

RESEARCH ARTICLE OPEN ACCESS

Combined Neutron- and Synchrotron-Based Diffraction Computed Tomography Studies of 18650-Type Lithium-Ion Cells

Vladislav Kochetov¹ | Dominik Petz^{1,2} | Tobias Hölderle^{1,2} | Volodymyr Baran³ | Alexander Schökel³ | Martin J. Mühlbauer⁴ | Michael Hofmann¹ | Olof Gutowski³ | Peter Staron⁵ | Ulrich Lienert³ | Vadim Dyadkin⁶ | Gavin Vaughan⁶ | Winfried Petry^{1,2} | Helmut Ehrenberg⁴ | Anatoliy Senyshyn¹

¹Heinz Maier-Leibnitz Zentrum (MLZ), Technische Universität München, Garching, Germany | ²Department of Physics, Chair for Functional Materials, TUM School of Natural Science, Technical University of Munich, Garching, Germany | ³Deutsches Elektronen Synchrotron DESY, Hamburg, Germany | ⁴Institute for Applied Materials (IAM-ESS), Karlsruhe Institute of Technology (KIT), Eggenstein-Leopoldshafen, Germany | ⁵Helmholtz-Zentrum Hereon, Geesthacht, Germany | ⁶Swiss-Norwegian Beamlines, ESRF-The European Synchrotron, Grenoble, France

Correspondence: Anatoliy Senyshyn (anatoliy.senyshyn@gmail.com)

Received: 25 March 2026 | **Revised:** 30 June 2026 | **Accepted:** 7 July 2026

Keywords: ageing | diffraction imaging | lithium-ion battery | neutrons | X-ray diffraction computed tomography (XRDCT)

ABSTRACT

The distribution of lithium in nickel-cobalt-aluminium (NCA) cathodes and graphite anodes of cylindrical (18650) high-energy lithium-ion batteries was studied using a combination of neutron- and synchrotron-based diffraction computed tomography (DCT). The in-plane lithium concentration in the positive and negative electrodes was assessed in situ at various nominal states of charge and health. DCT is one of the few techniques that enable non-destructive examination of electrochemical storage systems such as batteries. Several DCT configurations were explored, with probe-beam sizes ranging from millimetres to a few micrometres, demonstrating the method's capability across different length scales. The use of thermal neutrons as a probe in DCT was demonstrated for structural studies of lithium-ion batteries, where the results were compared to similar studies using high-energy photons. In addition, a region-of-interest approach for X-ray DCT was successfully demonstrated for the non-destructive characterisation of electrochemical storage systems.

1 | Introduction

Rechargeable lithium-ion batteries (LIBs) are essential for sustainable energy technologies due to their high gravimetric and volumetric energy densities, specific capacity, specific power and low self-discharge rates [1, 2]. While graphite remains one of the most commonly used materials for negative electrodes (anodes) in commercial LIBs today, silicon is increasingly added to modern cells to improve electrochemical performance [3, 4]. At the positive electrode side, there is a selection of cathode materials such as the olivine-type, like LiFePO₄, the spinel-type like LiMn₂O₄, and the frequently used delafossite-type LiCoO₂-based materials, where Co is substituted by other metals, e.g., Ni, Mn, Al, etc., resulting in

better accessibility, enhanced stability and safety, and a reduction of the overall costs [5]. The choice of electrode materials, as well as the morphology and chemical composition of various passive and active cell components, significantly impacts cell stability and performance [6]. These factors, along with cell design parameters, affect the transport of ions and electrons as well as the distribution and uniformity of lithium and electrolyte at macro-, micro- and nano-scale levels [7]. Consequently, this influences the performance, chemical stability and safety of LIBs.

Since LIBs are closed systems, quantifying the uniformity of charge distribution, reaction rates, current density and temperature gradients requires the application of non-destructive

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characterisation methods. Conventional approaches for in situ and operando characterisation include a range of imaging and spectroscopic techniques [8]. For studies of electrochemical systems, such as commercial battery cells, computed tomography (CT) and other high-energy X-ray- or synchrotron-based imaging techniques [9, 10] are widely utilised as these highly penetrative radiations enable in-depth analysis of LIBs' performance characteristics and dynamics under real operating conditions. In such instances, scattering-based methods focus on the uniformity of lithium distribution across cells and make it accessible [7, 11, 12]. X-ray diffraction CT (XRDCT) [13, 14] is a crucial technique due to its non-invasive nature, its high penetration depth of several centimetres, and its ability to provide both spatial and structural resolution. The effectiveness of XRDCT has been demonstrated in various scientific applications [15–20], particularly for the structural analysis of energy materials [21–24].

Neutron radiation typically serves as a complementary tool for characterising large samples, even surpassing its X-ray counterparts in certain contexts. The superior penetration depth of thermal neutrons empowers neutron powder diffraction (ND) studies of LIBs from low to high resolution. Different cell properties and operation aspects have been investigated using time- and spatially-resolved ND techniques, i.e., in situ and operando evolution of the crystal structure of electrode materials and its kinetics with different chemical compositions [25], state of charge (SOC) [26, 27], state of health (SOH) [28, 29], storage [30] and/or temperature [31]. Despite the challenges associated with sub-millimetre-sized neutron beams, the technique offers further advantages, including improved differentiation of transition elements, isotope contrast and enhanced sensitivity to light elements, enabling the observation of the electrolyte [32]. Conventional neutron CT and ND are routinely used, also in combination with X-ray (diffraction) CT [33]. However, it is advantageous to combine ND with tomographic data collection, such as neutron diffraction CT (NDCT), for which a first pilot experiment has recently been performed [34].

Steps towards higher-resolution operando studies on large (cm-sized) samples typically require long exposure times, generating large amounts of data whose collection, storage and handling are non-trivial. In many cases, focusing on a small region of interest (ROI) is sufficient for high-resolution analysis and requires less effort in terms of acquisition time and data volume. Such local tomography is an obvious approach for maintaining high resolution while focusing on small ROIs in larger samples. ROI X-ray CT, also known as local CT, is applied routinely nowadays [35, 36]. In our opinion, a similar approach may be applicable to diffraction-based CT without compromising generality. This has been demonstrated for millimetre-sized LIB electrodes [21] and a prototypical sample [34], but it has not yet been tested on a full-size 18650 battery cell.

