



In situ reflectance spectroscopy for state-of-lithiation mapping of porous silicon anodes

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HIGHLIGHTS

- *In situ* and ex situ reflectance spectroscopy evaluate SoL in porous Si anodes.
- Drift correction enables stable operando optical tracking during lithiation.
- Two-dimensional mapping reveals areal heterogeneity and homogenization.
- The method enables contactless quality control for prelithiation processes.
- Roughness and bandgap closure explain reflectance loss by scattering and absorption.

ARTICLE INFO

Keywords:

Spectroscopy
State of lithiation
In situ measurement
Silicon anode
Prelithiation
Quality control
In-line inspection

ABSTRACT

Silicon is a promising candidate for next-generation lithium-ion battery anodes due to its superior gravimetric capacity, derived from its alloying reaction with lithium. Given the correlation between electrochemical and optical properties, optical inspection provides a non-invasive method for probing lithiation dynamics. In this study, the state of lithiation (SoL) was systematically linked with optical response through both ex situ and *in situ* measurements. Reflectance consistently decreased as lithium content increased within the silicon lattice, a trend validated across multiple experimental setups. For instance, a spatial lithium gradient, introduced by varying electrode sizes in coin cells, was successfully visualized via reflectance mapping. This approach also suggests a scalable quality-control strategy for prelithiation, the process of alloying silicon with lithium. The areal lithiation uniformity of prelithiated silicon anodes was evaluated, demonstrating the potential of this spectroscopic method for real-time inspection in roll-to-roll manufacturing. Additionally, the physical mechanism of reflectance changes was explored. Surface roughness measurements and density functional theory (DFT) calculations indicate that reduced reflectance at higher SoL is due to increased light scattering from particle expansion and bandgap narrowing. Overall, this spectroscopic method enables both a fundamental understanding of silicon lithiation and a practical framework for industrial-scale quality control.

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<https://doi.org/10.1016/j.jpowsour.2026.240342>

Received 20 February 2026; Received in revised form 28 April 2026; Accepted 6 May 2026

Available online 14 May 2026

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1. Introduction

As global warming becomes an increasingly urgent challenge, developing effective strategies for carbon neutrality has become critical [1,2]. Large-scale electrification of transportation is an indispensable step for cutting emissions, but its widespread adoption depends on overcoming key battery performance challenges [3,4]. Balancing high energy density, fast charging, and long cycle life remains a barrier to mass-market penetration of electric vehicles (EVs). Although lithium-ion batteries (LIBs) are the leading mobile energy storage technology, further improvements in driving range, power delivery, and charging speed are needed to support continued growth.

One of the most effective ways to enhance battery performance is through material innovation [5,6]. Silicon has emerged as a promising anode material due to its high theoretical gravimetric (3579 mAh g^{-1} at 25°C) and volumetric capacity (2190 mAh cc^{-1} at 25°C) [7–9]. More recently, it has gained attention for its potential to enable high-power output and rapid charging [10]. These advantages arise from its high capacity, which increases flexibility in slurry formulation and anode sheet design, although practical realization of this flexibility also relies on binder development to ensure mechanical robustness and interfacial stability [11,12]. In particular, reducing the coating layer thickness facilitates faster charge and discharge processes, making silicon useful for multiple purposes.

The electrochemical behavior of silicon during lithiation differs from that of graphite, the latter being most widely used anode material since the commercialization of LIBs. Consequently, a detailed analysis and understanding of silicon's lithiation process are essential. Extensive research has been conducted on graphite anodes, exploring various aspects such as solid electrolyte interphase (SEI) formation and the kinetics of lithium intercalation [13–16]. Among the numerous methodologies developed, optical observation of the anode surface has proven to be an effective and practical approach for assessing the lithiation dynamics of graphite, owing to its inherent optical properties [17].

Previous studies have shown that the color of graphite changes depending on its state of lithiation (SoL) [18,19]. Specifically, as the open-circuit voltage (OCV) decreases relative to the lithium metal redox potential, graphite transitions from black to navy, red, and eventually yellow/gold. Apparent lithium diffusion rates have been analyzed, although it was pretty slow, under various conditions using optical microscopy [20,21]. Despite the increasing interest in silicon as an anode material, no studies have applied a similar approach to investigate its lithiation behavior. The primary challenge is that, unlike graphite, silicon does not exhibit distinct color transitions during lithiation but instead gradually darkens as lithium is incorporated [22]. Our observations confirm that this darkening effect occurs consistently across various silicon-based materials, including pure silicon, silicon alloys, and SiO_x . While these changes are difficult to detect through direct visual inspection or optical microscopy, they can be quantified by measuring reflectance within a specific wavelength range using a spectrometer. Chen et al. previously conducted a spectroscopic analysis of silicon as a function of lithiation degree; however, their study was performed on a silicon thin film rather than a commercial-type porous silicon electrode, resulting in different optical properties [23]. Similarly, Ghannoum et al. investigated reflectance changes in graphite using an *in situ* approach within the near-infrared (NIR) range, but their study was limited to graphite and did not extend to silicon [17].

For the first time, this study applies spectroscopic analysis to a commercial-grade, porous silicon-based electrode. A systematic correlation between reflectance and the SoL was established through a sequential approach: *ex situ* measurements, *in situ* analysis using a dual spectrometer system, and depth profiling to verify the homogeneity of lithium distribution across the electrode thickness. Furthermore, we demonstrated the feasibility of this method for large-scale electrode sheets, confirming its applicability to commercial manufacturing. In

addition, the possible underlying reasons that influence the reflectance were suggested by the roughened surface and the shrunk bandgap from density functional theory (DFT) calculations at a higher SoL.

This approach holds potential for integration into the prelithiation process, introducing additional lithium into the silicon anode before battery cell assembly. Prelithiation enhances the reversible lithium content within the cell, addressing the inherently low initial coulombic efficiency of silicon-based batteries [24–26]. As a result, this process enables higher energy density and improved cycle life [27,28]. Despite extensive research, the industrial adoption of prelithiation has been hindered by challenges related to scalability, including production throughput, economic feasibility, and compatibility with existing manufacturing infrastructure. One critical barrier to implementation is the lack of a practical quality control method; even if an efficient process is developed, its industrial application is limited without a reliable means of monitoring and ensuring outcome consistency.

The spectroscopic approach presented in this study offers a viable and scalable quality control solution for the prelithiation process. We demonstrated the feasibility of evaluating SoL uniformity across large anode areas through reflectance measurements. This technique enables a non-destructive, contactless, and rapid assessment of lithium distribution. More broadly, this study provides a conceptual framework and practical example for implementing spectroscopic analysis as an industrial-scale quality control tool in battery manufacturing.

2. Experimental section

2.1. Electrode production

Two types of anodes were used in this study: a silicon-graphite composite electrode (c-Si) and a pure silicon electrode (p-Si), with a gravimetric capacity of 927.5 mAh g^{-1} and $2739.7 \text{ mAh g}^{-1}$, respectively (first lithiation capacity). The c-Si electrode is commercially available from NEI Corporation (USA) under the name BE-150E [29]. It consists of 20 wt% silicon, 65 wt% graphite, 9 wt% polyacrylic acid (PAA) and carboxymethyl cellulose (CMC) as binders, and 6 wt% carbon mixture as a conductive agent, coated on a $10 \mu\text{m}$ -thick copper current collector. A slurry for the p-Si electrode was prepared in-house by mixing 88 wt% silicon (median particle size: $4.4 \mu\text{m}$, WACKER, Germany), 2 wt% graphite (TIMLEX, Imerys, France), 5 wt% lithium substituted PAA ($M_w = 240,000$, Acros Organics, Belgium), and 3 wt% CMC (2000 PA, DOW Chemical, USA), 2 wt% carbon black (Super P, Imerys, France) as a conductive agent. The slurry was dispersed in water and homogenized using a planetary mixer (pulverisette 7, Fritsch, Germany), and then immersed in an ultrasonic bath to minimize particle agglomeration. The prepared slurry was coated onto a $10 \mu\text{m}$ -thick copper foil at a rate of 5 mm s^{-1} by a coating unit (CUF 5, SUMET, Germany). After drying at room temperature, the electrode underwent calendaring with a linear pressure of 10 N mm^{-1} using a calendaring device (Labor-Kalander, SUMET, Germany). Additionally, the p-Si slurry was applied to a glass-fiber separator (GF/C, Whatman, UK) to produce a free-standing anode without a current collector beneath the coating layer. Due to the porous nature of the glass-fiber separator, part of the slurry components can infiltrate into the pore network, which may introduce a through-thickness distribution heterogeneity in the free-standing configuration. All anodes were cut by a high-precision electrode cutter (EL-CELL, Germany) and dried for at least 12 h at 80°C under vacuum using a Glass Oven B-585 (BÜCHI, Germany) before being transferred to a glove box for cell assembly.

Two different electrodes against the aforementioned anodes were used in this study: lithium metal (GELON, China) and a cathode composed of 90 wt% $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ (NMC622) as the active material, 5 wt% polyvinylidene difluoride (PVDF) as the binder, and 5 wt% Super P as the conductive agent, coated on a $16 \mu\text{m}$ -thick aluminium current collector. This electrode is commercially available (BE-54E, NEI Corporation, USA). The lithium metal chip was stored in an argon-filled

glove box and prepared for cell assembly by gently brushing its surface to remove the native passivation layer, typically formed by residual moisture, oxygen, or nitrogen. The NMC622 cathode was punched and dried under vacuum at 130 °C for at least 12 h using the same drying equipment as for the anodes.

