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# Surface engineering enables robust SEI growth towards a stable and efficient lithium-ion battery $\text{SiO}_x$ anode

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It is challenging to enhance the cycling stability and rate capability of  $\text{SiO}_x$  ( $0 < x < 2$ ) anodes for commercialization, due to the random and disordered growth of the solid electrolyte interphase (SEI) on the anode surface. Here, a surface engineering strategy was proposed to controllably regulate SEI formation. Through *in situ* constructing and densely coating a C–N network on the  $\text{SiO}_x$  nanoparticle surface, the surface energy and electronic structure were regulated, resulting in controlled growth of SEI components. The optimized SEI architecturally consists of inner  $\text{Li}_2\text{O}$  and an outer  $\text{LiF}/\text{Li}_2\text{CO}_3$  mixture. It not only enables mechanical robustness but also suppresses electrolyte decomposition, which significantly improves the  $\text{Li}^+$  transport kinetics at the electrode/electrolyte interfaces, resulting in a 4-fold reduced interfacial charge transfer resistance. Consequently, the anode exhibits outstanding electrochemical performance, with an initial reversible capacity of  $1674 \text{ mAh g}^{-1}$ . Moreover, the high capacities of  $1618 \text{ mAh g}^{-1}$ ,  $1274 \text{ mAh g}^{-1}$  and  $1114 \text{ mAh g}^{-1}$  were recorded at the 100th, 200th, and 300th cycles with a  $1 \text{ A g}^{-1}$  current density, respectively. The capacity retention rates were 96%, 83%, and 66%, respectively, which demonstrates good cycling stability. Besides, the rate capability approached  $888 \text{ mAh g}^{-1}$  at  $5 \text{ A g}^{-1}$  and recovered 98% as the current decreased to  $0.1 \text{ A g}^{-1}$ . More importantly, this method is low-cost, scalable, and uniform, making it suitable for large-scale industrial applications. This work provides a new way of producing high-performance  $\text{SiO}_x$  anodes; moreover, the scalable fabrication is promising for industrial applications.

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## 1 Introduction

The demand for high-energy-density lithium-ion batteries (LIBs) is rising with the development of electric vehicles and mobile electronic devices, but traditional graphite anodes cannot meet the increasing requirements, driving the need for research into high-capacity anodes.<sup>1–4</sup>  $\text{SiO}_x$  with medium volume expansion ( $\sim 113\%$ ) and outstanding reversible capacity ( $\sim 2000 \text{ mAh g}^{-1}$ ) has been looked at as an ideal candidate.<sup>5–7</sup> However, in actual applications, it is still limited by a low reversible capacity, short cycle lifetime, and poor rate performance.<sup>8</sup> All of these limitations have been demonstrated to derive from the solid electrolyte interphase, which forms on the

$\text{SiO}_x$  anode surface during the initial charge–discharge stage.<sup>9</sup> As reported, the SEI is constructed from organic and inorganic layers. The organic layer is composed of electrolytes and provides good elasticity and ion-free transport channels. Conversely, the inorganic layer consists of  $\text{Li}_2\text{CO}_3$ ,  $\text{Li}_2\text{O}$ , and  $\text{LiF}$ , respectively. Among them,  $\text{LiF}$  presents a high Young's modulus, which enables good mechanical properties, and  $\text{Li}_2\text{CO}_3$  exhibits good electron insulation, which can effectively suppress redox reactions.  $\text{Li}_2\text{O}$  shows poor electronic/ionic conduction and usually serves as a filler.<sup>10</sup> Their stoichiometry and spatial distribution not only affect the mechanical strength but also determine the ionic/electron diffusion/transport rate.<sup>11,12</sup> Thus, constructing an SEI with reliable mechanical performance, high ion conductivity, and electronic insulation is the key to obtaining ideal electrochemical performance.<sup>13–15</sup>

Currently, techniques such as anode surface engineering, artificial SEI, electrolyte design, and electrodeposition have been reported to regulate the SEI.<sup>16–21</sup> However, major bottlenecks persist, including: (1) complex, costly, and low-yield SEI manufacturing; (2) imprecise control over the molecular constituents and structure of the SEI; and (3) electrochemical and mechanical incompatibility with battery components. Among them, surface engineering, applied during the synthesis

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stage, is an effective strategy. It is carried out through coating, doping, and etching to regulate the surface atomic arrangement, electronic structure, surface energy, *etc.*, achieving controllable SEI composition, crystallinity, and spatial distribution.<sup>22</sup> Carbon coating is the most common route in surface engineering. It not only changes the surface properties but also relieves the volume expansion of  $\text{SiO}_x$ . Common methods include ball milling, chemical vapor deposition, sol-gel, and hydrothermal synthesis. Moreover, introducing heteroatoms into the carbon layer can further regulate its conductivity, surface energy, active sites, and electrochemical properties.<sup>23</sup> The reported heteroatoms include non-metal elements (B, N, P, S, F),<sup>24–28</sup> and metal elements (Mg, Co, Fe, Mo, Ti, Ag).<sup>29–34</sup> Although the positive effect of heteroatom-doping has been widely demonstrated, it was still limited from the perspective of controllable composition homogeneity, carbon layer thickness and densification. More importantly, its effect on SEI growth should be further clarified. Therefore, addressing these aspects is essential for accurately constructing a core-shell structure and ultimately enhancing the electrochemical performance of  $\text{SiO}_x$  is necessary.

To surmount the bottleneck of unregulated SEI formation limiting  $\text{SiO}_x$  anode commercialization, a dedicated surface engineering strategy is presented for controllable SEI modulation. This study establishes a new avenue for fabricating high-performance  $\text{SiO}_x$  anodes, laying a solid foundation for their large-scale industrial deployment.

## 2 Experimental section

### 2.1 The synthesis of the anode material

The anode composite material with 10 wt% N was prepared by using the solid-state reaction method. First, commercial  $\text{SiO}_x$  powder (purity  $\geq 99\%$ , particle size 100 mesh), melamine ( $\text{C}_3\text{H}_6\text{N}_6$ , AR) and glucose ( $\text{C}_6\text{H}_{12}\text{O}_6$ , purity  $\geq 99.5\%$ ) were mixed with a weight ratio of 6 : 2 : 2 and milled for 4 hours (zirconia balls, ethanol, 180 rpm). Then, the as-obtained powder was heated at 200 °C for 2 h in a vacuum drying oven. After that, the powder was milled for 1 h. Subsequently, the powder was sealed in a high-temperature furnace, heated at a rate of 5 °C  $\text{min}^{-1}$  to 900 °C under a  $\text{N}_2$  atmosphere (flow rate 50 sccm), and calcined for 1 h. Finally, the anode material was produced.

