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TOPICAL COLLECTION: HIGH-ENERGY BATTERY MATERIALS

X-Ray Computed Tomography (CT) Technology for Detecting Battery Defects and Revealing Failure Mechanisms

Yingjie Jiang[1] · Anqi Tian[1] · Li Yan[1] · Xueqi Du[2] · Lanmei Yang[2] · Li Li[1] · Jie Zhou[1] · Qi Wang[1] · Shuai Ruan[2,3] · Xinping He[2,3] · Yongqi Zhang[3,6] · Xiaoping Yu[4] · Yuanyuan Jiang[4] · Fangfang Tu[4] · Jiayuan Xiang[4] · Wangjun Wan[5] · Chen Wang[5] · Yang Xia[2] · Xinhui Xia[2,3] · Wenkui Zhang[2]

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Abstract

As the global lithium-ion batteries (LIBs) market continues to expand, the necessity for dependable and secure LIBs has reached an all-time high. However, the use of batteries is associated with a number of significant risks, including the potential for thermal runaway and explosions. The meticulous inspection of LIBs is not only essential for guaranteeing their quality and functionality, but also for ensuring their safety. This underscores the criticality of advanced inspection technologies. In contrast to traditional inspection technologies, industrial x-ray computed tomography (CT) scanning technology affords a non-destructive comprehensive, three-dimensional insight into the interior structure of a battery without the need for disassembly. It can make the inner LIBs structures visible through the housing and even batteries already installed in devices can be examined safely and accurately without being removed or opened. This capability is of critical importance for the identification of defects that could lead to battery failure or safety issues, and guide the optimization of LIBs with better safety and performance. This perspective review briefly summarize the comprehensive application of industrial CT in LIBs including battery materials, cells and modules. Finally, we further discuss the challenges and prospects of industrial CT for energy storage.

Keywords X-Ray computed tomography · lithium ion batteries · battery defects · failure analysis

Introduction

The urgent need to address energy saving and emission reduction on a global scale requires continuous exploration of potential solutions.[1,2] Lithium ion batteries (LIBs) are electrochemical energy storage devices that have been extensively employed in daily life.[3,4] They are widely acknowledged as pivotal devices facilitating the transition from finite fossil fuels to sustainable global energy production and utilization.[5] Currently, LIBs dominate the power source market

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✉ Shuai Ruan
1112101045@zjut.edu.cn

✉ Jiayuan Xiang
xiangjy@naradapower.com

✉ Wangjun Wan
wwj@zaiq.org.cn

✉ Xinhui Xia
cmxwy@zjut.edu.cn

[1] Information and Telecommunications Company, State Grid Shandong Electric Power Company, Jinan 250013, People's Republic of China

[2] College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, People's Republic of China

[3] Institute of Fundamental and Frontier Science, University of Electronic Science and Technology of China, Chengdu 611371, People's Republic of China

[4] Narada Power Source Co. Ltd, Hangzhou 310014, People's Republic of China

[5] Zhejiang Academy of Science and Technology for Inspection and Quarantine, Hangzhou 311215, People's Republic of China

[6] Chongqing Research Institute, Harbin Institute of Technology, Chongqing 401151, People's Republic of China

due to their high working voltage, large energy density, and good cycling stability, thereby exerting a profound influence on the overall quality of new energy equipment and energy storage systems.[6] Nevertheless, the safety concerns associated with internal defects and degradation mechanisms of LIBs have garnered increasing attention in recent years.[7] The efficiency of production, quality, and consistency of LIBs products are directly influenced by the detection technology and degree of automation employed in the manufacturing process.[8] Therefore, achieving precise and controllable detection of the internal structure of LIBs is crucial for quality control and overall performance optimization.[9] The selection of appropriate detection methods for visually identifying the internal structure and defects of batteries is crucial in establishing a comprehensive health monitoring, diagnostic, and early warning system for LIBs.[10] This will enhance the reliability of assessing the health and safety status of energy storage systems, ensuring the secure and stable operation of large-scale power stations with LIBs.[11]

