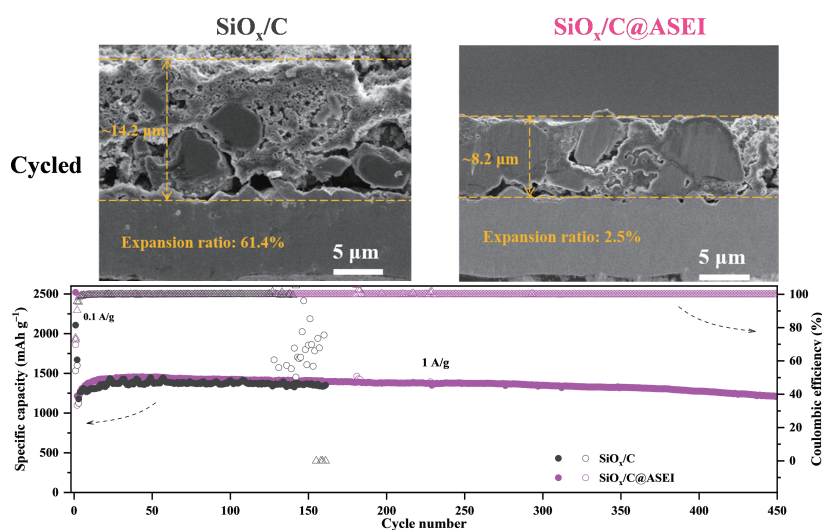


Graphical Abstract

In situ construction of highly resilient artificial solid electrolyte interphase for low-expansion silicon oxide anodes

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In situ polymerized artificial solid electrolyte interphase (ASEI) from a lithium maleate precursor stabilizes swelling-prone SiO_x/C anodes. The resilient interphase enables low-expansion electrode behavior and the formation of a thinner, more uniform solid electrolyte interphase, suppressing continuous electrolyte decomposition and Li consumption. This strategy enables long-life cycling at high areal capacities and enhances the stability of Ni-rich full cells.

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In situ construction of highly resilient artificial solid electrolyte interphase for low-expansion silicon oxide anodes

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ABSTRACT

Silicon oxide (SiO_x/C) anodes are fundamentally limited by large volume variations and repeated fracture of the solid electrolyte interphase (SEI), leading to continuous electrolyte decomposition and rapid loss of lithium (Li) inventory. Herein, we introduce a lithium maleate-derived unsaturated precursor to construct a highly resilient artificial SEI (ASEI) *in situ*. The precursor is compatible with slurry processing and undergoes interfacial reactions during the initial cycles, forming a conformal and mechanically robust interphase. Enabled by this resilient ASEI, the $\text{SiO}_x/\text{C}@\text{ASEI}$ anode shows negligible capacity decay at 1 A g^{-1} , maintains stable cycling under a high areal capacity ($\sim 3.1 \text{ mAh cm}^{-2}$) for 500 cycles, and improves the cycling stability of $\text{SiO}_x/\text{C}@\text{ASEI}||\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ (NCM811) full cells. Postmortem analyses directly verified reduced electrode expansion (2.5% vs. 61.4% swelling), suppressed cracking, the formation of a thin and uniform SEI (19.2 vs. 31.4 nm), and mitigated electrolyte decomposition. By decoupling SiO_x volume variation from repetitive interfacial reconstruction through a resilient ASEI, this study offers a scalable strategy for developing low-swelling SiO_x -based anodes for high-energy Li-ion batteries.

KEYWORDS

SiO_x/C anode, artificial SEI, interfacial reaction, low-expansion electrode, Li inventory loss

1 Introduction

High-energy lithium-ion batteries (LIBs) are essential for next-generation electric vehicles and grid-scale energy storage. However, further increases in energy density are increasingly constrained by the capacity limit of graphite anodes^[1–5]. Silicon (Si)-based anodes are widely regarded as one of the most promising alternatives because of their high theoretical capacity and low operating potential^[6–8]. Among them, SiO_x/C composites have attracted particular interest owing to their improved processability and superior cycling stability compared with pure Si, offering a more practical and manufacturable route toward high-capacity anodes^[9–12]. Nevertheless, the practical implementation of SiO_x/C anodes remains constrained by their pronounced chemo-mechanical degradation during cycling. Repeated lithiation and delithiation induce large volume changes, leading to particle pulverization and electrode swelling^[13–15]. This continuous fracture and reformation of the interphase repeatedly expose fresh reactive surfaces, thereby promoting ongoing electrolyte decomposition and loss of Li inventory^[16–18].

Extensive efforts have been devoted to mitigating the interfacial instability of SiO_x -based anodes, including

structural design, conductive carbon engineering, binder optimization, prelithiation, electrolyte and additive regulation, and artificial interphase construction^[19–24]. However, most of these approaches become less effective under the persistent mechanical stress induced by electrode swelling. Nanostructuring or sophisticated surface coatings can reduce tap density and complicate large-scale manufacturing, while electrolyte additives and prelithiation mainly alleviate the initial irreversible Li loss but cannot fully suppress the long-term rupture and reformation of the solid electrolyte interphase (SEI)^[25–28]. Likewise, *ex situ* artificial interfaces provide only temporary passivation and tend to crack or lose coverage as the electrode undergoes repeated volume changes^[29–33]. Notably, small-molecule polymer precursors containing unsaturated bonds (e.g., maleate-based species) have been reported to form protective artificial SEIs (ASEIs) on graphite anodes via electrochemical polymerization, demonstrating the potential of *in situ* interphase construction^[34–37]. However, extending this strategy to SiO_x -based anodes remains challenging. The interphase must provide effective chemical passivation and sufficient mechanical resilience to accommodate repeated volume changes at the electrode level during cycling^[38–41]. Therefore, there remains a critical need for

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a slurry-compatible, scalable interphase-engineering strategy that can withstand electrode volume variations while maintaining durable passivation, thereby minimizing continuous electrolyte decomposition and Li consumption.

Herein, we introduce a lithium maleate-derived precursor to construct a highly resilient ASEI on SiO_x/C anodes. The unsaturated small-molecule precursor is compatible with conventional slurry processing and can be uniformly dispersed throughout the electrode. During the initial cycles, it undergoes electrochemical polymerization to form a conformal interphase. The resulting ASEI buffers chemo-mechanical stress, maintains interparticle and electrode-current collector contact, and suppresses continuous electrolyte decomposition and Li consumption. Electrochemical evaluations in half-cell and Ni-rich full cells, together with multiscale postmortem analyses, demonstrate that this strategy provides a practical route toward low-swelling SiO_x anodes with reduced Li inventory loss.

