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# Overcoming Interfacial Bottlenecks in Commercial Hard Carbon Anodes: SEI Dynamics, Pre-sodiation Strategies, and Safety Protocols for High-Energy Sodium-Ion Batteries

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**Abstract — One of the most promising anode materials for high-energy sodium-ion batteries (SIBs) is hard carbon (HC); however, low initial Coulombic efficiency (ICE), unstable solid electrolyte interphase (SEI) formation, irreversible sodium loss, and sodium-plating risks at low potentials continue to limit its commercial implementation. The interfacial mechanisms controlling HC performance are critically examined in this paper, which also assesses methods for getting beyond these restrictions in realistic full-cell systems. The effects of surface imperfections, specific surface area, and open microporosity are highlighted in the link between HC microstructure, electrolyte breakdown, SEI composition, and sodium-ion transport. In addition, surface and artificial interphase engineering through atomic layer deposition, chemical vapour deposition, thermal-pyrolysis carbon coatings, and conductive protective layers is discussed for suppressing parasitic reactions and stabilising the interface; the mechanisms and operational triggers of sodium plating, such as high-rate charging, low temperature, and mass-transfer polarisation, are also examined; and finally, operando characterisation, scalable manufacturing, thermal management, and techno-economic considerations are identified as critical paths for converting HC-based SIBs into safe, long-lasting, and commercially viable high-energy cells**

**Keywords— Hard carbon, Sodium-ion batteries, Solid electrolyte interphase (SEI), Presodiation, Sodium plating, Interfacial engineering.**

## I. INTRODUCTION

The common non-graphitic material used in anodes of industrial full cells is hard carbon (HC), and the commercial implementation of high-energy sodium ion batteries (SIBs) would require replacement of the less common lithium with the abundant sodium [1, 2]. Nevertheless, the low initial Coulombic efficiency (ICE) of typically 70%–85% severely restricts the transition from lab-scale half-cells to commercial full cells, thus causing substantial irreversible loss of active sodium, preventing the return of ions and limiting the cell energy density [3, 4]. The disorderly structure of the hard carbon containing turbostratic graphene sheets, surface defects, and abundant pore spaces within the structure, facilitates the formation of an undesirable reduction of the electrolyte on the initial stages of sodiation [5, 6].

An SEI which is made up of organic alkyl carbonates as well as ionically conducting inorganic layers such as Na2CO3 and NaF forms heterogeneously at the interface between the electrode and the electrolyte, where the solvation sheath structures govern the initial decomposition of organic carbonate and ether solvents [7]. Conventional electrolytes using carbonates form a thick and brittle SEI which breaks repeatedly during cycling, leading to long-term electrolyte degradation as well as impedance increase according to operando analysis [8, 9]. The SEI formation can be engineered through the manipulation of the solvation sheaths and by adding additives in the electrolytes such as difluoro(oxalato)borate salts and fluoroethylene carbonate (FEC), which decomposes preferentially to form a robust, LiF-rich, low-impedence protective layer [10, 11]. Furthermore, in conjunction with the promotion of inorganic SEI layers from anions, the change of electrolytes from conventional carbonate-based electrolyte solutions to either ether-based or high-concentration local electrolyte solutions helps to facilitate fast interphase kinetics while ensuring closed-pore sodium deposition [12, 13]. In addition to surface chemistry, decreasing the open surface area whilst retaining the reversible low-voltage plateau capacity needs the manipulation of closed porosity and carbon precursor pyrolysis temperature [14]. The high energy edge defects can be effectively passivated through microstructure optimization methods such as chemical vapor deposition coating and heteroatom doping (N, P, S), which increase the ICE to commercially acceptable levels above 90% [15]. This is essential for any scalable full cell design to find an accurate relationship between the hard

carbon microstructure properties and SEI passivation efficiency [16, 17].

Pre-sodiation methods have proved to be crucial for the commercial implementation of SIB technology, preventing active sodium loss without any reduction in the capacity of the cathode [18, 19]. Prior to cell assembly, chemical pre-sodiation, which involves the use of sodium-polycyclic aromatic hydrocarbon complexes such as sodium biphenyl and sodium naphthalenide, leads to inject sodium into hard carbon at an ultra-fast rate, resulting to a rise in ICE up to 100% and the energy density of the full cell [20]. In addition, the additives of the cathode that can decompose during the first charge and deliver extra sodium ions (Na+) to the electrolyte pool include Na2C2O4, Na2O, and organometallic sodium salts [21].