Industrial computed tomography (CT) inspection is a non-contact technology that utilizes x-rays for the purpose of internal structure inspection.[2] By employing advanced scanning and reconstruction algorithms, it generates two-dimensional tomography images or three-dimensional images of objects, enabling clear, accurate, and intuitive visualization of the internal structure, composition, material, and defects in the inspected objects.[12] This technology is widely recognized as the most effective nondestructive testing and evaluation technology available today.[13] Currently, industrial CT testing is employed extensively in the LIB industry for non-destructive examination of internal structures and defects.[14] It serves as an indispensable tool for failure analysis and quality control purposes while providing an efficient and precise method for inspecting the internals during LIB manufacturing processes.[15] Furthermore, the application of industrial CT testing contributes to the enhancement of product design, improvement of production quality standards, and reduction of safety risks to a significant extent.[16] Consequently, it provides robust technical support in the development of new LIB products.[17]

This perspective review addresses a number of issues related to the deterioration mechanism of LIBs and provides a comprehensive analysis of the impact of CT non-destructive testing technology on the internal materials and structures of LIBs.[4,18] This review explores process optimization, quality control, and safety improvement from various levels, including battery materials, cells, packs, and modules.[19] It also offers an efficacious solution and theoretical foundation for non-destructive testing, failure analysis, and quality control of the internal structure and defects in LIBs.[20,21]

Results and Discussion

Principle of Industrial x-Ray Computed Tomography (CT)

The fundamental principle underlying industrial CT imaging involves acquiring internal information about an object by leveraging x-ray or gamma ray attenuation laws during their penetration, in which two-dimensional images, or radiographs, are created by x-rays in a cone-beam that passes through an object and projects radiographs onto a flat-panel detector (Fig. 1). During detection, a radiation source emits a focused beam that systematically scans distinct sections/levels of the target object, while an accompanying detector captures transmitted radiation.[22] Subsequently, these captured rays undergo conversion to visible light and subsequent transformation into electrical signals which are further digitized via an analog-to-digital converter prior to computer input.[23] Employing dedicated reconstruction algorithms, this computational system processes acquired data to generate comprehensive density distributions represented as two-dimensional or three-dimensional images.[24]

Flat panel CT detection is based on the principle of projection amplification, resulting in a decrease in sample resolution as its size increases.[25] To enhance image resolution, two common approaches are reducing x-ray focus and/or employing a higher resolution flat-panel detector.[26] However, these methods do not overcome the limitations

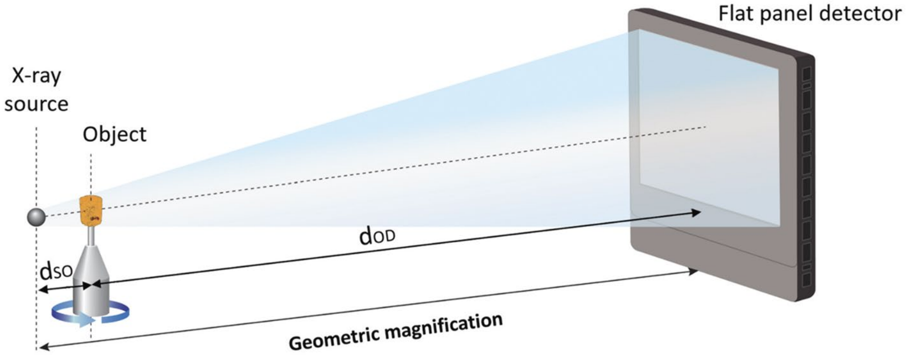


Fig. 1 Schematic of x-ray CT and x-ray microscopy (XRM; Xradia Versa; ZEISS) systems: a cone-beam x-ray CT setup with a flat-panel detector.

of geometric magnification for larger samples (> 25 mm diameter). An alternative option involves incorporating optical lenses after x-ray inspection to create an optically amplified image through scintillator lens–charge-coupled device (CCD) detector coupling (Fig. 2).[1] This configuration indirectly converts x-ray photons into charged signals by allowing the x-rays to interact with the scintillator, which then converts them into visible light.[27] The visible light is subsequently passed through an optical lens and projected onto a CCD for enlargement.[28] By implementing this strategy, the effective pixel size of the detector can be reduced by 150 nm, enabling XRM images with a spatial resolution of less than 700 nm.[29]