2 Experimental section

2.1 Preparation of the $\text{SiO}_x/\text{C}@$ ASEI electrode

The ASEI precursor solution (lithium maleate) was prepared by reacting maleic acid (Aladdin Reagent Co., Ltd.) with Li_2CO_3 (Aladdin Reagent Co., Ltd.) at a molar ratio of 1:1 in deionized water to obtain a 20 wt% aqueous solution. Based on this stoichiometric ratio, the precursor is expected to consist predominantly of dilithium maleate ($\text{Li}_2\text{C}_4\text{H}_2\text{O}_4$). For the pristine SiO_x/C electrode, SiO_x/C powder (specific capacity $\approx 2000 \text{ mAh g}^{-1}$; XFH New Energy Materials Co., Ltd.), conductive carbon black (Guangdong Canrd New Energy Technology Co., Ltd.), a carbon nanotube (CNT) dispersion (1 wt% CNT solids; XFH New Energy Materials Co., Ltd.), and sodium carboxymethylcellulose (CMC-Na; Guangdong Canrd New Energy Technology Co., Ltd.) were mixed in deionized water at a mass ratio of 70:18:2:10 to form a homogeneous slurry. The slurry was uniformly coated onto commercial Cu foil and dried at 80°C for 12 h. The dried electrodes were punched into 12 mm-diameter disks for subsequent cell assembly. The $\text{SiO}_x/\text{C}@$ ASEI electrodes were fabricated using the same procedure, except that the lithium maleate precursor solution was introduced at 1.5 wt% of the total solid content in the electrode slurry. Accordingly, the mass ratio of SiO_x/C powder, lithium maleate precursor (as a slurry additive), conductive carbon black, CNT dispersion (1 wt% CNT solids), and CMC-Na was adjusted to 68.5:1.5:18:2:10. For conventional half-cell testing, the active material loading on the Cu foil was $\sim 0.6 \text{ mg cm}^{-2}$. For high-areal-capacity testing, separate high-loading electrodes were prepared with an active material loading of 1.54 mg cm^{-2} , corresponding to an areal capacity of $\sim 3.1 \text{ mAh cm}^{-2}$.

2.2 Material characterization

Raman spectra were collected using a LabRAM HR800 (Horiba) equipped with a 532 nm laser. X-ray diffraction (XRD) patterns were recorded on a Rigaku D/MAX 2500/PC diffractometer using Cu K α radiation ($\lambda = 0.154 \text{ nm}$). Fourier transform infrared (FTIR) spectra were

acquired using a Bruker Vertex 70 spectrometer. The surface chemical compositions and SEI species were analyzed using X-ray photoelectron spectroscopy (XPS; Physical Electronics PHI 5802) with a monochromatic Al K α X-ray source. For cycled cells, the electrodes were disassembled and transferred to the XPS chamber under an Ar atmosphere to minimize ambient contamination. XPS depth profiling was performed via Ar^+ sputtering (2 kV) over a $2 \text{ mm} \times 2 \text{ mm}$ area. The morphologies and microstructures of the pristine and cycled electrodes were characterized using field-emission scanning electron microscopy (FE-SEM; HITACHI S-4800), cross-section polishing scanning electron microscopy (CP-SEM; Model 1061 SEM Mill), and transmission electron microscopy (TEM; FEI TECNAI G2 F30). An Ar-protected transfer and observation module was employed to reduce air exposure during sample transfer and analysis. Energy-dispersive X-ray spectroscopy (EDS) was used to map the elemental distributions. Three-dimensional surface topography and roughness were characterized via atomic force microscopy (AFM; Bruker Dimension Icon). Time-of-flight secondary ion mass spectrometry (TOF-SIMS) measurements were performed on a NanoTOF-2 instrument (ULVAC-PHI, Japan) equipped with a Bi_3^{2+} primary ion beam operated at 30 kV. Depth profiling was conducted using an Ar^+ sputtering beam (3 keV, 100 nA) at a sputtering rate of 0.1 nm s^{-1} . The analysis and sputtering areas were $100 \mu\text{m} \times 100 \mu\text{m}$ and $400 \mu\text{m} \times 400 \mu\text{m}$, respectively.

2.3 Electrochemical measurements

CR2032 coin cells were assembled in an Ar-filled glovebox with O_2 and H_2O levels below 0.5 ppm and tested using a LAND CT2001A battery testing system. The electrolyte (Suzhou DoDoChem) consisted of 1 mol/L LiPF_6 dissolved in ethylene carbonate/ethyl methyl carbonate/diethyl carbonate (1:1:1, v/v/v) containing 5 wt% fluoroethylene carbonate. A Celgard 2400 membrane was used as the separator. Unless otherwise specified, a fixed electrolyte volume of 50 μL was used for each cell. Galvanostatic charge-discharge cycling was conducted between 0 and 1.5 V. Cyclic voltammetry (CV) measurements were performed over the voltage range of 0.01–2.0 V, and electrochemical impedance spectroscopy (EIS) was conducted over the range of 10 mHz–100 kHz with an AC amplitude of 5 mV using a CHI660 electrochemical workstation. For full-cell assembly, the cathode slurry was prepared by mixing NCM811 (Guangdong Canrd New Energy Technology Co., Ltd.), conductive carbon black, and poly(vinylidene difluoride) (Guangdong Canrd New Energy Technology Co., Ltd.) at a weight ratio of 9:0.5:0.5 in *N*-methyl-2-pyrrolidone (Aladdin Reagent Co., Ltd.). The slurry was coated onto Al foil and dried in a vacuum oven for 12 h. The negative-to-positive capacity (N/P) ratio was controlled at 1.07. The full cells were cycled between 2.8 and 4.2 V for cycling tests (1 C = 180 mA g^{-1}). No additional activation or prelithiation step was applied before full-cell assembly.

3 Results and discussion

The successful incorporation of the ASEI precursor layer

on the SiO_x/C electrode was first verified via structural and chemical characterization. As shown in Fig. 1a, the pristine SiO_x/C electrode displays three prominent Raman bands at 1335, 1587, and 2662 cm^{-1} , corresponding to the D band (disordered carbon), G band (graphitic carbon), and 2D band, respectively. After the introduction of the ASEI precursor, additional weak Raman features appeared at $\sim 1300/1413$ and 1735 cm^{-1} . Because the bands at ~ 1300 and ~ 1413 cm^{-1} overlap with the carbon D-band region, they are regarded as supporting rather than definitive evidence for the presence of the precursor. To further support the assignment, the Raman spectrum of the pure ASEI precursor powder was collected. As shown in Fig. S1 in the Electronic Supplementary Materials (ESM), the precursor shows characteristic bands in similar spectral regions, which are assigned to C–O/carboxylate and C=O-associated vibrational modes of lithium maleate. These results indicate the incorporation of maleate-related oxygen-containing organic species into the SiO_x/C electrode^[42–44]. The phase composition was further examined via XRD (Fig. 1b). Both electrodes show three strong diffraction peaks at 42.5°, 50.5°, and 74.4°, corresponding to the Cu current collector. In addition, the $\text{SiO}_x/\text{C}@$ ASEI electrode exhibits several weak additional diffraction peaks in the low-angle region. The XRD pattern of the pure ASEI precursor powder (Fig. S2 in the ESM) shows characteristic diffraction peaks at 21°–22°, 24°–25°, and 28°–29°, which are in good agreement with these additional peaks observed for the $\text{SiO}_x/\text{C}@$ ASEI electrode. These results further support the successful incorporation of the lithium maleate precursor during slurry processing^[44].