In this contribution, we demonstrate the methodological application of various aspects of DCT techniques to similar-type cylinder LIBs. A set of techniques, including NDCT, spatially-resolved XRD with conical slits, and (ROI) XRDCT, was used to non-destructively look into the batteries' interior, continuing a series of investigations using diffraction and imaging techniques for battery research [34, 37, 38]. The results are organised by increasing characterisation resolution, examining the static distribution of lithium in the anode and cathode of cylindrical batteries at various length scales, ranging from millimetres to micrometres.

2 | Experimental Section

2.1 | Cell Chemistry and Preparation

For all measurements, commercial 18650-type cells (NNP – nickel oxide-based new platform, NCR18650B, Panasonic) were prepared for characterisation in three different states: fresh charged, aged charged and aged discharged. The studied cells were taken from the same batch (factory formed) and are characterised by an initial capacity of 3350–3400 mAh, an operational voltage window of 2.5–4.2 V, and a high specific energy in the 200–260 Wh/kg range [39]. To introduce substantial cell degradation, a cell, which will be further referred to as aged, was cycled up to 3000 times. Each electrochemical cycle consisted of a discharge phase at a constant current of 3.4 A (1 C) down to 2.5 V, followed by a charge phase with a constant current at 1.65 A (approximately 0.5 C) up to 4.2 V and a constant voltage phase, where charging is continued at 4.2 V until a cut-off current of 160 mA (approximately 1/20 C) is reached. A 15 min relaxation period was applied between the discharge and charging steps. After 3000 cycles, the aged cell's SOH was reduced to approximately 38% when measured at a discharge rate of 1 C and 79% when measured at the reference discharge rate of 1/8 C of the initial capacity of the fresh cell (corresponding to SOH of 100%). Electrochemical characterisations are shown in Figures S1 and S2.

2.2 | NDCT

The NDCT experiment was carried out with the standard setup of the materials science diffractometer STRESS-SPEC [40] (neutron source FRM II, Garching b. München), including sample environment with a translation and rotation stage, a beamforming primary and secondary slit systems, a 2D ^3He -PSD detector covering an area of $25 \times 25 \text{ cm}^2$ with $256 \times 256 \text{ px}^2$, and a beam-stop, see Figure 1a. The sample was irradiated by neutrons taken from a Ge 511 monochromator at a 66° take-off angle, giving rise to a neutron wavelength of $1.63 \text{ \AA}$. A neutron monitor was located in the direct (downstream) beam to facilitate the simultaneous collection of neutron attenuation data. The sample was translated perpendicular to the beam direction at a raster resolution of 1 mm. A Soller collimator of $5'$ in front of the sample was installed to limit the horizontal beam divergence in order to reach the desired spatial resolution. The 2D detector was located at a distance of 1 m from the sample, covering 16° in 2θ . It was centred at $25^\circ 2\theta$ with respect to the direct beam. The chosen 2θ region contains the characteristic first-order Bragg reflections corresponding to different phases of lithiated graphites. The small beam size of $1 \times 10 \text{ mm}^2$ ($H \times V$), along with the tight beam collimation, results in a quite small sample volume to be irradiated, thus limiting the counting rate. The collected 2D data were reduced using the program Steca² [41]. Examples of both are shown in Figure 1b. The nominal voxel size of the reconstruction is $1 \times 1 \times 10 \text{ mm}^3$.

2.3 | X-Ray CT

The cell layout was studied non-destructively by X-ray CT using a high-resolution Phoenix v|tome|x s 240 tomographic scanner. The direct tube's voltage was set to 130 kV and a current of $100 \mu\text{A}$. The measurements were performed using a cone-beam geometry with a detector having a sensitive area of $200 \times 200 \text{ mm}^2$.