### 2.2 Material characterization

Scanning electron microscopy (SEM) images were recorded by a TESCAN MIRALMS, and transmission electron microscopy (TEM) and high-resolution TEM images were collected with a JEOL JEM-2100F TEM. X-ray photoelectron spectroscopy (XPS) analysis was carried out on a K-Alpha. Fourier-transform infrared (FT-IR) measurements were performed on a Nicolet iS10 Fourier-transform infrared spectrometer. Inductively coupled plasma (ICP) measurements were carried out on a Thermo Scientific iCAP 7400 ICP-OES.

### 2.3 Coin cell assembly

The  $\text{CN-SiO}_x$ , Super P, and sodium alginate were mixed with a weight ratio of 7 : 2 : 1. The electrode slurry was prepared using deionized water as the solvent and coated on Cu foil. The loading was controlled from 0.8 to 1.2  $\text{mg cm}^{-2}$ . The as-obtained electrodes were dried in a vacuum oven at 60 °C for 12 h, and the electrodes were punched into discs with a diameter of 12 mm. The polished Li foil was punched into 14 mm diameter, and 200  $\mu\text{m}$  thick discs. CR2032 coin cells were assembled in an argon-filled glove box, using a lithium disc as the counter electrode and a PP separator with 80  $\mu\text{L}$  of electrolyte. After that, the batteries were rested at 25 °C for 12 h.

### 2.4 Electrochemical measurements

A CT-4008T-5V10mA-164 battery testing system was used to measure the galvanostatic charge/discharge of the coin cells. Cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), and Galvanostatic Intermittent Titration Technique (GITT) measurements were carried out on a CHI660E electrochemical workstation (Shanghai Chenhua Instrument Co., Ltd).

### 2.5 Theoretical calculations

All calculations were carried out using density functional theory (DFT) within the Vienna *Ab initio* Simulation Package (VASP). The electron exchange–correlation functional was treated using the Perdew–Burke–Ernzerhof generalized gradient approximation (PBE-GGA), and the ion–electron interactions were described by the projector augmented wave (PAW) method. Monkhorst–Pack *k*-point meshes with a spacing of 0.25  $\text{\AA}^{-1}$  were used for the N-doped graphite/graphite/ $\text{SiO}_x$ @SEI slab models. Geometry optimization was performed until the total energies (atomic forces) converged to  $10^{-4}$  eV (0.01 eV  $\text{\AA}^{-1}$ ) with a plane-wave energy cutoff of 400 eV.

## 3 Results and discussion

Fig. 1a displays the preparation process of the  $\text{SiO}_x$  anode with an N-doped carbon network and coating (named as CNS). Briefly, glucose ( $\text{C}_6\text{H}_{12}\text{O}_6$ ) and melamine ( $\text{C}_3\text{H}_6\text{N}_6$ ) were used as carbon and nitrogen sources, respectively. They were uniformly mixed with  $\text{SiO}_x$  powder and ball-milled to form a preliminary coating, followed by vacuum drying to remove their internal water molecules (dehydration reactions). During this process, preliminary C–N network formation occurred through the condensation of melamine and glucose. Subsequently, the mixture was further ball-milled to reduce the particle size and achieve a uniform coating of the carbon precursor. After that, it was calcined under an argon atmosphere for several hours. Finally, the CNS was produced; as a comparison, carbon-coated  $\text{SiO}_x$  (named as CS) was prepared using the same procedure but without nitrogen source addition. The microstructure was characterized by TEM. Fig. 1c shows that the CNS particles are below one micrometer in size. The high-resolution TEM showed partial lattice fringes (Fig. 1g), corresponding to the Si (111) plane, indicating that partially crystalline silicon exists within it. Fig. 1d, e, h and i display the elemental mapping images of