Two-electrode coin cells of the 2032-type were used for ex situ investigations. The electrolyte consisted of 1.0 M LiPF₆ dissolved in a 3:7 (w/w) mixture of ethylene carbonate (EC) and ethyl methyl carbonate (EMC) (99.9%, Solvionic, France), along with 10 wt% fluoroethylene carbonate (FEC) as an additive (99.9%, Solvionic, France). 90 µL of this electrolyte was injected into each cell. A dried glass-fiber separator (GF/C, Whatman, UK) was placed between the anode and cathode to prevent short circuits. All cell assembly was conducted in an argon-filled glove box (GS Glovebox Systemtechnik, Germany), maintaining O₂ and H₂O concentrations below 5 ppm and 0.1 ppm, respectively.

2.2. In situ optical cell

A specialized optical cell (Opto-10, EL-CELL, Germany) was used to enable *in situ* measurements. A 0.2 mm-thick sapphire glass window was selected after confirming its transmittance exceeded 90% across the wavelength range of interest. The lithium chip, separator, and electrolyte used were identical to those in the coin cell setup. An additional separator was introduced despite the anode slurry already being coated onto a separator, as the sparse distribution of active material within the separator pores often led to short circuits. A stainless-steel ring was employed as a current collector on top of the free-standing electrode. All assembly steps were conducted in the same glove box as for the ex situ investigations.

2.3. Lithiation of the anode

Following assembly, the cells were transferred to a battery cycler (BTS-4000, Neware, China) and underwent an 8-h wetting step before lithiation. The targeted SoLs were set at 10%, 20%, 30%, 40%, 50%, 60%, 80%, and 100%, with a lithium chip serving as the opposite electrode and lithium-ion source. Two types of working electrodes—c-Si and p-Si—were used in this half-cell configuration. Lithiation was carried out using a constant current (CC) of 0.08C until the specific capacity corresponding to each SoL target was reached, except for the 100% SoL case, which was lithiated until the potential dropped to 0.01 V vs. Li/Li⁺. For the *in situ* optical cell, lithiation was performed at 0.07C under CC mode until the potential reached 0.01 V vs. Li/Li⁺. This low C-rate was intentionally chosen to minimize polarization and concentration gradients and to obtain a quasi-equilibrium correlation between reflectance and the state of lithiation during the first lithiation. We note that this operando protocol was used to establish a robust baseline relationship and should not be interpreted as a requirement for practical inspection workflows.

2.4. Spectrometer system

After lithiation, the cells were returned to the glove box and disassembled. Residual salt and solvent from the electrolyte, as well as any adhered glass fiber from the separator, were removed using EMC. The samples were then thoroughly dried under vacuum at room temperature, mounted onto glass slides, and sealed. All samples were subsequently transferred to a dry room, where the spectrometer system was housed, with moisture levels maintained at a minimum dew point of −55 °C (equivalent to ~20 ppm at 1 bar and 22 °C). The time between cell assembly and ex situ measurement was minimized to prevent artifacts. Since reflectance values vary depending on the active material, measurement setup, and post-lithiation environmental conditions, normalized reflectance was adopted for most cases in this study.

Slightly different spectrometer configurations were used for ex situ and *in situ* measurements. For ex situ measurements, a spectrometer

(HDX-XR, Ocean Optics, USA) was coupled with a dual-source light source (DH-2000-BAL, Ocean Optics, USA) via a bifurcated optical fiber cable (OCF-108973, Ocean Optics, USA). This cable was connected to the probe at the other end. The light source included two bulbs: deuterium for the ultraviolet (UV) region and halogen for the NIR region. Due to the spectral characteristics of these sources, the intensity of visible light (Vis) was relatively weak (Fig. S1), resulting in a lower signal-to-noise ratio in the 430–630 nm range and reducing measurement reliability and reproducibility (Fig. S2). Consequently, reflectance values within this range were excluded from the analysis. To minimize signal sensitivity to variations in distance between the probe and sample, a collimating lens (74-UV-MP, Ocean Optics, USA) was used to generate a parallel beam. A high-reflectivity mirror (STAN-SSH, Ocean Optics, USA) was selected to ensure minimal signal loss and was used for calibration before every ex situ measurement. Reflectance measurements were conducted across 2068 data points over a wavelength range of 200–1100 nm. Each measurement was integrated over 0.015 s, with a boxcar width of 30, and averaged over 60 scans.

Since optical components, including the light source and cables, experience heating over time, leading to intensity drift, frequent calibration is necessary. In ex situ measurements, each data point is recorded within seconds, allowing frequent recalibration between measurements to compensate for drift. However, in *in situ* experiments, data acquisition must continue for extended periods (often tens of hours) to reach a quasi-equilibrium state. Since continuous calibration is not feasible, a secondary spectrometer (HR4 XR, Ocean Optics, USA) was incorporated to monitor and correct light source intensity fluctuations. If the intensity drift was uniform across all wavelengths, reflectance values would remain unaffected. However, probably due to the presence of two different light sources, intensity drift varies across the spectrum, necessitating a dual-spectrometer system to ensure accurate reflectance data.

2.5. Glow discharge–mass spectroscopy (GD–MS)

GD–MS analysis was performed by an Element GD Plus™ (Thermo Fisher Scientific, Germany). The lithiated anodes, prepared with varying lithium dosages, were carefully opened in an argon-filled glove box and transferred to a specially designed carrier to prevent exposure to ambient air before measurement.

GD–MS enables semi-quantitative analysis by determining the ion beam ratio (IBR), which is defined as the ratio of the signal intensity of a specific isotope to the total signal intensity of all isotopes within the sample matrix. The IBR is calculated as Equation (1):

$$IBR_{M,i}^A = \frac{I_A/f_A}{\sum_i (I_{M,i}/f_{M,i})} \quad (1)$$

IBR_{M,i}^A represents the ion beam ratio of isotope A in a sample with *i* matrix elements *M*. The term *I_A* refers to the intensity of isotope A, while *f_A* denotes its respective isotopic abundance. The same definitions apply correspondingly to matrix elements.

The porosity of lithium-ion battery electrodes presents challenges for plasma formation during GD–MS analysis. Sample inhomogeneity can lead to uneven plasma distribution, potentially affecting measurement accuracy and reproducibility. Although sampling time is correlated with the depth of material removal, it cannot be directly converted into depth. Consequently, depth profiles are reported as a function of time, with crater depth measured only after analysis. Therefore, in GD–MS measurements, time cannot be directly translated into depth.

2.6. Density functional theory (DFT)

Electronic-structure calculations with the help of density-functional theory (DFT) were conducted using the Vienna Ab initio Simulation

Package (VASP), version 5.4.4 [30–33]. To increase the accuracy of the PBE functional [34] in combination with the projector-augmented wave method [35,36], the HSE06 functional was used [37]. Furthermore, the D3 dispersion correction of Grimme and co-workers with zero damping was utilized to account for van der Waals interactions [38]. The kinetic energy cut-off of the plane wave expansion was set to 500 eV. The corresponding structures were optimized with respect to the forces,

utilizing a convergence criterion of 10^{-2} eV Å⁻¹ for the structural optimization and 10^{-6} eV for the electronic steps. The optimized structures were then used to subsequently predict the bandgap by inspecting the density of states.

