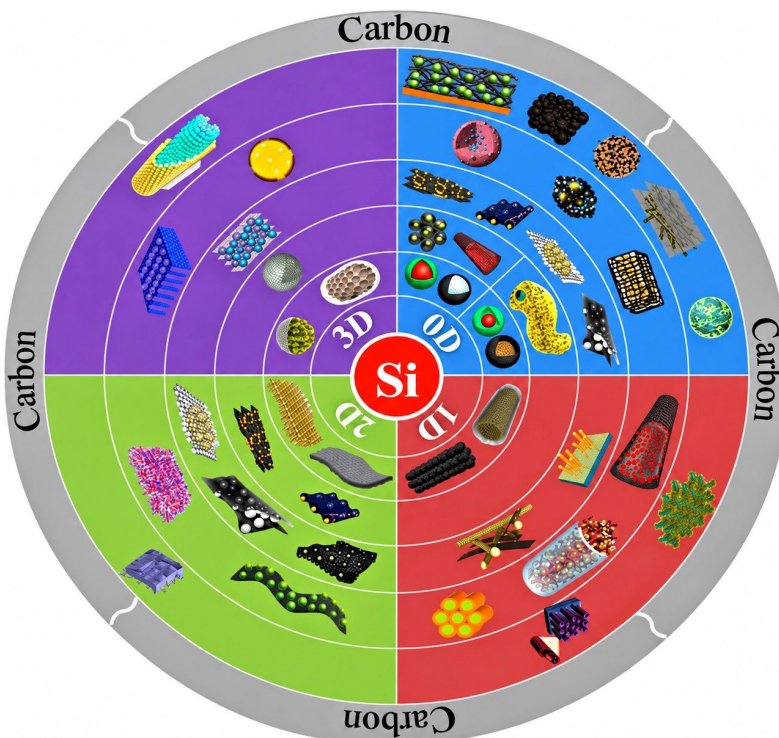


Figure 4. Diagram of recently published Si anode materials for LIBs [69].

2.2.1. Silicon-Carbon (Si-C) Composites

Silicon-carbon (Si-C) composites are currently considered as one of the most promising anode materials for lithium-ion batteries (LIBs). Although pure silicon is attractive due to its very high theoretical capacity 4200 mAh/g, the lithiation process causes a volume expansion of about 300%, which leads to particle breakage, reduced electrical conductivity, formation of unstable SEI layers and rapid capacity degradation. To mitigate these limitations, Si-C composites are prepared by combining silicon with various types of carbon materials such as graphite, amorphous carbon, graphene, CNTs, carbon nanofibers, etc. The carbon matrix absorbs the mechanical expansion of silicon, increases the electron conductivity and helps form a relatively stable SEI layer. This greatly reduces the tendency of silicon breakage, electrical isolation of the active material and repeated SEI reorganization. Various engineered architectures have been used in Si-C composites such as Si@C core-shell structures, yolk-shell (Si@void@C) designs, silicon embedding in porous carbon frameworks and carbon-coated silicon nanoparticles. In particular, the yolk-shell architecture provides internal voids for silicon expansion, resulting in intact particle structures and high long-cycle stability. Various synthesis techniques are used to prepare Si-C composites, such as ball milling,

pyrolysis, chemical vapor deposition (CVD), hydrothermal methods, etc. which help in controlling the structural design. Many Si-C composites with advanced architectures have shown reversible capacities exceeding 1000 mAh/g and excellent capacity retention over hundreds of cycles. Although they offer significantly improved performance compared to pure silicon, some challenges remain such as complex and expensive synthesis processes, relatively low initial Coulombic efficiency, difficulty in implementing high areal loading and challenges in integrating into practical full-cell configurations. Nevertheless, Si-C composites in combination with improved binders, electrolyte additives and scalable manufacturing methods are being considered as highly promising anode systems for next-generation high-energy-density batteries.

2.2.2. Si-Metal and Si-Metal Oxide Composites

Si-metal and Si-metal oxide composites have emerged as an effective strategy to overcome the intrinsic limitations of silicon anodes in lithium-ion batteries (LIBs), particularly the substantial volume expansion (300%) and mechanical pulverization that occur during lithiation and delithiation [74]. Incorporating metals such as Cu, Ni and Fe, or metal oxides including CuO, SnO₂ and Fe₂O₃, provides conductive, mechanically resilient matrices that buffer volumetric strain, stabilize the solid-electrolyte interphase (SEI) and maintain electrical continuity during cycling [75]. Metal oxides further contribute additional capacity through conversion or alloying reactions while promoting the formation of robust interfacial layers. Recent studies demonstrate that Si-CuO and Si-Fe₂O₃ composites can form interconnected nanoscale frameworks with enhanced structural integrity and improved electrochemical performance relative to pure silicon, particularly in terms of capacity retention and cycle stability [76] [77]. These materials are synthesized via scalable techniques such as high-energy ball milling, spray pyrolysis and solution-based routes, enabling tailored core-shell or heterostructured morphologies. Although challenges remain including interfacial reaction control, dispersion uniformity and mitigating inactive mass contributions Si-metal and Si-metal oxide composites continue to represent a promising pathway toward durable, high-power silicon-based anodes for next-generation LIBs [78].

2.2.3. SiO and SiO_x Composites

SiO and SiO_x composites have gained prominence as practical silicon-based anode materials that bridge the performance gap between elemental Si and SiO₂, offering a favorable balance between high reversible capacity and improved mechanical stability [79]. Their amorphous or sub-stoichiometric structures enable moderated volume expansion 150% - 200% and promote the formation of more stable solid-electrolyte interphase (SEI) layers compared to pure silicon, owing to the buffering effect of silicon oxide domains and their participation in both alloying and conversion reactions during lithiation [79]. When combined with conductive carbon frameworks such as graphite, graphene, amorphous carbon coatings or carbon nanotubes SiO_x composites exhibit enhanced electrical conductivity, re-

duced mechanical degradation and significantly improved cycling stability and first-cycle Coulombic efficiency [80]. Despite challenges, such as irreversible Li consumption due to the formation of lithium silicates and Li_2O having theoretical capacity (1600 - 2000 mAh/g), SiO and SiO_x composites are highly appealing for commercial LIB applications because of their reliability, manufacturability and compatibility with existing electrode fabrication processes [24] [81].

2.2.4. MOF-Derived and Polymer-Derived Silicon Composites

MOF-derived and polymer-derived silicon composites have emerged as highly effective strategies for addressing the intrinsic limitations of silicon anodes in lithium-ion batteries (LIBs), including poor electronic conductivity, unstable solid-electrolyte interphase (SEI) formation and severe 300% volumetric expansion during cycling [13] [82]. In MOF-derived systems, the carbonization of metal-organic frameworks produces hierarchically porous, high-surface-area carbon scaffolds capable of uniformly anchoring or encapsulating silicon nanoparticles, thereby enhancing ion transport, buffering mechanical strain and maintaining long-range electrical connectivity [83]. Polymer-derived carbon matrices originating from precursors such as polypyrrole (PPy), polyaniline (PANI), or phenolic resins offer similarly advantageous conductive and flexible frameworks that improve silicon dispersion and structural stability while enabling the formation of engineered architectures, including yolk-shell and core-shell designs [84]. These approaches collectively improve reversible capacity, mitigate electrode pulverization and stabilize SEI evolution, although challenges persist related to scalable synthesis, tap-density optimization and initial irreversible capacity loss. Overall, MOF- and polymer-derived silicon composites represent a versatile and promising route for achieving durable, high-performance silicon anodes suitable for next-generation LIB applications.

2.2.5. Silicon Nanoparticles (SiNPs)

Silicon nanoparticles have emerged as a highly promising anode material for next-generation Li-ion batteries due to their exceptional theoretical capacity of approximately 4200 mAh/g nearly ten times greater than that of traditional graphite anodes [13] [67]. Their nanoscale structure effectively mitigates the significant volume expansion 300% that occurs during lithiation, which otherwise leads to electrode pulverization and capacity fading [85]. Various synthesis strategies, including chemical vapor deposition, ball milling and solution-based methods, have been employed to fabricate Si nanoparticles with tailored morphologies and surface modifications. When combined with conductive matrices such as graphene, carbon nanotubes, or doped carbon coatings, silicon nanoparticles demonstrate improved cycling stability and rate capability. Despite ongoing challenges related to mechanical degradation and solid electrolyte interphase (SEI) instability, recent advances in nanostructuring and composite design have significantly enhanced their electrochemical performance, positioning silicon nanoparticles as a leading candidate for high-energy-density lithium-ion battery applications [86] [87].

2.2.6. Advanced Silicon-Carbon Architectures

Advanced silicon-carbon (Si-C) architectures are crucial for achieving high capacity, long cycle stability and high rate capability in next-generation lithium-ion batteries. Conventional Si-C composites often suffer from performance limitations due to the approximately 300% volume expansion of pure silicon and the unstable SEI layer [13] [88]. To address these issues, various advanced architectures such as core-shell, where the carbon shell absorbs the expansion of silicon to enhance SEI stability and conductivity yolk-shell, where silicon expands into the internal voids to maintain structural integrity three-dimensional (3D) hierarchical, porous networks, which ensure rapid electrolyte penetration and ion transport and nanostructured Si-C, which confines silicon to a small size to reduce mechanical stress have significantly enhanced the durability of silicon [89]. In addition, MOF-derived or biomass-derived Si-C architectures are particularly promising due to their high porosity, improved electrical conductivity and environmentally friendly fabrication process [13]. These advanced architectures, combined with SEI stability, mechanical tolerance, improved electronic conductivity and long cycle life, make silicon-carbon electrodes very promising for next-generation high-performance and safe battery technologies [89].