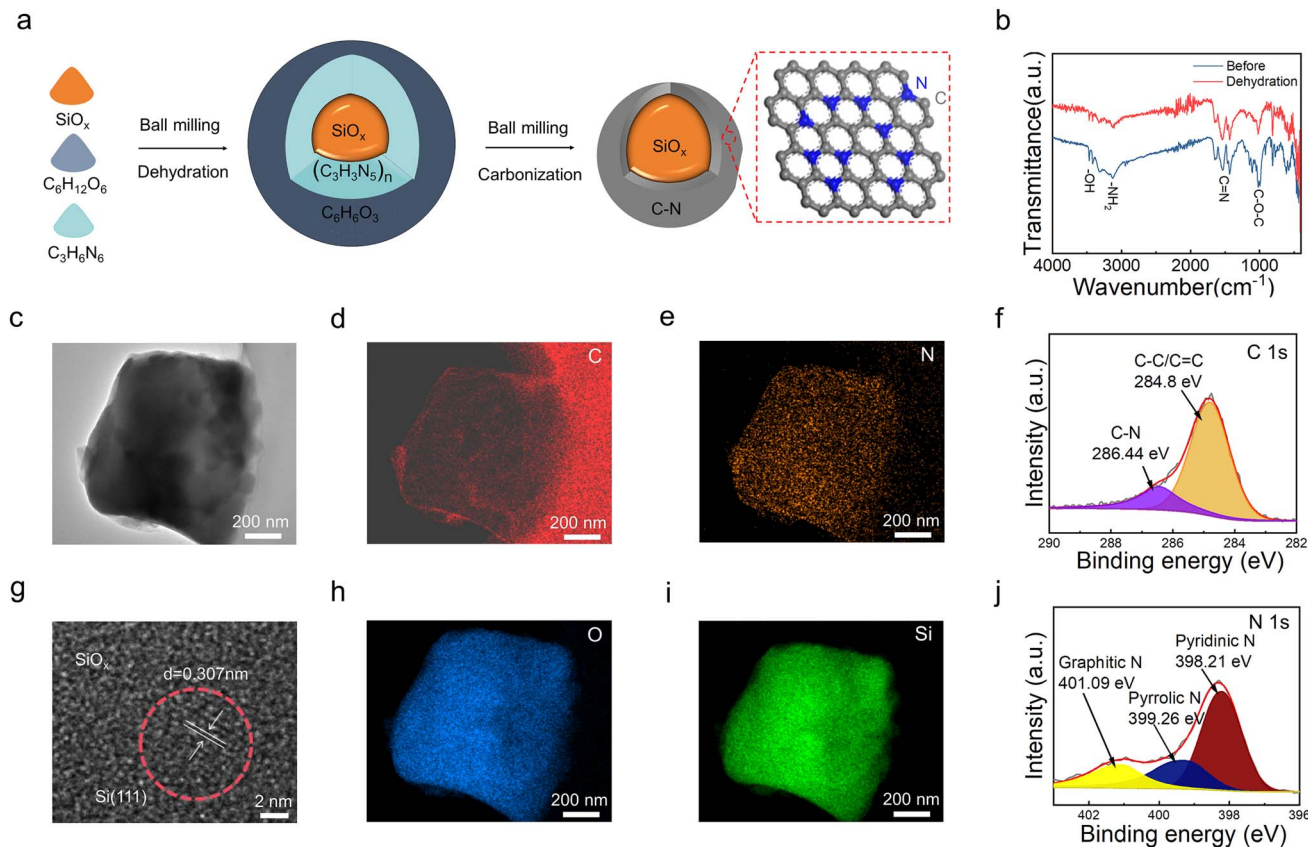


C, N, O, and Si in the CNS, respectively. This not only confirms that the C layer is fully and uniformly coated onto the  $\text{SiO}_x$  particles but also shows that the N element is uniformly dispersed on the C layer. This suggests that the *in situ* dehydration process at the particle scale was the key to forming a dense and uniform coating. The removal of  $-\text{OH}$  and  $-\text{NH}_2$  groups leaves behind ether bonds ( $-\text{C}-\text{O}-\text{C}-$ ) and  $\text{C}=\text{N}$  groups (Fig. 1b). The formed polymer and  $\text{C}=\text{N}$  bonds enable complete encapsulation of the  $\text{SiO}_x$  particles and facilitate the incorporation of N atoms into the carbon framework, preventing their escape at high temperatures.

Then, X-ray photoelectron spectroscopy (XPS) was used to investigate the elemental valence states. The C 1s spectrum was fitted with two characteristic peaks located at 284.8 eV and 286.44 eV (Fig. 1f), corresponding to C-C and C-N bonds.<sup>35–37</sup> This result indicates that the carbon in the composite material exists in  $\text{sp}^2$  and  $\text{sp}^3$  hybridization states, along with residual functional groups, and the N 1s spectrum shown in Fig. 1j has three peaks at 398.21 eV, 399.26 eV, and 401.09 eV, which can be attributed to pyridine-N, pyrrole-N, and graphitic-N, respectively.<sup>38</sup> These results suggest that N-doping has been introduced into the carbon layer. These two peaks (284.8 eV and 286.44 eV) correspond to the amorphous and graphite carbon, respectively. The Si 2p spectrum (Fig. S1) displays characteristic peaks that appear at 99.2 eV ( $\text{Si}^0$ ) and 102.9 eV ( $\text{Si}^{2+}$ ), further

evidencing the presence of crystalline silicon. Furthermore, structural, morphological, and compositional evaluations of the as-prepared samples from large-scale batches confirm their good consistency (Fig. S2).

Fig. 2a shows the cyclic voltammetry (CV) curve of CNS at  $0.1 \text{ mV s}^{-1}$ , and it can be seen that the oxidation and reduction peaks with good symmetry appear at around 0.61 V and 0.66 V, respectively. Compared to the CS, which presents two oxidation peaks at 0.38 V/0.54 V and one reduction peak at 0.15 V (Fig. S3), the sharper peaks of CNS indicate easier redox reactions and higher electrochemical activity. The higher response current corresponds to improved reaction kinetics. Moreover, the smaller potential difference between the oxidation and reduction peaks suggests better stability and reversibility of the SEI.<sup>39</sup> Fig. 2b shows the rate capability at different current densities ranging from 0.1 to  $5 \text{ A g}^{-1}$ . As shown, the CNS has a discharge capacity of  $2000 \text{ mAh g}^{-1}$ ,  $1600 \text{ mAh g}^{-1}$ ,  $1200 \text{ mAh g}^{-1}$ , and  $888 \text{ mAh g}^{-1}$  at the current densities of 0.1, 0.5, 1, 2, and  $5 \text{ A g}^{-1}$ , respectively. Moreover, as the current densities recover to 0.1, the discharge capacity is  $1900 \text{ mAh g}^{-1}$ , retaining 98% of its initial value. In comparison, the CS and  $\text{SiO}_x$  show poor discharge capacity and recovery rate, indicating that the CNS has higher capacity and rate performance. Additionally, the charge/discharge curve of CNS is shown in Fig. 2c, and presents a high initial reversible capacity approaching  $3000 \text{ mAh g}^{-1}$ .



**Fig. 1** (a) Schematic illustration of the synthesis and structural features. (b) Fourier transform infrared spectroscopy before and after dehydration condensation treatment. (c) HAADF-STEM images of CNS. (d–i) Elemental mapping images of C, Si, N, and O. (g) HRTEM images of CNS. (f and j) High-resolution XPS spectra of C 1s and N 1s.

