Application of nano-coated silicon-based materials in lithium-ion anodes

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Abstract:

In recent years, Silicon-based anode have became one of the most promising candidates for next-generation lithiumion batteries due to its superior electrical performance, materia; abundance and environmental friendly property. However, it still has so many disadvantages, such as volume expansion (~300%), unstable solid-electrolyte interphase (SEI), and rapid capacity fading hinder their commercialization. This paper evaluates three advanced nano-coating strategies, Si-C, Si-metal oxide (Al₂O₃), and Si-polymer (x-PAN), to address these limitations. First, Si@Graphene composites exhibit exceptional capacity (3578 mAh/g) and rate capability (975 mAh/g at 5 A/g) but suffer from graphene defects and complex synthesis, while Al₂O₃ coatings reduce SEI growth and lithium loss by 37%, yet their brittleness and insulating nature limit energy density. In contrast, crosslinked Si-polymer anodes achieve 76.2% capacity retention after 1,000 cycles and high-rate performance (1880 mAh/g at 50C), but face scalability challenges due to pinhole formation and costly processing. Such key findings and comparisons highlight trade-offs between performance, manufacturability, and cost. Hybrid designs (e.g., conductive/oxide multilayers) and scalable techniques (e.g., dry electrode processing) are recommended to bridge these gaps. This analysis provides actionable insights for developing commercially viable silicon anodes in high-energy lithium-ion batteries for electric vehicles.

Keywords: Si anodes; nano-coating; lithium-ion battery.

1. Introduction

Nowadays, the development of technology such as green energy and supported policy which push toward carbon neutrality accelerated the advancement of new energy vehicles (NEVs) [1]. Many types of

new batteries and technologies have been innovated. Among a variety of battery options, high-performance lithium-ion batteries (LIBs) play an important role, given to the fact that people expect it can deliver higher energy density, longer cycle life, faster charging capabilities, and greater safety, while still

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can handle the cost-effectiveness for large-scale commercial use. In this case, those demands drive the research into novel electrode materials, particularly at the anode. Right now, the graphite, as the most commonly used traditional anode material, has several limitations that hinder their performance in next-generation lithium-ion batteries. First, graphite anode has safety risk under fast charging. At low temperature, lithium may plate on the surface of graphite, which will lead to dendrite growth, increasing the risk of short-circuiting [2]. Second, it is incompatible with high voltage cathodes, such as nickel-rich layered oxides (e.g., NMC811) or lithium-rich metal oxides, which operate above 4.3 V vs. Li/Li⁺. This is because under this elevated voltage, the electrolyte becomes unstable [3]. Lastly, and the most important one, the graphite anodes are approaching their theoretical capacity limit (372 mAh/g), which constrains further improvements in battery performance. It is obvious to find that graphite alone cannot fulfill future energy storage requirements as EVs demand longer driving ranges and reduced charging times [4]. In this case, bottleneck of conventional anode materials becomes one of the most critical challenges in the development of LIBs for researchers.

Under this circumstance, the silicon-based material stands out and attract significant attention among world industry as next generation anode materials for lithium-ion batteries given to several reasons [2]. First, compared with graphite, the silicon-based material has a silicon boasts an ultra-high theoretical capacity of approximately 4200 mAh/g, which is about more than ten times the energy capacity. This property makes silicon be one of the most promising candidates for achieving higher energy density since it can reduce the weight for EVs and have longer range. In addition, the silicon is the second most abundant element in the Earth's crust, and is readily available worldwide unlike cobalt or nickel. Its extraction process and manufacturing process is well-established in semiconductor industry. Thus, those facts make silicon be really cost-effective for anode materials. Lastly, from environmental perspective It is less toxic compared to many transition metals used in batteries.

However, although silicon-based anode material brings out lots of benefits for using, it still has some limitations by several critical challenges [3]. First, and also the most prominent issue is that the silicon has significant volume expansion about 300% during lithiation, which might cause mechanical stress, and this stress would lead to pulverization of the electrode, loss of electrical contact, and rapid capacity fading over repeated charge-discharge cycles. Moreover, the multiple expansion might disrupt the solid electrolyte interphase (SEI). SEI is layers that is a thin, passivating layer that forms spontaneously on the

surface of the anode in lithium-ion batteries during the initial charge cycles. It can prevent reactions between the electrolyte and the anode, acting like a protective shield, so it would lead to unstable cycling and excessive consumption of electrolyte. All those issues would contribute to safety concerns.