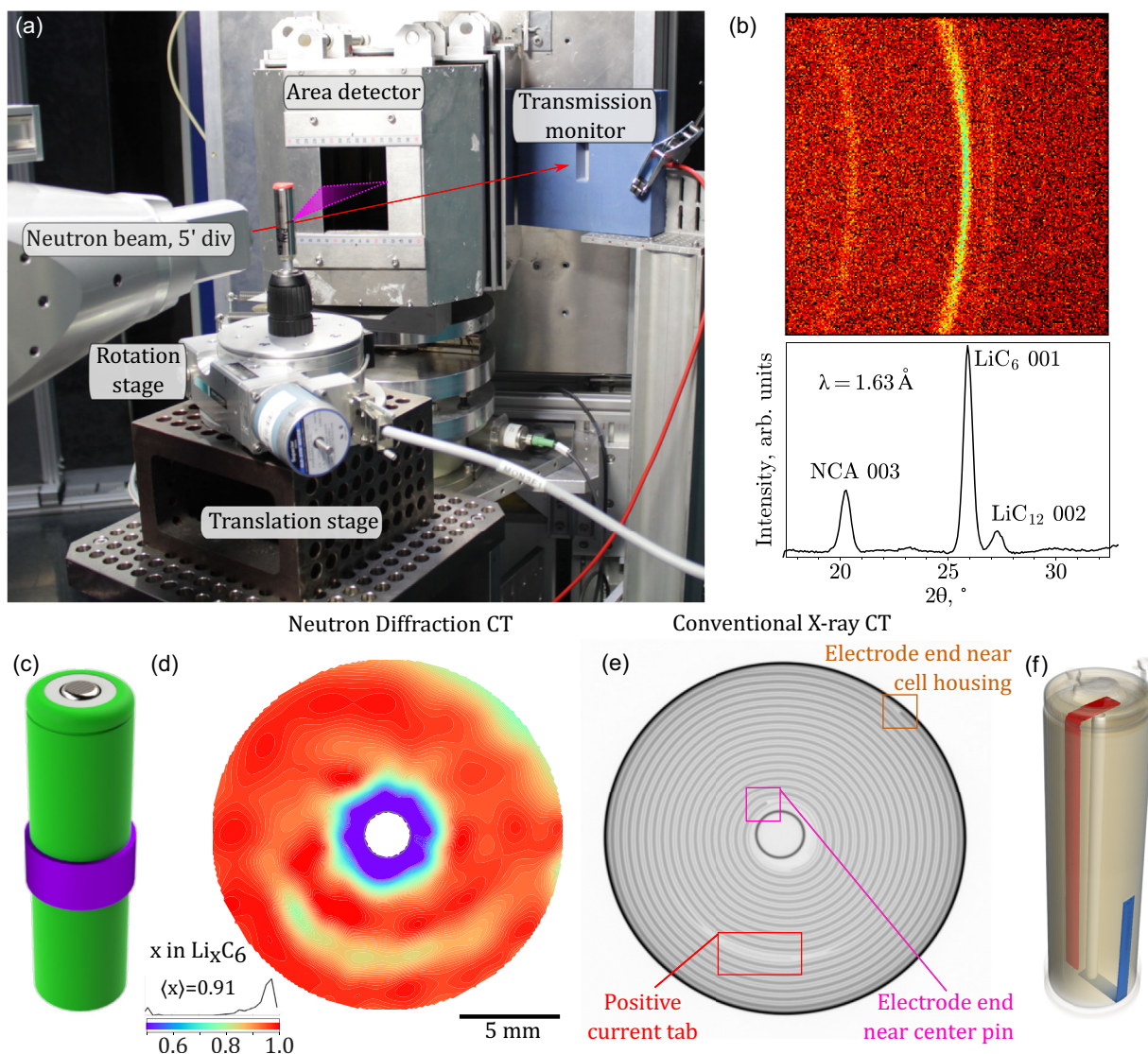


FIGURE 1 | Details of the NDCT experiment. (a) Photo of the NDCT setup at instrument STRESS-SPEC; (b) exemplary 2D data collected on the neutron area detector and radially integrated diffraction pattern; (c) illustration of the probed plane in measurements with a neutron beam of 1×10 ($H \times V$) mm^2 ; (d) lithium distribution inside the anode with a nominal voxel size of $1 \times 1 \times 10 \text{ mm}^3$ obtained from the NDCT reconstruction (histogram distribution of x is shown with a solid line above the colour bar) and (e,f) Reference middle-plane slice and full volume reconstruction of the cell obtained from conventional X-ray CT with a voxel size of $40 \mu\text{m}$ [38].

($1000 \times 1000 \text{ px}^2$). The magnification was adjusted to achieve an effective pixel size of about $40 \mu\text{m}$. In total, 1001 X-ray radiographs were collected, each corresponding to an average of three single exposures of 1000 ms. Between consecutive radiographs, two single exposures were skipped. The reconstruction, analysis and final data visualisation were performed with the Phoenix datos|x software, ImageJ [42] and VGStudio MAX from VolumeGraphics, respectively. Data from these measurements have been partially published previously in ref. [38] and used in this article for illustration purposes.

2.4 | Spatially-Resolved XRD Using Conical Slits

A spatially-resolved diffraction experiment with conical slits was performed at the HEMS beamline (P07) at PETRA III (DESY, Hamburg) [43]. The experiment employed a goniometer, allowing

for sample translation and rotation, a PerkinElmer XRD1621 fast area detector ($200 \times 200 \mu\text{m}^2$ pixel size, 2048×2048 pixel area), and a slit system [44] positioned in front of the cell. An optimal wavelength of $0.208 \text{ \AA}$ (59.6 keV), matching the 001 reflection from LiC_6 and the 002 reflection from LiC_{12} with the first opening of the conical slits, was chosen. With such a configuration of the slits shaping the incoming photon beam to a size of $100 \times 100 \mu\text{m}^2$, a needle-like gauge volume of about $0.1 \times 0.1 \times 1.03 \text{ mm}^3$ was formed, and data were collected on a polar mesh (Figure S3), with steps of 1 mm in radial direction of the cell and 30° for the rotational steps in angular direction.

2.5 | XRDCT

Several synchrotron-based XRDCT experiments were conducted at the powder diffraction beamline P02.1 [45] at PETRA III (DESY,

Hamburg), operating with high-energy X-rays at 60 keV (0.207 Å). The instrumental setup was similar across all XRDCT experiments, consisting of a source with a needle beam and a fast area 2D detector, as sketched in Figure S4a. The sample was translated along an axis perpendicular to the beam direction and rotated after each complete translation scan. A PerkinElmer XRD1621 detector was positioned at a sample-to-detector distance of 2 m. A beam size of $0.5 \times 0.5 \text{ mm}^2$ and different translation steps of 0.25 and 0.5 mm were employed, yielding reconstruction voxel sizes of $0.25 \times 0.25 \times 0.5 \text{ mm}^3$ and $0.5 \times 0.5 \times 0.5 \text{ mm}^3$, respectively. For comparison and illustration (Figure 2), data sets were binned to 0.5 and 1 mm steps by averaging consecutive diffractograms.

2.6 | High-Resolution XRDCT

High-resolution XRDCT was conducted at beamline ID15A (ESRF, Grenoble) [46] using a $30 \mu\text{m}$ -sized beam with a wavelength of 0.133 Å ($\sim 93 \text{ keV}$). The PerkinElmer 2D detector was used to collect diffraction data, which were then integrated using the Bubble package [47]. The nominal voxel size is $30 \mu\text{m}$. Reconstructed images were virtually unrolled by transformation into polar coordinates using the OpenCV Python library [48].

Additional high-resolution measurements on the aged cell were performed at beamline P07 at PETRA III (DESY, Hamburg), using a monochromatic beam with a wavelength of 0.169 Å (73.3 keV) and a spot size of $30 \times 2 \mu\text{m}^2$ (H $\times$ V). Data were collected using a DECTRIS EIGER2 X CdTe 4 M detector with a translational step size of $19 \mu\text{m}$ for the horizontally mounted cell. Attenuation data were acquired using a silicon diode installed at the beamstop position and were used for the absorption correction. The nominal voxel size of the reconstruction is $19 \times 19 \times 30 \mu\text{m}^3$.

A further high-resolution ROI XRDCT experiment was conducted at the Swedish Materials Science Beamline P21.2 [49] at PETRA III (DESY, Hamburg), which covered a translational range—ROI—of $480 \mu\text{m}$ using a small needle beam of $4 \mu\text{m}$. The scattered photons with a wavelength of 0.230 Å (54 keV) were

collected on a PILATUS2 $\times 2 \text{ M}$ detector. Nominal voxel size of the reconstruction is $4 \times 4 \times 10 \mu\text{m}^3$.

3 | Results and Discussion

The studied LIB adopts an NCA cathode ($\text{Li}_y\text{Ni}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$, $0 < y < 1$, which we will denote as $\text{Li}_y(\text{NCA})$) and graphite anode chemistry with aluminium and copper current collectors, a polypropylene (PP/PE) separator, and a liquid organic electrolyte composed of a 72:5:23 DMC:EC:EMC mixture containing LiPF_6 salt [32]. The internal cell layout was revealed by absorption-based X-ray CT, as shown in Figure 1e,f. It adopts a typical jelly-roll design, including the centre pin, housing, current tabs and electrode sheets. The geometrical cell layout plays a crucial role in determining cell performance and the uniformity of lithium distribution across both electrodes, which can be evaluated using diffraction techniques.