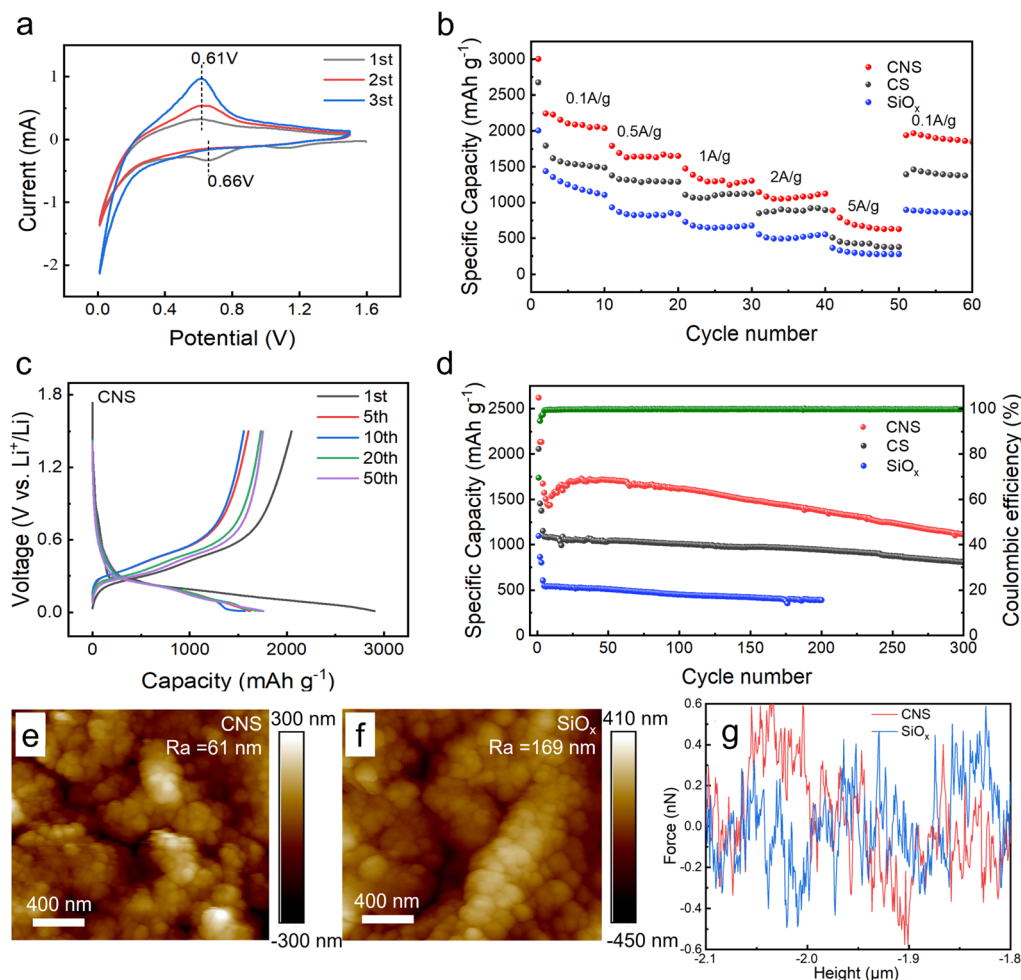


Fig. 2 (a) CV profiles of CNS at  $0.1 \text{ mV s}^{-1}$ . (b) Rate performance. (c) Discharge-charge voltage curves of CNS at  $100 \text{ mAh g}^{-1}$ . (d) Cycling performance at  $1.0 \text{ A g}^{-1}$ . (e) AFM height map of the CNS electrode. (f) AFM height map of the  $\text{SiO}_x$  electrode. (g) Force-distance curves of CNS and  $\text{SiO}_x$ .

Although the lower initial coulombic efficiency ( $\sim 70\%$ ) was significantly reduced, the capacity of  $1680 \text{ mAh g}^{-1}$  at the 5th cycle was recorded. Moreover, it shows good coincidence at the 10th, 20th, and 50th cycles, indicating the excellent cycling stability of CNS. In contrast, the CS shows a smaller initial reverse capacity ( $\sim 2000 \text{ mAh g}^{-1}$ ), initial coulombic efficiency ( $\sim 67\%$ ), and steady-state capacity ( $\sim 1230 \text{ mAh g}^{-1}$ ) (Fig. S4). Similarly, the  $\text{SiO}_x$  shows poor initial reverse capacity ( $\sim 1700 \text{ mAh g}^{-1}$ ), initial coulombic efficiency ( $\sim 62\%$ ), and capacity ( $\sim 900 \text{ mAh g}^{-1}$ ) (Fig. S5). Both carbon and N-doped carbon coating on the  $\text{SiO}_x$  particles have significantly increased the cycling stability and reverse capacity, which is in agreement with the results reported above. This is because carbon coating on  $\text{SiO}_x$  buffers expansion, stabilizes the SEI and enables electron transport, while nitrogen doping boosts the active sites for  $\text{Li}^+$  intercalation and conductivity. Their synergy enhances the cycling stability and reversible capacity.<sup>40</sup> Fig. 2d shows the cycling performance of CNS, CS, and  $\text{SiO}_x$ . The CNS retains a high specific capacity and smooth charge/discharge cycles within 300 times. The specific capacity was  $1600 \text{ mAh g}^{-1}$ ,  $1400 \text{ mAh g}^{-1}$ , and  $1140 \text{ mAh g}^{-1}$  at the 100th, 200th, and 300th

cycles, respectively. The capacity was 96%, 84%, and 67%, respectively. Moreover, the coulombic efficiency was maintained over 99%, indicating good stability. In comparison, the CS also shows good stability and maintains about 70% capacity after 300 cycles, but a lower specific capacity ( $1000 \text{ mAh g}^{-1}$ ). Meanwhile, the  $\text{SiO}_x$  shows both poor specific capacity ( $600 \text{ mAh g}^{-1}$ ) and stability.

Besides, the characteristic capacity curve of the CNS electrode shows a notable recovery capacity around the initial 20 cycles, which is intricately linked to the dielectrophoresis effect that is derived from the C-N coating layer.<sup>41</sup> The dielectrophoresis effect could be described briefly as follows: as the current during the charging process increases from  $0.1\text{C}$  to  $1\text{C}$ , the electrode rapidly expands, causing some of the alloy phase  $\text{Li}_x\text{Si}$  to detach from the copper foil surface and enter the electrolyte, forming a neutral state. Subsequently, the irregular electric field causes it to move back to the electrode surface and re-work from the neutral state, resulting in capacity recovery. N-doping enables the good conductivity of CNS, which results in the generation of localized non-uniform electric fields during the charge-discharge stage. Meanwhile, the  $\text{SiO}_x$  and CS did not