Electrochemical and direct contact presodiation methods have proven to be equally precise in establishing a desired level of sodium loading [22]. The thermal stability and failure modes of hard carbon anode materials during extreme conditions of operation constitute an important safety aspect along with interfacial capacity rejuvenation [23, 24]. Thermal instability and runaway of sodiated hard carbon occur due to the exothermic decomposition of unstable SEI film at temperatures ranging from 80 to 120 degrees Celsius [25]. Further reactions include reactions of retained sodium within graphitic interlayer cavities and organic solvents. Localized sodium plating at the hard carbon anode may lead to a violent outburst of gases and extreme internal short circuiting due to overcharging or high currents, leading to extremely high temperatures more than 600 °C [26, 27]. Nonflammable fluorinated solvents, ionic liquid additives, and self-healing artificial electrolyte layers are used in combination to increase the threshold temperature of SEI decomposition above 200 °C while avoiding the formation of exothermic gases to develop safe protocols [28]. This is now feasible to detect interfacial decomposition prior to failure because of thermal management systems in combination with acoustic/thermal sensing and BMS overcharge protection [29, 30]. Finally, these technical barriers can be addressed through low-cost precursor selection, scalable chemical or additive-based presodiation, and intrinsically safe electrolyte interphase systems, paving the way for commercial use of high-energy sodium-ion batteries in grid energy storage and electric vehicle applications [13, 31].

The presence of severe instability at the interface during early and subsequent cycles greatly impedes the utilization of hard carbon anode for high-energy density sodium-ion batteries [32, 33]. Formation of a composite and heterogeneous solid electrolyte interphase (SEI) film arises from the electrochemical breakdown of ester- and ether-type electrolytes at very low potential ranges approaching 0V vs Na/Na+ [34-36]. The SEI film forms and dissolves continuously because of the high solubility and low mechanical stability of the sodium salts forming the interphase when compared to the corresponding lithium salts resulting in irreversible loss of active sodium and consumption of the electrolyte [37-39]. In addition, the slow process of Na+ ion desolvation and diffusion leads to overpotential, resulting in localized deposition of metallic sodium at 0 V [40-42].

Despite much work on the topic, the existing literature mainly focuses on specific approaches, including special coatings, binder modification, or local electrolyte addition [43-45]. Hence, a substantial knowledge gap still exists regarding a general concept of the link between the processing of electrode materials and the cell performance in terms of atomic-level interface behavior [23, 46]. The present review fills this gap by providing a holistic view on the connection of fundamental SEI dynamics, scaling presodiation process of the whole cell and safety operation criteria for practical high-energy sodium-ion batteries [24].

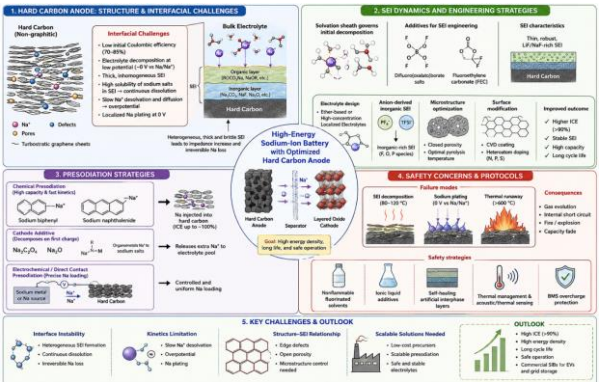


Fig 1. Integrated Framework for Overcoming Interfacial, Kinetic, and Safety Challenges in Hard Carbon Anodes for High-Energy Sodium-Ion Batteries

## II. SOLID ELECTROLYTE INTERPHASE (SEI) MECHANISMS ON HARD CARBON

### 1. Thermodynamics and Kinetics of SEI Formation

Lowering of the reduction potential ofx the electrochemical cell below the energy levels of LUMO of organic solvents and sodium salts [47-49] is the main driver behind the thermodynamics of SEI formation on hard carbon electrodes. This leads to rapid and nonspontaneous electron transfer causing decomposition of electrolyte components at low potential of operation [50, 51]. At the same time, the microstructure properties of carbon host material affect the

kinetic aspects of SEI nucleation and growth [52-54]. During the first sodiation step, high SSA (specific surface area) accelerates continuous decomposition of the parasitic electrolyte [6], while open micropores and defects of the edges of the carbon particle create high-energy reactive sites for permanent entrapment of Na+ ions [55, 56]. For limiting initial irreversible capacity loss and forming a stable and ionic conducting interphase structure for practical applications of sodium-ion batteries, it is very important to limit the open porosity and defective surface structures and retain closed internal porosity [15, 57, 58].