The technical advantages of industrial CT include: (1) high resolution, which can provide very high image resolution, so that the small internal structure can be clearly displayed; (2) non-destructive testing, which, due to the use of x-rays or other forms of radiation, will not cause any damage to the object being tested, which is particularly important for the detection of items with high value or that cannot withstand damage; and (3) CT is widely used in various industries, such as aerospace, automotive, electronics, material science, and other fields, for detecting internal defects of components, dimensional measurement, assembly verification, etc.[30]

With the development of the technology, industrial CT systems are being continuously upgraded to provide faster scan speeds and higher image quality. In addition, there are many different types of industrial CT, including x-ray tomography (XCT), Compton scattering tomography (CST), and Mossbauer effect tomography, which are mainly used for real-time detection of industrial in-line processes and exploration of large industrial components.[31] Compared with traditional x-ray inspection and ultrasonic inspection, industrial CT has the characteristics of high spatial resolution and non-destructive testing, which can quickly, accurately, and intuitively find the internal defects of products, such as castings, plastic parts, and ball grid arrays.[32] In summary, industrial CT, as an efficient means of non-destructive testing, plays an important role in modern manufacturing and other industries, which not only improves the accuracy of product quality control but also helps to improve production efficiency and to reduce costs.[33]

Applications of Industrial CT Detection in LIBs Industry

CT is a stereoscopic imaging technology that enables three-dimensional detection of the internal structure of batteries without any blind spots, allowing for comprehensive assessment of various components such as pole plates, pole ears, coated electrode materials, and battery shells. Furthermore, it facilitates evaluation of overall battery structure including packaging and welding outcomes. By examining internal structures and measuring cell dimensions, this technology optimizes production processes and enhances product quality. For instance, it can identify internal deformations and metal impurities in lithium batteries manufactured through winding or laminating processes. Additionally, industrial CT assists manufacturers in identifying and resolving potential safety risks associated with batteries by detecting issues like internal defects, manufacturing process problems, or abnormal temperatures to improve safety and reliability. In cases where batteries exhibit failure symptoms such as increased internal resistance or capacity attenuation due to poor electrode contact,, or uneven electrolyte distribution caused by improper assembly or gas generation during operation, can be analyzed using industrial CT without disassembling the battery. Overall, industrial CT detection technology plays a crucial role in the LIB manufacturing sector. The subsequent section will present the application of CT detection technology in LIBs, encompassing material analysis, cell characterization, and module evaluation.

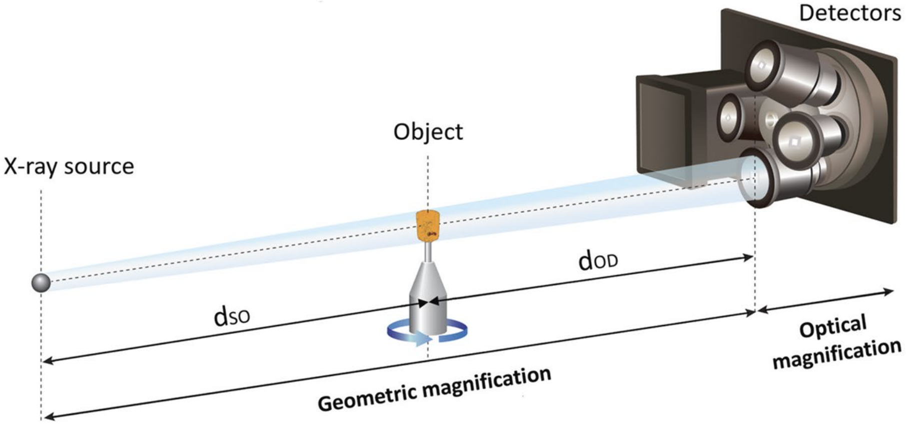


Fig. 2 Schematic of a two-stage magnification architecture in a 3D XRM featuring adjustable objectives with varying optical magnifications. The integration of geometric and optical magnification enhances the resolution capabilities of XRMs, surpassing those of flat-panel x-ray CT systems.