For the anode, the relative lithiation value x in Li_xC_6 is determined based on the partial phase concentrations of LiC_6 , LiC_{12} , and low-lithiation $\text{Li}_{x < 0.5}\text{C}_6$ phases. These concentrations are estimated by analysing the corresponding diffraction peak intensities. For the cathode, the lithium content y in $\text{Li}_y(\text{NCA})$ is calculated from the positions of the diffraction peaks 003 and 101 of the NCA material. The relationship between lithium occupation y in $\text{Li}_y(\text{NCA})$ and SOC has already been established by ex situ ND and inductively coupled plasma optical emission spectroscopy [50].

Using spatially-resolved ND [38], it was demonstrated that, for this type of cell in its charged state, the anode lithiation exhibits local minima in the middle of the cell, specifically around the centre pin and at the positive current tab positions. The method of NDCT, illustrated in Figure 1a, facilitates tomographic data collection with a rectangular gauge volume cross-section of $1 \times 1 \text{ mm}^2$, compared to the rhombic cross-section of 9.48 mm^2 used in the referenced spatially-resolved ND experiment. The inhomogeneities of lithiation in the anode of the charged fresh

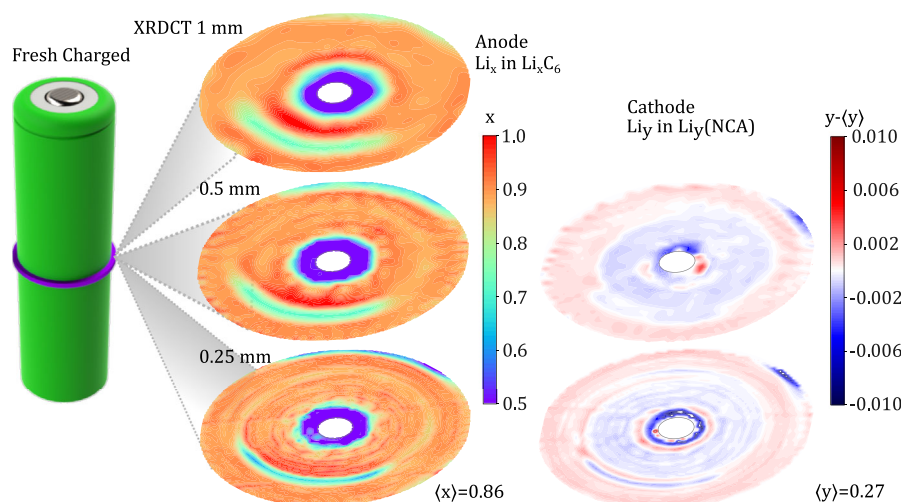


FIGURE 2 | XRDCT (beamline P02.1) reconstructions acquired from the fresh charged cell with different translational steps of 0.25, 0.5 and 1 mm: Lithium distribution in the anode and corresponding deviation of the NCA lithiation from the mean value $\langle y \rangle$.

cell reveal a distinct pattern, as observed with NDCT (see Figure 1d). At SOC 100%, the anode lithiation reaches levels of 90%–100% in most areas of the cell, clearly reflecting the high-energy type of the studied cells, while decreasing to 50%–70% at the centre, near the positive current tab, and partially at the edge of the cell close to the cell housing. This variation can clearly be correlated with the cell's geometry, as lower lithiation tends to occur at the ends of the electrode stripes. The mean value of anode lithiation $\langle x \rangle = 0.91$ in the charged state obtained with NDCT aligns with the findings from the spatially-resolved ND experiments reported in refs. [38, 51].

Currently, achieving a spatial resolution of less than 1 mm in neutron diffraction imaging remains challenging. The application of time-of-flight techniques, combined with a 90° detector bank geometry, offers better prospects than monochromatic neutrons for the shape and dimensions of the gauge volume. However, with XRDCT, this level of resolution is more easily accessible. An alternative for diffraction imaging could be spatially resolved XRD using conical slits, similar to spatially resolved ND. In spatially resolved diffraction experiments, diffraction patterns are collected at selected positions or along 2D or 3D scan paths on a predefined mesh. Consequently, the measured signal represents the diffraction response integrated along the beam path through the sample, and interpretation may be affected by the superposition of information from different depths. To reduce spurious contributions, a secondary collimation—downstream of the sample—is needed. Furthermore, owing to the experimental geometry, the gauge-volume cross-section in spatially-resolved experiments is typically rhombic, whereas the effective voxel in XRDCT is approximately rectangular (or square in the reconstructed slice) as a result of the tomographic reconstruction workflow. However, measurements conducted with conical slits [37] revealed that this kind of synchrotron technique suffers from low count statistics. As a result, it requires long exposure times, which complicate the collection of high-resolution data at large grid densities (see Figure S3). An XRDCT experiment was performed (Figure 2), utilising a comparable 1 mm scanning step to further validate the NDCT technique. On the qualitative level, similar results were obtained, whilst certain discrepancies were observed between the data obtained from these two methods on the quantitative scale. They can be attributed to a non-identical SOC of the cell, caused by the storage conditions after charging, leading to a lower total lithium content in the anode, with a mean value of 0.86 measured by X-rays (as shown in Figure 2, compared to the “neutron” value of 0.91). Another experimental difference to consider is the beam's vertical dimension. For neutrons, the vertical size of the beam cross-section was set to 10 mm in order to improve counting statistics by a larger irradiated sample volume, while for X-rays, a height of 1 mm was chosen for the beam. This discrepancy results in neutron measurements averaging over a larger volume of the cell.