FTIR spectra provide complementary evidence for the presence of the introduced precursor (Fig. S3 in the ESM). After incorporation of the ASEI precursor, the $\text{SiO}_x/\text{C}@$ ASEI electrode shows enhanced absorption bands related to oxygen-containing organic functional groups. The broad absorption bands in the ranges of 2350–2560

and 2900–3100 cm^{-1} are attributable to O–H-related vibrations of adsorbed water and surface hydroxyl species, respectively. The absorption bands in the 1600–1710 cm^{-1} region are assigned to maleate-related vibrational modes, where contributions from C=C, carbonyl, and carboxylate groups may overlap^[44,45]. These observations further support the presence of maleate-derived functional groups in the modified electrode. XPS was then conducted to investigate the surface chemical states and bonding environments. For the pristine SiO_x/C powder, the high-resolution Si 2p spectrum (Fig. S4a in the ESM) can be deconvoluted into four components centered at ~ 100.0 , 102.3, 103.9, and 105.2 eV, corresponding to Si^+ , Si^{2+} , Si^{3+} , and Si^{4+} , respectively, reflecting the typical mixed-valence nature of SiO_x ^[46,47]. The C 1s spectrum of the SiO_x/C powder (Fig. S4b in the ESM) contains peaks assigned to C–C/C=C/C–H (284.8 eV), C–O (286.2 eV), and C=O (288.4 eV). The O 1s spectrum (Fig. S4c in the ESM) can be fitted with components corresponding to oxygen-containing species, including carbonyl and Si–O groups, further indicating the abundance of oxygen-containing surface functionalities. For the electrodes, the C 1s spectrum of the pristine SiO_x/C electrode (Fig. S5 in the ESM) can be deconvoluted into five components assigned to C–C/C=C/C–H (284.8 eV), C–O (286.1 eV), C=O (287.3 eV), O=C–O/–COO (288.8 eV), and CO_3^{2-} (290.5 eV)^[48]. In comparison, the high-resolution C 1s spectrum of the ASEI precursor powder (Fig. 1c) shows a dominant O=C–O/–COO component at 288.6 eV, consistent with the lithium carboxylate structure of lithium maleate^[20]. After incorporation of the ASEI precursor, the C 1s spectrum of the $\text{SiO}_x/\text{C}@$ ASEI electrode (Fig. 1d) shows increased contributions from oxygen-containing carbon species. As summarized in the detailed C 1s peak-fitting results (Table S1 in the ESM), the relative area of the C–O component increases from 14.1% for the pristine SiO_x/C electrode to 16.7% for the $\text{SiO}_x/\text{C}@$ ASEI electrode,

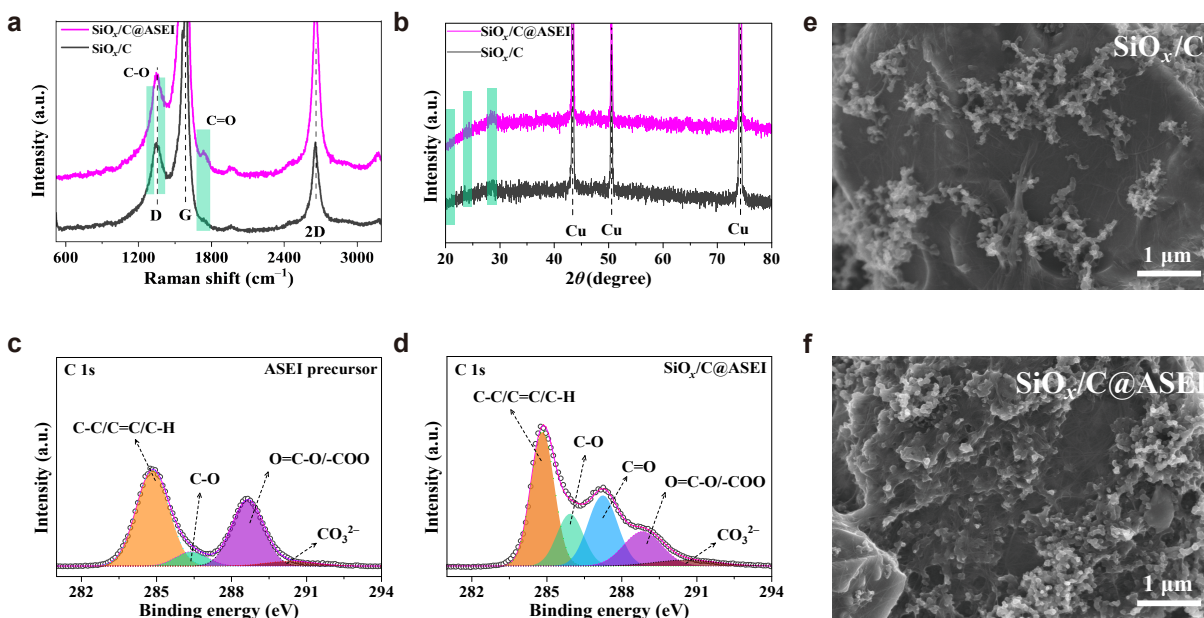


Figure 1 Structural and morphological characterization of the SiO_x/C and $\text{SiO}_x/\text{C}@$ ASEI electrodes. Raman spectra (a) and XRD patterns (b) of SiO_x/C and $\text{SiO}_x/\text{C}@$ ASEI electrodes. High-resolution C 1s XPS spectra of the ASEI precursor powder (c) and the $\text{SiO}_x/\text{C}@$ ASEI electrode (d). SEM images of the SiO_x/C (e) and the $\text{SiO}_x/\text{C}@$ ASEI (f) electrodes.

whereas that of $\text{O}=\text{C}-\text{O}-\text{COO}$ increases from 12.3% to 15.8%. Moreover, the O 1s spectra (Fig. S6 in the ESM) show enhanced contributions from $\text{O}=\text{C}-\text{O}-\text{COO}$ and $\text{C}-\text{O}$ species in the $\text{SiO}_x/\text{C}@\text{ASEI}$ electrode. These changes confirm the increased presence of oxygen-containing lithium carboxylate species after precursor incorporation.

Taken together, the Raman, XRD, FTIR, and XPS results demonstrate the successful incorporation of the lithium maleate precursor into the SiO_x/C electrode. The electrode morphology and the uniformity of the precursor distribution on the electrode were further examined using SEM and elemental mapping. As shown in Fig. S7 in the ESM, the pristine SiO_x/C powder consists of particles with sizes of 2–10 μm , and the corresponding elemental maps confirm the homogeneous distribution of Si, C, and O. For the pristine SiO_x/C electrode, the conductive additives (carbon black and CNTs) are uniformly distributed throughout the electrode matrix (Fig. 1e and Fig. S8 in the ESM), forming a continuous conductive network. After incorporation of the ASEI precursor, the $\text{SiO}_x/\text{C}@\text{ASEI}$ electrode shows a more uniform surface morphology, with nanoscale deposits evenly distributed across the electrode surface (Fig. 1f and Fig. S9 in the ESM). The elemental

maps of Si, C, and O remain homogeneous after precursor incorporation, suggesting that the lithium maleate precursor is uniformly distributed within the electrode without obvious elemental segregation. Notably, the ASEI in this study is not a thick coating layer that can be directly identified via SEM before cycling. Instead, it is a functional artificial interphase formed *in situ* from the lithium maleate precursor incorporated into the slurry during the initial electrochemical cycles, as evidenced by the electrochemical performance and post-cycling interfacial characterization. The incorporation of this precursor is expected to reduce the exposure of reactive sites on the SiO_x/C surface and guide subsequent interfacial evolution toward the formation of a highly stable and resilient ASEI.