Battery Material Analysis

CT technology enables a comprehensive 3D visualization of battery components, facilitating the examination of electrode pore structure, particle morphology and surface degradation, dendrite formation, intra-granular cracking, and short-circuit dendrite morphology in solid electrolytes, as well as other alterations in material structure that may contribute to accelerated capacity decline and impact the operational reliability of batteries.[21–24] Paul et al. detected and quantified defective particles by using low-resolution imaging, which has greatly improved material statistics (Fig. 3).[34] Daniele et al. proposed a study on nano-CuO anodes of LIBs using a combination of electrochemical measurements and off-site x-ray CT at the nano-scale level (Fig. 4).[7] The electrodes undergo transformation reactions with Li^+/Li at voltages around 1.2 and 2.4 V during discharge and charging, respectively. The 3D nano-CT imaging reveals significant recombination of CuO particles and precipitation of Li^+ conductive films suitable for battery applications.[7] The CT detection technique enables the identification of material type and composition, observation of the impact of charge and discharge variations on materials, and exploration into the growth and morphological evolution of lithium dendrites in lithium metal batteries. Miao et al. propose a novel laminated pressure in situ x-ray CT imaging device for the detection of microstructure evolution at the interface during electrochemical processes (Fig. 5).[35]

The relationship between the electrochemical properties of lithium metal batteries and the interfacial morphology is effectively investigated using this in situ technique. It can be observed that the growth and morphological evolution