Applying the 1 mm translation step, an agreement of the distributions obtained by ND and XRDCT is unambiguously observed from Figures 1d and 2. With the use of “oversampling,” i.e., by reducing the translational step size in the XRDCT data acquisition down to 0.5 mm and then to 0.25 mm (see Figure 2), finer details of lithiation become apparent for both the negative and positive electrodes. Examples of collected diffraction data at different SOC and SOH of the studied cells (averaged over the examined plane) are presented in Figure S4b. The calculated mean lithium concentration $\langle x \rangle$ in

the anode was observed to be highest for the fresh charged state (Figure 3a–c). Cell ageing leads to a reduction of the mean lithium concentration $\langle x \rangle$ inside the anode in the charged state, i.e., the observed mean lithiation value dropped to $\langle x \rangle = 0.68$, which is about 79% of the initial value $\langle x \rangle = 0.86$ of the fresh charged cell. This reduction is directly correlated with the remaining cell capacity of around 80% for the aged cell. The observed decrease in lithium concentration in the charged state after ageing is associated with the loss of active lithium, a phenomenon previously reported with various ageing protocols in 18650-type cells using $\text{LiCoO}_2|\text{C}$, $\text{NCA}|\text{C}$ and $\text{NMC}|\text{C}$ chemistries [52, 53]. Additionally, the lithium concentration (probed in the charged state) decreases at approximately 5 mm from the cylinder centre, indicating a circular zone of reduced electrochemical activity. This location matches the position of the positive current tab. On the contrary, in the discharged state shown in Figure 3c, these positions around the current tab and at the end of the electrode exhibit small amounts of lithium residue in the graphite anode. In Figures 2 and 3, the interpolated reconstructions from the middle cross-sectional plane of the cell are presented. It is worth noting that the studied cell is characterised by a degree of height homogeneity along its height as revealed by measurements at three selected planes (Figure S5).

The lower row in Figure 3 illustrates the deviation of the lithium content from the calculated mean value $\langle y \rangle$ inside the cathode. It is worth noting that while the anode lithiation spans $x \in [0, 1]$, for the cathode, the range is given by $y \in [0.23, 0.85]$ (see Figure S8 for the relationship between the lattice parameters and the lithium content). The upper limit of y (or its deviation from 1) can be associated with lithium losses during cell formation and degradation. In the discharged state, the mean cathode's lithiation level of 0.79 reflects the amount of active lithium in the cathode and, therefore, the available capacity [54]. However, the degree of cathode delithiation in the charged state is not expected to exhibit the same direct correspondence with capacity fade, showing approximately the same amount of retained lithium in the fresh and aged states, 0.27 and 0.25, respectively. The variation of the lithiation y inside the cathode is ~10% for the discharged state and ~1% for the charged state. As expected, the inhomogeneities observed in the charged state are more pronounced for the aged cell compared to those of the fresh cell (Figure 3e,d), which is directly attributed to cell degradation and the accompanying augmentation of the heterogeneous state. Furthermore, certain differences in the inhomogeneity patterns are observed at the current tab, separating the inner and outer windings as reflected in the distribution histograms. It is essential to note that, in reality, the variation in cathode lithiation can exceed 1% for charged cells and 10% for aged, discharged cells.

To quantify lithium distribution in each voxel, we assume a uniform lithiation state (or SOC) within the voxel, yielding an averaged value according to a one-component model. A more complex situation arises when several phases of the (de)lithiated cathode are present in the observed region/volume, leading to partial volume averaging. For example, in the charged state, there are contributions from both low-lithium content (corresponding to SOC = 100%) and high-lithium content, i.e., the discharged/aged cathode state. Unfortunately, the achieved resolution, along with the overall quality of the reconstructed cathode signal (see Figure S6), does not permit analysis in the frame of a two-component model [55]. Nevertheless, the

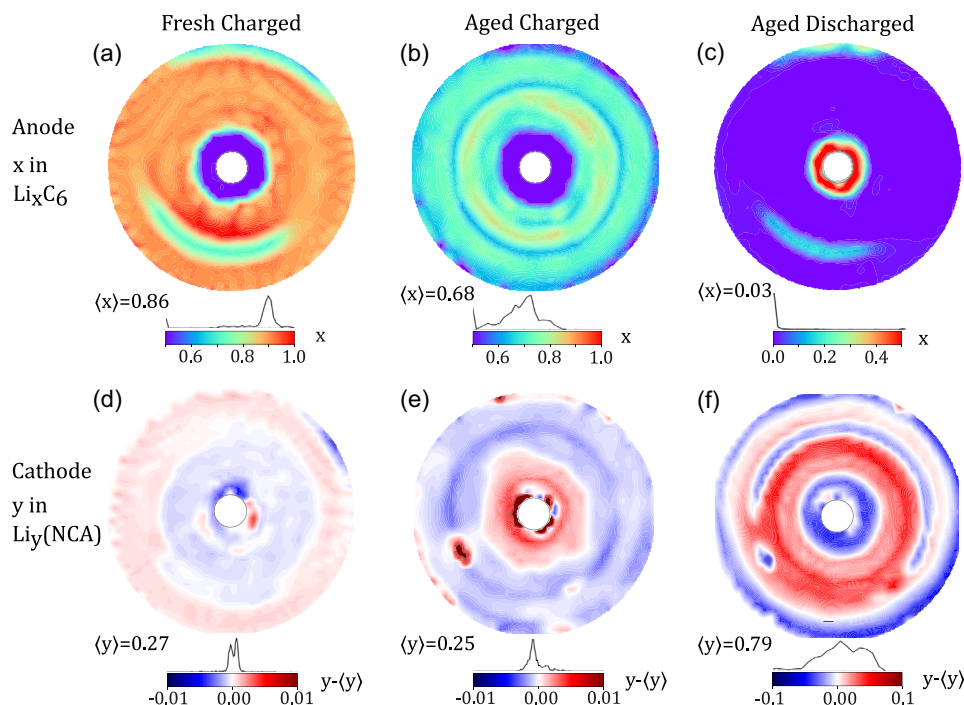


FIGURE 3 | Lithiation distribution (XRDCT data, translational step size 0.5 mm, beamline P02.1) in (a) the anode of the charged fresh cell, along with the data for (b) a charged and (c) discharged fatigued cylinder-type cell with SOH of 80%. (d–f) The corresponding distribution of the Li content in the cathode $y - \langle y \rangle$ (displayed as the deviation from the mean value). The histogram distributions are shown with solid lines above the colour bars.

lithiation distributions obtained from the single-component analysis can at least be considered and discussed qualitatively.