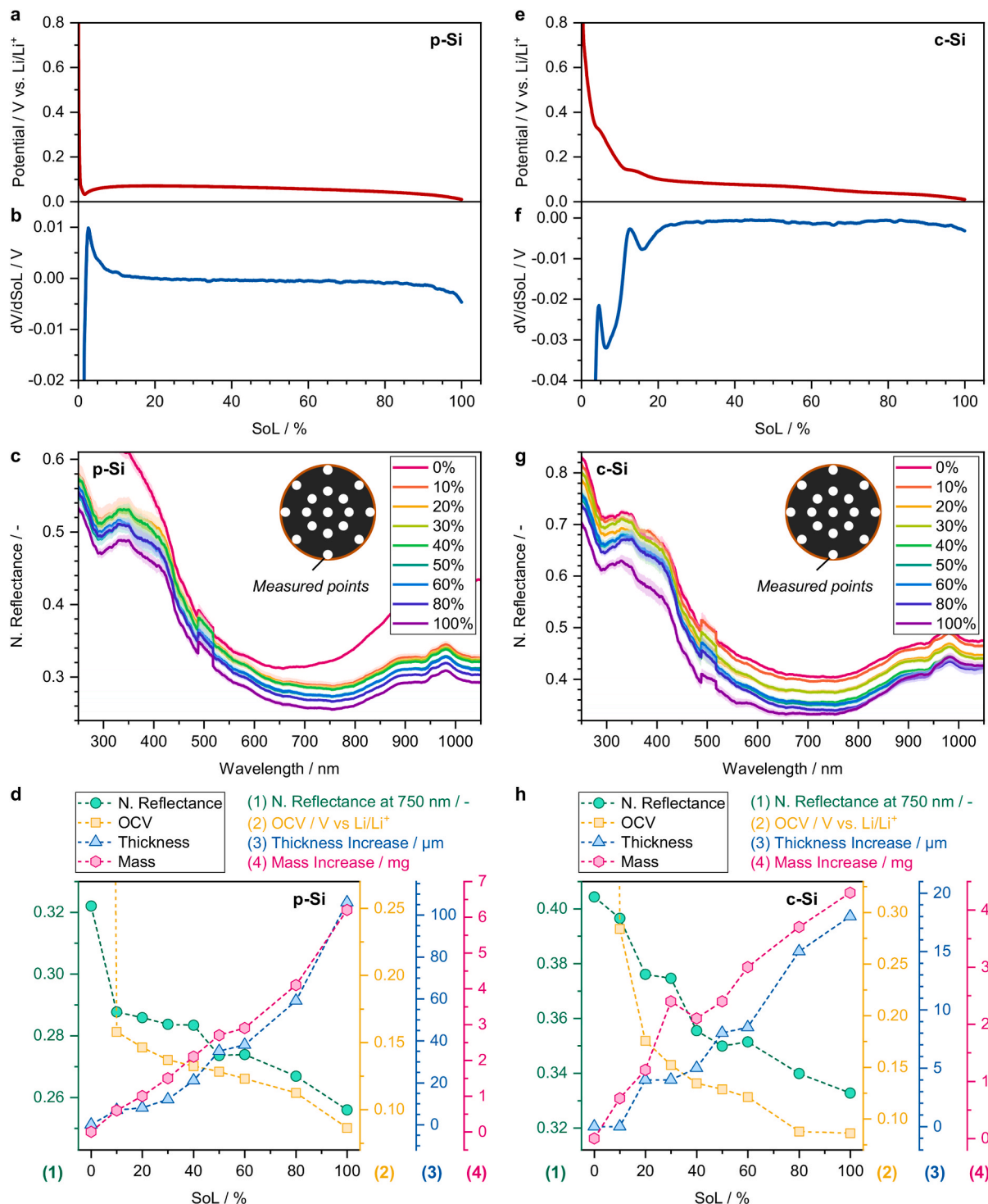


Fig. 1. Electrochemical and optical measurement results of (a–d) the p-Si anode and (e–h) the c-Si anode. The plots sequentially show: (a, e) potential profiles versus SoL, (b, f) differential voltage analysis, (c, g) normalized reflectance spectra over the 250–1050 nm wavelength range, and (d, h) four parameters plotted as functions of SoL—normalized reflectance at 750 nm, OCV, electrode thickness increase, and mass increase. Insets in (c) and (g) indicate the 17 measurement locations on each anode sample.

3. Results and discussion

3.1. Ex situ characterization

While the primary focus of this study is to investigate the optical behavior of pure silicon (p-Si) anodes, a silicon-graphite composite (c-Si) electrode was also examined to evaluate the applicability of the proposed methodology under more practical and commercially relevant conditions. The lithiation voltage profiles for both electrodes are shown in Fig. 1a and e.

The p-Si electrode exhibits an initial open-circuit voltage of approximately 2.8 V vs. Li/Li⁺, followed by an abrupt potential drop at the onset of lithiation. This sharp decline is attributed to the inherently slow lithiation kinetics of silicon [39]. Subsequently, a broad voltage plateau is observed, corresponding to a series of phase transitions during the formation of various Li_xSi alloys. In contrast, the c-Si electrode displays a higher initial potential of 3.1 V vs. Li/Li⁺, with two short voltage plateaus between 0.11 and 0.3 V, which are associated with lithium intercalation into the graphite component [14]. Below 0.1 V vs. Li/Li⁺, the potential decreases more gradually, indicating the coexistence of lithiation reactions in both silicon and graphite [40,41].

To further elucidate the lithiation mechanisms, differential voltage analysis (DVA) was performed, as shown in Fig. 1b and f. Local maxima or flat regions in the differential voltage (dV/dSoL) vs. SoL profiles typically correspond to phase transitions and active lithiation steps. For the p-Si electrode, a broad plateau appearing beyond 10% SoL suggests that silicon undergoes lithiation at a relatively constant potential during the rest of the SoL region. In contrast, the c-Si electrode exhibits two distinct peaks at approximately 5% and 12% SoL, which are absent in the p-Si profile. Based on the analysis of Fig. 1a and b, where the primary lithiation of silicon occurs near 0.1 V vs. Li/Li⁺, these early-stage peaks in Fig. 1f are attributed to lithium intercalation into graphite. The subsequent flat region appearing beyond 20% SoL corresponds to concurrent lithiation of both silicon and graphite phases [14]. Electrodes at various SoLs were prepared by adjusting the lithiation time under a constant current (CC) protocol, as the lithiated capacity is linearly proportional to processing duration under CC conditions. Upon reaching the target SoL, cells were disassembled in an argon-filled glove box, and the lithiated anodes were mounted onto glass slides for optical analysis. Samples were then transferred to a dry room with a dew point maintained below −55 °C to prevent humidity-induced artifacts during measurement. A custom-built optical fixture was employed to ensure consistent and fixed distances between the probe, sample surface, and reference mirror. To assess the lateral homogeneity of lithiation, reflectance measurements were conducted at 17 distinct positions on each electrode, as shown in the insets of Fig. 1c and g. To eliminate ambient noise, background light intensity was subtracted from both sample and reference spectra. Reflectance values (*R*) were calculated according to Equation (2), where *I*_{sample} represents the signal intensity from the sample, and *I*_{mirror} denotes light source intensity measured using the reference mirror. The intensity of ambient light (*I*_{amb}) was obtained using the reference mirror in the absence of the light source.

$$R = \frac{(I_{\text{sample}} - I_{\text{amb}})}{(I_{\text{mirror}} - I_{\text{amb}})} \quad (2)$$

Fig. 1c and g presents the average normalized reflectance spectra of the p-Si and c-Si electrodes, respectively, over the 300–1100 nm wavelength range. The shaded regions denote the standard deviation across the 17 measurement points. In both cases, reflectance decreased progressively with increasing lithium content. The small standard deviation across spatial points indicates good lithiation uniformity and reliable signal response across the electrode surface. Fig. 1c and g illustrate the evolution of reflectance at 750 nm as a function of SoL, alongside changes in OCV, electrode thickness, and mass for both p-Si and c-Si anodes. As confirmed in Fig. S3, the reflectance at

approximately 750 nm exhibited the strongest correlation with OCV across the measured spectral range, although variations in reflectance trends across different wavelengths were relatively minor. Therefore, we use 750 nm as a single representative wavelength to provide a practical and consistent metric for correlation and spatial mapping throughout the manuscript. We emphasize that this wavelength is intended as a guideline for our setup rather than a universally optimal choice, and users may adjust around this region depending on their specific optical configuration and electrode composition.

In the case of the p-Si anode, the reflectance decreases sharply at the initial stages of lithiation and then gradually declines, closely mirroring the OCV profile (Fig. 1d). In contrast, due to the large volume expansion associated with silicon lithiation, the thickness of the electrode increases as more lithium is incorporated into the silicon lattice (Fig. 1d). Similarly, the mass of the electrode increases with SoL as lithium alloying with silicon progresses, and additional electrolyte decomposition products accumulate on the surface of the active material. The increases in both volume and mass are approximately linear with respect to SoL, in contrast to the non-linear behavior observed in reflectance and OCV.

Fig. 1h presents the evolution of reflectance, OCV, thickness, and mass for the c-Si anode. As indicated in Fig. 1e and f, the voltage drop observed up to 20% SoL is attributed primarily to lithium intercalation into graphite rather than silicon lithiation. This likely explains why the reflectance at 10% SoL does not show a noteworthy decrease from its initial value. Although the mass fractions of silicon and graphite in the composite are 20% and 65%, respectively, their contributions to the total capacity are approximately 72% for silicon and 28% for graphite, owing to the much higher specific capacity of silicon. This implies that silicon becomes the dominant active material beyond 20% SoL, particularly given that the initial 20% SoL is primarily achieved through graphite intercalation. Consequently, in an SoL range above 20%, the reflectance trend of the c-Si electrode closely resembles that of p-Si, despite the differing optical properties of silicon and graphite [17,18]. Considering that silicon exhibits far greater volume expansion than graphite, the absence of a thickness increase at 10% SoL (Fig. 1h) is consistent with graphite being the primary active material at the early stage of lithiation.

An additional factor influencing the reflectance response is exposure to ambient conditions. Fully lithiated graphite exhibits a yellow/gold hue, which rapidly darkens upon exposure to dry air—particularly in the case of composite electrodes containing silicon. This explains the absence of a reflectance peak near the yellow visible light region (~580 nm), despite minimizing the time interval between sample unsealing and measurement. Furthermore, the low signal reliability in this spectral range may also have contributed to the lack of a detectable peak, as demonstrated in Fig. S2.

In summary, the correlation of normalized reflectance with electrochemical and physical parameters across varying SoL confirms that the reflectance of silicon-based electrodes systematically decreases with increasing lithium content. These ex situ measurements highlight the potential of optical reflectance as a contactless method for monitoring lithiation dynamics in silicon anodes.