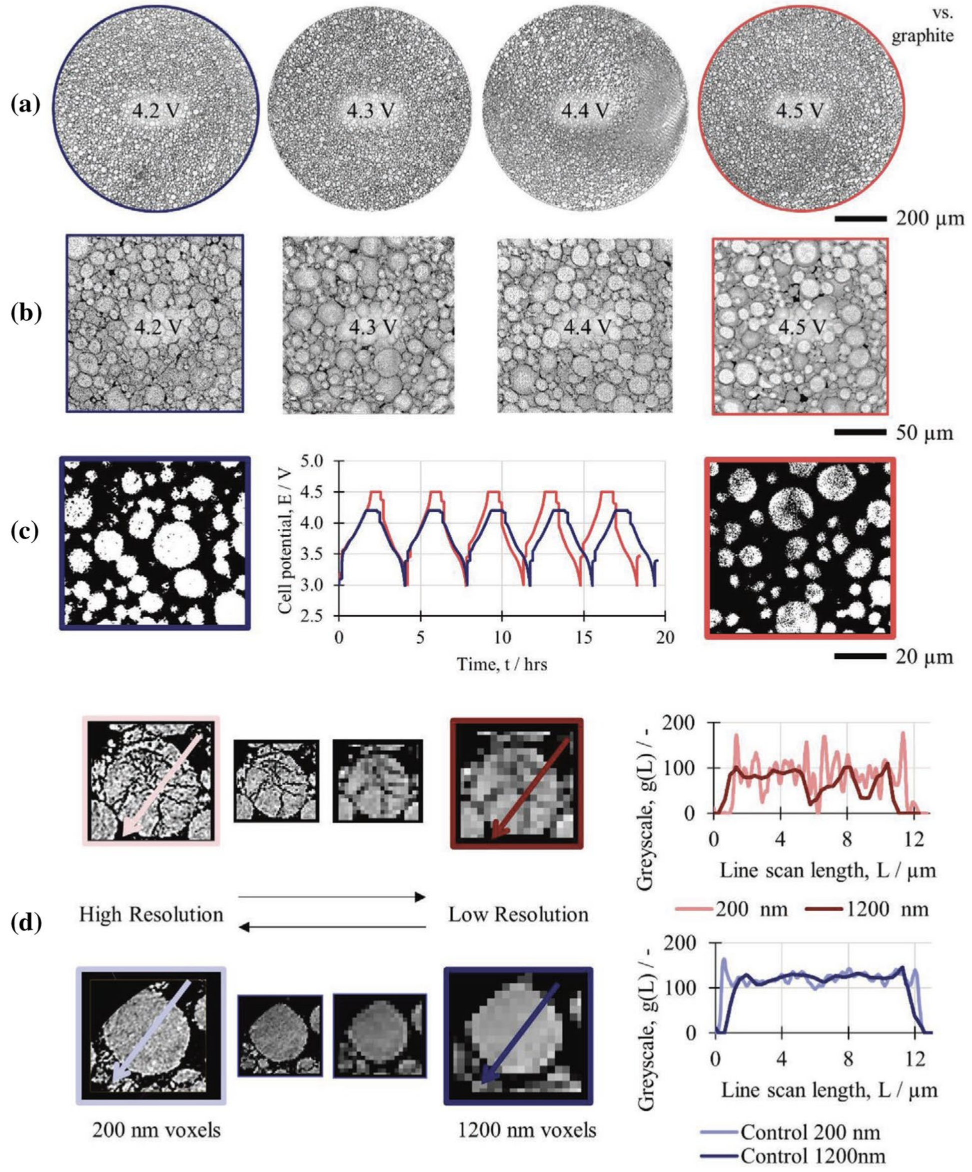
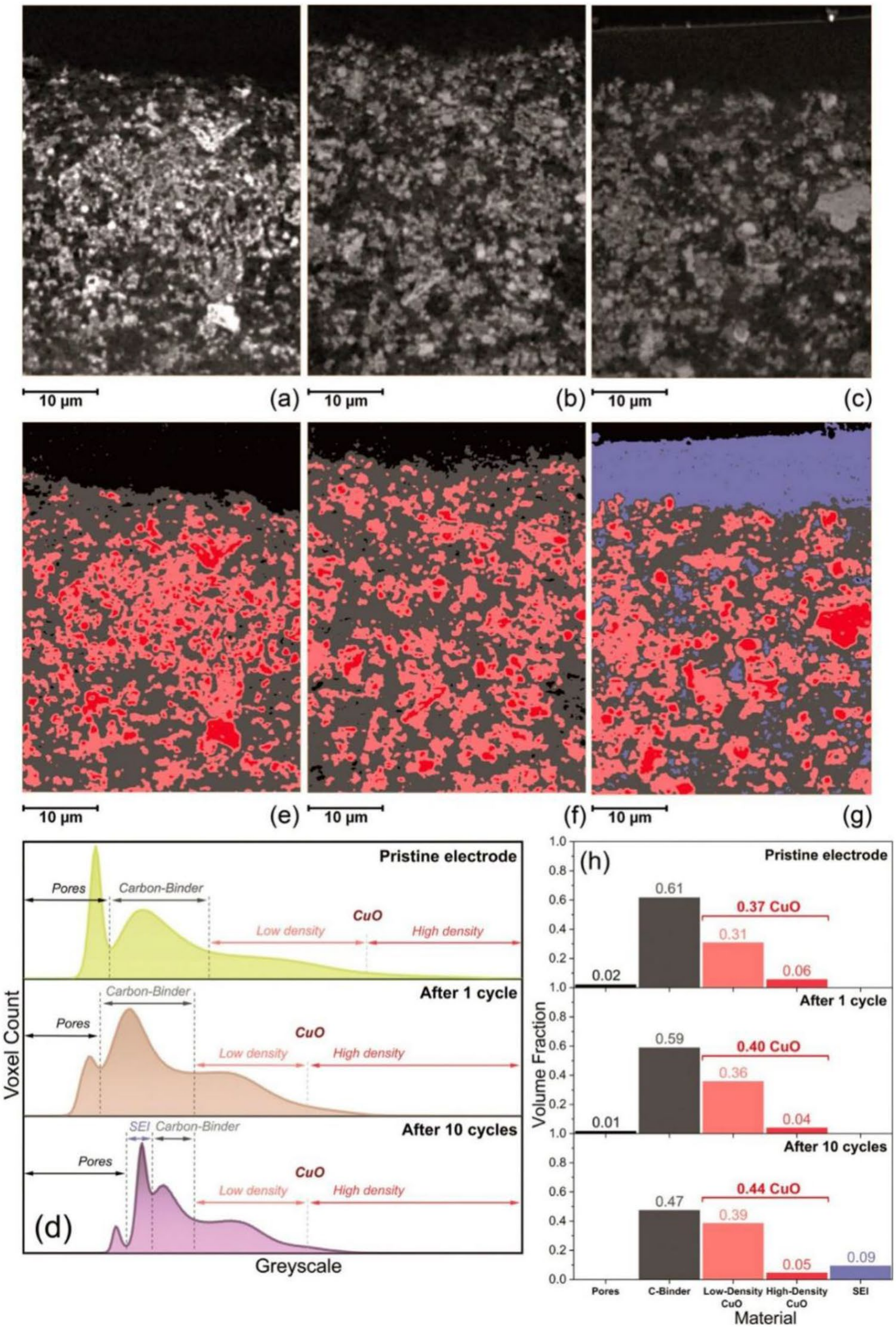