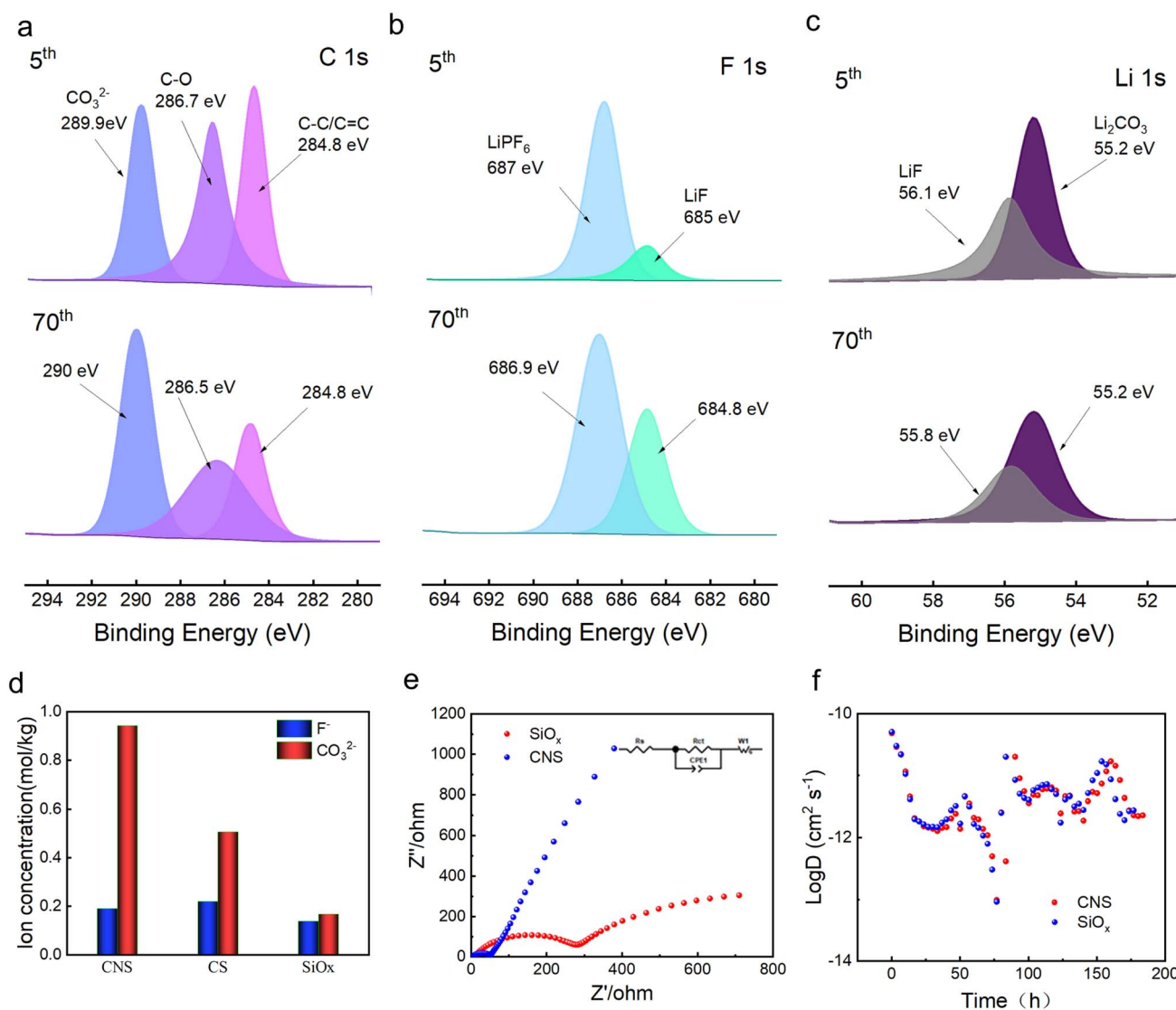




exhibit capacity fluctuation, which could contribute to their poor conductivity and inability to generate sufficient irregular electric fields. As shown in Fig. 2e and f, the surface roughness of CNS was  $\sim 61$  nm, while the surface roughness of  $\text{SiO}_x$  was 169 nm. This indicates that the CNS morphology is more uniform. The force–height curve (Fig. 2g) suggests that the CNS has a higher slope between the force maximum and zero points, which indicates higher hardness and elastic modulus, and is higher than that of  $\text{SiO}_x$ .<sup>42</sup>

The CNS shows superior electrochemical performance compared to both  $\text{SiO}_x$  and CS. To investigate the mechanism, further characterizations were conducted. First, the electrodes after the 5th and 70th cycles were disassembled. Then, the surface elemental valence states were tested using XPS. The C 1s spectrum after the 5th cycle could be fitted to three peaks located at 284.8 eV, 286.7 eV, and 289.9 eV, respectively (Fig. 3a). They were assigned to C–C/C=C, C–O, and  $\text{CO}_3^{2-}$ , respectively,

where the C–C/C=C and C–O were derived from the decomposition products of the organic electrolyte, and the  $\text{CO}_3^{2-}$  was attributed to the  $\text{Li}_2\text{CO}_3$  of the SEI inorganic layer. In comparison, the  $\text{CO}_3^{2-}$  peak after the 70th cycle has increased 0.1 eV. The two fitted F 1s spectra after the 5th cycle were observed at 687 eV and 685 eV (Fig. 3b), which could be attributed to  $\text{LiPF}_6$  and LiF, respectively. After the 70th cycle, they decreased by 0.2 eV. The two Li 1s peaks after the 5th cycle were located at 56.1 eV and 55.2 eV (Fig. 3c), which could be attributed to LiF and  $\text{Li}_2\text{CO}_3$ , respectively. Moreover, after the 70th cycle, they decreased by 0.3 eV. The shifted element binding energies for C 1s, F 1s and Li 1s suggest that LiF and  $\text{Li}_2\text{CO}_3$  gradually form a robust and heterogeneous interface during cycling, enhancing electron transfer between the two phases.<sup>43</sup> It also indicates that the composition and spatial distribution of the SEI are not constant, but dynamically evolve during the cycling process.<sup>44</sup> However, the Li 1s peaks of  $\text{SiO}_x$  show three peaks