In order to overcome those limitations, several strategies are used. Researchers found that nano-coating can get better performance. In specific, the surface of the silicon particles can be reinforced to accommodate mechanical stress while enhancing electrical conductivity. The coating can increase the surface area of electrode, allowing more uniform flux during cycling. This strategy really stabilizes the SEI and buffers the volume change for silicon-based anode. In addition, the nano-coating can also make anodes highly suitable for fast charging application which need high current density and structural integrity are required. For example, Kim et al. in 2021 developed a carbon coated Si nanoparticle and achieved 85 % capacity retention after 300 cycles at 1C rate, significantly outperforming uncoated Si anodes[5].

This paper discusses the current implementation strategies of various nano-coated silicon-based materials for use as anode materials in lithium-ion batteries for new energy vehicles (NEVs). The paper presents three specific case of different coating types- Si-C Coating Materials, Si-Polymer Coating Materials and Si-Metal Oxide Coating Materials, comparing their respective advantages and limitations from both material and synthesis perspectives, and concludes with tailored recommendations based on performance needs and scalability.

2. Case

2.1 Si-C

Si-C coating material refers to silicon particles encapsulated or conformally layered with carbon-based media, such as amorphous carbon, hard carbon, graphene sheets, or conductive carbon nanotube (CNT) networks. Si-C coating material has several functions. First, it has high intrinsic electronic conductivity, which can create an efficient percolation path that lowers the overall electrode resistance and improves rate capability. In addition, it has mechanically resilient carbon shell, acting as elastic buffer that absorbs the ~300 % volumetric expansion of silicon during lithiation. Third, this coating also serves as a semi-permeable barrier, so less contact between silicon surface and electrolyte, thereby reducing the growth of SEI.

2.2 Si-metal oxides

Si-metal oxide coating materials consist of silicon particles enveloped by nanometer-thick layers of oxides such as SiO₂, TiO₂, Al₂O₃, ZrO₂, or Li-conductive phosphates. The coatings for such metal are commonly fabricated though sol-gel condensation, atomic layer deposition (ALD), etc. Such technique can precisely control the thickness of layer. These inorganic functions as a chemically inert and mechanically rigid scaffold not only can restrict silicon pulverization but also can permit lithium-ion diffusion through inherent nanopores or engineered grain boundaries. In addition, given to physical blocking for the electrolyte penetration, the oxide shell can retard continuous SEI thickening and limit parasitic side reactions. As manufacturing costs for ALD and scalable sol-gel reactors decline, Si-metal oxide coatings are poised to play a pivotal role in commercial high-silicon anode architectures for long-life automotive and aerospace batteries.

2.3 Si-Polymer

Coating materials use elastomeric, ion-conductive, or redox-active polymers to encapsulate or cross-link silicon particles within a flexible matrix. Typical media include polyacrylic acid (PAA), polyimide, polyvinylidene fluoride-hexafluoropropylene (PVDF-HFP), conductive polyaniline derivatives, and self-healing polyurethane networks. The polymers are introduced through in-situ polymerization on silicon surfaces-layer by layer dip-coating, or solution casting followed by thermal cross-linking. The polymer network really accommodates silicon's expansion and contraction, preserving electrode integrity and electrical contact over hundreds of cycles and re-

ducing the growth of SEI. In addition, compared with inorganic coating such as Silicon Dioxide or Aluminum Oxide, polymer layers offer superior ductility, low density, and easy processability at low temperatures.

3. Analysis and Challenge

3.1 Si-C

In general, the synthesis of core-shell structured Si@ Graphene composites primarily involved electroless deposition and catalytic growth methods[6]. For starters, surface treatment of Si Nanoparticles was needed. In specific, in order to remove surface oxides about 0.5 Si nano-particles were dispersed in a mixture of HF(hydrogen fluoride), deionized water, and methanol. Then moving to Ni electroless plating, mixing 0.1 mol/L NiCl, 0.56 mol/L NH4Cl, 0.03 mol/L C₆H₈O₇, 0.3 mol/L NaH₂PO₂ to form the solution and adjusted to 8.5 P.H with NH₃·H₂O and coated at 50°C for 10 min. In next step, the Ni-coated Si need to disperse in triethylene glycol/NaOH solution and heated to 185°C about14 h then annealed at 450°C under Ar for 60 min to achieve Graphene Growth. Lastly, the Ni layer was completely etched using a HNO₃/HCl/HF mixture, yielding the final 3D Si@Graphene composite with uniform graphene wrapping as confirmed by TEM imaging. The Si@Graphene composites is evaluated through coin cells with an electrode composition of 60% active material, 20% acetylene black, and 20% sodium alginate binder, achieving a mass loading of 0.5 mg/cm² on Cu foil [6]. The procedure for the preparation of Si@Graphene is shown in Figure 1 below