These results motivated high-resolution XRDCT experiments, capable of yielding details of lithium distribution in 18650-type batteries at a finer length scale down to several micrometres. With high-resolution XRDCTs utilising raster steps of 30 or $\sim 20 \mu\text{m}$, every single layer of the fully charged LIB's double-coated electrodes ($>150 \mu\text{m}$) could be resolved for the fresh and aged cells. Studies were performed at the middle height of the cell, i.e., the centre of the rolled electrodes, revealing the spiral structure of the rolled double-layer anode. Figure 4a shows the deviation of lithiation from the estimated mean value $\langle x \rangle$ of 0.95 inside the anode, while the cathode in Figure 4b shows a low mean lithiation of $\langle y \rangle = 0.238$ in the fully charged state. It has to be noted that the observed mean value of lithiation of $\langle x \rangle = 0.95$ inside the anode of the fresh cell is higher than the one obtained with 0.5 mm resolution (Figure 3a, $\langle x \rangle = 0.86$) due to differences in the storage conditions. The results of high-resolution XRDCT have been found in satisfactory agreement with the mean lithiation values obtained from neutron measurements ($\langle x \rangle = 0.91$ for the fresh cell). The corresponding high-resolution XRDCT results for the aged cell are presented in Figure 4c,d. The mean values of lithiations, $\langle x \rangle = 0.70$ and $\langle y \rangle = 0.26$, are found in excellent agreement with low-resolution XRDCT measurements, $\langle x \rangle = 0.68$ and $\langle y \rangle = 0.25$. The amount of information in $660 \times 660 \text{ px}^2$ (fresh) and $1000 \times 1000 \text{ px}^2$ (aged) lithiation distribution maps is rich. Some details are highlighted by zoomed-in insets in Figure 4a–d. The histograms in Figure 4e,f show the different distributions of lithiation levels for the fresh and aged cells. Figure 5 shows the lithiation values x and y along a virtually unrolled electrode stack. The thickness-averaged electrode lithiations and trends are shown as a function of winding number along the unrolled double-layer

spiral, which can be linked to position along the electrode stripe. The numbers (1) to (6) correlate to the approximated the positions of the insets in Figure 4.

Systematically lower lithium concentrations in the anode are observed at the inner (inset 1) and outer (inset 6) ends of the electrodes and at the radial position close to the positive current tab—located at the 11th spiral winding (insets 3 and 4). This in situ observation qualitatively agrees with ex situ diffraction measurements, where the anode stripes were physically extracted, unrolled and scanned [56]. The highest remaining lithium in the cathode was detected near the positive current tab, around the 11th spiral turn (insets 3 and 4). Because of the missing anode coating, this region of the cathode is inactive or less active. The missing anode coating also explains the low lithium content at the same position in the anode. On the other hand, the lowest amount of cathode's lithium is observed at the electrodes' ends (insets 1 and 6) as the outer coating of the anode misses a counter electrode and introduces an excess of graphite. Variations in cathode lithiation are generally small within 1% for the fresh cell and 2% for the aged one (Figure 4f), while the anode shows significant lithiation inhomogeneities (Figure 4e), starting with a low concentration of $\langle x \rangle = 0.5$ in the cell centre¹, followed by increasing lithiation levels up to $\langle x \rangle = 1.0$. Ageing led to an overall decrease in the anode's lithiation as seen in the middle of the cell (inset 2) and at the cell's edge (inset 5). It has to be noted that due to the high resolution, the distribution histograms of lithiation inside the anode show two maxima corresponding to LiC_{12} (more in the aged cell) and LiC_6 (prevailing in the fresh cell), rather than a Gaussian-like distribution as observed in low-resolution XRDCT. The domains of homogenous lithiation inside the cathode are still smaller than the achieved resolution (20 to 30 microns), leading to spatial averaging and Gaussian-like distributions.