**2. Composition and Morphological Evolution**

Models using mosaic or bilayer structures consisting of an outer organic-rich phase, made up of sodium alkyl carbonates, and an internal inorganic-rich layer composed of Na2CO3 and NaF have been commonly utilized to characterize the composition and evolution of SEI films on hard carbon electrodes [59-61].

The interphase film created by the ester electrolyte is normally organic-rich with high solubility of sodium salts, rapid dissolution kinetics, and mechanical degradation upon repeated cycling of the electrochemical process [62-64]. In contrast, the ether electrolyte leads to a mechanically flexible and thinner inorganic-rich interphase film formation, exhibiting slower dissolution kinetics and activation energy barrier for ion transfer across the interface being reduced significantly [65-67]. Thus, to form a coherent, flexible, and chemically robust interphase film which prevents the continuous consumption of electrolyte is a challenge [68-70].

**3. Degradation and Self-Discharge Mode**

Degradation of interphase through thermal instability, SEI formation, and chemical dissolution are some of the primary reasons for self-discharge and which considerably affect the overall stability and lifetime of hard carbon anode-based high-energy sodium-ion battery systems [68, 71].

Higher temperatures promote exothermic reactions of organic compounds in the interphase, which cause further degradation of the electrolyte and increase the parasitic losses on the surface of the sodiated carbon [52, 72, 73]. SEI growth is caused by solvent diffusion through microcracks in the interface layer and further electron tunnelling through the thinning passivating layers [74, 75]. Besides that, the enhanced solubility of the sodium salts in organic carbonates facilitates the constant dissolution of the SEI, leading to spontaneous desodiation from hard carbon layers and closed pores, resulting in accelerated self-discharge and permanent reduction in capacity.

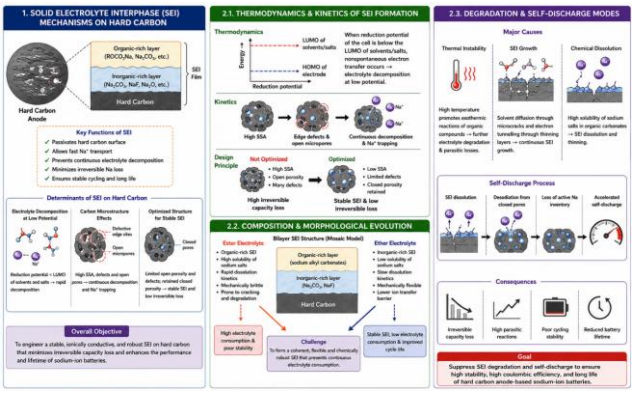


Fig 2. Comprehensive schematic illustration of SEI formation, evolution, degradation, and self-discharge mechanisms on hard carbon anodes for sodium-ion batteries.

## III. PRESODIATION TECHNOLOGY FOR ACTIVE NA+ LOSS COMPENSATION

**1. Chemical Presodiation**

Prior to assembly of cells, presodiation via the use of organometallic complex solutions, such as sodium biphenyl, sodium naphthalenide, and sodium alkoxyaluminum hydrides, enables fast and controllable transfer of electrons and sodium ions into the hard carbon anode. In this way, surface defects can be passivated, while an inorganic-dominated solid electrolyte interface (SEI) is pre-formed, resulting in maximization of initial Coulombic efficiency (ICE) to 100% [20, 76, 77]. At the same time, electrochemical forces due to short circuiting are employed to provide an exact amount of active Na+ to the carbon host through the direct contact presodiation technique utilizing physical pressure application and thin films of sodium metal [22, 78]. In the case of scalable high-energy sodium-ion full cells, the absence of solvent contamination and reduced hazards in the process due to direct contact methodologies provide enhanced reaction uniformity and rapid kinetics [79-81]. This provides a supplementary processing method to make up for the initial sodium consumption.