Fig. 1 Schematic illustration shows the procedure for the preparation of Si@Graphene [6]

The Si@Graphene composite exhibited superior electrochemical performance compared to bare Si in the area of cycling stability, rate capability, and structural integrity. First, Si@Graphene composite has high initial capacity&cycling stability with the following supported data: the initial discharge capacity is about 3578 mAh g-1 and after 100 cycle, it still has 1909 mAh g⁻¹. The Coulombic efficiency is about 76.9%,stabilizing at >99% after 10 cycles. Secondly, Si@Graphene composite has excellent rate capability. It has 2757 mAh g⁻¹ at 0.2 A g⁻¹, 2148 mAh g⁻¹ at

1 A g⁻¹ and 975 mAh g⁻¹ at 5 A g⁻¹ while pure Si only has 80 mAh g⁻¹. Third, it has Suppressed Volume Expansion & Stable SEI. In specific, after 50 cycles, Si@Graphene composite electrode swelling: +17.8 µm about 160%, while pure Si has +29.8 µm (300%) . The SEI increase from 40.06 Ω to 49.45 Ω after 30th–50th cycles,while pure Si surges from 78.40 Ω to 161.70 Ω [6].

The Si@Graphene composite synthesis exhibits intrinsic limitations across multiple dimensions. Structurally, the thin graphene coating (~4.74 nm) contains defects (ID/

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IG=0.16) and incomplete Ni etching, which compromise mechanical stability and electrical conductivity[6]. Chemically, oxygen functional groups on graphene and trapped Li+ in defects lead to irreversible capacity loss (initial CE=76.9%) [7]. Process-wise, the multi-step synthesis (electroless plating, catalytic growth, acid etching) presents scalability challenges, while hazardous HF etching raises environmental concerns [8]. Furthermore, the nanoscale architecture demands exceptional environmental stability, restricting practical applications under extreme conditions [9]. These combined material and manufacturing constraints highlight the need for alternative catalysts, defect engineering, and greener processing methods to realize the composite's full.

3.2 Si-metal oxides

In general, the preparation of Al₂O₃-coated silicon thin films primarily involved magnetron sputtering. For the first part of the synthesis, it start with the Silicon Thin Film Deposition.200 nm Si films were deposited on smooth Cu foil via magnetron sputtering. Finally, Al2O-3coatings were deposited via reactive sputtering with an Ar/O₂ (1:1) mixture at 5×10^{-3} mbar using 80 W DC power (The coating thickness controlled 1.5 nm (30s), 3 nm (60s), and 5nm (90s). For testing, it has two cell configuration:Li||Si half-cells with Si/Al₂O₃ as the working electrode and Li foil as the counter electrode, and Si||LFP full-cells with Si/Al₂O₃ as the anode and LiFePO₄ as the cathode. The cycling setup is 3 cycles at 0.1s in formation and 97 cycles at 0.5C (voltage ranges: 0.05-1 V for halfcells, 2.5–3.7 V for full-cells) for Aging [10]. The SEM of pristine silicon thin film as show in Figure 2[10].

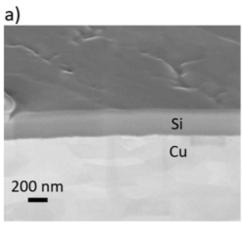


Fig. 2 FIB prepared cross section SEM image of the pristine silicon thin film. A dense structure with no visible porosity can be observed [10]

The Al₂O₃-coated silicon thin-film electrodes had signifi-

cant improvement compared with pure silicon in cycling stability and lithium inventory retention. First, it reduced capacity fade. In Si||LFP full-cells, the 5 nm Al₂O₃ coating (SiA5) reduced capacity fade per cycle by 64% (from 14 mAhg⁻¹ for bare Si to 5.4 mAhg⁻¹), which significantly enhancing long-term cycling stability[10]. Secondly, it preserve lithium inventory, the loss of lithium inventory (LLI) decreased by 37% in SiA5||LFP cells (from 90 μ Ah to 57 μ Ah), attributed to suppressed SEI growth. Moreover, given to the fact that the Al₂O₃ coating fractured during initial lithiation, it eliminate over potential in subsequent cycles. Compared with carbon coating, Al₂O₃ outperformed 40 nm carbon coatings in reducing LLI (37% vs. 22%), though it was less effective at mitigating active material loss [10].