The electrochemical effects of the ASEI were evaluated by comparing the SiO_x/C and $\text{SiO}_x/\text{C}@\text{ASEI}$ electrodes in both half and full cells. The first-cycle CV curves (Fig. 2a) exhibit an additional electrochemical feature at 1.5–1.7 V, indicating an early interfacial reaction associated with the lithium maleate precursor. Because CV alone cannot directly confirm polymerization of the C=C bonds, FTIR spectra were collected before and after the initial cycle. As shown in Fig. S10 in the ESM, the electrolyte-exposed

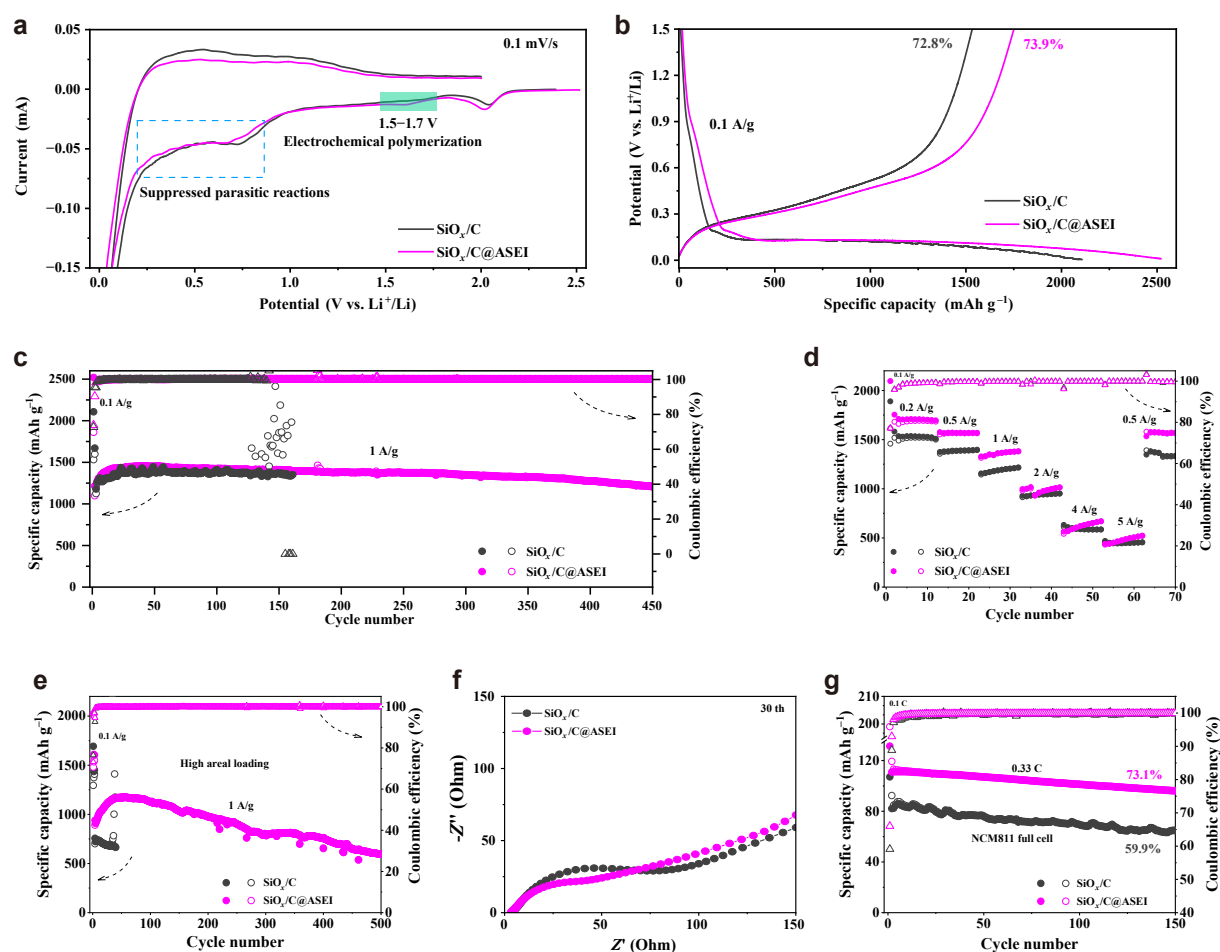


Figure 2 Electrochemical performance of SiO_x/C and $\text{SiO}_x/\text{C}@\text{ASEI}$ electrodes. (a) Initial CV curves at 0.1 mV s⁻¹. (b) Initial charge-discharge profiles at 0.1 A g⁻¹. (c) Long-term cycling performance and corresponding Coulombic efficiency (CE) at 1 A g⁻¹. (d) Rate capability and corresponding CE from 0.2 to 5 A g⁻¹. (e) Cycling performance and corresponding CE of high-areal-loading electrodes at 1 A g⁻¹. (f) EIS spectra of SiO_x/C and $\text{SiO}_x/\text{C}@\text{ASEI}$ electrodes after 30 cycles. (g) Long-term cycling performance and corresponding CE of $\text{SiO}_x/\text{C}||\text{NCM811}$ and $\text{SiO}_x/\text{C}@\text{ASEI}||\text{NCM811}$ full cells at 0.33 C.

$\text{SiO}_x/\text{C}@\text{ASEI}$ electrode shows a more evident broad absorption band at $\sim 1620\text{ cm}^{-1}$ than the electrolyte-exposed SiO_x/C electrode. This band is attributable to $\text{C}=\text{C}$ /carboxylate-associated vibrational modes of the lithium maleate precursor. After the initial cycle, the intensity of this band decreases markedly, suggesting the consumption or transformation of $\text{C}=\text{C}$ -containing groups during the early stages of interfacial evolution. Combined with the CV results, these observations support the involvement of lithium maleate in ASEI formation, likely through interfacial polymerization or oligomerization of its unsaturated structure. In addition, the $\text{SiO}_x/\text{C}@\text{ASEI}$ electrode shows a low cathodic current in the low-potential region (highlighted in Fig. 2a), indicating suppressed parasitic reactions and reduced irreversible Li consumption during the initial formation of the interphase. Consistent with this observation, the initial charge–discharge profiles (Fig. 2b) indicate improved initial reversibility for the $\text{SiO}_x/\text{C}@\text{ASEI}$ electrode. Accordingly, the $\text{SiO}_x/\text{C}@\text{ASEI}$ electrode delivers a higher reversible charge capacity of 1750 mAh g^{-1} and an initial Coulombic efficiency (CE) of 73.9%, compared with 1530 mAh g^{-1} and 72.8%, respectively, for the pristine SiO_x/C electrode. These results suggest that the ASEI precursor layer reduces irreversible Li consumption by promoting the formation of a highly uniform and stable interphase while suppressing uncontrolled surface reduction reactions. The long-term cycling performance was subsequently evaluated at a current density of 1 A g^{-1} (Fig. 2c). To optimize the lithium maleate precursor content, electrodes containing different amounts of the precursor were systematically compared. As shown in Fig. S11 in the ESM, the electrochemical performance strongly depends on the precursor content. Compared with the pristine SiO_x/C electrode, the electrode containing 1.0 wt% precursor shows improved cycling stability; however, its CE begins to fluctuate after prolonged cycling, indicating insufficient long-term interfacial stabilization. By contrast, increasing the precursor content to 2.0 wt% leads to a noticeable decrease in reversible capacity, suggesting that excessive precursor introduces an increased fraction of electrochemically inactive material and exacerbates interfacial polarization. Therefore, a precursor content of 1.5 wt% was selected as the optimum because it provides the best balance between reversible capacity and long-term cycling stability. After the initial cycle, the $\text{SiO}_x/\text{C}@\text{ASEI}$ electrode delivers a relatively lower specific capacity of $\sim 1098.5\text{ mAh g}^{-1}$, which can reasonably be attributed to an electrode activation process. This activation is likely associated with progressive electrolyte wetting and interfacial stabilization, the gradual utilization of electrochemically accessible active sites within the composite electrode, and the establishment of efficient ion and electron transport pathways across the interphase. Following this activation, the reversible specific capacity increases and remains as high as 1205.3 mAh g^{-1} even after 450 cycles, indicating that the ASEI-regulated interphase effectively preserves electrode integrity and electrochemical accessibility during prolonged cycling. By contrast, the pristine SiO_x/C electrode displays pronounced CE fluctuations accompanied by a progressive increase in the apparent reversible capacity during cycling. This behavior is likely caused by multi-