Table 4. Comparative overview of composites and hybrid anode materials.

Composites	Synthesis/Structure	Cycle capacity	Rate capability & ICE (%)	Performances
Ni-based (NiO/NiO-C) [90] [91]	NiO nanoarrays on 3D Ni foam with carbon coating	389 mAh/g at 0.5 C > 100 cycles with stable area capacity	2.11 and 1.76 mAh/cm ² at 0.2 C and 0.5 C	C boosts conductivity, structural integrity, first-cycle loss, NiO low conductivity
Ni-Co mixed oxides (NiO/Co ₃ O ₄ /rGO) [92] [93]	Hollow hierarchical microspheres, rGO encapsulation on carbon cloth	1845 mAh/g at 0.1 A/g after 100 cycles, high capacity at 100 cycles	Tested at 0.1 A/g	rGO improves conductivity, buffers volume change, Stability beyond 100 cycles
Transition metal oxide hybrid (MnO ₂ @CNT fabric) [94]	Binderless hybrid: MnO ₂ nanostructures directly grown on CNT fiber fabric	1100 mAh/g at 25 mA/g, 97% retention over 1500 cycles at 5 A/g	500 mAh/g at 5 A/g & 97.5%	Good mechanical conductivity, strong collector coupling ultrastable cycling, Forming complexity control & cost
Hybrid (SnO ₂ /NiO nanotubes + Ag) [95]	1D hollow nano-tubes, porous walls, Ag nanoparticle decoration	>1000 mAh/g, Long-cycle stability	Ag enhances kinetics and conversion reversibility	Hollow design, Ag high conductivity, assists Li ₂ O conversion, Ag cost
Si/liquid metal (LM/Si) [96]	Self-healing LM/Si nanocomposite maintains interfacial contact	2300 mAh/g at 500 mA/g, 968 mAh/g after 1500 cycles at 8 A/g	360 mAh/g at 20 A/g (55°C) & 96%	Mechanical self- repair, high ICE, cycle endurance, LM cost, reactions, stress durability
Si/SiO composite (SiO/C/G) [34]	Carbon coating and graphite network, ternary composite with interfacial contact	963 mAh/g, 950 mAh/g after 100 cycles	Good rate response across tested currents & 70% - 80% for SiO-based	C/G improve conductivity, mechanical support, Lower capacity, irreversible loss

Table 4 summarizes the electrochemical performance of Si-C composites re-

ported in recent analyses. Transition-metal oxides and self-healing composites have been investigated to improve anode capacity and cycling stability. Carbon-coated NiO nanoarrays grown on three-dimensional Ni foam delivered $389 \text{ mAh}\cdot\text{g}^{-1}$ for over 100 cycles at 0.5C, with areal capacities of 2.11 and $1.76 \text{ mAh}\cdot\text{cm}^{-2}$ at 0.2C and 0.5C, respectively [90] [91]. The carbon coating improves conductivity and structural integrity, addressing the intrinsically poor conductivity of NiO. Hollow NiO/Co₃O₄ microspheres encapsulated with reduced graphene oxide achieved $1845 \text{ mAh}\cdot\text{g}^{-1}$ after 100 cycles at $0.1 \text{ A}\cdot\text{g}^{-1}$ [92] [93]. Here, rGO facilitates electron transport and buffers volume changes, although longer-term stability requires further evaluation. Binder-free MnO₂ nanostructures grown directly on CNT fabric delivered $1100 \text{ mAh}\cdot\text{g}^{-1}$ at $25 \text{ mA}\cdot\text{g}^{-1}$ and retained 97% capacity over 1500 cycles at $5 \text{ A}\cdot\text{g}^{-1}$ [94]. Hollow SnO₂/NiO nanotubes decorated with Ag nanoparticles provided over $1000 \text{ mAh}\cdot\text{g}^{-1}$, with Ag enhancing reaction kinetics and conversion reversibility, although its cost may limit scalability [95]. A self-healing liquid-metal/Si composite achieved $2300 \text{ mAh}\cdot\text{g}^{-1}$ at $500 \text{ mA}\cdot\text{g}^{-1}$ and retained $968 \text{ mAh}\cdot\text{g}^{-1}$ after 1500 cycles at $8 \text{ A}\cdot\text{g}^{-1}$ [96]. Finally, the SiO/C/graphite composite delivered $963 \text{ mAh}\cdot\text{g}^{-1}$ and retained $950 \text{ mAh}\cdot\text{g}^{-1}$ after 100 cycles, benefiting from the conductive and mechanically supportive carbon-graphite network [34].

2.3. Alloy and Intermetallic Composite Anodes

Alloy and intermetallic composites notably tin (Sn), antimony (Sb) and silicon-based intermetallics offer a balance between capacity and volume expansion. Tin-based composites, for instance, have a theoretical capacity of around 990 mAh/g and exhibit faster lithiation kinetics [97]. Alloying Sn or Sb with carbon or metals like nickel can enhance conductivity and buffer volume changes. A report of Sn-Sb nanoparticles embedded in carbon nanofibers exhibiting stable cycling over 600 cycles with less than 10% capacity fade, solving the common problem of electrode degradation. However, challenges include optimizing alloy composition for mechanical stability and maintaining good electrical contact over prolonged cycling.

2.3.1. Tin-Based Composite Anodes

Tin-based composite anodes remain a very active frontier in Li-ion battery research because of their high theoretical capacity for example; SnO₂ and Sn-based electrodes can in principle far surpass graphite. But until recently, the large volume changes (up to 300%) associated with lithiation/delithiation of Sn led to rapid capacity fade and poor cycle life [97]. However, suggest design strategies that substantially mitigate these issues. For instance, a 2025 report exposed that a binder-free SnO₂ thin-film electrode on copper, optimized via post-deposition annealing, delivered very high reversible capacity (1430 mAh/g) and retained 1200 mAh/g after 500 cycles, attributed to a hierarchical nanostructured morphology and a stable SnO₂-Cu interface that buffers volume change and supports interfacial Li-storage beyond classic alloying/dealloying [98]. Another recent advance (2024)

demonstrated that simply narrowing the cycling potential window (0 - 1.0 V vs Li/Li⁺) and using an ether-based electrolyte significantly improves the cycling stability of commercial SnO₂ nanopowders the anode delivered 650 mAh/g for 100 cycles [99]. This suggests that controlling the electrochemical window and electrolyte environment rather than relying solely on complex nanocomposites can meaningfully suppress the conversion-reaction-induced degradation [99] [100]. Hybrid/composite strategies also remain promising for example, embedding SnO₂ in a reduced-graphene-oxide (rGO) matrix improves the mechanical resilience of the electrode during cycling, with one study reporting 598 mAh/g after 200 cycles at 1 A/g [101]. Modern Sn/SnO₂-based anodes can now approach much more practical performance high reversible capacity, stable long-term cycling and mitigated mechanical failure when composite design, interface engineering, potential window control and electrolyte optimization are judiciously combined.

2.3.2. Antimony- and Bismuth-Based Composites

Antimony (Sb) and to a lesser extent bismuth (Bi) based composites have recently regained attention as high-capacity alloy-type anodes owing to Sb's theoretical capacity (660 mAh/g) and Bi's high volumetric density and potential to moderate volume-change stress. For example, a recent work reported a Sb@N-doped carbon fibers (Sb@NCF) free-standing electrode that delivered 675 mAh/g initially and retained 480 mAh/g after 300 cycles at 400 mA/g, with good rate capability (420 mAh/g at 1 A/g) [102]. Similarly, a BiSb/C composite synthesized via high-energy mechanical milling showed stable reversible capacities of 583, 466, 433 and 425 mAh/g at 500 mA/g after 100, 300, 500 and 1000 cycles, respectively [103]. These composite designs embed nanosized Sb or BiSb within conductive carbon matrices, which buffer volume expansion, preserve electrical connectivity and stabilize the solid-electrolyte interface (SEI), thereby suppressing pulverization and capacity fade [104]. Nevertheless, challenges remain: many of these studies employ thin electrodes with low mass loading, which may overestimate performance and underrepresent issues relevant for practical, high-loading cells [105]. First-cycle Coulombic efficiency (ICE) is often suboptimal due to Li consumed in SEI formation and side reactions long-term structural stability under high areal loading remains uncertain. Translating Sb/Bi composites into commercially viable LIB anodes will likely require further advances in electrode formulation, scalable synthesis, and electrode-level engineering to ensure consistent performance in full cells [106].

2.3.3. Silicon-Alloy and Intermetallic Composites

Silicon-alloy and intermetallic composites have emerged as a promising strategy to mitigate the severe volume expansion and poor intrinsic conductivity that limit pure Si anodes. By embedding Si within conductive alloy or intermetallic matrices such as Cu-Si, Ni-Sn, and Si-Al and combining these with carbon coatings, the structural integrity and electrochemical stability of the anodes are significantly enhanced. For example, N-doped carbon-coated Si-Cu composites prepared by

ball milling and thermal treatment form stable alloy/porous carbon structures that buffer Si volume changes and facilitate Li⁺ diffusion, yielding high capacities (>1000 mAh/g after hundreds of cycles) with improved coulombic efficiency (>90%) at 1 A/g [107]. Intermetallic matrices such as Ni₃Sn₄ and Ni₃Sn₂ serve as effective mechanical buffers when combined with Si and carbon, demonstrating low capacity decay (<0.05%/cycle) and high coulombic efficiencies (99.6%) while maintaining reversible capacities above 500 mAh/g [108]. In similarly engineered Si/Al carbon-coated composite thin films, the carbon layer enhances electrical conductivity, accommodates volumetric strain and suppresses side reactions, leading to stable cycling and respectable rate performance (579 mAh/g at 3 A/g). These compositional and architectural innovations not only improve electronic and ionic transport but also stabilize the solid electrolyte interphase and retard particle pulverization, collectively driving better cycling durability and practical viability for next-generation lithium-ion batteries.