Given to the content above, it is clear that Al₂O₃ coating demonstrates clear benefits in mitigating SEI growth and lithium inventory loss. However, this approach still meeting several critical limitations which must need practical implementation. For starters, Al₂O₃ coating had mechanical stability issues, given to the figure below, the 5 nm Al₂O₃ coating fractures during initial lithiation. This cause fresh Si expose to electrolyte and would affect long term protection, which might result in more material loss compared with carbon coatings [11]. In addition, there was a trade-off between protection and energy density. Although Al₂O₃ outperforms carbon in reducing LLI, its insulating nature inherently limits capacity utilization [10]. Lastly, Magnetron sputtering is a high-vacuum technique with low throughput and high equipment costs, making it impractical for mass production [9]. Based on the information above, it was obvious that Al₂O₃ coating strategy was good at targeted SEI suppression but it still was not a good solution. Several things could be done in the future such as hybrid coating to balance ionic /electronic conductivity and mechanical resilience and use Pre-lithiation techniques to compensate for irreversible LLI.

3.3 Si-Polymer

In general, the preparation of ultrathin crosslinked polyacrylonitrile (x-PAN) coatings on silicon nanoparticles involves emulsion-assisted polymer coating and thermal crosslinking. Polymer synthesis are separated into two kinds: PANVDC copolymer and PANVDA. For PANVDC copolymer, Acrylonitrile (AN) and vinylidene chloride (VDC) (85:15 molar ratio) were polymerized in water at 30°C using APS/SBS initiators. The synthetic route is shown in Figure 3 below [12]. For PANVDA, the chlorine in PANVDA were replaced with azides by reacting with NaN3 in DMF at 65°C Then moving to the coating process, the Si were first dispersed in toluene which mixed

with PANVDA/DMF solution and then emulsified in F108/formamide surfactant solution (12,000 rpm, 2 min).

After that, the solvent need to evaporated to 70°C to form tetrazole-linked x-PAN.

y pasting a Si NP@x-PAN slurry (60 wt% Si NP@x-PAN powder as the

mixture was then added into the reaction medium for copolymeria

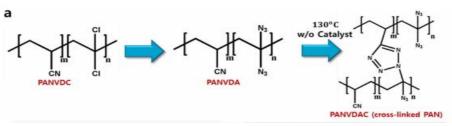


Fig. 3 characterization of x-PAN [12]

The testing for ultrathin crosslinked polyacrylonitrile (x-PAN) coatings on silicon nanoparticles need three steps. The slurry for electrode fabrication was prepared with 60% Si NPs@x-PAN, 20% Super P, 20% CMC binder and coated on Cu foil (~1.0 mg/cm² loading). CR2032 coin cells with Li metal counter electrode and 1M LiPF₆ in EC/DEC (1:1) + 10% FEC + 1% VC electrolyte for cell assembly. The cycling rate test was charged at 0.2C, discharged at 0.1-50C and long cycling was 0.5C/0.5C for 1000 cycles. The Si NPs@x-PAN anode exhibited exceptional electrochemical performance in three different aspects: ultrahigh cycling stability, superior rate capability and commercial viability. First, Si NPs@x-PAN anode still 76.2% capacity retention after 1,000 cycles at 0.5C, which was much better than purse Si (typically <20% after 100 cycles) [12].

Si NPs@x-PAN anode superior capability reflect by the fact that it can deliver delivers 1,880 mAh/g at 50C, which due to its enhanced wettability and Unimpeded Li⁺ diffusion through the ultrathin x-PAN layer. Lastly, Si NPs@x-PAN anode had commercial advantages. It could achieve areal capacity of ~2 mAh/cm² which meeting industry requirement. It also had high initial coulombic efficiency (86.4%), indicating minimal irreversible Li loss [12].