ple concurrent processes, including gradual electrode activation, electrolyte wetting, and changes in the local current distribution. However, considering the pronounced CE fluctuations together with the severe structural and interfacial degradation revealed by the postmortem analyses, repeated SEI fracture and reconstruction, persistent electrolyte decomposition, and particle pulverization resulting from large volume variations are also likely to contribute significantly to this behavior.

To further clarify the role of the $\text{C}=\text{C}$ bond in lithium maleate, lithium succinate (LS) was used as a saturated lithium dicarboxylate control. LS contains lithium carboxylate groups but lacks a polymerizable $\text{C}=\text{C}$ bond. As shown in Fig. S12 in the ESM, the $\text{SiO}_x/\text{C}@LS$ electrode does not show improved cycling stability compared with the pristine SiO_x/C electrode. Instead, it delivers a low capacity and undergoes rapid degradation within 150 cycles. This result indicates that the lithium carboxylate groups are insufficient to construct an effective ASEI. Without the $\text{C}=\text{C}$ bond, LS cannot undergo early interfacial polymerization or oligomerization and may instead form an ineffective or overly resistive interfacial layer. Therefore, the superior cycling stability of the $\text{SiO}_x/\text{C}@ASEI$ electrode highlights the critical role of the $\text{C}=\text{C}$ bond in enabling the formation of a stable and mechanically compliant artificial interphase. Moreover, the corresponding voltage profiles (Fig. S13 in the ESM) further show enhanced reversibility for the $\text{SiO}_x/\text{C}@ASEI$ electrode during cycling, consistent with the formation of a stabilized interphase that mitigates mechanically induced interfacial degradation. Rate capability measurements were further conducted to evaluate the ion/electron transport behavior of the electrodes (Fig. 2d). The $\text{SiO}_x/\text{C}@ASEI$ electrode shows comparable to slightly improved rate performance relative to the pristine SiO_x/C electrode over a current density range of $0.2\text{--}5\text{ A g}^{-1}$, retaining capacities of ~ 1707 and $\sim 643\text{ mAh g}^{-1}$ at 0.2 and 4 A g^{-1} , respectively. Although the capacity difference in the short-term rate test is not pronounced, the corresponding voltage profiles (Fig. S14 in the ESM) show reduced polarization for the ASEI-modified electrode. This result suggests that the ASEI mainly improves interfacial charge-transfer kinetics and reduces polarization, rather than substantially changing the intrinsic bulk transport of the SiO_x/C electrode. This interpretation is further supported by the high-current-density cycling test at 2 A g^{-1} (Fig. S15 in the ESM). Under this condition, the $\text{SiO}_x/\text{C}@ASEI$ electrode exhibits negligible capacity fading over 250 cycles, whereas the pristine SiO_x/C electrode undergoes continuous capacity decay and becomes unstable within ~ 100 cycles, further confirming the superior interfacial resilience enabled by ASEI treatment. Importantly, the ASEI strategy also shows strong potential under practical operating conditions. For high-loading electrodes with an areal capacity of $\sim 3.1\text{ mAh cm}^{-2}$, the $\text{SiO}_x/\text{C}@ASEI$ electrode maintains a reversible specific capacity of $\sim 600\text{ mAh g}^{-1}$ after 500 cycles under extended cycling conditions (Fig. 2e). By contrast, the SiO_x/C electrode undergoes severe capacity degradation within 40 cycles. This result indicates that the ASEI is particularly effective under high-stress conditions, where SiO_x/C electrodes are more susceptible to cracking, loss of electrical

contact, and repeated interphase reconstruction. EIS measurements were conducted to probe the evolution of interfacial resistance. Compared with the SiO_x/C electrode, the $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode exhibits a smaller semicircle diameter in the high-frequency region after the first cycle (Fig. S16 in the ESM) and after prolonged cycling (Fig. 2f), confirming reduced charge-transfer resistance and more favorable interfacial kinetics enabled by the ASEI coating. The consistently low impedance from the initial stage to long-term cycling demonstrates that the ASEI-derived interphase suppresses continuous interfacial degradation, which is a key contributor to progressive polarization and Li inventory loss in SiO_x -based anodes. To further clarify the role of lithium carboxylate groups in the precursor, a maleic acid-based control electrode was also examined after cycling. As shown in Fig. S17 in the ESM, the $\text{SiO}_x/\text{C}@ \text{maleic acid}$ electrode exhibits substantially higher charge-transfer resistance than the SiO_x/C and $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrodes. This result indicates that a Li-free organic acid-derived interface is unfavorable for Li^+ transport. By contrast, the low resistance of $\text{SiO}_x/\text{C}@ \text{ASEI}$ suggests that the lithium carboxylate groups in lithium maleate are important for constructing a Li^+ transport-favorable artificial interphase. The interfacial stabilization effect of the ASEI was further validated in full cells employing NCM811 as the cathode. As shown in Fig. S18a in the ESM, the NCM811|| $\text{SiO}_x/\text{C}@ \text{ASEI}$ full cell exhibits smaller voltage hysteresis and a higher initial CE of 66% than the corresponding NCM811|| SiO_x/C counterpart (59.1%) at 0.1 C, indicating reduced interfacial polarization during both lithiation and delithiation processes. This interfacial advantage translates into enhanced long-term full-cell stability. Under 0.33 C cycling conditions (Fig. 2g), full cells employing the $\text{SiO}_x/\text{C}@ \text{ASEI}$ anode deliver higher reversible capacity and a slower decay trend, retaining 73.1% of the initial capacity after 150 cycles, whereas cells using the SiO_x/C anode

retain only 59.9%. The voltage profiles during cycling further confirm reduced polarization in the $\text{SiO}_x/\text{C}@ \text{ASEI}$ -based full cell (Fig. S18b in the ESM), consistent with the low impedance values and suppressed continuous electrolyte decomposition. Overall, these results demonstrate that the lithium maleate-derived ASEI precursor enables rapid interphase regulation during the initial cycling stage and maintains an electrochemically robust interphase throughout cycling. Such an interphase can better accommodate repeated SiO_x/C volume changes, thereby reduce continuous SEI reconstruction and minimizing active Li consumption, which collectively improves half-cell durability and full-cell capacity retention.