3.2. In situ characterization

Although ex situ experiments successfully revealed the overall trend of reflectance changes across the full range of SoL, they remain limited in understanding the dynamic behavior of silicon particles during lithiation. While ex situ techniques are capable of providing high-resolution post mortem analysis and detailed structural and electrochemical characterization, they cannot capture real-time phenomena, and the disassembly process may alter the sample state. Additionally, ex situ methods struggle to replicate the realistic operating conditions of LIBs. In contrast, *in situ* measurements are often preferred for studying dynamic processes and transient phenomena within LIB systems, while ex situ techniques are more suitable for post-cycling evaluations [42,43].

Given that this study focuses on the reflectance behavior of silicon, *in situ* measurements were performed using the p-Si anode. As previously described, a dual-spectrometer system was implemented to compensate for temporal fluctuations in light source intensity. Fig. 2a demonstrates that the intensity of the deuterium light source gradually increased over time, while that of the halogen source decreased, introducing distortions in the recorded reflectance signal. Since these fluctuations are non-linear and vary over time, they cannot be corrected simply by applying a uniform scaling factor. Moreover, external factors such as ambient temperature and the alignment of optical components influenced the degree of signal drift.

To address this, a second spectrometer (Spectrometer 2 in Fig. 2b) was deployed to continuously monitor the light source intensity at a given time t , denoted as $I_{source,t}$. A correction factor, $k_{corr}(t)$, was determined by dividing $I_{source,t}$ by the initial light source intensity at the beginning of the measurement ($t = 0$), $I_{source,0}$. To eliminate background noise, the ambient light signal within the optical fiber ($I_{amb,fiber}$), measured with a closed shutter at $t = 0$, was subtracted from the light source intensities, as defined in Equation (3).

$$k_{corr}(t) = \frac{I_{source,t} - I_{amb,fiber}}{I_{source,0} - I_{amb,fiber}} \quad (3)$$

The corrected reflectance value at time t , denoted as $R_{corr}(t)$, was obtained by multiplying $k_{corr}(t)$ with the uncorrected reflectance ($R_{uncorr}(t)$), acquired from Spectrometer 1 in Fig. 2b, as shown in Equation (4). In this case, $I_{mirror,0}$ and $I_{amb,mirror}$ were recorded once prior to the start of lithiation, with and without the reference mirror in place, respectively.

$$R_{corr}(t) = k_{corr}(t) \cdot R_{uncorr}(t) = \left(\frac{I_{source,t} - I_{amb,fiber}}{I_{source,0} - I_{amb,fiber}} \right) \left(\frac{I_{sample,t} - I_{amb,mirror}}{I_{mirror,0} - I_{amb,mirror}} \right) \quad (4)$$

Table 1 summarizes the physical meaning of each parameter used in the reflectance calculation, along with the corresponding measurement method and timing.

Fig. 3a illustrates a schematic of the internal structure of the *in situ* cell, incorporating a free-standing anode. Reflectance data was collected during the first lithiation process and plotted over the wavelength range of 250–1050 nm as a function of the SoL. The observed trend in reflectance closely matches that obtained from *ex situ* measurements, which exhibits a sharp initial decrease followed by a more gradual decline beyond approximately 10% SoL (Fig. 3b). For comparison, Fig. S4 presents the results obtained using a single spectrometer. Unlike

Table 1

List of parameters used for deriving the corrected reflectance, along with their physical meaning, associated spectrometer, and measurement conditions.

Parameters	Physical Meaning	Acquired by	Shutter	Measured at	Measured with
$I_{source,0}$	Light source intensity	Spectrometer 2	Opened	$t = 0$	Direct connection
$I_{source,t}$	Light source intensity	Spectrometer 2	Opened	$t > 0$	Direct connection
$I_{amb,fiber}$	Ambient light intensity within the fiber	Spectrometer 2	Closed	$t = 0$	Direct connection
$I_{sample,t}$	Reflected light intensity by the anode	Spectrometer 1	Opened	$t > 0$	Anode
$I_{mirror,0}$	Reflected light intensity by the mirror	Spectrometer 1	Opened	$t = 0$	Reference mirror
$I_{amb,mirror}$	Ambient light intensity measured with the mirror	Spectrometer 1	Closed	$t = 0$	Reference mirror

the consistent downward trend in Fig. 3, the reflectance values in Fig. S4 exhibit minor fluctuations with SoL, likely caused by uncorrected drift in the light source intensity over time.

The reflectance at 750 nm was further analyzed and compared with the corresponding anode potential. Fig. 3c presents both the differential voltage and the differential reflectance as functions of SoL. The electrochemical data reveal a long voltage plateau beginning at approximately 5% SoL, indicating a continuous and stable lithiation process across a broad SoL range. The differential reflectance follows a similar pattern, with a slight shift of approximately 5% toward higher SoL. This offset is likely attributable to overpotential effects, as the measured voltage corresponds to dynamic (under-load) conditions rather than OCV. Moreover, the free-standing electrode architecture (slurry-coated separator) may accentuate kinetic inhomogeneity, which can further increase the overpotential under dynamic conditions. Additionally, in

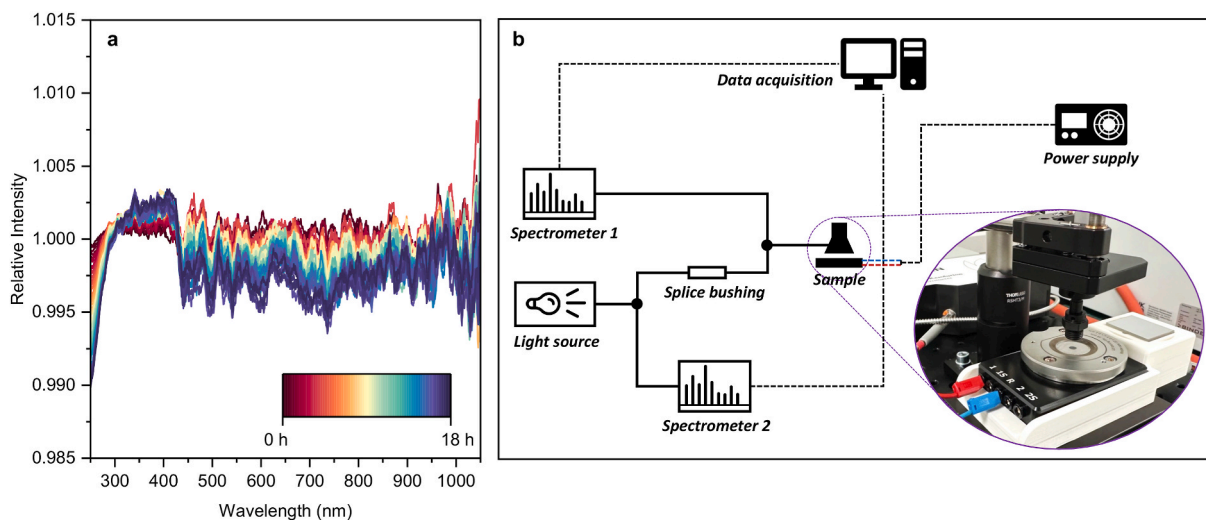


Fig. 2. (a) Intensity drift of the light source over an 18-h period. The UV-range intensity decreases, while the IR-range intensity increases with time. (b) Schematic of the dual-spectrometer system. Spectrometer 1 analyzes the reflected signal from the sample, while Spectrometer 2 monitors the temporal intensity drift of the light source for real-time correction.

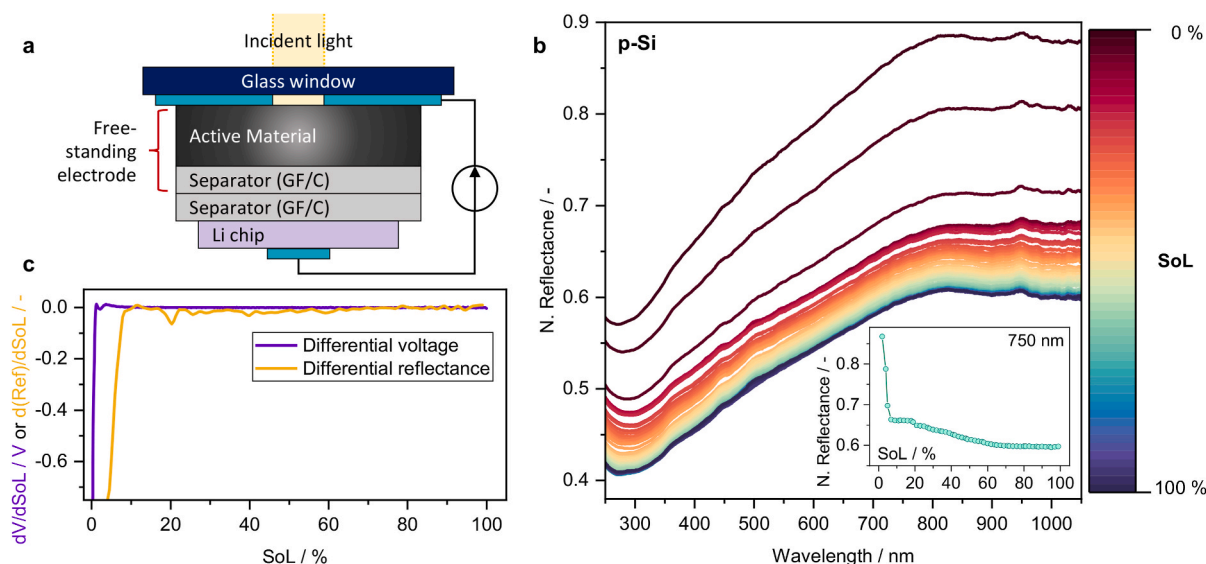


Fig. 3. Experimental setup and results of the *in situ* analysis. (a) Cross-sectional schematic of the optical cell. All components are compressed using a spring to ensure stable electrical and mechanical contact. (b) Normalized reflectance of the p-Si anode during lithiation. The inset displays the trend of reflectance at 750 nm according to SoL (c) Differential voltage and differential reflectance plotted as a function of SoL.