Just like above two coated silicon, the Si NPs@x-PAN anode still faced several intrinsic and practical challenges although it demonstrates exceptional cycling stability and rate capability. First, The ultrathin (~3 nm) crosslinked PAN layer may exhibit two dfects which might affect its performance: the nanoscale pinholes formed during incomplete crosslinking, which enable localized electrolyte penetration and subsequent silicon degradation, and second, non-uniform thickness distribution create uneven stress patterns during lithiation cycles [13]. In addition, the coated silicon is not stable and the it is not cost-effective given to the fact that the emulsion based coating need precise control of the solvent evaportation and must need stay in crosslinking conditions (130 °C, Ar), which was difficult in industry for mass production. The high puirty Si NPs were also cost a lot compare with graphene [11]. For manufacturing perspective, it is not a good option compared with other such as Al₂O₃, but its elasticity provides a unique advantage over brittle alternatives. So in the future, the researchers should focus on optimizing crosslinking density to eliminate pinholes and develop cheaper, scalable coating techniques for broader adoption

4. Summary and Evaluation of Si-Based Anode Materials

Table 1. Comparison of various Si-based anodes

Type	Advantages	Disadvantages
Si-C	 I High capacity: 3578 mAh g⁻¹ initial discharge. I Stable cycling: 1909 mAh g⁻¹ after 100 cycles. I Fast charging: 975 mAh g⁻¹ at 5 A g⁻¹. I Volume control: Only +17.8 μm swelling (vs. +29.8 μm for pure Si). Robust SEI: Minimal resistance growth. 	 I Graphene defects: ID/IG = 0.16 reduces conductivity. I Low initial CE: 76.9%. I Complex synthesis: Multi-step, hazardous (HF). Ni residues: Incomplete etching harms performance.
Si-metal oxide	 <i>l</i> Less fade: 64% lower capacity loss in full-cells. <i>l</i> Lithium retention: 37% less Li loss. <i>l</i> SEI stability: Limits electrolyte decomposition. Crack-tolerant: Coating adapts post-fracture. 	I Brittle coating: Cracks expose Si to electrolyte. I Insulating: Lowers energy density. Expensive: Sputtering is hard to scale.

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Si-polym		<i>l</i> Long life: 76.2% capacity after 1,000 cycles.	<i>l</i> Pinholes: Cause localized degradation.
	Ci malayman	<i>l</i> High-rate: 1,880 mAh g ⁻¹ at 50C.	<i>l</i> Hard to make: Needs precise solvent control.
	Si-polymer	l Ready for industry: 2 mAh/cm² areal capacity.	l Costly: Ultra-pure Si NPs.
		Elastic: Handles Si expansion better than oxides.	Scaling issues: Crosslinking limits mass production.

Based on the Table. 1 above, it is obvious to see that silicon-based anode materials have emerged as promising candidates for next-generation lithium-ion batteries due to several different reasons, although they still have many flaws. Si@Graphene, Si-metal oxide (Al₂O₃-coated), and Si-polymer (x-PAN-coated)—exhibits distinct advantages and limitations that must be carefully evaluated for practical applications.

For starters, Si-C demonstrates extremely outstanding electrochemical performance. For instance, its initial capacity is really high which around 3578 mAh g⁻¹. In addition, it has excellent charging rate capability about 975 mAh g⁻¹ at 5 A g⁻¹.. It also has stable cycling given to the fact that the capacity still has 1909 mAh g⁻¹ after 100 cycles. The graphene coating effectively mitigates volume expansion (+17.8 µm vs. +29.8 µm for pure Si) and stabilizes the solid-electrolyte interphase (SEI). Besides the Si-C have many different advantages, it still has many disadvantages. First, the presence of graphene defects, indicated by an ID/IG ratio of 0.16, reduces its electrical conductivity. In addition, the initial coulombic efficiency is relatively low at 76.9%, which limits energy storage performance. There are also lots of complex synthesis process, involving multiple steps and hazardous chemicals such as hydrofluoric acid (HF). Lastly, the incomplete etching always leave nickel residues, negatively impacting the overall performance. To overcome these limitations, the researchers should pay more attention on hybrid material design. In specific, for graphene defects (ID/IG = 0.16), it would be better to optimize the synthesis process. For Low initial Coulombic efficiency, Pre-lithiation or electrolyte additives would be good choices to stabilize SEI. For complex manufacturing, it need to explore dry etching. All in all, Focusing on scalable, safer synthesis process is the best way to improve the performance for Si-C.