To elucidate the benefits of the ASEI in regulating electrode-level interfacial expansion, the morphological and structural evolution of SiO_x/C and $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrodes after 30 cycles was systematically investigated using SEM, EDS mapping, CP-SEM, and large-area AFM measurements. For the cycled SiO_x/C electrode, pronounced surface cracking is clearly observed in the top-view SEM image (Fig. S19a in the ESM), accompanied by abundant flaky deposits covering the electrode surface. The corresponding elemental maps (Figs. S19b–S19e in the ESM) reveal the presence of F-containing species distributed across the surface, suggesting that these deposits largely originate from electrolyte decomposition products and fractured SEI fragments. Meanwhile, the gaps between adjacent SiO_x particles become increasingly evident, implying progressive loss of particle-particle contact during repeated expansion and contraction. This structural degradation is further corroborated by cross-sectional analysis. The pristine SiO_x/C electrode exhibits a thickness of $\sim 8.8 \mu\text{m}$ (Fig. 3a), whereas after cycling, the thickness increases to $14.2 \mu\text{m}$ according to CP-SEM analysis (Fig. 3b), corresponding to a high electrode swelling ratio of 61.4%. To improve the reliability of the thickness analysis, additional CP-SEM images were collected from

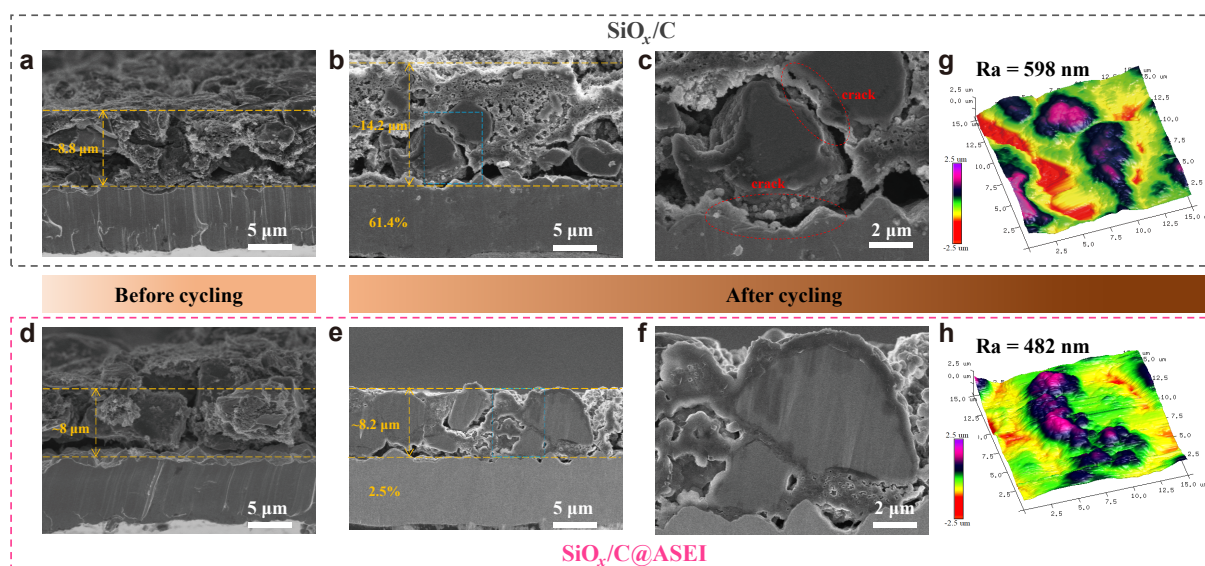


Figure 3 Electrode-level interfacial expansion and morphological evolution. (a) Cross-sectional SEM image of the pristine SiO_x/C electrode. (b) CP-SEM image and (c) corresponding magnified image of the cycled SiO_x/C electrode after 30 cycles. (d) Cross-sectional SEM image of the pristine $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode. (e) CP-SEM image and (f) corresponding magnified image of the cycled $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode after 30 cycles. AFM surface morphology images of cycled (g) SiO_x/C and (h) $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrodes after 30 cycles.

different regions of the cycled electrode. As shown in Fig. S20 in the ESM, the SiO_x/C electrode shows thicknesses of 14.4, 13.8, and 15.9 μm , corresponding to expansion ratios of 63.6%, 56.9%, and 80.9%, respectively. Based on these four measured regions, the average thickness expansion ratio of the cycled SiO_x/C electrode is calculated to be $65.7\% \pm 10.5\%$. Moreover, the magnified CP-SEM image (Fig. 3c) shows prominent cracks both between particles and at the particle-current collector interface, indicating the disruption of electrical percolation pathways. Elemental mapping of the cross-sectional region (Fig. S21 in the ESM) further shows that the interparticle voids are filled with decomposition products, notably F-containing species, leading to a porous and mechanically weakened internal structure. Such severe swelling-induced structural disruption and persistent reactions result in inferior reversibility and cycling stability of the SiO_x/C electrode, as repeated SEI fracture and reconstruction continuously consume active Li. By contrast, the cycled $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode exhibits markedly improved morphological integrity. The top-view SEM image (Fig. S22a in the ESM) shows a clean surface with substantially reduced accumulation of decomposition products, and the corresponding EDS mappings (Figs. S22b–S22e in the ESM) indicate that the SiO_x/C active particles remain clearly distinguishable with better-maintained particle–particle contact. The pristine $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode has a thickness of $\sim 8.0 \mu\text{m}$ (Fig. 3d). After cycling, its thickness only slightly increases to $\sim 8.2 \mu\text{m}$ (Fig. 3e), corresponding to a swelling ratio of 2.5%. Besides the representative region showing an expansion ratio of 2.5%, measurements from additional regions reveal thicknesses of 8.1, 8.3, and 8.7 μm , corresponding to expansion ratios of 1.25%, 3.75%, and 8.75%, respectively (Fig. S23 in the ESM). The average expansion ratio is only $4.1\% \pm 3.3\%$, confirming that the lithium maleate-derived ASEI effectively mitigates electrode swelling during repeated lithiation–delithiation. Consistently, the CP-SEM image of the cycled $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode (Fig. 3f) shows no obvious inter-particle cracking, and both particle–particle contacts and particle–current collector interfaces remain intact. The corresponding elemental mapping (Fig. S24 in the ESM) does not show extensive voids filled with decomposition products, suggesting that the ASEI-regulated interphase suppresses electrolyte decomposition and prevents the formation of a loose, porous internal structure. Overall, these results indicate that the ASEI enables the formation of a mechanically resilient interphase on SiO_x/C particles. This interphase accommodates volume changes, preserves electrical contact throughout the electrode, and mitigates continuous Li loss associated with repeated interphase reconstruction.

To further validate electrode-level structural evolution over a large lateral scale, AFM was performed on cycled electrodes over $15 \mu\text{m} \times 15 \mu\text{m}$ regions (Figs. 3g and 3h). The representative cycled SiO_x/C electrode exhibits a distinctly roughened and uneven surface with a roughness of $R_a = 598 \text{ nm}$ (Fig. 3g), consistent with the extensive cracking and decomposition-product accumulation observed via SEM. Additional AFM measurements from different regions show similar R_a values of 587 and 580 nm (Fig. S25 in the ESM), confirming severe surface

roughening of the pristine electrode after cycling. By comparison, the cycled $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode shows a smoother surface with a reduced roughness of $R_a = 482 \text{ nm}$ in the representative region (Fig. 3h), while additional regions show R_a values of 495 and 514 nm (Fig. S26 in the ESM). Based on these measurements, the average R_a value decreases from $588 \pm 9 \text{ nm}$ for the SiO_x/C electrode to $497 \pm 16 \text{ nm}$ for the $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode. Notably, the AFM scans were collected over relatively large areas of $15 \mu\text{m} \times 15 \mu\text{m}$, enabling a more comprehensive evaluation of surface roughening than smaller areas such as $5 \mu\text{m} \times 5 \mu\text{m}$ that are often used in AFM analyses. The lower roughness of the $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode indicates suppressed surface disruption and more uniform interfacial evolution compared with the SiO_x/C electrode.