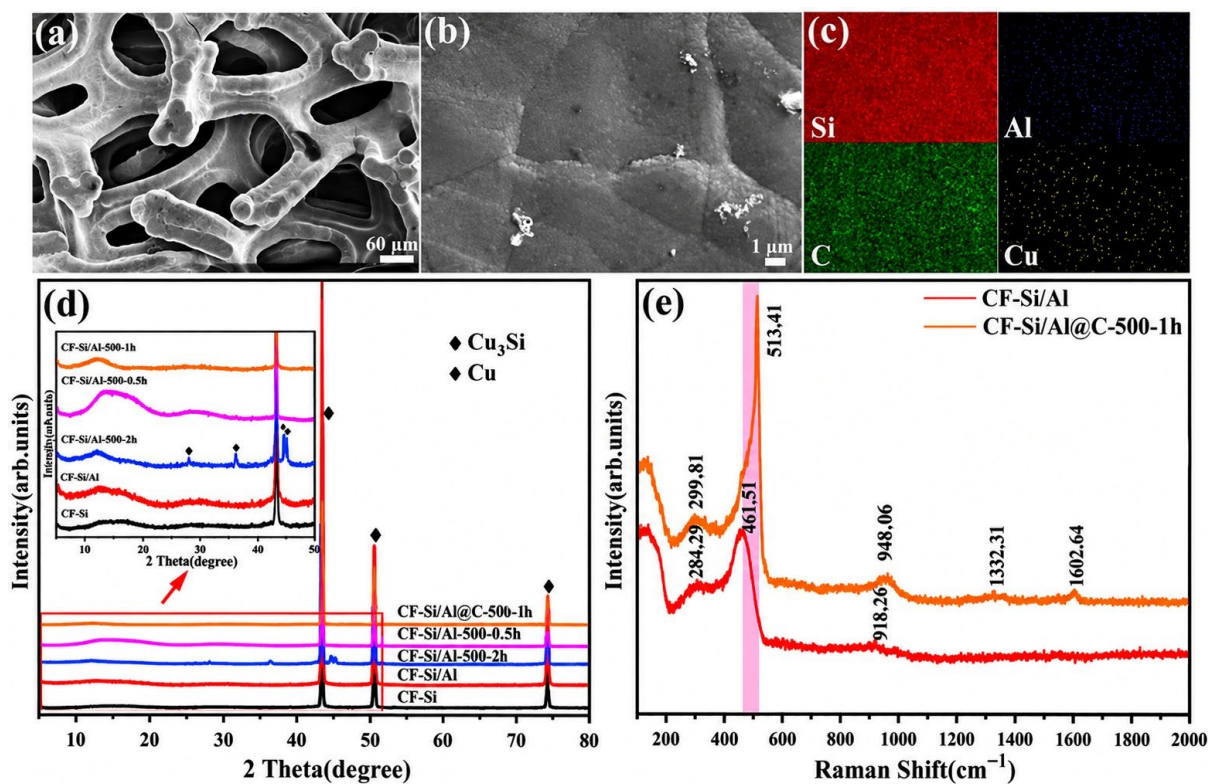


Figure 5. (a)-(c) SEM and EDS mapping of CF-Si/Al@C-500-1h, (d) XRD patterns of CF-Si-based samples, (e) Raman spectra of CF-Si/Al and CF-Si/Al@C-500-1h [109].

Figures 5(a)-(c) shows that CF-Si/Al@C-500-1h retains the interconnected porous carbon foam structure and exhibits uniform Si/Al deposition and homogeneous Si, Al, C, and Cu distributions [109]. This architecture provides strong interfacial contact and space to accommodate silicon volume changes during cycling. The XRD patterns in **Figure 5(d)** identify crystalline Cu and Cu₃Si phases, confirming alloying and phase evolution during thermal treatment. The Raman

spectra in **Figure 5(e)** confirm silicon-related vibrations and characteristic carbon bands, indicating preservation of the conductive carbon framework after coating and heat treatment. Collectively, these structural and compositional features support improved conductivity, mechanical stability, and electrochemical performance.

2.3.4. Manganese-Based Alloy Composites

Manganese monoxide (MnO) is an attractive conversion-type anode material for lithium-ion batteries because of its high theoretical capacity of approximately 755 mAh/g, low cost, natural abundance, and environmental compatibility. Unlike alloying-type anodes, MnO stores lithium predominantly through a reversible conversion reaction involving the reduction of MnO to metallic Mn accompanied by Li_2O formation. However, its practical application is limited by intrinsically poor electronic conductivity, substantial volume changes, and structural degradation during repeated lithiation/delithiation, resulting in capacity fading and limited rate capability [110].

To address these limitations, MnO has been extensively integrated with conductive carbonaceous matrices. MnO nanoparticles embedded within three-dimensional graphene and N-doped carbon networks (NC/MnO/rGO) exhibit enhanced conductivity and structural buffering, delivering reversible capacities of approximately 1360 mAh/g at 0.2 A/g over 150 cycles [111]. Similarly, porous N-containing carbon/MnO composites achieve capacities of approximately 1000 mAh/g with 98% retention after 200 cycles, attributed to abundant active sites, enhanced Li^+ transport, and improved electronic conductivity [112]. MnO incorporated into three-dimensional carbon networks has also demonstrated capacities exceeding 1060 mAh/g at 0.2 A/g and approximately 720 mAh/g after prolonged cycling, highlighting the effectiveness of carbon frameworks in accommodating conversion-induced mechanical strain and improving reaction kinetics [113].

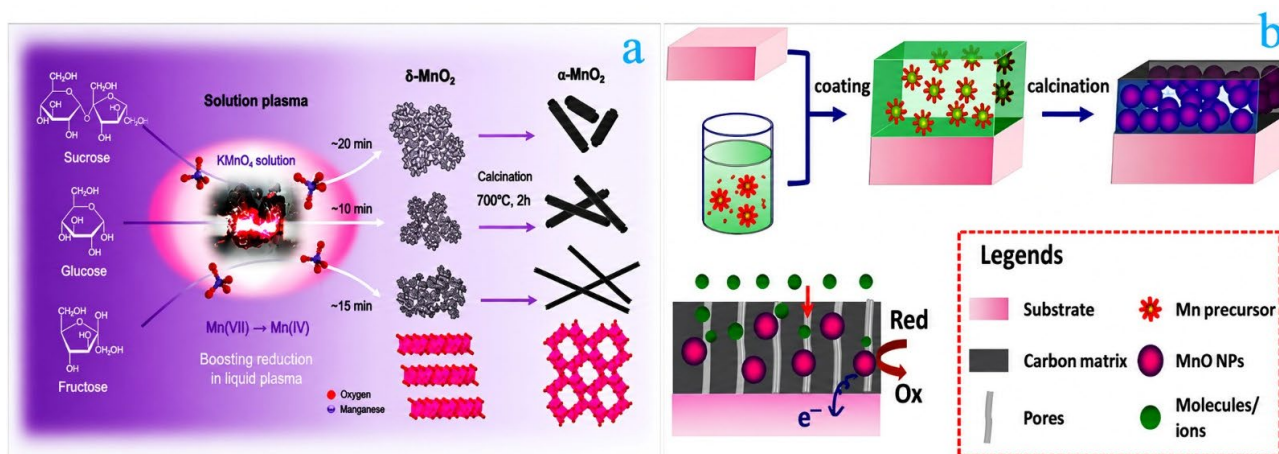


Figure 6. (a) Schematic of $\delta\text{-MnO}_2$ synthesis and $\alpha\text{-MnO}_2$ formation [114], (b) Schematic of MnO@C nanocomposite growth on substrate [115].

Figure 6(a) presents the controlled synthesis and phase transformation of man-

ganese oxides through solution-plasma processing followed by calcination [114], whereas **Figure 6(b)** illustrates the in-situ formation of MnO@C nanocomposites on a conductive substrate through precursor coating and thermal treatment [115]. Collectively, these studies demonstrate that MnO-carbon systems should be classified as conversion-type composite anodes rather than alloy or intermetallic composites. Their improved electrochemical performance arises primarily from conductive carbon networks, nanoscale structural engineering, and effective accommodation of conversion-induced volume changes. Within the scope of this review, these conversion-type systems are considered comparative benchmarks for Si-C anodes, particularly for evaluating capacity, structural stability, conductivity, and cycling durability.