Secondly, Si-metal oxide (Al₂O₃) is excellent in suppressing SEI growth and reducing lithium inventory loss (37% improvement), making them suitable for long-term cycling. In addition, the 5 nm Al₂O₃ coating significantly decreases capacity fade. It only has 64% loss in full cells. The SEI for Si-metal is stable which limits electrolyte decomposition. However, although it has so many advantages, its disadvantages are also vital and need more improvement just like Si-C. First, the coating's brittleness leads to mechanical failure during lithiation, exposing fresh

Si to electrolyte and accelerate degradation. Also, given to its insulating property, it reduce its energy density in the battery system. From manufacturing perspective, the entire procedure for coating is really expensive and hard to mass produce, which limit its commercial viability. To overcome these disadvantages, there are some innovative techniques could be develop in the future. For brittle coating, it can use multilayer design. In specific, using graded oxide coatings (e.g., SiO₂/TiO₂ nanolayers). While the inner layers accommodate expansion while outer layers stabilize SEI. The coating such as oxide-coated Si nanowires also can resolve the issue of insulating inherent property. Lastly, to solve the problem of manufacturing, it would be better to develop patial ALD systems for continuous oxide coating on Si powders. These approaches balance performance improvements with practical manufacturability, addressing the core limitations while leveraging existing battery production infrastructure.

Lastly, Si-polymer (x-PAN-coated) anodes stand out for their ultra-long cycle life (76.2% capacity retention after 1,000 cycles) and remarkable rate performance (1,880 mAh g⁻¹ at 50 C). It is also really suitable for industrial applications given to its 2 mAh/cm² areal capacity which meet the industry standard. Lastly, The elastic property of it allow it handling the Si expansion circumstance better than oxide-based materials. However, Si-polymer (x-PAN-coated) anodes still have some defects. Pinholes can lead to localized degradation, also the mass production is hard given to the fact that it need precise solvent control and Ultra-pure Si NPs are really expansive. Lastly, crosslinking issues hinder large-scale production. There are many possible way to overcome the limitations of Si-polymer (x-PAN-coated) anodes. For the formation of Pinholes, using interface engineering would be applicable. In specific, applying protective coatings (e.g., carbon, conductive polymers) to reduce electrolyte side reactions. The manufacturing process can also be simplified. For example, finding alternative blinder like water-soluble or less toxic binders (e.g., CMC, alginate) to reduce solvent dependency. Using cheaper Si composites instead of pure Si can also reduce the costs.

5. Conclusion

The silicon-based anode truly shows its potential as an alternative material for lithium-ion batteries based on its

own excellent properties, offering unparalleled energy density to meet the demands of electric vehicles and renewable energy storage. The study illustrate three different type of Si-based anode-carbon encapsulation (Si-C), metal oxide coatings (Al₂O₃), and elastic polymer matrices (x-PAN) by comparing there advantages and disadvantages and giving suggestions to mitigate silicon's inherent challenges.

The Si-C composites have excellent capacity and rate performance also good at fast charging, but it still require defect-free graphene and safer and cheaper synthesis methods. Future work should focus on pre-lithiation and dry processing to improve initial Coulombic efficiency (currently 76.9%) and scalability, therefore satisfying the manufacturing standard.

The Al₂O₃ coatings effectively stabilize SEI and reduce lithium inventory loss for long term cycling but the brittle coating might expose Si to electrolyte. Hybrid designs could balance mechanical resilience and conductivity, while atomic layer deposition optimization may reduce costs.

Si-polymer anodes demonstrate superior cycling stability (76.2% retention after 1,000 cycles) and elasticity but face pinhole defects and high purity requirements. Emulsion-based coating refinements and alternative binders (e.g., water-soluble polymers) could enhance manufacturability.

In summary. all three strategies above are not perfect, cannot fully resolving the limitations of Si. Following priorities are critical. First, combining conductive protective and elastic phases to address multiple failure modes (hybrid coatings). Second, advancing compatible techniques to replace batch processes like magnetron sputtering. Lastly, utilizing lower-purity silicon sources and integrate recycling loops for sustainable production. It is obvious that the combinations of material innovations and industrial-scale processes will unlock potential of Si. By addressing both technical and manufacturing challenges, silicon-based batteries can play a transformative role in achieving a sustainable energy future.

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