Based on the above results, these multiscale characterizations consistently demonstrate that the ASEI strategy significantly suppresses electrode swelling and structural degradation of SiO_x/C electrodes during cycling. By maintaining internal electrical contact and reducing the accumulation of electrolyte decomposition products, the ASEI-treated electrode effectively limits the buildup of electrochemically inactive species and minimizes continuous Li consumption, in agreement with the enhanced CE stability and cycling durability observed from electrochemical measurements.

To further elucidate the role of the ASEI strategy in regulating SEI chemistry and structural evolution during cycling, depth-resolved XPS, three-dimensional TOF-SIMS, and TEM analyses were performed on electrodes after 50 cycles. For the cycled SiO_x/C electrode, the surface C 1s spectrum (0 nm, Fig. 4a) can be deconvoluted into four components located at 283.1 eV (C–Li), 284.8 eV (C–C/C–H), $\sim 287.0 \text{ eV}$ (C–O), and $\sim 289.4 \text{ eV}$ (C=O). Notably, the C–Li component rapidly disappears with increasing sputtering depth, indicating that this low-binding-energy feature is confined to the outermost SEI region rather than the bulk electrode matrix. Such surface-localized Li–C/organolithium-like species are typically associated with severe interfacial reduction and unstable organic fragments generated during uncontrolled electrolyte decomposition on electronically conductive but defective carbon domains^[49,50]. Meanwhile, a carbonate-related component assigned to $\text{Li}_2\text{CO}_3/\text{ROCO}_2\text{Li}$ becomes evident at higher binding energy ($\sim 290 \text{ eV}$) with increasing sputtering depth^[51], consistent with the evolution of the O 1s spectra (Fig. 4b), where the $\text{Li}_2\text{CO}_3/\text{ROCO}_2\text{Li}$ -related signal ($\sim 530.5 \text{ eV}$) becomes increasingly pronounced upon sputtering. These features suggest that, for the SiO_x/C electrode, continuous SEI fracture and reconstruction lead to the accumulation of decomposition products on the surface and the retention of a considerable amount of electrolyte-derived species within the SEI. By contrast, the cycled $\text{SiO}_x/\text{C}@ \text{ASEI}$ electrode shows markedly different depth-dependent chemical characteristics. First, the C–Li component is absent in the C 1s spectra (Fig. 4a), indicating that the ASEI effectively suppresses the formation of unstable surface Li–C/organolithium-like species. Second, the electrolyte decomposition-related $\text{Li}_2\text{CO}_3/\text{ROCO}_2\text{Li}$ contribution becomes significantly weaker and diminishes with increasing sputtering depth in both the C 1s and O 1s spectra (Figs. 4a and 4b), suggesting substantially reduced

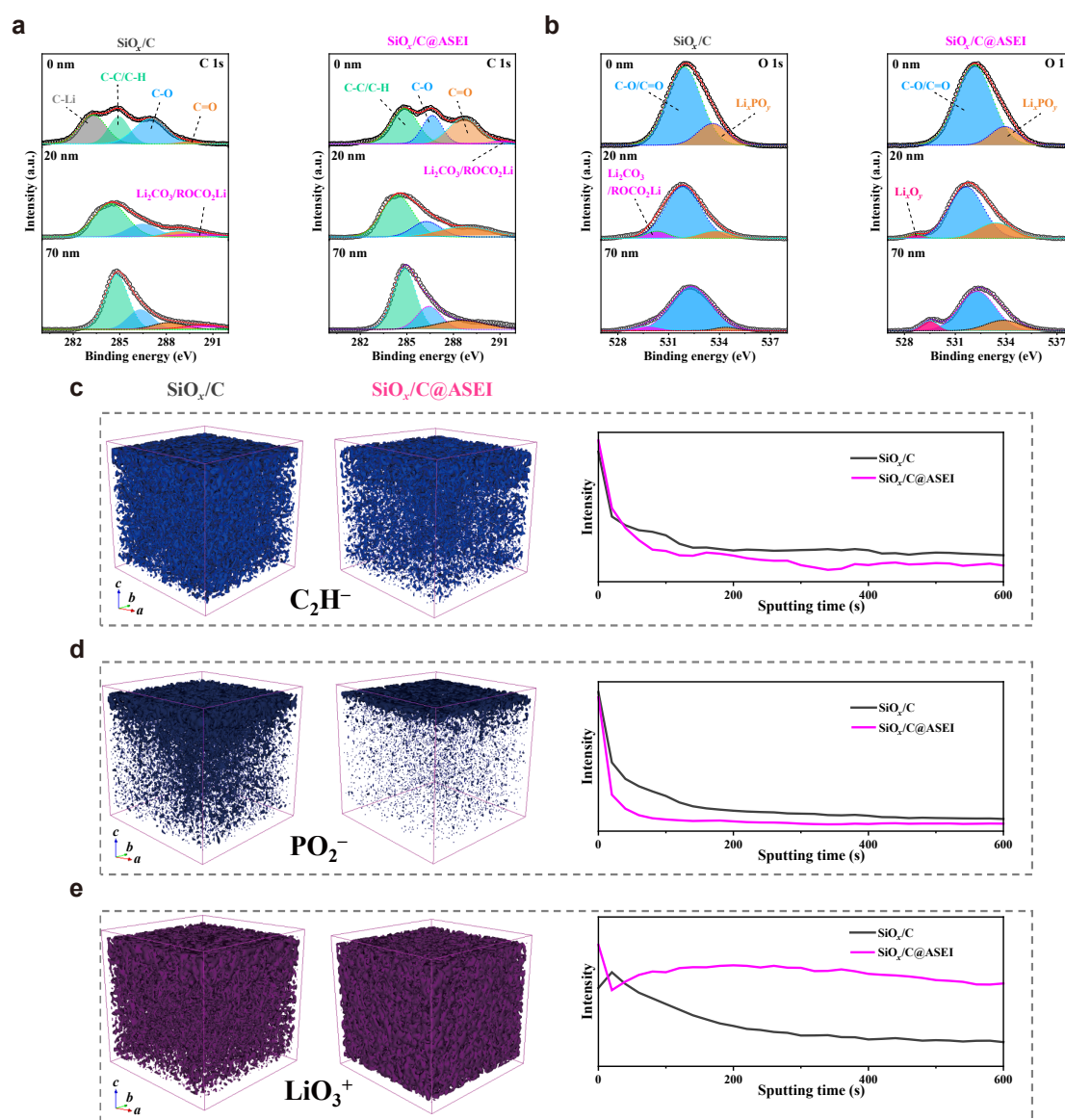


Figure 4 Interphase composition and spatial distribution of SiO_x/C and $\text{SiO}_x/\text{C}@ASEI$ electrodes after cycling. Depth-resolved XPS spectra of C 1s (a) and O 1s (b) for the SiO_x/C and $\text{SiO}_x/\text{C}@ASEI$ electrodes after 30 cycles. Three-dimensional TOF-SIMS maps and corresponding depth profiles of C_2H^- (c), PO_2^- (d), and LiO_3^+ (e) for the SiO_x/C and $\text{SiO}_x/\text{C}@ASEI$ electrodes after 30 cycles.