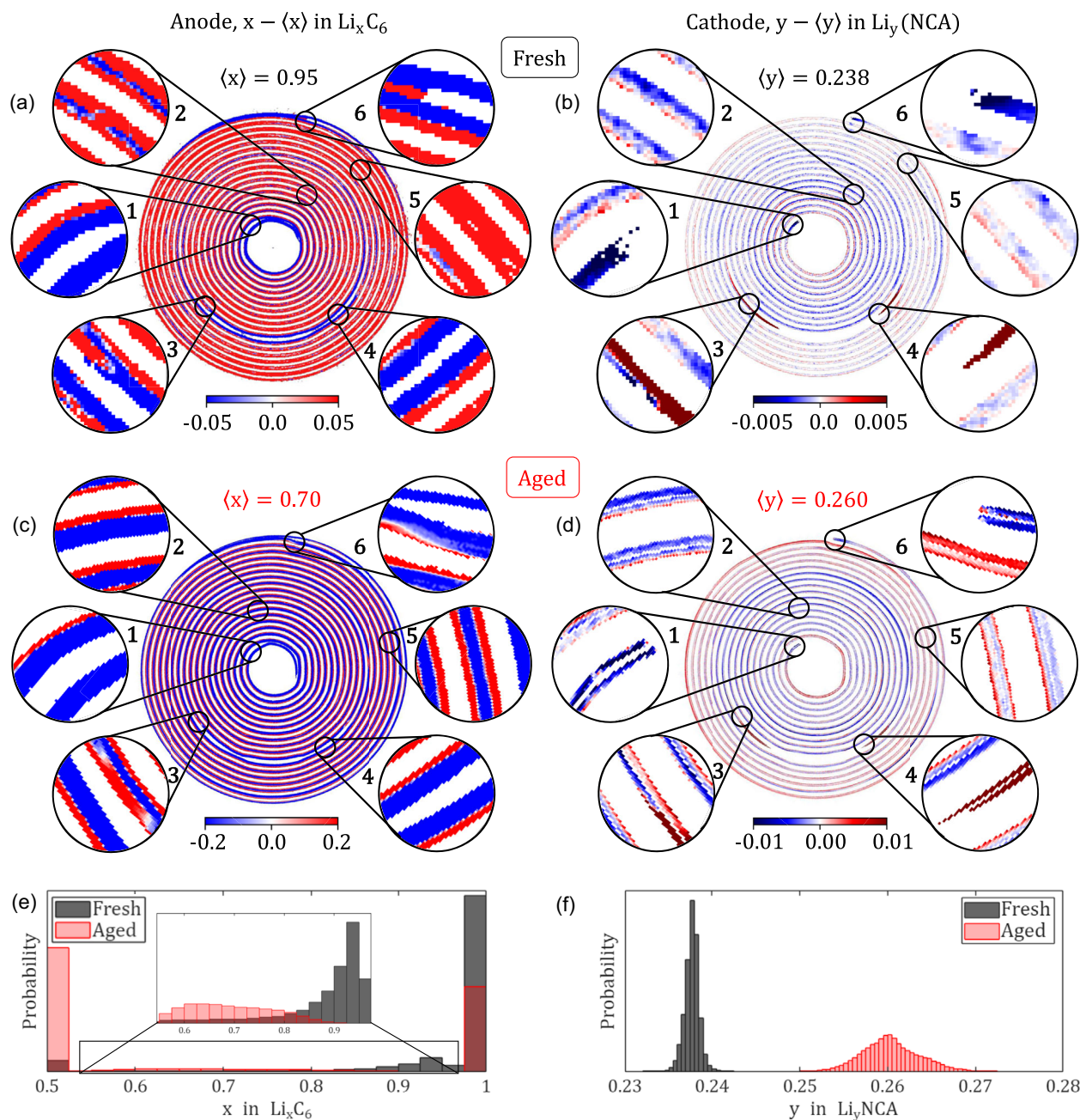


FIGURE 4 | High-resolution XRDCT of (a,b) the fresh (translational step 30 μm , beamline ID15A) and (c,d) the aged cell (translational step 19 μm , beamline P07), both in the charged state, where (a,c) show the deviation of lithiation from the mean value $\langle x \rangle$ inside the graphite anode and (b,d) the deviation of the lithiation from the mean value $\langle y \rangle$ inside the NCA cathode. The histograms show distributions of lithiation levels inside (e) the anode and (f) the cathode for fresh and aged cells.

The general effects of ageing mentioned above also hold along the length of the electrodes: an overall decrease in lithiation inside the anode, as seen in the black versus red lines in Figure 5a,b, however, preserving characteristic features of the distribution along the electrode length. On the contrary, aged cathodes retain more lithium in the charged state than fresh ones do. Achieving high spatial resolution enables the examination of the variation in lithium concentration throughout the thickness of the double layers of both the anode and cathode. In the anode, the lithium concentration is lower on the concave inner side (blue) and higher on the convex outer side (green), predominantly near the cell centre (Figure 5a,b). In contrast, the cathode exhibits higher lithium concentrations

on the concave side and lower concentrations on the convex side, as illustrated in the insets 1, 2, and 5 of Figure 4b,d. Part of the observed behaviour can be attributed to the electrical connection scheme of the electrodes [38], where a general drop of the lithiation level x towards the centre of the cell is expected due to the resistive network of the cell.

The unrolled lithiation patterns for the inner and outer stripes support these observations, as shown in Figure 5c. Despite the trends for the fresh cell being qualitatively similar—specifically, there is systematically lower lithiation at the inner anode and outer cathode (see Figure S7 for a zoom-in view)—the differences are significantly

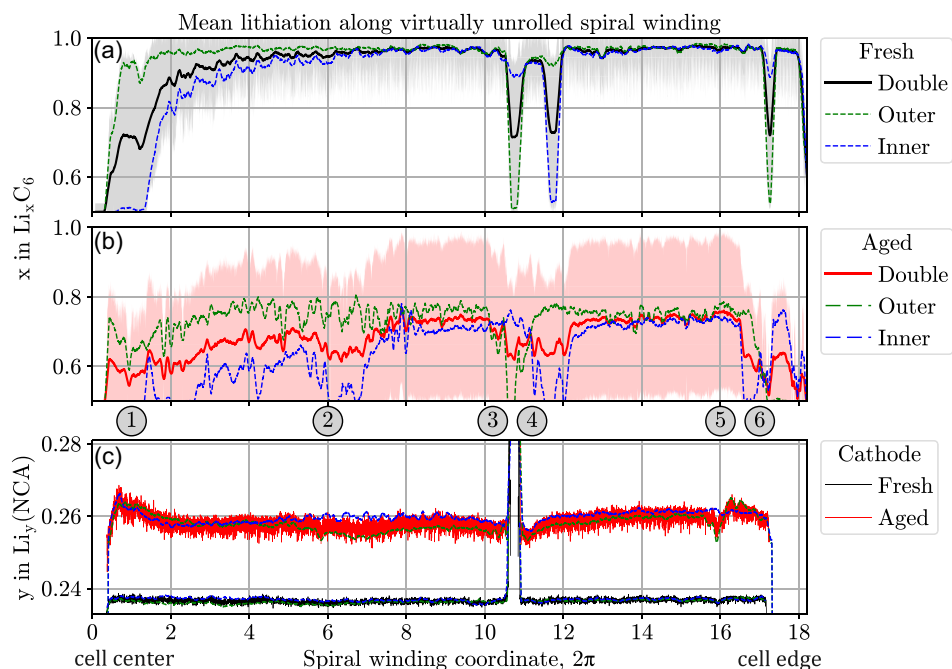


FIGURE 5 | Mean lithiations x and y along the virtually unrolled (a,b) anode and (c) cathode double electrodes for fresh (black) and aged (red) cells. Trend lines of averaged lithiations along inner concave (blue) and outer convex (green) layers are shown along the corresponding double layer, which is the sum of both. Corresponding variations in the fresh cell's cathode are significantly lower than those inside the aged one (see Figure S7). The y -values are given as they are, while the smoothed trend lines, along with their standard deviation (shaded areas), are plotted for x -values. Numbers in circles correspond to the positions of the insets in Figure 4.

smaller, indicating that the fresh cell can be considered more balanced. The mismatch between layers becomes more pronounced with cell ageing, while the trend remains similar in the charged state—more lithium remains in the inner cathode and outer anode stripes. Different balancing of “concave anode–convex cathode” and “concave cathode–convex anode” interfaces can potentially be attributed to a surface/volume effect of spiral winding. The volume difference between two consecutive layers, e.g., 150 μm , can be estimated to be $\sim 1.0\%$ at a radial distance of 2 mm from its centre and 0.1% at 8.5 mm (close to its edge), consistent with the order of the observed lithiation differences in the cathode layers. On the other hand, the inhomogeneities in lithium distribution and the lower overall concentration near the cell centre can be attributed to greater mechanical degradation of the positive electrode. High-resolution CT imaging has revealed mechanical or intercalation-induced cracks within the cathode [57]. Additionally, slight variations across

the double layer can be caused by misalignment of the electrodes during cell assembly.

Further, high-resolution XRDCT was applied to detect inhomogeneities on an even smaller length scale, specifically at the interface between cathode, separator and anode. The application of local XRDCT to large, highly absorbing and scattering samples is a non-trivial issue (see discussion in the Supporting Information: Figure S9). The lateral padding of sinograms with averaged edge values is a simple technique that does not require additional measurements [58, 59]. The reconstruction approach is primarily guided by ref. [60], which demonstrates the practical applicability of conventional local CT.