**Fig. 3** (a–c) Comparison of the high-resolution XPS spectra of C 1s, F 1s, and Li 1s after the 5th and 70th cycles of CNS. (d) Comparison of the  $\text{F}^-$  and  $\text{CO}_3^{2-}$  concentrations at the first activation of  $\text{SiO}_x$ , CS, and CNS. (e) EIS comparison between CNS and CS before cycling. (f) Comparison of the GITT profiles and calculated D of CNS and  $\text{SiO}_x$ .



corresponding to  $\text{Li}_2\text{O}$ ,  $\text{Li}_2\text{CO}_3$  and  $\text{LiF}$ , respectively (Fig. S6). This indicates that the N-doped carbon coating in CNS effectively suppresses the formation of  $\text{Li}_2\text{O}$ .<sup>45</sup>

As reported,  $\text{Li}_2\text{O}$  was a byproduct derived from the reaction between the electrolyte and impurities (such as EC, and residual  $\text{SiO}_x$ ), which is inconducive to  $\text{Li}^+$  transport and mechanical properties. Regarding the CS electrode, the  $\text{Li}_2\text{O}$  is not detected after the 5th cycle, but it was found at the 70th cycle (Fig. S7), which suggests that the reduction process causes  $\text{Li}_2\text{O}$  to be continuously generated, while the outer layer of  $\text{Li}_2\text{CO}_3$  and  $\text{LiF}$  becomes loose and partially consumed. To accurately obtain the content of carbonate and fluoride in the SEI, ion chromatography was performed. The SEI layer was washed with KOH and methanesulfonic acid to remove the residual surface organic electrolyte, and to prepare the test sample, it was volumetrically made up to volume with pure water, shaken and sonicated for 1 h, and finally diluted with pure water. As shown in Fig. 3d, for  $\text{CO}_3^{2-}$ ,  $\text{CNS} > \text{CS} > \text{SiO}_x$ , while for  $\text{F}^-$ ,  $\text{SiO}_x < \text{CNS} < \text{CS}$ . This indicates that the  $\text{Li}_2\text{CO}_3$  in the SEI was the critical component for electrochemical performance, due to its moderate ionic conductivity and good chemical stability. To investigate the charge transfer capability of the SEI, Fig. 3e displays the Nyquist plots of the CNS and CS electrodes. The  $R_{ct}$  value was fitted with

the equivalent circuit shown in Fig. 3e inset, and it shows a value of  $60.56 \, \Omega$  and  $240 \, \Omega$  for CNS and  $\text{SiO}_x$ , respectively. The N-doping obtained a quarter of the charge transfer resistance, which indicates significantly improved conductivity.<sup>46</sup> Furthermore, the Galvanostatic Intermittent Titration Technique (GITT) was employed to evaluate the  $\text{Li}^+$  transport kinetics of the SEI, and the diffusion coefficient ( $D$ ) was calculated using eqn (1).<sup>47,48</sup>

$$D = \frac{4}{\pi\tau} \left( \frac{m_B V_M}{M_B S} \right)^2 \left( \frac{\Delta E_S}{\Delta E_\tau} \right)^2 \quad (1)$$

Fig. 3f shows the diffusion coefficient of CNS and CS with logarithm value. It indicates that the value for the CNS is higher than that for the CS. This indicates that the SEI of the CNS has a better ion diffusion ability. Based on the above results and discussion, we conclude that the CNS significantly improves the reversible capacity, rate performance, and long-term cycling stability, owing to the formation of a well-compacted  $\text{LiF}/\text{Li}_2\text{CO}_3$  layer on the same plane. This not only enables higher electron conductivity but also maintains a higher ion diffusion coefficient (Fig. 4a). Meanwhile, for the  $\text{SiO}_x$  alone, the SEI presents a loose distribution, which readily facilitates the growth of lithium dendrites, resulting in poor electron conductivity and ion

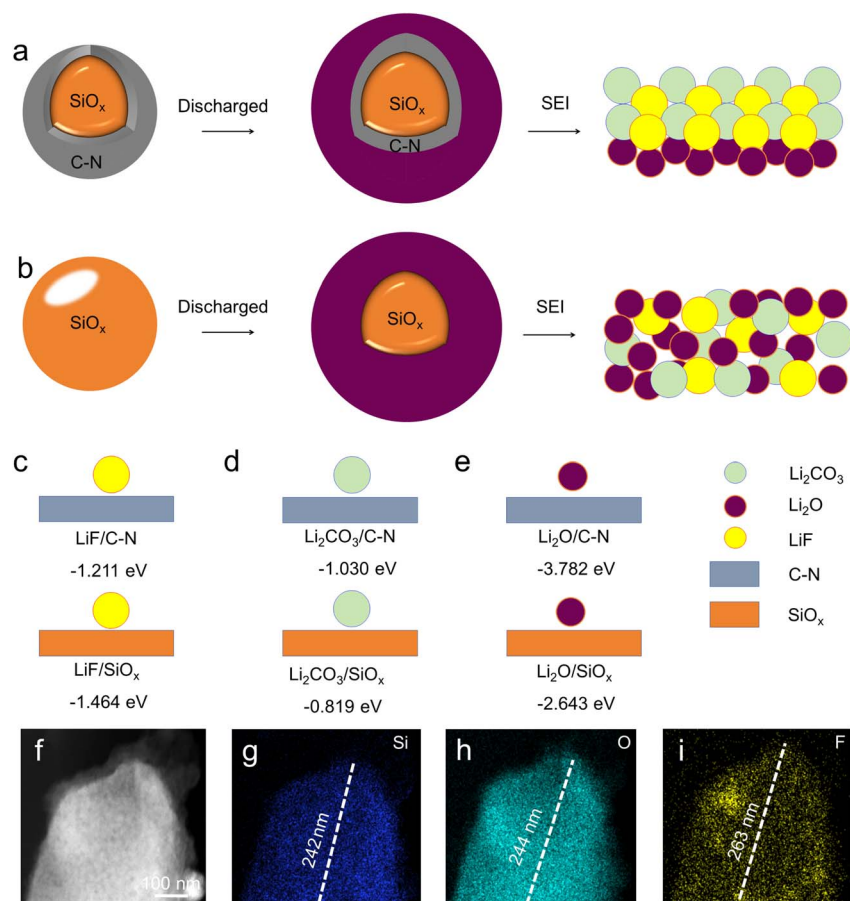


Fig. 4 (a) Schematic diagram of the interface of CNS during discharge. (b) Schematic diagram of the interface of  $\text{SiO}_x$  during discharge. (c–e) Schematic diagram of the energy values corresponding to  $\text{LiF}/\text{Li}_2\text{CO}_3/\text{Li}_2\text{O}$  at the interface of CNS and  $\text{SiO}_x$ . (f) HAADF-STEM image of a CNS sample. (g–i) Elemental mapping images of Si, O, and F.



diffusion coefficient (Fig. 4b). During cycling, the SEI was easily damaged due to the electrode volume expansion, causing rapid growth of lithium dendrites.<sup>49</sup> Thus, the reversible capacity, rate performance, and long-term cycling stability deteriorate.