2.3.5. Iron-Based Alloy Composites

Iron oxides such as Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$ are promising LIB anodes due to high theoretical capacities (1000 mAh/g), abundance and low cost, but their low conductivity, slow Li^+ diffusion and large volume changes limit performance. Recent studies show that integrating Fe oxides with carbon matrices, nanostructuring and doping significantly improves stability and kinetics. For example, Fe_3O_4 @C nanocomposites with hierarchical carbon layers achieve high reversible capacities (1647 mAh/g over 170 cycles) and excellent rate performance, while Fe_3O_4 /CNT composites maintain 683 mAh/g at 1 A/g after 100 cycles. These strategies enhance electron transport, buffer volume expansion and suppress nanoparticle aggregation, making iron-based composites strong candidates for high-performance lithium-ion battery anodes [116] [117]. **Figure 7** illustrates the lithiation

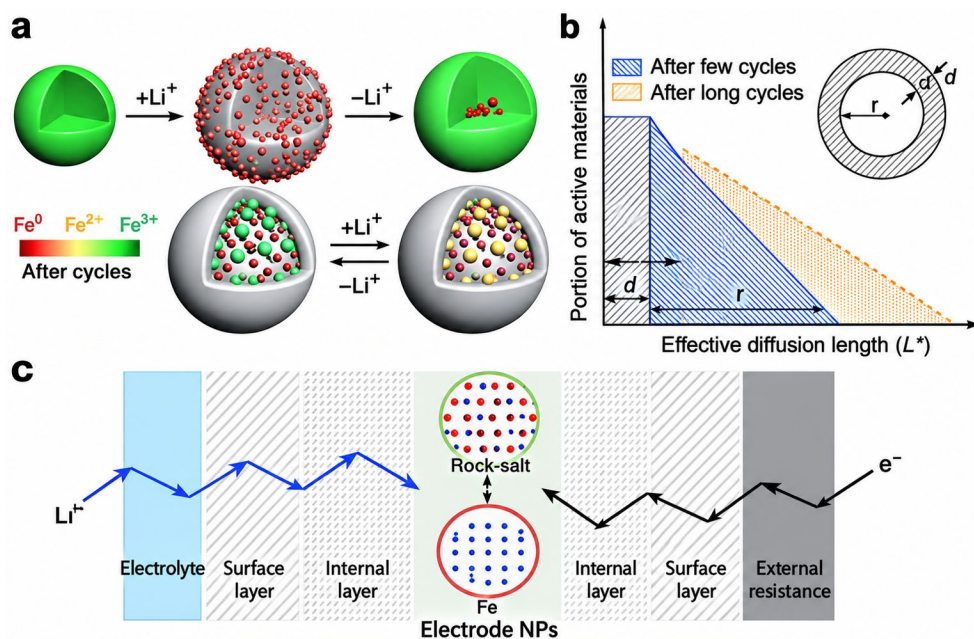


Figure 7. Schematics of Li in conversion-type electrodes: (a) phase evolution in Fe_3O_4 electrode, (b) surface layers and diffusion length growth over cycles, (c) Li^+ and electron diffusion paths for electrochemical activation [119].

and delithiation mechanisms of Fe-based conversion electrode nanoparticles, showing the progression of the reaction front from the surface toward the core. The evolving internal phases that include Fe^0 embedded in Li_2O or “rock-salt” matrix, affect active material utilization over cycling. Surface and internal layers each impose distinct Li^+ and electron transport resistances that increase with repeated volume change and phase transformation, contributing to capacity fade and polarization. These behavior patterns have been confirmed in recent nanoscale electrode studies that link structural evolution to electrochemical performance and cyclability [118] [119].

It is summarized in Sections 2.3.1 to 2.3.5, the alloy or intermetallic and transition metal oxide anodes demonstrate that combining conductive matrices, nanoscale engineering, controlled electrochemical environments and interface optimization can significantly enhance reversible capacity, rate capability and cycling stability. These strategies provide a foundation for the design and implementation of next-generation high-performance LIB anodes.

2.4. Conversion-Type Composite Anodes

Conversion-reaction composites based on transition metal oxides and sulfides such as Fe_2O_3 , Co_3O_4 and MoS_2 exhibit very high capacities due to multi-electron transfer reactions. These composites, often paired with carbonaceous materials, improve electrical conductivity and buffer volume changes during cycling [120]. To overcome limitations such as poor electronic conductivity, slow lithium-ion diffusion and large volume changes, these materials are commonly integrated with conductive matrices, carbon, CNTs, graphene or MOF-derived frameworks. This strategy increases electron transport, stabilizes the solid-electrolyte interphase (SEI), buffers volumetric strain to improve cycling stability and rate performance. For instance, MoS_2/rGO composites have demonstrated specific capacities exceeding 1000 mAh/g with excellent rate capabilities [121]. These composites effectively solve the limited capacity problem but face challenges like voltage hysteresis, low initial Coulombic efficiency and capacity fading due to particle agglomeration and SEI instability.

2.4.1. Transition-Metal Oxide Composites

Transition-metal oxide composites combine TMOs (MnO_2 , Co_3O_4 , NiO , Fe_2O_3 and TiO_2) with conductive supports like oxides with carbon, CNTs, graphene offer multi-electron conversion reactions while mitigating the low conductivity, large volume expansion and poor rate capability [122]. Composite designs significantly enhance performance for example, ZIF-derived $\text{ZnO}/\text{Co}_3\text{O}_4@\text{CNTs}$ achieve 1156 mAh/g after 200 cycles at 200 mA/g with only 0.54% capacity loss per cycle, while N-doped MnO/carbon composites deliver 1000 mAh/g with 98% retention after 200 cycles [112] [123]. Similarly, $\text{MnO}_2@\text{CNT}$ fabrics maintain 97% of their initial capacity (1100 mAh/g) over 1500 cycles, while $\text{Co}_3\text{O}_4@\text{N-doped graphene aerogels}$ and $\text{NiO}@\text{MXene}/\text{C}$ composites receive 1320 mAh/g after 300 cycles and 900 mAh/g after 500 cycles, respectively, demonstrating the role

of conductive and porous frameworks in improved kinetics and structural stability [124] [125]. Despite these advances, challenges include low initial Coulombic efficiency, scale-up difficulties, and limited performance at high areal loadings, though TMO composites remain strong candidates for high-capacity, long-life LIB anodes.

2.4.2. Transition-Metal Sulfide Composites

Transition-Metal Sulfide (TMS) composites, sulfides such as ZnS, FeS_x, MoS₂ and CoS with conductive matrices, such as carbon, CNTs, polymers, or doped carbon shells, offer high theoretical capacities while addressing intrinsic limitations, poor conductivity, interface instability and volume expansion. Core-shell or matrix-supported designs enhance electrochemical performance for instance, a Zn-Co-Fe-S@N-doped carbon polyhedron exhibits 966.6 mAh/g after 100 cycles at 100 mA/g and maintains 499 mAh/g at 2 A/g after 120 cycles, with the core-shell structure that accommodates volume changes and N-doped carbon to improve conductivity [126]. Similarly, ZnS/CNT composite with 10 nm ZnS nanoparticles anchored to CNT bundles delivered 333 mAh/g at 2 A/g over 4000 cycles, demonstrating long-term stability and high-rate capability [127]. Remaining challenges include high first-cycle irreversible capacity due to large surface area, potential structural degradation under extended high-rate cycling, and synthesis complexity. However, TMS composites are promising for next-generation high-rate, high-capacity, and flexible LIBs, and may also be adapted for solid-state systems with optimized conductivity and interface stability.

2.4.3. Multi-Anion Composite Systems

Multi-anion composites, comprising two or more distinct anionic species (O²⁻, F⁻, S²⁻, and phosphate-related anions) within a single electrode framework, have emerged as a promising strategy to enhance lithium-ion storage performance. These materials exploit interdependent mechanisms, including intercalation, conversion, and anion redox activity, to improve capacity, structural stability, and ionic/electronic transport. Recent studies demonstrate the efficacy of this approach. For instance, high-entropy oxyfluoride anodes, such as Li_{0.5}(Co_{0.2}Cu_{0.2}Mg_{0.2}Ni_{0.2}Zn_{0.2})_{1.5}O_{1.5}F_{0.5}, demonstrate enhanced reversible capacities (562 mAh/g) and improved rate capability, due to synergistic interactions between oxide-fluoride species and entropy-stabilized lattices [128]. Again, fluorine incorporation in oxyfluoride-based cathodes mitigates lattice degradation and oxygen loss, leading to superior cycling stability. Challenges remain, including significant first-cycle irreversible capacity loss due to solid-electrolyte interphase formation, complex control over anion distribution during synthesis, and issues related to scalability and reproducibility [129]. Nonetheless, the tunable nature of multi-anion architectures provides an electrochemically versatile materials for high-capacity, stable electrodes suitable for next-generation Li-ion and solid-state batteries.

In summary, the conversion-type composite anodes (Sections 2.4.1 to 2.4.3), *i.e.*

multi-metal and multi-anion composites provide a bridge among different cations and anions to improve capacity, rate performance, and lattice stability. High-entropy oxyfluorides and core-shell sulfide-carbon architectures represent this approach towards achieving high-capacity, high-rate, and durable anode designs suitable for both conventional and solid-state batteries. Collectively, these advanced anode materials integrate structural design, conductive frameworks, alloying, and multi-component chemistries that can overcome conventional limitations of high-capacity electrodes, providing the way for long-life, practical and energy-dense LIBs.

2.5. Phosphorus-Based and Elemental Composite Anodes

Phosphorus-based anodes, specially red phosphorus (red-P) and black phosphorus (BP), have attracted significant interest as high-capacity anode materials for Li-ion Na-ion batteries due to their very high theoretical capacity of around 2596 mAh/g [130] [131]. Pristine red phosphorus suffers from extremely low electronic conductivity and large volume expansion during lithiation or sodiation, leading to mechanical pulverization and rapid capacity decay [132] [133]. Embedding red phosphorus within conductive porous carbon, carbon matrices, carbon nanotubes (CNTs) and graphene mitigates these issues by enhancing electron transport and providing mechanical buffering, enabling significantly improved cycling stability and rate capability [134] [135].