**2. Electrochemical Presodiation**

The hard carbon anodes may be electrochemically presodiated through either direct internal short-circuiting with respect to a sodium metal sacrifice or careful galvanostatic pre-cycling within an auxiliary half-cell configuration. Such processes facilitate controlled presodiation extent along with simultaneous interphase development [19, 22, 76, 78]. While the auxiliary cell pre-cycling technique provides excellent controllability over the cut-off voltage and presodiation extent, it suffers from excessive electrolyte wastage during cell dismantling, costly capital investment, and considerable

manufacturing difficulty [19, 82, 83]. While the internal short-circuiting arrangement ensures an easier and cheaper operation route, it suffers from risks associated with thermal management and current density inhomogeneity [81]. However, slow kinetics of ion transport, tolerance for precise alignment, sensitivity to air contact, and safety hazards associated with the use of large quantities of reactive pre-sodiated carbon films pose serious scalability restrictions for both of these electrochemical methodologies used in continuous roll-to-roll fabrication lines of electrodes [84, 85].

### 3. Sacrificial Cathode/Anode Additive

The first charge cycle witnesses the irreversible oxidation of organic salts such as sodium oxalate Na2C2O4 as well as cathode and anode presodiation additive materials such as inorganic Na2O and Na3N, which help in providing additional Na+ ions into the active material storage to directly address the consumption of hard carbon SEI without complex pre-assembly of the cell [86-88]. While the oxidation products of oxide-containing materials such as Na2O are solid in nature and need to be properly balanced to avoid metal dissolution and electronic isolation, other materials such as Na2C2O4 and Na3N form gaseous products (CO2 and N2, respectively) in the electrochemical activation process that needs to be carefully degassed [89, 90]. It remains critical for maintaining structural stability and efficient initial Coulombic efficiency of commercial sodium-ion full-cells [79, 86]

### 4. Comparative Benchmarking

A number of presodiation approaches provide a controlled compromise between process difficulty, ICE enhancement, and industrial scale-up challenges to overcome the problem of active Na+ ion loss due to SEI formation in industrial hard carbon anodes [84, 86, 87]. Chemical methods, involving anode exposure to organometallic compounds, show excellent capacity compensation and medium process difficulty, giving an increase in ICE of nearly 100%. However, they present significant industrial scale-up challenges in terms of chemical stability, reactant behavior, and organic solvents safety [91]. Spontaneous redox reactions, together with direct physical presodiation involving metallic sodium foil or powder, provide a moderate-to-high ICE improvement level [92, 93]. Nevertheless, the complicated process of implementation makes it dangerous for use due to safety and fire risks [94, 95]. Sacrificial cathode additive, on the other hand, has relatively simple process due to easy integration into conventional slurry mixing and coating processes [96, 97]. Nevertheless, its capacity to enhance ICE level is moderate and is limited by excess weight and potential gas evolution as a result of irreversible electrochemical reaction.

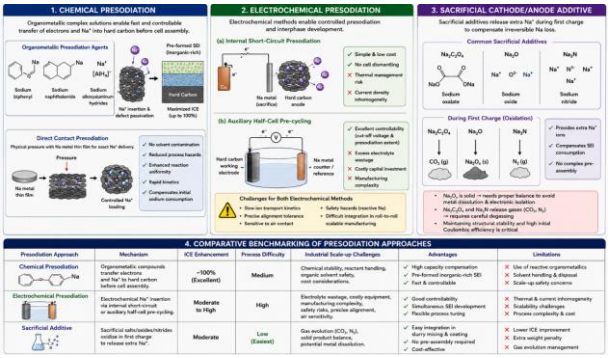


Fig 3. Comparative overview of presodiation strategies for hard carbon anodes in sodium-ion batteries: mechanisms, initial Coulombic efficiency enhancement, and practical challenges

## IV. SURFACE COATINGS AND ARTIFICIAL INTERPHASE PASSIVATION

### 1. Atomic/Molecular Layer Deposition (ALD/MLD)

One such highly controllable technique is ALD/MLD (atomic and molecular layer deposition), in which conformal layers of materials such as Al2O3 and TiO2 may shield the reactive sites at the interface, inhibit continuous electrolyte decomposition, and facilitate the formation of thinner and more stable inorganic-rich SEI films [48, 98-101]. Although PEALD $TiO_2$ layers provide excellent uniformity and low charge-transfer resistance upon $Na^+$ insertion, recently published papers have shown that the application of $Al_2O_3$ layers on the surface can increase initial Coulombic efficiency and cycle stability due to lower side reactions. In general, surface and interface modification has been recently recognized as one of the most effective techniques to address the problem of unstable SEI and poor initial Coulombic efficiency of HC [102-106]. Any future studies must focus on developing cost-effective deposition methods that retain the protective properties of the thin artificial SEI films due to the fact that, although ALD/MLD offers highly precise film thickness, conformality, and atomic layer deposition, the commercialization process is largely constrained by the use of specific hardware, repeat deposition cycles, chemical reagents, and high costs [107-109]