To further understand the components and spatial distribution formation of the SEI during the activation stage, density functional theory (DFT) calculations were performed. The adsorption energies of LiF/Li<sub>2</sub>CO<sub>3</sub>/Li<sub>2</sub>O on N-doped graphite, graphite, and SiO<sub>x</sub> were calculated, respectively. The smaller absorption of energy corresponds to easier formation. As a result, the adsorption energy was  $-3.78$  eV,  $-1.21$  eV, and  $-1.03$  eV for CNS. This indicates that Li<sub>2</sub>O was more likely to form in the inner layer of the SEI, while LiF and Li<sub>2</sub>CO<sub>3</sub> tend to form a dense and compact outer layer, effectively sealing the electrode surface, in agreement with the XPS results. Meanwhile, for SiO<sub>x</sub>, the adsorption energies of Li<sub>2</sub>O, LiF, and Li<sub>2</sub>CO<sub>3</sub> were  $-2.64$  eV,  $-1.46$  eV, and  $-0.81$  eV, respectively, which

shows an obvious gradient energy distribution and may cause a loose arrangement of the three components (Fig. 4c and d). Moreover, for CS, the adsorption energies of Li<sub>2</sub>O, LiF, and Li<sub>2</sub>CO<sub>3</sub> were  $-0.025$  eV,  $-0.027$  eV, and  $-0.04$  eV, respectively (Fig. S8). The nearly identical adsorption energies indicate random and disordered growth. All these results indicate that the surface composition and spatial distribution can be controlled. After that, we performed the TEM element mapping of the CNS after SEI formation (Fig. 4f–i), and the distribution ranges for Si, O, and F were 242 nm, 244 nm, and 263 nm, respectively. The smaller range of O than F indicates that the Li<sub>2</sub>O was distributed in the inner layer, and LiF was located in the outer layer. Based on the results above, we conclude that the SEI structure is Li<sub>2</sub>O/LiF@Li<sub>2</sub>CO<sub>3</sub>.

The reaction kinetics were investigated by cyclic voltammetry (CV) at scan rates ranging from 0.1 to 0.6 mV s<sup>-1</sup> within a potential window of 0.01–1.5 V (Fig. 5a). The redox peaks were