2.5.1. Red Phosphorus-Carbon Composites

Red phosphorus-carbon (red-P@C) composites combine high-capacity phosphorus with conductive various carbon matrices, such as graphite, porous carbon, carbon films, carbon nanotubes, etc., are used to mitigate mechanical degradation and electrical isolation. The carbon component provides electronic conductivity and mechanical support, while phosphorus offers a very high theoretical capacity of approximately 2596 mAh/g [130] [136]. The composites overcome the two main drawbacks of pure red phosphorus, such as its very low electrical conductivity and high severe volume expansion during lithiation/delithiation that enable stable cycling with minimal capacity fading. Several recent works illustrate these improvements, a fluffy carbon-coated red phosphorus composite made by ball milling red phosphorus with expanded rice-derived carbon. For instance, ball-milled red-P coated with expanded rice-derived carbon achieved 1655.9 mAh/g at 0.05 C and 1230 mAh/g at 1 C with 96% retention over 1000 cycles [137]. A red-P or nitrogen-doped carbon composite (RPNC) showed 1453 mAh/g after 100 cycles at 300 mA/g, at higher rate (1200 mA/g) it still delivered 1178 mAh/g. The analysis on the red-P composite anodes for Li-ion batteries, reported 800 mAh/g with 99.7% coulombic efficiency with minimal thickness changes (e.g., 7% for 4.5 mAh/cm²) indicate its potentiality for practical applications [138]. In addition, flexible red-P embedded cross-linked structural carbon films achieve 903 mAh/g after 640 cycles at 100 mA/g and 460 mAh/g after 1000 cycles at 2 A/g, with nearly 100% Coulombic efficiency [139]. Collectively, these designs demonstrate that

carbon (coatings) matrices improve electron transport, reducing mechanical failure, accommodate volumetric expansion and stabilize the SEI, enabling high-capacity, long-life red-P anodes.

2.5.2. Black Phosphorus Composites

Black phosphorus (BP) possesses a layered structure of lithium-ion insertion and relatively higher intrinsic conductivity compared to red-P, but still undergoes significant volumetric expansion during cycling. Black phosphorus (BP) has attracted significant attention as a high-performance anode material for LIB due to its high theoretical capacity of 2596 mAh/g, that facilitates Li^+ insertion and relatively good electrical conductivity [140]. However, BP suffers from substantial volume expansion during lithiation and delithiation, which can lead to mechanical grind, formation of unstable solid electrolyte interphase (SEI) and rapid capacity fading. To address these challenges, recent studies have focused on BP-carbon composites that enhance conductivity, stabilize the BP structure and buffer volume changes. It is observed that the BP embedded with conductive carbon matrices, such as graphite, carbon nanotubes (CNTs), or porous carbon, as well as forming carbon coatings, reinforce mechanical integrity and interfacial stability. The effectiveness of these approaches were demonstrate and observed a BP/graphite/CNT composite (BP/G/CNTs) synthesized via ball milling exhibited an initial capacity of 1375 mAh/g at 0.15 A/g and retained 1031.7 mAh/g after 450 cycles, while maintaining 508.1 mAh/g after 3000 cycles at 2 A/g [141]. Similarly, a black phosphorus/hollow porous carbon (BP/HPC) composite showed a capacity of 350 mAh/g after 1000 cycles at 1000 mA/g, significantly outperforming pure carbon under the same conditions [142]. Another study demonstrated that forming stable phosphorus-carbon bonds in BP-graphite composites resulted in an initial discharge capacity of 2786 mAh/g at 0.2 C and 80% capacity retention after 100 cycles, with excellent rate capability across various C-rates [143]. BP-carbon composites improve performance by enhancing electron transport through the carbon matrix, buffering volume expansion with porous or flexible carbon structures and stabilizing the SEI through engineered interfaces. Despite these advantages some challenges are noted, like long-term stability at high current densities is still limited, initial coulombic efficiency is often reduced due to SEI formation and irreversible reactions.

2.5.3. Other Elemental Composites

Composites based on tellurium (Te), germanium (Ge), silicon-telluride (Si_2Te_3) and Te/red phosphorus with carbon have emerged as promising anodes, offering higher conductivity, moderate volume expansion and favorable lithium storage pathways compared to pure Si or P. Layered $\text{Si}_2\text{Te}_3@\text{C}$ composites have demonstrated high reversible capacities of 816 mAh/g (559 mAh/cm³) at 100 mA/g, retaining roughly 97% of capacity after 200 cycles with high Coulombic efficiency in lithium-ion battery tests [144]. Germanium oxide-based carbon composites (GeO_2/C and $\text{GeO}_2\text{-Ge/C}$) have also been investigated as high-capacity anode ma-

terials, with some reports showing reversible capacities approaching or exceeding 747 mAh/g after hundreds of cycles and even higher values in nanostructured Ge-rich composites due to alloying/conversion reactions, though specific cycling performance depends on composite design and testing conditions [145]. Te-rP-C composites provide 734 mAh/g at 100 mA/g, high initial Coulombic efficiency (72% - 80%) and excellent high-rate performance (580 mAh/g at 10 A/g). Carbon matrices enhance conductivity, buffer volume changes, and stabilize the SEI [132] [146]. Challenges include moderate first-cycle Coulombic efficiency, performance often tested at low areal loadings or thin electrodes, and material cost or scarcity (Ge, Te). Nonetheless, these composites provide a favorable balance of capacity, conductivity, rate capability, and structural durability, making them suitable candidates for design and scaling of high-energy, high-power lithium-ion batteries.

It is summarized in sections 2.5.1 to 2.5.3, alloy/intermetallic composites, conversion-type TMOs/TMSs and phosphorus/elemental composites jointly provide strategies to mitigate the intrinsic challenges of high-capacity anodes. The design principles are analyzed to improve capacity, cycle life and rate performance, based on the embedding active materials into conductive matrices, nanostructuring to mitigate volume changes, interfacial engineering to stabilize the SEI and hybridizing multiple elements or phases. These approaches collectively provide high-capacity anodes as important candidates for next-generation LIBs with enhanced energy density, rate capability and durability.

2.6. Organic and Polymeric Composite Anodes

Organic and polymeric composite anodes have emerged as promising alternatives to traditional inorganic materials in lithium-ion batteries (LIBs), due to their flexibility, sustainability and structural tunability. However, their practical application is restricted due to the low conductivity, solubility in electrolytes and limited cycle life. To overcome these challenges, composite strategies combining organic materials with conductive frameworks such as carbon nanotubes (CNTs), graphene and conductive polymers are widely studied. Conductive polymer-inorganic composites, in particular, have indicated the ability to barrier changes while increasing electronic transport. For instance, polymers like polypyrrole (PPy), polyaniline (PANI) and PEDOT offer mechanical flexibility and conductivity while buffering volume changes. It is noted that PPy-Fe₃O₄ composite achieved 720 mAh/g over 200 cycles, whereas PEDOT-SnO₂ composite delivered 500 mAh/g at 2 A/g with >98% Coulombic efficiency [147] [148].

Redox-active organic materials such as quinones and imide-containing frameworks offer inherently high theoretical capacities, whereas their practical use is hindered by dissolution in conventional electrolytes and low intrinsic electronic conductivity, resulting in rapid capacity decay and poor rate of performance. A key strategy to overcome these limitations is the design of covalent organic frameworks (COFs) with built-in redox centers and composite architectures that improve electronic and ionic transport. For instance, β -ketoenamine-linked COFs

grown in situ on carbon nanotubes (TP-3J-COF@CNT and TP-3Q-COF@CNT) exhibited maximum reversible capacities of approximately 1020 mAh/g and 731 mAh/g, along with excellent cycling stability exceeding 1400 and 3000 cycles, respectively, due to synergistic interactions between COF redox sites and conductive CNT networks [149]. Moreover, advanced composites such as covalently bonded 3D COF/Ti₃C₂ MXene heterostructures showed high reversible capacities of 490 mAh/g at 0.1 A/g and extraordinarily long cyclability, retaining significant capacity after 10000 cycles at 1 A/g, owing to enhanced conductivity and structural stability from the MXene support [150]. Despite these advances, challenges remain in achieving higher intrinsic electronic conductivity, improved electrolyte compatibility, enhanced thermal stability and scalable synthesis methods, underscoring the importance of further molecular design and electrode engineering for the development of COF-based lithium-ion battery anodes.

2.6.1. Conductive Polymer-Inorganic Composite Anodes

Conductive polymer-inorganic composite anodes represent an emerging strategy for improving the electrochemical and mechanical stability of lithium-ion battery electrodes. In these systems, conductive polymers can provide continuous electron-transport pathways, strengthen interfacial contact, and accommodate mechanical stresses generated by active materials during repeated lithiation and delithiation. When combined with high-capacity inorganic anode materials, particularly silicon and conversion-type metal oxides, polymer matrices can suppress particle aggregation, maintain electrode integrity, and reduce electrical isolation caused by volume changes. Conductive polymers such as polyaniline, polypyrrole, and PEDOT-based materials are particularly attractive because their flexible frameworks can simultaneously enhance electrical conductivity and provide structural confinement. Nanoscale inorganic particles embedded within conductive polymer networks can further shorten Li⁺ diffusion pathways and increase electrochemically accessible active sites. Accordingly, polymer-inorganic composite design provides a useful approach for balancing capacity, conductivity, mechanical durability, and cycling stability. However, practical implementation requires optimization of polymer content, interfacial adhesion, active-material loading, electrode density, and scalable fabrication. Within this review, only polymer-inorganic systems directly functioning as composite anodes are considered, while polymer-ceramic solid electrolytes and electrolyte-focused systems are excluded.