Figure 6b,c shows the results of synchrotron-based XRDCT on a small ROI of 480 μm diameter chosen at a radial distance of

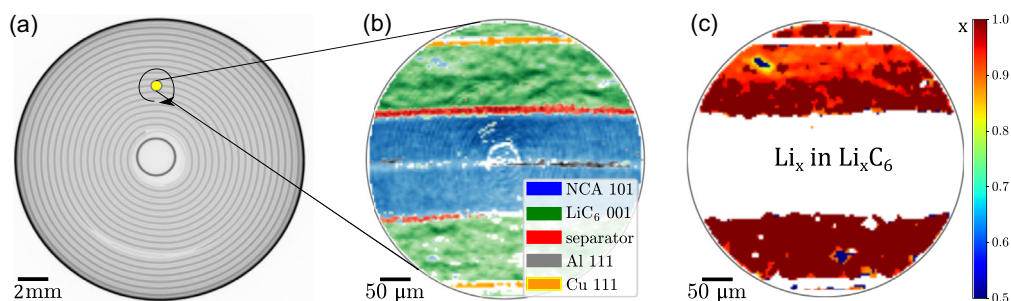


FIGURE 6 | ROI XRDCT (beamline P21.2) with a 4 μm beamsize: (a) reference cell layout from CT with the ROI of 480 μm in diameter marked by the yellow circle; (b) overlaid reconstructions from different phases. Colours correspond to one-colour colourmaps, which were used for the reconstruction of each reflection and (c) anode lithiation applying the colour scheme for the value x .

4.5 mm from the centre of the cell (Figure 6a). The reconstructions of five different phases are shown in Figure 6b, allowing for a clear distinction of the layered structure of the electrodes, whose width is approximately 100 μm : one anode layer, a double-layer of the cathode and another anode layer. The reconstruction of the anode lithiation revealed grains of lower and higher lithiation. A lithium-depleted region is observed close to the copper current collector, showing a significantly lower lithiation of the graphite, which is expected since the ROI was chosen at a position of lower lithiation in the middle of the cell, see insets 3 and 4 in Figure 4a. An increased lithium concentration is observed at the separator side in agreement with ref. [37]. An accurate analysis of the cathode peak positions was not possible due to the poor quality of the reconstructed data. Additionally, the half-tomo configuration of the experiment (i.e., collecting projections over 0° – 180°) causes artificial peak shifts for large samples [61], which introduce additional artefacts.

4 | Conclusion

A series of non-destructive techniques based on synchrotron and neutron radiation was applied to probe the interior of a cylinder-type LIB at different spatial resolutions. The NDCT experiment performed on 18650-type LIB represents an upgrade over earlier spatially resolved ND experiments, which faced challenges such as low count rates due to a restricted gauge volume and short integration times. These findings underscore the promising application of NDCT in battery research, complementing—and in some cases exceeding—the capabilities of X-ray-based approaches. Although the use of sub-millimetre-sized neutron beams presents technical challenges, the method offers several unique advantages over XRDCT, including the investigation of large-format cells, improved differentiation of transition elements in cathode materials, and greater sensitivity to isotopes and light elements, making it particularly relevant for electrolyte-related studies. The distribution of electrolyte in Li-ion cells is particularly challenging to investigate with X-rays because of the electrolyte's low contrast. In contrast, NDCT offers enhanced sensitivity and is therefore well suited for measuring electrolyte distribution, as demonstrated in frozen cells using spatially resolved ND [32]. Key factors for the success of future NDCT experiments include high beam collimation, high neutron flux, and advanced detector technology and neutron optics. The potential for further development and utilisation of NDCT is evident, particularly with the prospect of higher neutron fluxes at facilities such as the European Spallation Source [62].

Synchrotron-based DCT significantly outperforms NDCT in terms of spatial resolution. The current study particularly demonstrates the qualitative distinction of inhomogeneities in aged and fresh LIBs with sub-millimetre resolution. The demonstrated potential of ROI XRDCT represents a significant advancement towards achieving micrometre-resolution in centimetre-sized samples. The time required to measure a single cross-sectional slice remains a significant obstacle to implementing DCT for comprehensive 5D characterisation (3D spatial + phase + time) of batteries, although substantial progress in the technique's speed has recently been made [63]. Given the high-intensity flux, micrometre-sized high-energy beam and millisecond exposure times available at modern synchrotron facilities, high-quality (ROI) XRDCT slices can now be acquired within minutes, making true operando studies feasible.

The trade-off between acquisition time and spatial resolution depends on the characteristic length scale of the heterogeneities under investigation. Microscopic phenomena, such as the influence of electrode delamination and particle cracking, require smaller beam sizes of less than 100 μm , comparable to the thickness of individual electrode layers. Nevertheless, (sub-)millimetre-scale scanning already yields sufficient resolution to assess the overall lithium distribution, allowing for a distinction of peculiarities in lithiation, for instance, in the vicinity of current tabs.

Acknowledgments

The authors gratefully acknowledge the financial support provided by the Heinz Maier-Leibnitz Zentrum (Technische Universität München) and German Federal Ministry of Education and Research (BMBF project 05K16VK2 and 05K19VK3). The authors acknowledge DESY (Hamburg, Germany), a member of the Helmholtz Association HGF, for the provision of experimental facilities. Parts of this research were carried out at PETRA III. Data was collected using beamlines P02.1, P07 and P21.2 provided by DESY Photon Science and P07 operated by Helmholtz-Zentrum Hereon. Beamtime was allocated for proposals I-20150544 and I-20160368 at P02.1, proposal I-20160368 at P07-Hereon, proposal I-20250477 at P07, and proposal I-20210735 at P021.2 of Deutsches Elektronen-Synchrotron DESY. The authors acknowledge the European Synchrotron Radiation Facility (ESRF) for the provision of synchrotron radiation facilities under proposal number MA-2298 using beamline ID15A. Neutron data have been acquired at STRESS-SPEC (Proposal p12667) of MLZ FRM II. The authors wish to thank the Facilities for the allocated beamtime.

Open Access funding enabled and organized by Projekt DEAL.

Funding

This study was supported by Bundesministerium für Bildung und Forschung (05K16VK2, 05K19VK3).

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

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

Endnote

¹ 0.5 is the minimum value due to the used evaluation.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section. The graphs of electrochemical cycling, the technical details of neutron- and synchrotron-based experiments, the data processing procedure, and the additional cited references [64–75] can be found in the Supporting Information.