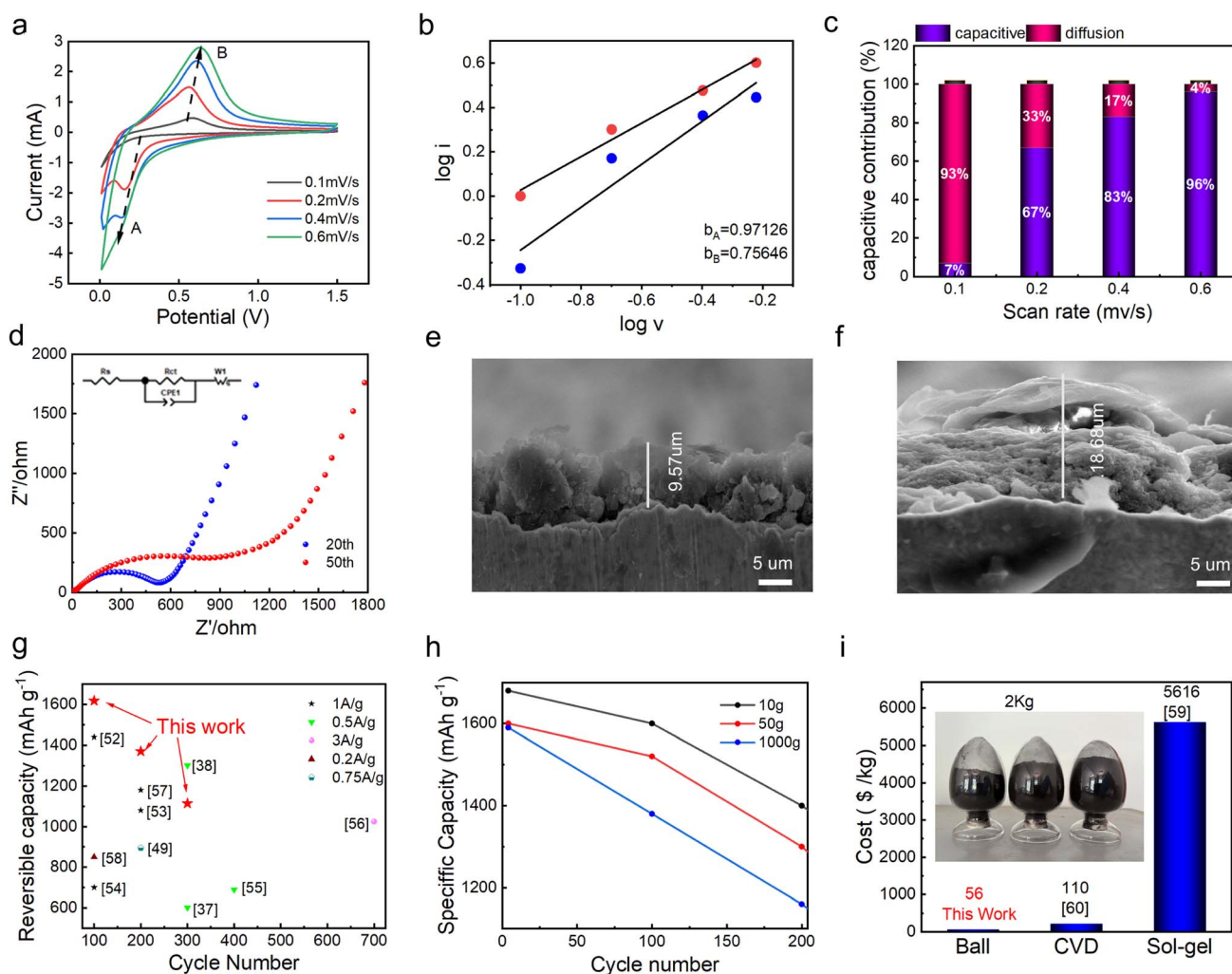


Fig. 5 (a) CV curves of CNS at different scan rates. (b)  $\log i$  vs.  $\log v$  plots for electrode kinetics analysis. (c) Capacitive and diffusion contributions to charge storage at various scan rates. (d) EIS comparison of CNS after 20 and 50 cycles. (e) Cross-sectional SEM image of the CS electrode after cycling. (f) Cross-sectional SEM image of the CNS electrode after cycling. (g) Capacity retention rate of this work compared with previous studies. (h) Specific capacity of CNS at different preparation scales (10, 50, and 1000 g) during cycling. (i) Production cost per kilogram for samples prepared by ball milling, CVD, and sol-gel methods.

observed at  $\sim 0.6$  V and  $\sim 0.2$  V, corresponding to the lithium insertion/extraction in the anode. Moreover, as the scan rate rises, the peak current and potential difference increase, indicating that the process is dominant from electrode polarization, and the repeatable curve indicates good rate response ability. Furthermore, the CV curves were fitted with  $\log i - \log v$  (Fig. 5b), and the relationship between peak current and scan rate can be described with eqn (2)–(4):<sup>50</sup>

$$i = av^b \quad (2)$$

$$\log i = \log a + b \log v \quad (3)$$

$$i(v) = k_1 v + k_2 v^{1/2} \quad (4)$$

The obtained slopes ( $b$ ) for the reduction and oxidation peaks were  $b_A = 0.97126$  and  $b_B = 0.75646$ , respectively. A  $b$ -value close to 1 indicates a capacitively controlled electrode process. This means that lithium ions could rapidly move on the anode surface, which results in high rate and fast-charging performance of the LIBs. Fig. 5c shows the contribution ratios of the capacitive and diffusion-controlled processes at different scan rates. As the scan rate increases, the capacitive percentage increases from 7% to 96%, confirming that lithium-ion storage becomes dominated by surface-controlled mechanisms at high rates.<sup>46</sup> This endows fast charging, which helps to mitigate the rate limitations typical of silicon-based materials.<sup>51</sup> Fig. 5d displays the Nyquist plots of CNS electrode after the 20th and 50th cycles. The charge transfer resistance ( $R_{ct}$ ) was obtained by fitting the data with the equivalent circuit shown in the inset of Fig. 5d, yielding values of 532  $\Omega$  and 806  $\Omega$  after the 20th and 50th cycles, respectively. Moreover, the  $R_{ct}$  and the impedance related to the solid-state diffusion process (reflected by the Warburg impedance and low-frequency region) indicate that the CNS anode maintains good structural integrity and interface stability, with no severe side reactions at the electrode/electrolyte interface. Fig. 5e and f show the cross-sectional SEM images of the CNS and  $\text{SiO}_x$  electrode after cycling. The CNS electrode has a smaller thickness of 9.57  $\mu\text{m}$ , maintaining a compact structure. This indicates that volume expansion is effectively suppressed. The  $\text{SiO}_x$  electrode presents a thickness of 18.68  $\mu\text{m}$ , exhibiting severe structural agglomeration and volume expansion. Fig. 5g shows the statistics and comparison of the reversible capacity at different cycling times with the literature.<sup>37,38,52–58</sup> The CNS presents competitive reversible capacities and stability of  $\sim 1600$   $\text{mAh g}^{-1}$  at 100 cycles,  $\sim 1400$   $\text{mAh g}^{-1}$  at 200 cycles, and  $\sim 1100$   $\text{mAh g}^{-1}$  at 300 cycles, respectively, demonstrating the superiority of the surface structure design method to regulate SEI performance. Beyond cycling stability and capacity retention, practical industrial application requires consideration of scalability, cost, and production efficiency. Here, for further description, Fig. 5h is given to illustrate the scalability advantages, showing the specific capacity of CNS samples prepared at 10 g, 50 g, and 1000 g scales. After 4 cycles, their specific capacity is 1680  $\text{mAh g}^{-1}$ , 1600  $\text{mAh g}^{-1}$ , and 1590  $\text{mAh g}^{-1}$ , respectively. In addition,

the sample prepared at the scale of 1000 g at 200 cycles still maintains a specific capacity of 1160  $\text{mAh g}^{-1}$ . This proves that the capacity decay of the materials is within a reasonable and acceptable range during mass production and has the potential to adapt to industrial mass production. Fig. 5i shows the expense per kilogram of CNS compared with that of CVD and sol-gel methods.<sup>59,60</sup> The inset in Fig. 5i shows a photograph of the CNS samples, with costs of 56\$, 110\$ and 5616\$ per kilogram respectively. This implies that the CNS has a cost advantage in terms of expense per kilogram, indicating that the preparation strategy of CNS may be conducive to large-scale application.

## 4 Conclusions

In conclusion, we have reported a surface engineering strategy for controllable growth of the SEI on the  $\text{SiO}_x$  anode. Through *in situ* constructing and densely coating a C–N network on the  $\text{SiO}_x$  nanoparticle surface, the surface energy and electronic structure were regulated, resulting in controlled growth of SEI components. The optimized SEI architecture features an inner layer of  $\text{Li}_2\text{O}$  and an outer mixture of  $\text{LiF}$  and  $\text{Li}_2\text{CO}_3$ . It not only enables mechanical robustness but also suppresses the electrolyte decomposition, which significantly improves the  $\text{Li}^+$  transport kinetics at the electrode/electrolyte interfaces, resulting in a 4-fold reduced interfacial charge transfer resistance. Consequently, the anode exhibits outstanding electrochemical performance, and this method is low-cost, scalable, and uniform, making it suitable for industrial applications.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Supplementary information (SI) is available. See DOI: <https://doi.org/10.1039/d5ra08407e>.

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