2.6.2. Organic Small-Molecule and COF-Based Composite Anodes

Organic small molecules and covalent organic frameworks (COFs) have attracted increasing attention as composite anode materials because of their structural tunability, multiple redox-active sites, low density, and potentially high lithium-storage capacity. Organic small molecules can provide high theoretical capacities but commonly suffer from dissolution, low intrinsic electronic conductivity, and rapid capacity fading [151]. Incorporating these molecules into conductive carbon frameworks can improve electrical transport, restrict dissolution, and enhance

structural stability. COFs provide additional advantages through ordered porous structures, tunable chemical functionalities, and well-defined ion-transport channels. For example, β -ketoenamine-linked TP-3J-COF@CNT and TP-3Q-COF@CNT composites produced through in-situ growth on carbon nanotubes exhibit improved conductivity and structural integrity, delivering specific capacities of approximately 1020 and 731 mAh/g, respectively, together with extended cycling stability of 1400 and 3000 cycles [152] [153]. These results demonstrate that coupling redox-active organic frameworks with conductive carbon networks can effectively overcome limitations associated with isolated organic electrodes. Nevertheless, scalable synthesis, electrode density, electronic conductivity, structural stability, and long-term electrochemical performance remain important challenges. Electrolyte-oriented polymer-inorganic hybrids and solid-electrolyte systems are therefore excluded from this subsection to maintain a clear focus on composite anode materials.

2.6.3. Redox-Active Polymer Composites

Redox-active polymers are broadly classified into two categories based on their chemical structures: (1) redox-active pendant-bearing polymers and (2) redox-active group-embedded polymers, the latter of which includes conjugated conductive polymers. In redox-active group-embedded polymers, the polymer backbone itself is electrochemically active and constructed from redox-capable monomers [154]. In contrast, redox-active pendant-bearing polymers feature redox-active moieties attached as side chains to non-conductive backbones, resembling grafted polymer architectures. These structural differences lead to distinct chemical and physical properties, redox mechanisms and electrochemical performances, making it essential to tailor their use in energy storage systems [155]. As illustrated in **Figure 8**, redox-active polymers encompass a diverse range of structures, including group-embedded conductive polymers such as PANI, PEDOT, PPy and PEDOT: PSS, as well as pendant-bearing systems incorporating quinone, imide, carbazole, triphenylamine, ferrocene and stable radical groups like nitroxyl and galvinoxyl. When integrated into composite systems with conductive additives, inorganic fillers, or porous hosts, these polymers can overcome limitations such as low conductivity, mechanical weakness, or sluggish redox kinetics [156]. Conductive polymer composites on the other hand improve electronic transport and structural robustness, while pendant-bearing polymer composites utilize tunable pendant groups to achieve higher capacities and faster charge-transfer behavior. By leveraging this structural versatility within composite architectures, redox-active polymer composites exhibit enhanced ionic/electronic conductivity, improved rate capability and superior cycling stability, providing highly promising materials for advanced LIBs applications.

The Sections 2.6.1-2.6.3 depict that the composite anodes based on conductive polymers, organic molecules and covalent organic frameworks (COFs) are emerged as promising solution to enhance the performance of LIB, while considering the intrinsic limitations of organic and polymeric materials, such as low electronic

conductivity, limited cycle life and solubility in electrolytes. Overall, these sections identified the critical role of structural design, conductive scaffolds, and hybrid architectures to analyze the high-capacity, stable, polymeric anodes and long-lived organic, while the performance and commercialization of the electrode remain key challenges.

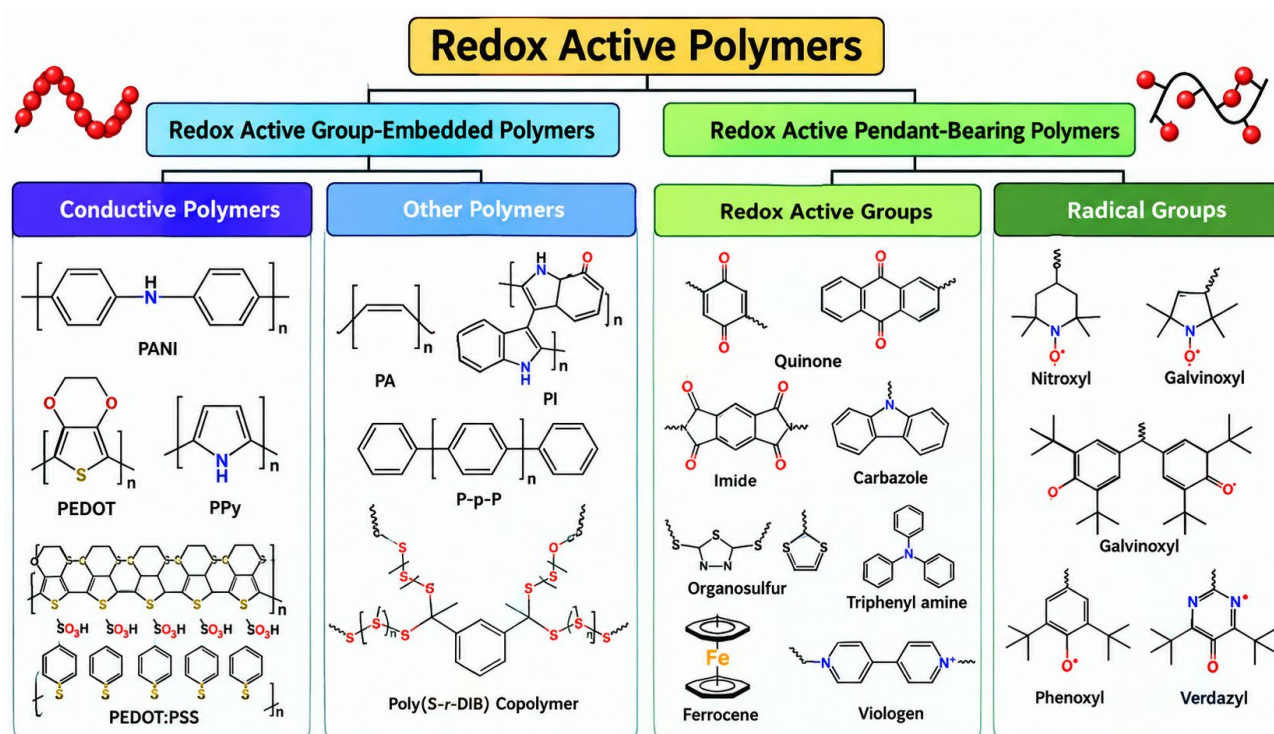


Figure 8. Classification and structures of redox-active polymers and groups [162].

2.7. Emerging and Multifunctional Composite Systems

Emerging multifunctional composite anodes, including MOF-derived materials, MXene hybrids and liquid-metal-based systems are expanding electrode design by offering tunable porosity, enhanced ionic/electronic conductivity and structural adaptability. MOF-derived Si/C composites with protective carbon shells from MOF precursors have shown high lithium storage, for instance, a Cu-MOF@Si/C anode delivering 725 mAh/g with 96.9% initial Coulombic efficiency and stable rate capability over 300 cycles [157]. In addition, MOF-derived heterostructures like N,S co-doped carbon matrix-encapsulated Cu₂S nanoparticles, exhibited 513 mAh/g after 1000 cycles at 1 A/g, illustrating the role of MOF templates in improving conductivity and cycling stability [158]. MXene hybrid anodes, like Ti₃C₂T_x, combined with carbon or metal oxides, provide enhanced electronic transport and structural integrity, achieving high capacities with good retention due to synergistic effects between MXene layers and conductive partners [159].

2.7.1. Liquid Metal-Active Material Composites

Liquid metal-active material composites have attracted interest as LIB anode ma-

terials because of their fluidic and self-healing properties that accommodate the large volume changes of high-capacity materials such as silicon and tin. For instance, eutectic gallium-indium-tin (EGaInSn) liquid metal nanoparticles used as anode material to deliver specific capacity of 474 mAh/g and retained 77% over 500 cycles at 0.1 A/g, due to self-healing and continuous electrical contact at the liquid phase [160]. Liquid-metal-enhanced composites such as LM/Ag nanowires@MoS₂ showed 468 mAh/g over 2000 cycles at 1 A/g with self-healing behavior and improved Li⁺ diffusion kinetics [161]. Carbon-encapsulated EGaIn nanoparticles achieved 644 mAh/g after 800 cycles at 1 A/g, demonstrating the conductive encapsulation to enhance cycle life [162]. These studies highlight how liquid metals can maintain structural integrity and stable interfaces during cycling. However, challenges such as controlling liquid metal dispersion to avoid aggregation, optimizing mechanical properties without sacrificing strength and addressing cost and sustainability of Ga/In, sources remain significant challenges to commercialization.