electrolyte decomposition within the inner SEI region. Importantly, an additional O 1s component centered at ~ 529 eV emerges upon sputtering of the $\text{SiO}_x/\text{C}@ASEI$ electrode (Fig. 4b), which is attributable to Li_xO_y -type species, implying the formation of an oxygen-rich inner SEI. Such an oxygen-rich inner interphase is generally considered beneficial for improving interfacial robustness and maintaining stable ion-transport pathways, especially when mechanical stress caused by SiO_x volume changes is mitigated. The suppressed salt-decomposition signatures after ASEI treatment are further confirmed by the P 2p and F 1s depth profiles. As shown in Fig. S27 in the ESM, the intensities of P–O/P=O (~ 134 eV) and P–F (~ 136.5 eV) components of the $\text{SiO}_x/\text{C}@ASEI$ electrode are significantly lower than those of the SiO_x/C electrode throughout the measured depth range, indicating suppressed LiPF_6 -derived decomposition^[25]. Consistently, the F 1s spectra (Fig. S28) show a reduced LiF contribu-

tion (~ 684.8 eV) for the $\text{SiO}_x/\text{C}@ASEI$ electrode compared with the SiO_x/C electrode, suggesting that ASEI suppresses excessive F-containing decomposition and prevents the formation of a highly F-enriched interphase. Importantly, extensive and nonuniform accumulation of F-rich decomposition products is typically associated with severe continuous salt decomposition and repeated interphase reconstruction, which are detrimental to long-term cycling stability and Li inventory retention. The above results are further supported by 3D TOF-SIMS mapping and depth profiling. Compared with the SiO_x/C electrode, the $\text{SiO}_x/\text{C}@ASEI$ electrode exhibits markedly lower C_2H^- (Fig. 4c), PO_2^- (Fig. 4d), and PO_3^- (Fig. S29 in the ESM) signal intensities throughout the depth profile, and these signals decrease more rapidly with increasing sputtering depth. Because C_2H^- and phosphate-related fragments are characteristic of organic SEI residues and LiPF_6 -derived decomposition products, respectively, these results indi-

cate that the ASEI suppresses electrolyte and salt decomposition within the SEI. Conversely, the oxygen-rich inorganic SEI signature represented by LiO_3^+ shows a distinctly different trend (Fig. 4e)^[51]. For the $\text{SiO}_x/\text{C}@$ ASEI electrode, the LiO_3^+ signal remains consistently stronger throughout the sputtering depth, whereas it decreases more clearly for the pristine SiO_x/C electrode. This comparison indicates that the ASEI promotes the formation of an oxygen-rich inorganic interphase in the inner region, consistent with the Li_xO_y species identified in the depth-resolved XPS O 1s spectra^[52]. Together, these results indicate that the ASEI reduces the heterogeneous distribution of decomposition products while promoting the formation of a more uniform, oxygen-rich, inorganic-dominated inner interphase. TEM and EDS were further employed to directly visualize the SEI thickness and elemental distribution after cycling. As shown in Fig. S30a in the ESM, the cycled SiO_x/C electrode forms a relatively thick and irregular SEI with a thickness of ~ 31.4 nm. The corresponding high-angle annular dark-field scanning TEM and elemental maps (Fig. S31 in the ESM) reveal extensive C- and F-rich surface coverage and a nonuniform elemental distribution, indicating the formation of a heterogeneous SEI caused by persistent decomposition and repeated interface reconstruction. By contrast, the $\text{SiO}_x/\text{C}@$ ASEI electrode exhibits a much thinner and smoother SEI with a thickness of only ~ 19.2 nm (Fig. S30b in the ESM). The scanning TEM elemental maps (Fig. S32 in the ESM) show a more uniform elemental distribution and a notably reduced F-rich contribution, consistent with suppressed salt decomposition. Such an interphase is beneficial to facilitate ion transport while maintaining mechanical integrity, especially for SiO_x -based anodes that undergo repeated volume changes. Overall, these results indicate that ASEI suppresses interfacial side reactions and promotes the formation of a thin, highly uniform SEI with an oxygen-rich inner region.

Herein, the ASEI does not refer to a single, well-defined compound but rather to an artificial interphase formed from the lithium maleate precursor during electrochemical cycling. The precursor is expected to consist predominantly of $\text{Li}_2\text{C}_4\text{H}_2\text{O}_4$, an unsaturated organic lithium salt containing both a C=C bond and lithium carboxylate groups. The unsaturated backbone is expected to facilitate the formation of a more compliant organic framework, while the lithium carboxylate groups are expected to render the interphase less electronically insulating than a conventional organic layer. Therefore, the ASEI can be regarded as an organic lithium salt-derived artificial interphase that accommodates volume change-induced stress, suppresses side reactions, and reduces repeated SEI fracture and reconstruction during long-term cycling. For comparison, representative interphase-regulation strategies reported for SiO_x anodes are summarized in Table S2 in the ESM^[25,29,31,38,39]. As shown, the lithium maleate-derived ASEI developed in this study achieves low electrode swelling, stable long-term cycling with high reversible specific capacity, and the formation of a relatively thin SEI.

4 Conclusion

Herein, a lithium maleate-derived ASEI strategy is

proposed to stabilize practical SiO_x/C anodes by simultaneously regulating interfacial chemistry and preserving electrode-level mechanical integrity. Benefiting from the ASEI, the $\text{SiO}_x/\text{C}@$ ASEI electrode exhibits improved initial reversibility and markedly enhanced cycling stability under a wide range of operating conditions, including long-term cycling, high-rate operation, high-current cycling, and high-areal-loading conditions. It further enables improved cycling durability and reduced polarization in NCM811 full cells. Postmortem analyses at the electrode level directly verify the low-expansion behavior of the $\text{SiO}_x/\text{C}@$ ASEI electrode, with an electrode swelling ratio of only 2.5% compared with 61.4% for the pristine SiO_x/C electrode. Depth-resolved XPS, 3D TOF-SIMS, and TEM analyses further reveal the formation of a thin and uniform SEI (19.2 nm vs. 31.4 nm), accompanied by suppressed electrolyte decomposition and an oxygen-rich inner region. Overall, the ASEI effectively mitigates repeated SEI fracture and reconstruction induced by SiO_x volume changes, thereby suppressing continuous parasitic reactions and minimizing active Li consumption. This study highlights a scalable interphase-engineering strategy for developing low-swelling, low-Li-loss SiO_x -based anodes for high-energy LIBs.

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Author contribution statement

Donghui Zhao is responsible for conducting the experiments, data analysis, and writing the original draft. Yu Bai is responsible for part of the data processing. Hongfang Song is responsible for part of the manuscript writing. Baohua Li is responsible for supervision, project administration, and funding acquisition. Yunshan Zheng is responsible for supervision, resources, and reviewing & editing the manuscript. All the authors have approved the final manuscript.

Data availability

The data that support the findings of this study are available in the supplementary material of this article.

Declaration of competing interest

Donghui Zhao, Yu Bai, Hongfang Song are the employees of Fujian XFH New Energy Materials Co., Ltd. Baohua Li is an Associate Editor of this journal, but he was not involved in the peer review or decision of this article. The other author declares no conflicts of interest.

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Use of AI statement

During the preparation of this work, the authors used ChatGPT for language polishing in order to improve the language and readability. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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