our *in situ* setup, electrochemical data were recorded at a much higher temporal resolution, whereas reflectance spectra were acquired every 15 min. Under constant-current lithiation, this corresponds to approximately 1.76% SoL per 15 min; therefore, during the first 30 min required to obtain the first two optical data points, the cell already reaches approximately 3.52% SoL. This difference in sampling interval and time alignment can contribute to an apparent horizontal offset when comparing derivative features ($dV/dSoL$ versus $dR/dSoL$).

Despite these minor deviations, the data clearly confirm that the reduction in reflectance from the silicon particles is closely correlated with lithiation progress, thereby validating the *ex situ* findings. One challenge of the *in situ* configuration is the occasional entrapment of gas generated during SEI formation. This trapped gas can increase reflectance locally, interfering with accurate signal acquisition and reducing measurement reliability. Beyond gas entrapment, SEI formation itself may also influence the optical signal by changing the near-surface microtexture and the local effective optical constants of the electrode surface. Although we do not attempt to isolate this contribution quantitatively, we note it here as a plausible secondary factor during the early stage of the first lithiation.

3.3. Scanning depth

Although a method for quantitatively determining the degree of lithiation in silicon has been validated in previous sections, it remains uncertain whether the obtained reflectance data accurately represent information throughout the entire electrode thickness. Given that battery electrodes typically exhibit a porous structure and a thickness on the order of tens of micrometers, it is likely that the reflectance signal predominantly originates from near-surface regions. This limitation stems from the complex optical interactions occurring within the porous medium, including reflection, refraction, and scattering, which attenuate the incident light while it penetrates an electrode. Moreover, since silicon particles used in anodes range in diameter from sub-micron to tens of microns, the transmittance of visible light through a single particle is inherently low. Therefore, it is improbable that photons penetrate deeply enough to interact with particles located deeper in the electrode, particularly those near the current collector.

Fig. S5 manifests the transmittance trend as a function of wavelength for a uniform silicon film. When assuming a simplified scenario where light penetrates a uniform silicon film of thickness equivalent to the

mean particle diameter in this study ($D_{50} = 4.4 \mu\text{m}$), the transmittance at 750 nm under vacuum at 300 K is approximately 0.20. In the UV-vis range, the transmittance is even lower due to the intrinsic optical properties of silicon, such as high imaginary part of the refractive index (more absorption) [44].

In real anode architectures, where silicon forms a porous network of discrete particles, the attenuation of incident light becomes significantly more amplified. Upon interacting with individual particles, light undergoes Mie scattering, resulting in random redirection. Additionally, diffuse reflectance caused by a huge refractive index mismatch between silicon and air further impedes light penetration into the material. These scattering and reflection phenomena occur repeatedly throughout the porous structure, introducing substantial complexity to any predictive modelling of transmittance.

Nevertheless, even the transmittance calculated under this idealized planar condition suggests considerable attenuation of incident light as it passes through multiple particles. Consequently, light reflected from deeper regions within the anode is unlikely to reach the spectroscopic probe, convincing that the measured reflectance signal is mainly derived from the near-surface region of the electrode.

Experimental data support the aforementioned assumption. Two c-Si anodes were prepared using different substrates: copper foil and double-sided adhesive tape (05338, Tesa SE, Germany) affixed to a mirror, as illustrated in Fig. 4a. Reflectance measurements were then performed and compared, as shown in Fig. 4b. If the incident light had penetrated the full thickness of the c-Si electrode and reached the underlying substrate, differences in reflectance would be expected due to the distinct optical properties of the two substrates. However, the resulting reflectance spectra showed only minor variations within the standard deviation range, indicating no difference between the two cases.

This observation confirms that the incident light does not transmit through the entire electrode but is instead reflected primarily from the surface. Therefore, the reflectance signal is limited to shallow depths within the electrode. Despite this limitation in probing depth, the validity of the measurement is supported in the following section by examining the vertical distribution of lithium across the electrode thickness.

3.4. Homogeneous lithium distribution across the electrode depth

If lithium is uniformly distributed along the electrode depth, this

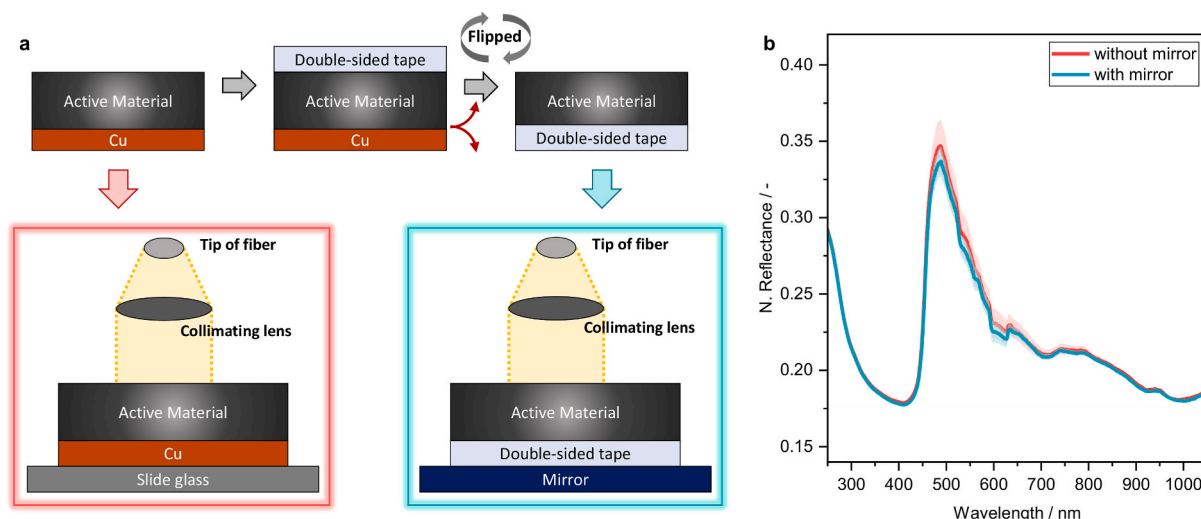


Fig. 4. (a) Schematic illustration of sample preparation for reflectance analysis using two substrate types: a copper foil and a polymer tape combined with the reference mirror. Clean detachment of the electrode coating was achieved using double-sided adhesive tape. (b) Normalized reflectance of the c-Si anode using the copper substrate (without mirror) and polymer tape/mirror substrate (with mirror).

would strongly support the assumption that surface-localized reflectance measurements can reliably represent the overall lithiation state. Our previous study demonstrated an almost consistent lithium distribution along the depth direction across all investigated SoLs using ion beam analysis [45]. Although a locally elevated lithium concentration was observed near the outermost area, the ratio of lithium content at the surface to that within the bulk remained constant. Given the nature of the measurement, particularly the brief exposure to air, the localized surface enrichment was likely attributed to the formation of LiOH, supported by a concurrent increase in both oxygen and hydrogen concentrations. Additionally, the lithium distribution appeared to correlate with the silicon content. Nevertheless, this observation still indicates that electrodes with high surface lithium content also contained proportionally high lithium concentrations deeper within the electrode. In this study, an additional approach was employed to validate this conclusion further. GD-MS was used to obtain depth profiles of lithium distribution in lithiated c-Si anodes. Two samples were analyzed: one

lithiated to 20% SoL and the other to 100% SoL. The resulting profiles, shown in Fig. 5, indicate that lithium concentration remained constant as a function of etching time in both samples, which corresponds to depth, thereby corroborating the previous ion beam analysis. As described in the Experimental section, the IBR of a given element is decided by a matrix element within the same sample; thus, cross-sample comparisons do not yield reliable quantitative information. In this context, the IBR reflects relative changes in elemental abundance along the depth of the anode. At higher states of lithiation (SoL), where the lithium content in lithium silicide (Li_xSi_y) is greater, lithium may be less readily ionized under the measurement conditions. This could explain the relatively lower lithium IBR observed in the fully lithiated (100% SoL) sample, accompanied by a corresponding increase in the carbon IBR.