2.7.2. MOF-Based and MOF-Derived Hybrid Composites

MOF-based and MOF-derived hybrid composites have emerged as versatile anode materials for lithium-ion batteries by combining high porosity, tunable pore architecture and abundant active sites to improve ion diffusion and electron transport. Parent MOFs provide controlled morphology and high surface area, but more commonly, MOF-derived materials produced via pyrolysis yield porous carbon matrices embedded with uniformly dispersed metal/metal oxide nanoparticles that enhance conductivity and structural stability. As for example, a CoMOF-derived Si@C core-shell composite exhibited a reversible capacity of 1493 mAh/g after 400 cycles and 648 mAh/g after 1200 cycles at 4 A/g, owing to uniform Si distribution and a robust carbon shell that buffers volume changes [163] and MOF-derived MnO/C composite delivered a high specific capacity of 1295 mAh/g at 500 mA/g after 300 cycles with significant retention [164]. Heteroatom-doped carbon encapsulated Cu₂S nanoparticles from MOF precursors achieved 513 mAh/g after 1000 cycles at 1 A/g, due to enhanced interface and conductivity [158]. Additionally, ZnO nanoparticles wrapped in N-doped carbon on CNTs from ZIF-derived MOF/CNT hybrids achieved 850 mAh/g at 100 mA/g with strong cycle stability [165]. Despite these promising results, challenges remain in controlling active site uniformity, optimizing synthesis for scalable production and improving initial Coulombic efficiency for practical applications.

2.7.3. MXene-Based Hybrid Anodes

MXene-based hybrid anodes have gained significant attention for lithium-ion batteries due to their exceptional electrical conductivity, hydrophilicity, tunable surface chemistry and ability to form interconnected conductive networks to enhance electron transport and ion accessibility. MXenes combined with high-capacity materials such as silicon and metal oxides help buffer volume expansion and improve cycling stability. For example, Si/MXene composite papers achieved a high

reversible capacity of 2118 mAh/g at 200 mA/g after 100 cycles, with excellent rate performance of 1672 mAh/g at 1000 mA/g and 890 mAh/g at 5000 mA/g, owing to the conductive layered of MXene's framework that accommodates silicon expansion [166]. MXene-metal oxide hybrids also show notable improvements: a SnO₂@MXene composite anode delivered 678 mAh/g at 2.0 A/g over 500 cycles due to synergistic conductivity and mechanical buffering from the MXene host [167]. Additionally, 3D hybrid structures such as SiNPs anchored on carbon foam coated with MXene demonstrated 1658 mAh/g after 100 cycles at 0.1C, retaining 857 mAh/g after 500 cycles at 0.5C, providing improved structural stability [159]. Despite these advantages, challenges such as MXene restacking, surface oxidation and controlling composite architecture persist, limiting ion accessibility and long-term stability.

2.7.4. Multi-Component Hybrids

Multi-component hybrid anodes combine three or more functional materials (carbon frameworks, metal/metal oxide nanoparticles, silicon or alloying elements, and conductive additives) to synergistically improve lithium-ion battery performance, while enhancing electrical conductivity, structural stability and lithium-ion transport. For instance, a ZnO/MnO@porous carbon nanofiber composite exhibited a specific capacity of 425 mAh/g at 5 A/g and maintained 315 mAh/g after 2000 cycles at 1 A/g, attributed to the 3D porous carbon network and dual metal oxide synergistic effects to mitigate structural collapse [168]. Ternary Si@C-CNT/carbon sheet composites achieved discharge capacities up to 2982 mAh/g at 0.5 A/g with 1488 mAh/g after 300 cycles, where carbon nanotubes and sheets provided mechanical flexibility and improved conductivity to restrict silicon's volume expansion [169]. Additionally, Co₃O₄-ZnO-g-C₃N₄-GO-Ag hybrid nanomaterials maintained stable capacity over 500 cycles at high current densities, demonstrating that multi-component hybrids with conductive carbon and heteroatom dopants can enhance rate capability and cycling stability [170]. Key challenges include precise control of component distribution, interfacial compatibility and scalable synthesis for practical deployment of multi-component hybrid anodes in next-generation LIBs.

2.7.5. Structurally Integrated Multifunctional Composite Anodes

Structurally integrated multifunctional composite anodes combine electrochemically active materials with conductive and mechanically resilient components to simultaneously address volume expansion, limited electronic conductivity, structural degradation, and unstable electrode interfaces. For silicon-based systems, integrating silicon with carbonaceous frameworks, conductive polymers, porous matrices, or mechanically adaptive networks can maintain electrical contact and accommodate repeated volume changes during lithiation and delithiation. Such multifunctional architectures can also provide shortened ion-diffusion pathways, improved electron transport, enhanced structural integrity, and more stable solid-electrolyte interphase formation. Their effectiveness therefore arises from the co-

ordinated functions of the active material, conductive network, and mechanically stabilizing framework rather than from electrolyte modification. Rational control of particle size, porosity, interfacial bonding, conductive pathways, and active-material loading is essential for achieving high capacity and long-term cycling stability.

Overall, the multifunctional composite anodes discussed in Sections 2.7.1-2.7.5 demonstrate how structural adaptability, electronic conductivity, interfacial engineering, and ion-accessible architectures can address major limitations of conventional anodes. Future development should prioritize high active-material loading, scalable fabrication, structural durability, cost-effectiveness, and stable electrode interfaces under practical operating conditions. Solid-state electrolytes, gel-polymer electrolytes, lithium plating, and electrolyte-specific transport properties are excluded from these sections unless they directly provide evidence for the performance of the composite anode itself.

3. Overview of Various Composite Anode

Table 5 summarizes representative anode materials and composite strategies developed to balance specific capacity, cycling stability, and rate capability in lithium-ion batteries (LIBs). Graphite remains the dominant commercial anode because of its low cost, abundance, stable cycling, high initial Coulombic efficiency (ICE), and mature manufacturing infrastructure. However, its theoretical capacity is limited to $372 \text{ mAh}\cdot\text{g}^{-1}$, and lithium plating may occur during high-rate charging or low-temperature operation [171]. Nanostructured silicon offers a substantially higher capacity of approximately $3600 - 4200 \text{ mAh}\cdot\text{g}^{-1}$ but undergoes 300% - 400% volume expansion, particle pulverization, unstable solid-electrolyte interphase (SEI) formation, and rapid capacity fading [172] [173]. SnO_2 has a theoretical capacity of approximately $1491 \text{ mAh}\cdot\text{g}^{-1}$, although large volume changes, mechanical degradation, and low ICE restrict its practical application [97]. Graphene-based anodes offer high electrical conductivity, tunable interlayer spacing, and capacities approaching $744 \text{ mAh}\cdot\text{g}^{-1}$, but their performance can be limited by sheet restacking [174]. Mn_3O_4 /graphene hybrids can deliver approximately $900 \text{ mAh}\cdot\text{g}^{-1}$, although conversion-reaction hysteresis and structural instability remain concerns [175]. Lithium metal provides an exceptional theoretical capacity of $3860 \text{ mAh}\cdot\text{g}^{-1}$ but presents serious dendrite-formation and safety risks [176]. LTO supports rapid charging and stable cycling but has a relatively low capacity of $175 \text{ mAh}\cdot\text{g}^{-1}$ and reduces full-cell energy density because of its high operating potential. Sn-Sb and Sn-Cu alloys provide intermediate capacities of approximately $600 - 800 \text{ mAh}\cdot\text{g}^{-1}$ but experience volume changes, irreversible reactions, and capacity fading [177].

Among composite systems, incorporating carbon into NiO improves electrical conductivity and structural integrity, whereas Ni-Co mixed-oxide/rGO hybrids achieve higher capacities through synergistic redox activity and improved accommodation of volume changes. However, their long-term cycling performance re-

quires further investigation. Binder-free MnO_2/CNT fabrics demonstrate excellent durability, retaining approximately 97% of their capacity after 1500 cycles at a high current density because of their mechanical robustness and strong integration with the current collector. Ag-decorated SnO_2/NiO hollow nanotubes combine internal void space with improved conductivity, thereby enhancing lithium-storage kinetics and reversibility; nevertheless, noble-metal cost and synthesis complexity may restrict scalability. Liquid-metal/Si composites provide high ICE, adaptive interfaces, and improved accommodation of silicon expansion, while $\text{SiO}/\text{C}/\text{graphite}$ composites generally achieve greater cycling stability through carbon and graphite buffering, although at the expense of specific capacity.

Table 5. Comparative overview of anode materials for LIBs.