### 2. Chemical Vapor Deposition (CVD) And Thermal Pyrolysis Coating

Forming an amorphous/soft-carbon film that seals the exposed micropores, reduces the excessive surface reactivity, and inhibits electrolyte degradation and irreversible consumption of $Na^+$ ions at the beginning of the cycling process, CVD and thermal-pyrolysis coating provide efficient surface modification approaches for industrially relevant hard-carbon

(HC) electrodes [99, 101, 110-112]. The carbon coatings derived from pitch/asphalt materials are able to convert the open pores to closed pores and increase low-potential $Na^+$ storage [111, 113, 114], while CVD method using pyrolyzed products of acetonitrile is found to seal the open micropores and reduce the specific surface area of HC, thereby hindering excessive SEI formation and increasing the initial Coulombic efficiency [111]. Recent experimental studies have also indicated that nanoscale carbon coatings can enhance cycle stability and rate performance, stabilize the electrode/electrolyte interface, and reduce surface defects [115-117]. These results are in agreement with recent review articles that stress the importance of maintaining an appropriate balance between accessible surface area, pore opening/closing ratio, and reactivity at interfaces to improve the performances of HCs as well as the energy density [118-120]. On the one hand, too much carbon depositing on HCs could limit their available storage sites and the diffusion of $Na^+$ ions, whereas on the other hand, inadequate carbon coating would not be able to protect the reactive surfaces, hence controlling the thickness and uniformity of the coating is vital [115]

## V. RISK MITIGATION: SODIUM PLATING NEAR 0 V VS. NA/NA+

### 1. Physics of Low-Potential Anode Operation

The electrochemical window between the process of nanopore filling with sodium and quasi-metallic Na clusters and that of metallic Na nucleation is narrow due to the fact that the low-potential plateau of hard carbon (HC), typically observed below ~0.1 V relative to $Na/Na^+$, is an important requirement to achieve a high capacity of sodium storage and yet is located quite near the thermodynamic potential for $Na^+$ formation (0 V) [101, 105, 107, 121]. Recent studies have shown that the process of sodiation is responsible for the formation of quasi-metallic Na clusters within the confined space of the nanopores, which can later overcome the nucleation barrier and form metallic Na [26, 122]. These factors increase the risk due to the slowness of $Na^+$ diffusion and the development of concentration polarization, leading to the local anode voltage being driven close to 0 V. This means that while the optimization of low voltage plateau capacity must take into account the limit for safe plating of sodium, and not assume that all plateau capacity is reversible.

### 2. Operational Triggers for Plating

In cases where the charging condition creates substantial polarisation effects leading to the reduction in anodic potential towards 0 V vs $Na/Na^+$, there is a strong tendency for sodium plating at hard-carbon (HC) electrodes. This is particularly the case when the rate of charging is high enough that there will be a kinetic limitation of sodium ion diffusion into HC, thus increasing concentration gradient within the electrode [26, 105, 121]. Rapid charging may lead to increased polarisation through the charge transfer, ohmic, and mass transfer process [123]. Due to the effect of high values of concentration and activation overpotentials, which become even higher as a result of low electrolyte conductivity, slower $Na^+$ desolvation and diffusion, and high interfacial resistance, the temperature dependence poses an additional challenge to the safety of sodium metal batteries [124-126]. Moreover, mass transfer polarisation depends on electrode thickness, tortuosity, pore structure, N/P ratio, and electrolyte characteristics. It is also shown experimentally that under conditions of sodiation and long charging, initially stored or quasi-metallic Na clusters can become metallic Na deposits on the HC surface. Hence, all these factors must be addressed for effective prevention

### 3. Interfacial Strategies to Suppress Dendrite/Plating Growth

In order to prevent localized $Na^+$ accumulation and preferential deposition sites, interfacial engineering can suppress sodium plating on hard-carbon (HC) anodes by creating $Na^+$-conductive interphases that homogenize ionic flux and regulate the nucleation barrier at highly sodiated surfaces [26]. Recent artificial-interphase studies on sodium electrodes show that ion-conductive phases like NaCl, $Na_3P$, and sodium formate can reduce the critical nucleation overpotential and distribute Na nucleation more uniformly across the electrode surface. Surface-affinity engineering offers a complementary strategy because sodiophilic functional groups and defect sites can lower the critical nucleation overpotential.