These findings support using the reflectance value as a valid proxy for the overall SoL in silicon-based anodes. Given the porous nature of the electrode, such uniform lithium distribution is plausible, assuming

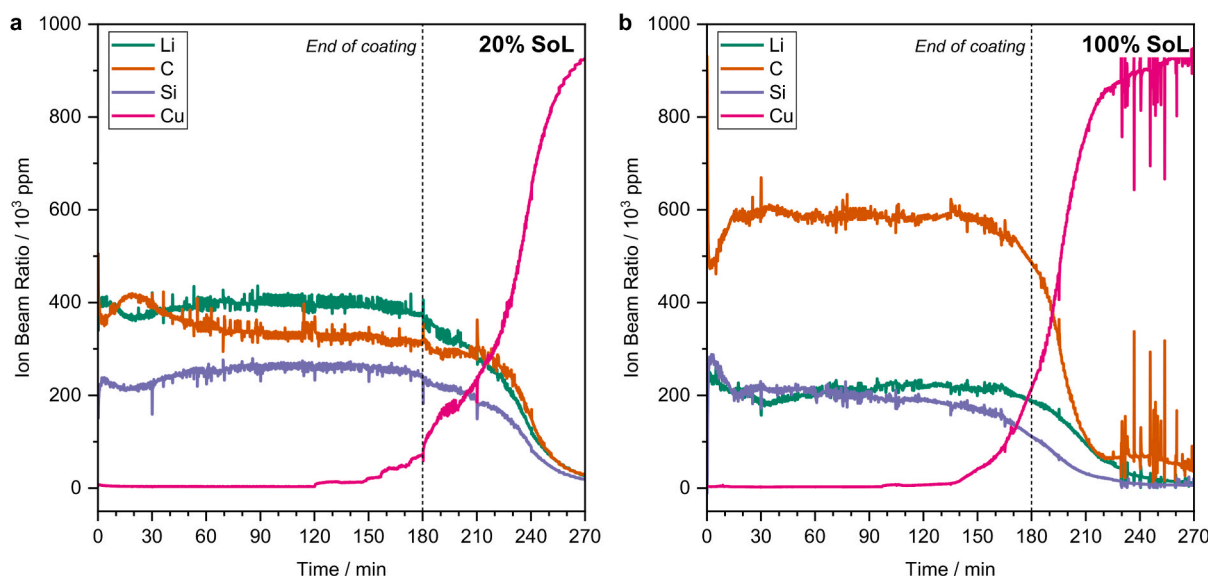


Fig. 5. IBR profiles with respect to etching time for c-Si anodes at (a) 20% SoL and (b) 100% SoL. A sharp increase in copper concentration after ~180 min suggests full removal of the active material layer, exposing the current collector. Due to the difference in matrix composition (e.g., variations in Li/Si phase composition) between the samples, the IBR profiles should be interpreted within each individual sample. Direct quantitative comparison across two samples is not valid.

the active material is well dispersed and the electrode is thoroughly wetted with electrolyte prior to lithiation. When all active material particles are in contact with the electrolyte, lithium can be inserted regardless of their position within the electrode. Although local inhomogeneities caused by trapped air, impurities, or uneven mixing can lead to uneven lithiation or non-uniform current density, the enhanced lithium diffusivity in lithiated silicon mitigates these effects and supports homogeneous lithium distribution throughout the electrode. Accordingly, the method is expected to be most reliable for well-wetted porous electrodes and operating conditions where severe through-thickness gradients are minimized. In contrast, discrepancies may arise under conditions such as non-uniform wetting, thick electrodes or rate-induced transport limitations that generate pronounced depth gradients, as well as in the presence of thick or highly scattering surface films or strong surface reactions that modify the near-surface optical response. Therefore, our approach should be regarded as a practical SoL proxy for the studied electrode architecture rather than a universal technique for depth-resolved lithium quantification.

3.5. Application of the spectroscopic inspection

An inhomogeneous lithium distribution was intentionally induced in a coin cell configuration by using electrodes of different diameters: a 16 mm c-Si anode and a 10 mm lithium chip, as illustrated in the inset of Fig. 6a. The anodes were discharged to 0.01 V vs. Li/Li^+ and left in the coin cell for either 1 h or 3 h. A relatively high C-rate (0.2C) was applied to minimize diffusion during lithiation. Following disassembly, both anodes were rinsed with EMC, the linear carbonate solvent used in the

electrolyte. A 2-h delay occurred between disassembly and the reflectance measurements (Table 2).

Fig. 6a presents the average reflectance values across multiple points (solid lines) for each sample, along with their standard deviations (shaded areas). The measured points were identical to those shown in the inset of Fig. 1c or 1g. Across most of the wavelength range, the sample rested for 3 h exhibits reflectance values that are either identical to or slightly lower than those of the 1-h rest sample. However, the noticeably narrower shaded region indicates reduced areal variation in lithiation. The box plot in Fig. 6b confirms that the reduced standard deviation after 3 h is not the result of a few outliers but rather reflects tighter clustering of reflectance values around the mean. Fig. 6c and d displays spatial reflectance maps of the anode surface, visually representing lithium distribution. As previously established, lower reflectance corresponds to a higher degree of lithiation. Accordingly, these maps reveal that lithium was initially concentrated near the center of the anode and gradually diffused toward the outer regions over time.

Table 2

Fabrication procedure for producing non-uniformly lithiated anodes.

Step	1-h rest	3-h rest
Lithiation	Li ($\varnothing$ 10 mm) c-Si anode ($\varnothing$ 16 mm) 0.2C up to 0.01 V vs. Li/Li^+	
Rest	1 h	3 h
Disassembly	-	-
Interval	2 h	2 h
Measurement	Reflectance at 17 points	

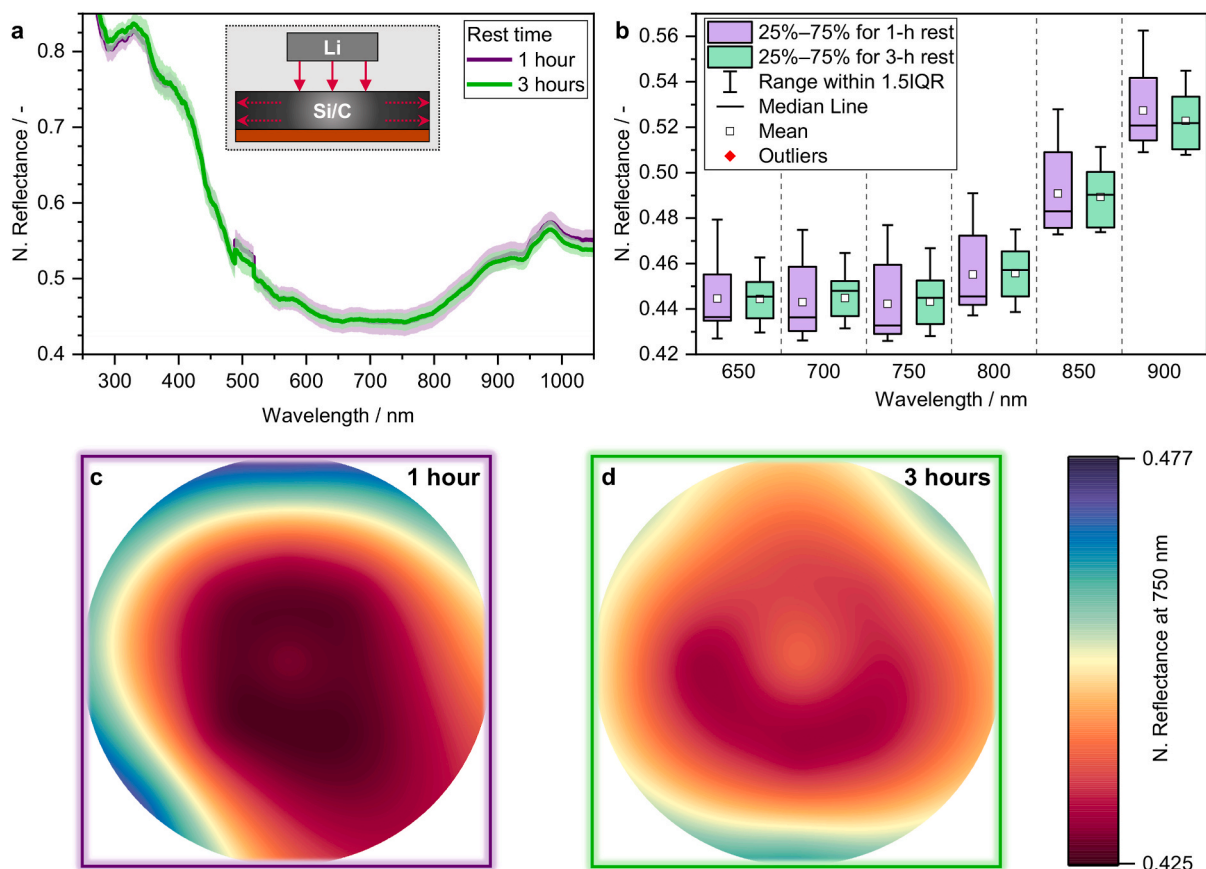


Fig. 6. (a) Normalized reflectance of c-Si anodes subjected to different resting times after lithiation with a smaller-area lithium metal opposite electrode. Solid lines denote average reflectance, and shaded areas represent standard deviations. Inset shows the schematic of the coin cell configuration used for lithiation. (b) Box-and-whisker plots of reflectance values for both samples. The box represents the interquartile range (IQR), spanning from the first quartile (Q1) to the third quartile (Q3), while the whiskers extend to the furthest data points within $1.5 \times \text{IQR}$ from each quartile. (c, d) Areal maps of normalized reflectance for (c) 1-h and (d) 3-h resting samples. Values between measured points were interpolated.