Materials	Capacity (mAh/g)	Advantages	Limitations
Graphite (G) [171]	372 (theoretical for LiC_6)	Abundant, low-cost, stable cycling, well-established	Low capacity limits energy density, Li plating risk at high rates/low temperature
Si (nano-structured) [172] [173]	3600 - 4200 (pristine Si up to 4200), practical depends on design	Extremely high capacity, abundant	300% - 400% volume expansion, pulverization, poor cycle life unless nanostructured/composited
Tin oxide (SnO_2) [97]	Theoretical 1491, practical 650 - 1000	High theoretical capacity, operates at safe potentials	Severe volume change, particle pulverization, low ICE%
Gr-based (2D carbon) [174]	744 (few-layer Gr, composite designs often >372 vs G)	High conductivity, tunable spacing, buffers volume changes	Lower capacity than alloy/conversion types, restacking, experimental maturity
Mn_3O_4 , Gr hybrid [175]	900 (optimized $\text{Mn}_3\text{O}_4/\text{Gr}$ hybrids)	High capacity, environmentally benign	Conversion mechanism hysteresis, capacity fade, unstable structure
Lithium metal [176]	3860 (1 g Li stores 3860 mAh)	Ultimate capacity, lowest potential	Dendrites, unstable SEI, safety risks
Lithium Titanate (LTO, $\text{Li}_4\text{Ti}_5\text{O}_{12}$) [177]	175, energy density 177 Wh/L typical	Fast charging, long cycle life, dendrite-free spinel	Low voltage plateau (2.4 V) reduces energy density, higher cost

Comparisons based only on gravimetric capacity and cycle number may overestimate the practical potential of advanced anodes. Commercially relevant evaluation should also consider areal mass loading, areal capacity, electrode density, ICE, prelithiation requirements, electrolyte quantity, and full-cell performance. High capacities obtained using low mass loadings or highly porous electrodes may not translate into high volumetric or cell-level energy densities. Similarly, low ICE consumes cyclable lithium and reduces full-cell capacity, making controlled prelithiation particularly important for silicon-rich and other high-capacity anodes. Practical testing should therefore employ realistic electrode loadings, sufficiently dense electrodes, limited electrolyte quantities, and appropriate negative-to-positive electrode capacity ratios. Performance demonstrated in lithium-metal half-cells should ultimately be validated in full cells using practical cathode loadings and electrolyte-to-capacity ratios.

Overall, **Table 5** demonstrates that high specific capacity alone is insufficient to

establish commercial viability. The meaningful assessment of next-generation anodes, particularly Si-C composites, requires simultaneous consideration of areal capacity, electrode density, ICE, prelithiation, electrolyte consumption, full-cell performance, cycling durability, manufacturability, scalability, and cost. These metrics provide a more rigorous basis for determining whether laboratory-scale improvements can be translated into commercially competitive LIB technologies.

4. Challenges and Prospects

Composite anode materials, while promising for next-generation lithium-ion batteries, face several critical challenges that must be addressed to realize their full potential. Carbon-based composites suffer from limited theoretical capacity and relatively slow Li-ion diffusion that restricts their energy density and rate capability. Silicon-based composites, despite their extraordinarily high capacity, experience severe volume expansion during lithiation, leading to particle fracture, unstable solid electrolyte interphase (SEI) and rapid capacity fading. Incorporating metals or metal oxides with silicon can buffer some of these effects, but the complexity of synthesis, partial irreversible reactions and interface instability still pose significant obstacles. SiO_x composites offer improved stability but at the cost of lower capacity and complex electrochemical reactions that reduce initial Coulombic efficiency. Metal-organic framework (MOF)-derived composites bring tunable porosity and high surface area, yet their synthesis is often complicated and initial efficiency suffers from side reactions. Alloy and intermetallic composites such as those based on Sn, Sb, or Bi face mechanical degradation due to volume changes and phase transformations that impact cycling stability and rate performance. Transition metal oxides and sulfides offer high capacity via conversion reactions but struggle with low conductivity, voltage hysteresis, polysulfide dissolution, and capacity fading. Phosphorus-based composites exhibit similar volume expansion issues as silicon and poor conductivity, limiting their practical use. Organic and polymeric composites are attractive for their flexibility and sustainability but suffer from poor conductivity, dissolution in electrolytes and limited cycle life. Lithium-metal host composites promise the highest capacities but are plagued by dendrite growth, safety concerns, and challenges in scalable host fabrication. Liquid metal composites provide self-healing properties but face high cost and material aggregation problems. MXene-based hybrids deliver excellent conductivity and fast kinetics but must overcome restacking and stability issues. Finally, multi-component hybrids harness synergistic effects from multiple materials but are challenged by synthesis complexity, interfacial compatibility and reproducibility. Addressing all these challenges through advanced material design, novel synthesis methods, and interface engineering remains crucial for advancing composite anodes toward commercial viability.

The future development of composite anode materials for lithium-ion batteries is poised to address the critical challenges of energy density, cycling stability and safety through innovative material design and engineering. One promising direc-

tion lies in the synthesis of multifunctional composites that integrate high-capacity elements like silicon or phosphorus with conductive carbon matrices, enabling enhanced mechanical resilience and improved electron/ion transport. Advanced nanoarchitectures such as core-shell structures, 3D porous frameworks and hierarchical composites are expected to effectively mitigate volume expansion and stabilize the solid electrolyte interphase (SEI), thereby prolonging cycle life. Emerging materials like MXenes and metal-organic framework (MOF)-derived composites offer exceptional conductivity and tunable porosity, showing great potential for boosting rate capability and facilitating rapid lithium diffusion. Furthermore, the integration of liquid metal components could impart self-healing properties, enhancing the durability of high-capacity anodes. Progress in scalable and eco-friendly synthesis methods, coupled with computational modeling and machine learning approaches, will accelerate the rational design and optimization of composite anodes. Importantly, pairing these composites with solid-state electrolytes holds promise for safer batteries by suppressing lithium dendrite formation and enhancing interfacial stability. Overall, continued interdisciplinary research and innovation will be essential to realize next-generation composite anode materials that meet the rigorous demands of electric vehicles, portable electronics and grid-scale energy storage.

5. Conclusions

The accelerating deployment of renewable energy systems has intensified the demand for high-performance and sustainable energy-storage technologies. Among next-generation anode materials for lithium-ion batteries (LIBs), silicon-carbon (Si-C) composites are particularly promising because they combine silicon's high theoretical capacity with the electrical conductivity and structural resilience of carbon. Carbon matrices can mitigate key limitations of silicon, including severe volumetric expansion, low intrinsic electrical conductivity, and unstable solid-electrolyte interphase (SEI) formation, providing a pathway toward high-energy-density and long-cycle-life LIBs.

This review critically examines recent advances in Si-C composite anodes, emphasizing synthesis strategies, structure-property relationships, electrochemical performance, and sustainability. Other anode classes, including metallic, intermetallic, conversion-type transition-metal oxides, phosphorus-based, organic, and multidimensional composites, are considered selectively as comparative benchmarks. Particular attention is given to void-engineered, yolk-shell, porous and hierarchical architectures, MXene-containing composites, MOF-derived structures, and graphene- and carbon nanotube networks designed to accommodate volume changes, enhance charge transport, and stabilize electrode interfaces.

Importantly, sustainability is evaluated using specific criteria rather than electrochemical performance alone. These include the origin and availability of silicon and carbon precursors, particularly opportunities to utilize biomass-, waste-, or recycled-material-derived feedstocks; energy demand associated with synthesis,

including high-temperature treatment, milling, and multistep processing; solvent consumption and toxicity; material yield and process efficiency; and the potential for scalable, low-waste manufacturing. Consideration is also given to material recoverability, recycling compatibility, and end-of-life management, including the potential environmental burden and resource recovery from spent Si-C electrodes. These criteria enable comparison of whether improved electrochemical performance is achieved without disproportionate increases in energy, material, or environmental costs.

The review therefore assesses Si-C architectures across capacity retention, rate capability, Coulombic efficiency, mechanical integrity, electrode density, scalability, cost, and sustainability. Key research gaps and future directions are identified to promote resource-efficient synthesis, responsible precursor selection, improved material recovery, and manufacturing-compatible electrode designs. Overall, this integrated performance-sustainability framework provides a more rigorous basis for evaluating Si-C composite anodes and guiding their development toward durable, scalable, and environmentally responsible next-generation LIBs.

Author Contributions

Hasanuzzaman Aoyon: Conceptualization, Methodology, Investigation, Data curation, Formal analysis, Visualization, and Writing original draft. Himangshu Bhowmik: Conceptualization, Methodology, Supervision, Project administration, Validation, and Writing, review and editing. Mohammad Asaduzzaman Chowdhury: Methodology, Validation, Formal analysis, and Writing, review and editing. Mandira Bhowmik: Literature review, Visualization, review and editing. Shaon Talukdar: Conceptualization, Methodology, Supervision, Validation, and review. Md Masud Rana: Investigation, Data curation, Formal analysis, and Visualization. Md Mostafizur Rahman: Validation, review and editing. All authors reviewed and approved the final version of the manuscript.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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List of Abbreviations

A/g—Current density	LiBOB—Lithium bis(oxalato)borate
BP—Black phosphorus	LiPF ₆ —Lithium hexafluorophosphate
C—Carbon	mAh/cm—Areal capacity
C-rate—Charge/discharge rate	mAh/g—Gravimetric specific capacity
CE—Coulombic efficiency	MOF—Metal-organic framework
CNT—Carbon nanotubes	MOx—Metal oxides
COF—Covalent organic framework	NiO—Nickel oxide
CVD—Chemical vapor deposition	MSx—Metal sulfides
EC—Ethylene carbonate	NG—Nanographene
Fe ₃ C—Cementite or iron carbide	N/P—Negative/Positive capacity ratio
Fe ₃ O ₄ —Magnetite or black iron oxide	PAA—Polyacrylic acid
G—Graphite	PVDF—Polyvinylidene fluoride
GO—Graphite oxide	PVA—Polyvinyl alcohol
Gr—Graphene	rGO—Reduced graphite oxide
GrO—Graphene oxide	rGrO—Reduced graphene oxide
C@SiP—C-coated Si-P composite	red-P—Red phosphorus
ICE—Initial Coulombic efficiency	SEI—Solid-electrolyte interphase
Li—Lithium	Si—Silicon
Li ⁺ —Lithium ion	Si@—Silicon encapsulated
Li ₂ CO ₃ —Lithium carbonate	TM—Transition metal