This idea is especially pertinent to HC, where the low-voltage plateau can produce quasi-metallic Na inside nanopores prior to metallic Na nucleating externally. Operando X-ray studies have verified that plating is linked to highly sodiated regions and the formation of pseudo-metallic Na [127], and recent mechanistic research has identified nucleation and subsequent surface deposition as separate phases regulated by current density and interfacial conditions.

Therefore, a promising way to maintain uniform Na transport and increase the safety margin against plating during high-rate operation is to combine $Na^+$-conductive artificial interphases, controlled sodiophilic surface chemistry, optimized pore structures, and electrolyte-derived SEI formation.

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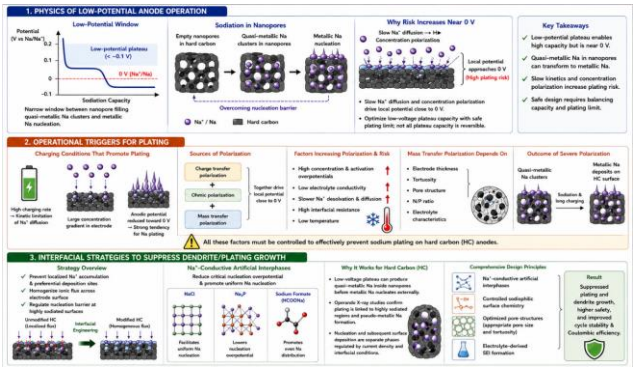


Fig 4 Mechanisms, Risk Factors, and Interfacial Engineering Strategies for Suppressing Sodium Plating on Hard Carbon Anodes in High-Energy Sodium-Ion Batteries

**Challenges and Limitations**

The lack of direct, real-time understanding of SEI formation, restructuring, and degradation under realistic operating conditions is a significant unresolved challenge for commercial hard-carbon anodes. While operando/in-situ TEM, X-ray scattering, NMR, XPS, and related methods have started to distinguish $Na^+$ insertion, nanopore filling, SEI evolution, and sodium plating, further development of cryogenic TEM and complementary spectroscopic techniques is necessary to capture highly reactive interphases without changing their native structure [128-130]. The usefulness of multimodal characterization for resolving these associated processes has already been demonstrated by recent in-situ TEM and operando NMR experiments, which have previously revealed plating, dendritic development, SEI composition, and quasi-metallic sodium storage [131]. Scalable interfacial treatments that provide quantifiable improvements in initial Coulombic efficiency, energy density, and cycle life without undermining the intrinsic cost advantage of SIBs will be necessary for commercialization, as advanced post-treatments like atomic layer deposition and presodiation must be justified at the manufacturing level against their additional equipment, processing, energy, and material costs [40]. While cell-level studies demonstrate that SIB cost competitiveness is still heavily dependent on increasing energy density and reducing the amount of HC, electrolyte, and other components required per kWh, techno-economic analyses suggest that biomass-derived HC could be produced at roughly \$1.58–1.72 $kg^{-1}$ at large scale.

## VI. CONCLUSION

In conclusion, regulating the electrode–electrolyte interface, making up for irreversible sodium consumption, and avoiding low-potential deterioration are critical to the commercialization of high-energy sodium-ion batteries utilizing hard carbon anodes. Because the high surface area, defects, and open micropores of hard carbon encourage parasitic electrolyte reactions, the formation and ongoing evolution of the SEI continue to be significant causes of low initial Coulombic efficiency, electrolyte consumption, impedance growth, and capacity loss. Surface reactivity can be decreased while maintaining the closed-pore storage responsible for high reversible capacity using techniques including electrolyte and solvation-shell engineering, pore-structure optimization, CVD and pitch-derived carbon coatings, and artificial interphases. Although chemical, electrochemical, and sacrificial-additive approaches still require improvements in safety, process control, cost, and manufacturing compatibility, presodiation offers an additional way to improve full-cell energy density and compensate for irreversible $Na^+$ consumption. Crucially, the hard-carbon plateau's close closeness to 0 V vs. $Na/Na^+$ establishes a tiny safety margin where metallic sodium plating can be encouraged by high-rate charging, low-temperature operating, and mass-transfer polarization. Therefore, robust Na-conductive interphases, regulated surface chemistry, operando characterization, scalable presodiation, and sophisticated thermal and battery-management systems must be integrated for future advancements. Transforming hard-carbon sodium-ion technology from laboratory-scale demonstrations into safe, long-lasting, and commercially viable high-energy cells would ultimately require combining interfacial engineering with manufacturability and techno-economic factors.

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