The methodology developed in this study demonstrates strong potential for integration with the prelithiation process to evaluate lithium distribution homogeneity [46]. Prelithiation, commonly employed to introduce additional lithium into the anode prior to cell assembly, serves to compensate for lithium losses associated with the formation of SEI. Since prelithiation is performed during the initial lithiation phase, the findings of this study are directly applicable. Furthermore, because reflectance changes occur more prominently at low SoL levels and prelithiation typically aims for lithiation in the 10–20% SoL range, this spectroscopic analysis becomes particularly effective for assessing prelithiated electrodes.

The feasibility of this concept was validated using a larger-format anode lithiated through a roll-to-roll, continuous prelithiation process, drawing upon the previous experience [27]. To assess the uniformity of this process, the prelithiated electrode sheet was punched into 16 individual pieces, as shown in Fig. 7a. Reflectance measurements were conducted on each piece, followed by electrochemical evaluation via coin cell assembly with lithium metal to determine reversible capacity and OCV. The results are compared in Fig. 7b. Except for a few outliers, the reflectance trends correlated well with OCV and showed an inverse relationship with delithiation capacity, consistent with earlier observations in this study. Minor deviations from the trend are likely attributed to the time interval between reflectance and electrochemical measurements, which may have introduced environmental degradation or contamination, especially by moisture. Since variations in reflectance values are more critical when evaluating the homogeneity of lithium distribution within the same sample, absolute reflectance was used in this case.

Fig. 7c presents the linear relationship between OCV and reflectance. While the coefficient of determination (R^2) is not particularly high, a clear proportional trend is evident. In future implementations, minimizing the time between disassembly and measurement and controlling surface exposure are expected to improve correlation strength and quantitative reliability. In addition, increasing the number of spatial sampling locations would improve the statistical robustness of the regression and the quantitative reliability of the interpolated map. For real-world applications, it is also advantageous to establish the reflectance–SoL calibration under controlled ex situ conditions (as in Fig. 1), where electrodes can be prepared at defined SoL values and a larger number of replicates can be collected to obtain a more reliable regression. Furthermore, independent quantitative lithium analysis at selected locations, such as inductively coupled plasma optical emission spectroscopy (ICP-OES), would provide additional cross-validation and further strengthen the reflectance-based calibration.

Based on the derived linear relationship, an OCV mapping was constructed, as shown in Fig. 7d. The OCV map here is presented as an illustrative, semi-quantitative visualization based on the observed correlation in Fig. 7c and should not be interpreted as a precise OCV prediction for each location under the present dataset and handling conditions. Nevertheless, OCV is a widely used and robust indicator of lithiation state; therefore, spatial variations in OCV can serve as a practical proxy for assessing areal SoL uniformity after prelithiation.

With this interpretation, integrating reflectance measurements with the prelithiation process offers a promising route for in-line, non-destructive, continuous, and contactless inspection of lithiation uniformity. As illustrated in Fig. 7e, this approach provides a practical pathway for implementing quality control in roll-to-roll prelithiation systems. Moreover, it holds the potential to be incorporated into a closed-loop feedback system, enabling real-time adjustment of process parameters based on inspection results, thereby enhancing overall process reliability and consistency.

In practical implementations, measurement speed is primarily constrained by the process window (e.g., web speed and lead time), rather than by the spectroscopic readout itself. In our ex situ configuration, spectra are acquired with short integration times and can be further accelerated by reducing averaging, increasing illumination intensity,

and optimizing detector sensitivity in the wavelength region of interest. Moreover, a narrow-band approach using a single wavelength (or a small set of wavelengths around the most sensitive region) could significantly reduce acquisition time and simplify hardware integration. These strategies imply a trade-off between sensitivity (signal-to-noise) and throughput, which can be tailored to the required spatial resolution and inspection objectives.

Meanwhile, prelithiated anodes typically experience a loss of reversible lithium over time due to increased reactivity during storage [47]. This spectroscopic approach has the potential to monitor the degradation behavior of prelithiated anodes by tracking changes in optical properties over time.

3.6. Underlying principles for the reflectance changes

Two possible explanations were investigated to elucidate the fundamental mechanism underlying the observed decrease in the reflectance of silicon particles during lithiation. The first hypothesis considers that the volume expansion of silicon upon lithium insertion may induce surface and structural changes, potentially increasing diffuse reflection and light scattering. To examine this effect, the surface morphology of anodes at various SoL levels was analyzed using a laser scanning microscope (VK-9700K 3D, Keyence, Japan). Fig. 8a presents the trend in surface roughness, represented by the arithmetical mean height (S_a), which quantifies the average deviation in height compared to the mean plane. The change in S_a observed for the p-Si anode across different SoLs likely corresponds to the pronounced increase in electrode thickness shown in Fig. 1d. The c-Si anode also exhibited a general increase in surface roughness with increasing SoL, although the variation was less visible compared to p-Si and was accompanied by a relatively high standard deviation. At 10% SoL, the S_a value of the c-Si anode did not show a measurable increase, which aligns well with the unchanged electrode thickness and a small reflectance decrease observed in Fig. 1h. These changes in surface roughness are further supported by the 3-dimensional surface profile, as presented in Fig. S6.

A more important factor reducing reflectance is likely the modification of the electronic band structure induced by lithium incorporation. Lithium doping introduces additional energy levels by contributing electrons to the conduction band or creating localized states within the bandgap. As a result, the energy difference between the valence and conduction bands is reduced, effectively narrowing the bandgap. This bandgap shrinkage increases the probability of photon absorption, as electrons can now move across a broader range of energy levels, thereby supporting the observed reduction in reflectance upon lithiation. Fig. 8b illustrates the calculated density of states (DoS) for both pure silicon and the fully lithiated state of silicon ($\text{Li}_{15}\text{Si}_{14}$), along with their respective atomic structures. While intrinsic silicon exhibits a bandgap of approximately 1.1–1.2 eV, lithium silicide displays no bandgap, consistent with prior studies [48].

Additionally, introducing free charge carriers, such as electrons, through lithium doping enhances photon–electron interactions across a broad wavelength range, facilitating intra-band electron transitions. Lithiation may also entail structural defects (e.g., cracks, grain boundaries) or the amorphization of crystalline silicon [49–52]. In the amorphous phase, silicon lacks the periodic atomic arrangement characteristic of its crystalline counterpart. This increased structural disorder enhances internal light scattering within the material, thereby further contributing to the reduction in reflectance. Considering the extent of change in both surface roughness and electronic band structure, the observed variation in reflection across the visible to NIR range is well supported by the experimental results.

It should be noted that a rigorous quantitative separation of roughness-driven scattering and intrinsic absorption is challenging for thick, porous composite electrodes, because the measured reflectance is influenced by coupled factors such as porosity, packing density, evolving microtexture, and the effective optical constants of the surface

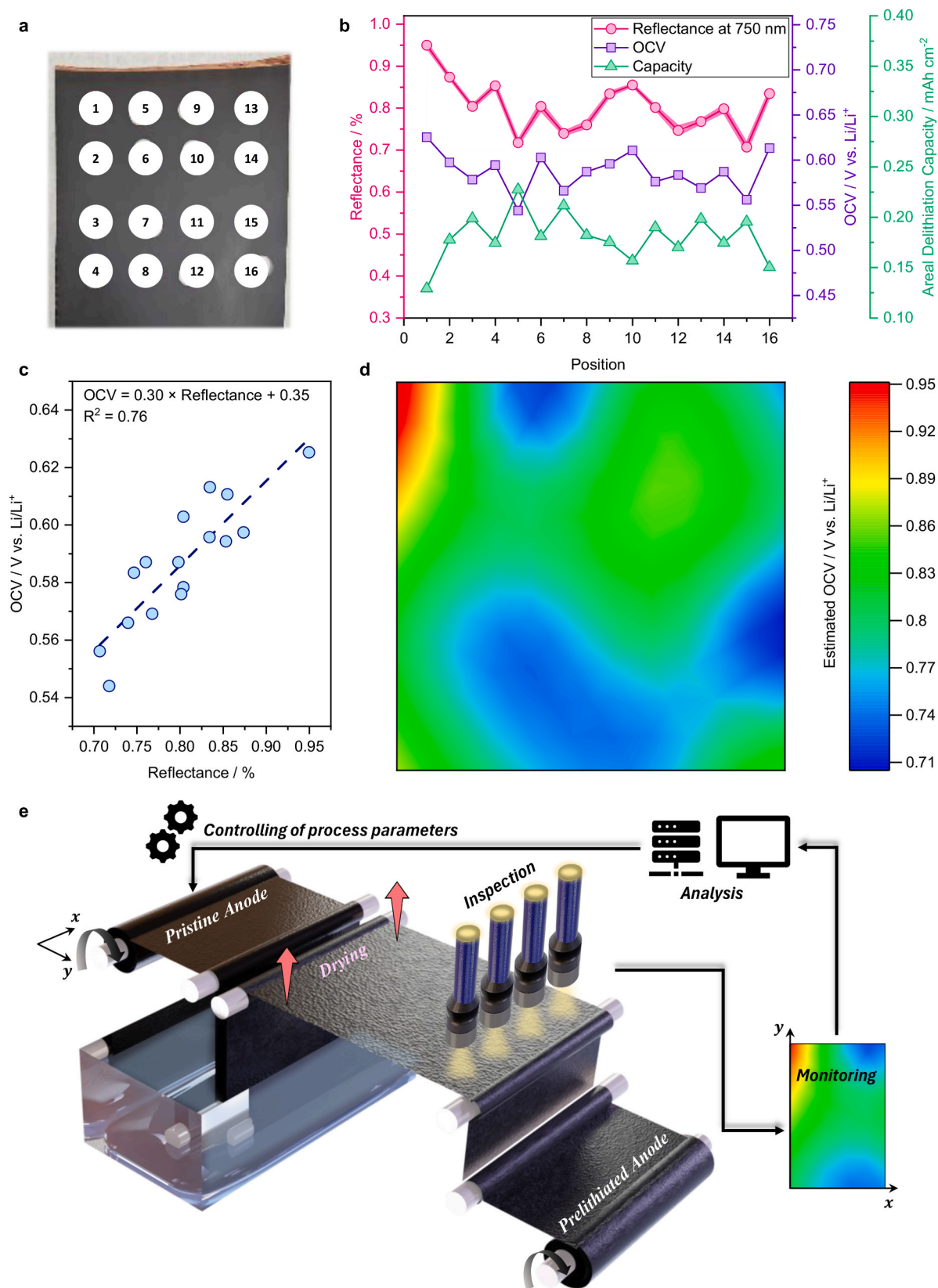


Fig. 7. (a) Prelithiated anode sheet processed via roll-to-roll method, with 16 sampling locations marked. (b) Comparison of reflectance, OCV, and areal delithiation capacity at each point. (c) Linear relationship between reflectance and OCV, with fitted regression equation. (d) Estimated OCV map derived from reflectance data using the fitted equation. Values between measured points were interpolated. (e) Schematic diagram of in-line and real-time spectroscopic inspection combined with an electrochemical prelithiation process. The data monitoring can be performed after drying of the prelithiated anode, as an example.

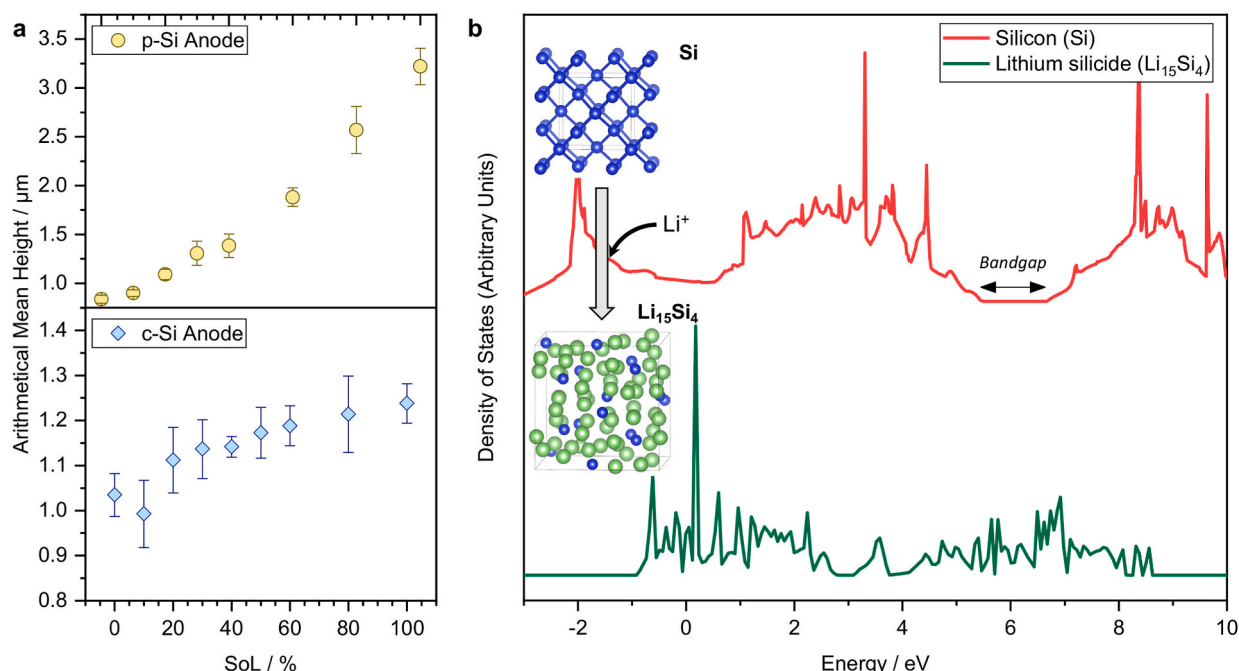


Fig. 8. (a) Mean values and standard deviation of arithmetical mean height of the p-Si and c-Si anodes depending on SoL. (b) Calculated density of states of crystalline silicon and lithium silicide (Li₁₅Si₄). The insets show the corresponding atomic structures of each material. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

near region. Therefore, in this work, we focus on a mechanistically supported interpretation rather than a fully deconvoluted optical model. The observed increase in surface roughness with SoL supports an enhanced scattering contribution, while the DFT results indicate band structure evolution toward bandgap narrowing and eventual closure at high lithiation, which is consistent with increased photon absorption.

4. Conclusion

The optical response of porous silicon-based anodes was systematically examined as a function of the SoL using complementary *ex situ* and *in situ* reflectance spectroscopy. Reflectance decreased with increasing lithiation and followed trends consistent with the electrochemical state, with the most robust correlation observed in the long-wavelength region for our setup (approximately 700–900 nm), whereas shorter wavelengths were more susceptible to reduced signal quality and subtler contrast.

To enable reliable long-duration operando measurements, we implemented a dual-spectrometer scheme to correct non-uniform light-source drift during slow lithiation. Although the optical penetration depth in porous silicon electrodes is limited, as supported by transmittance considerations and the substrate-insensitivity test, GD-MS depth profiling indicated approximately homogeneous lithium distribution across electrode thickness under the investigated conditions, supporting the use of surface-oriented reflectance as a practical proxy for bulk SoL.

These findings were successfully applied to a practical case involving the prediction of lithium distribution. Reflectance measurements were conducted on anodes lithiated using a smaller-area lithium opposite electrode. The resulting data effectively visualized the areal distribution of lithium and captured diffusion effects over time. Furthermore, a contactless OCV map of the prelithiated anode was generated by correlating local reflectance values with electrochemical measurements. Although this map should be regarded as semi-quantitative, it enables visualization of areal SoL variations and demonstrates the potential of the method for assessing prelithiation uniformity.

Finally, the reflectance change can be rationalized by combined

contributions from lithiation-induced microstructural evolution (increased surface roughness and scattering) and electronic-structure changes (enhanced absorption associated with bandgap narrowing/closure at high lithiation). Overall, this spectroscopic framework provides a non-destructive and contactless route for SoL tracking and heterogeneity mapping in silicon-based anodes, and it offers a foundation for process-relevant inspection concepts when paired with standardized calibration and further validation.

CRedit authorship contribution statement

Hyunsang Joo: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Software, Validation, Writing – original draft. **Malina Helling:** Data curation, Investigation, Methodology, Validation, Writing – review & editing. **Damian Mroz:** Data curation, Investigation, Methodology, Validation. **Nicha Wongkaew-Lenz:** Data curation, Investigation, Writing – review & editing. **Sascha Nowak:** Data curation, Investigation, Methodology, Supervision. **Sun-Je Kim:** Conceptualization, Investigation, Methodology, Supervision, Writing – review & editing. **Martin Winter:** Funding acquisition, Supervision, Writing – review & editing. **Egbert Figge-meier:** Funding acquisition, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Hyunsang Joo reports financial support was provided by Federal Ministry of Research, Technology and Space of Germany. Hyunsang Joo has patent #WO2022248222A1 pending to Forschungszentrum Jülich GmbH. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors are highly grateful to the German Federal Ministry of

Research, Technology and Space (BMFTR) for funding this work within the projects "PräLi-Präliithierung von Elektroden" (13XP0238A) and "InProMal-Integrierte und beschleunigte Prozessoptimierung zur Herstellung präliithierter Elektroden für Energiespeicher mit Methoden des Maschinellen Lernens" (13XP0532B). In addition, we sincerely thank Dr. Eunmi Im for graphical support, Dr. Akihiro Yamashita for scientific discussion, and the Wacker Team, including Dr. Stefan Haufe, Dr. Johanna Poschenrieder, and Dr. Ricarda Kloth, for providing silicon material and practical advice regarding electrode manufacturing.

Simulations were performed with computing resources granted by RWTH Aachen University under project rwth1450.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jpowsour.2026.240342>.

Data availability

Data will be made available on request.

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