Fig. 3 Using x-ray-CT to assess electrode- and particle-level degradation in NMC811 cathodes taken to various upper cut-off voltages: (a) 3D grayscale volume rendered at the electrode level (1.57 μm isotropic voxel length); (b) 3D grayscale volume renders at the particle level (206 nm isotropic voxel length); and (c) 2D grayscale ortho-slices taken from 4.2- and 4.5-V structures with the same grayscale threshold applied to display grayscale variations; accompanied by the electrochemical cell potential plotted with respect to time; (d) two examples of a defective (red) and pristine (blue) particle and their intra-particle grayscale variation with respect to imaging resolution (Color figure online).

Fig. 4 Ex situ x-ray nano-CT study of the CuO electrode: (a–c) cross-sectional slices extracted in a plane orthogonal to the current collector for (a) pristine CuO, (b) CuO after 1 cycle, and (c) CuO after 10 cycles. (d) Grayscale histogram for the datasets. (e–g) Four-phase segmentation of the slices in panels (a–c) for (e) pristine CuO, (f) CuO after 1 cycle, and (g) CuO after 10 cycles (black pores; *gray* carbon-binder; light red low density CuO; dark red high-density CuO); voxel size 63 × 63 × 63 nm^3; dataset size 600 × 600 × 800 $voxel^3$; (h) phase volume fractions of cropped datasets (600 × 600 × 600 $voxel^3$) (Color figure online).



of lithium dendrites are directly influenced by reactor pressure. Within the critical pressure range, Li dendrites transition from acicular to island-like structures, growing from tip to base. Furthermore, appropriate pile pressure can effectively inhibit the growth of lithium dendrites. This innovative in situ CT imaging technique provides a feasible method for elucidating interface microstructure evolution, electrochemical-pressure complexation, and safety aspects pertaining to next-generation rechargeable batteries.[35]

Cell Analysis

During the charge–discharge cycle of LIBs, electrode deformation can result in localized high strain and significant capacity degradation. In numerous applications,

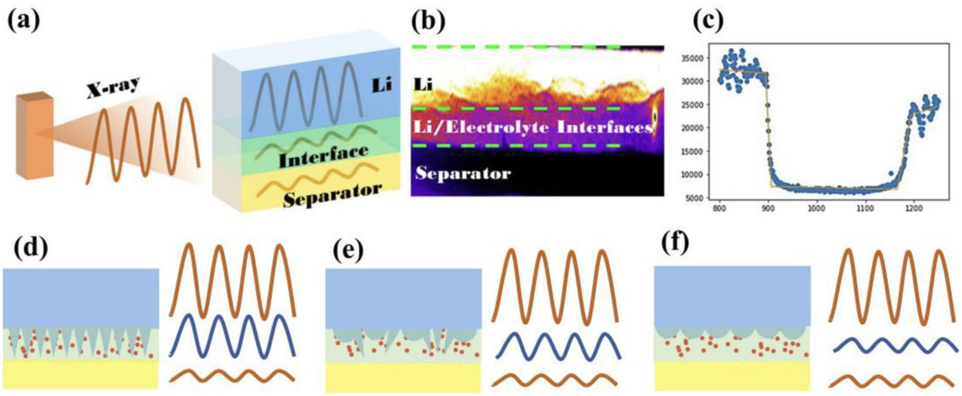


Fig. 5 (a) Schematic of x-ray transmitting through different elementals, reflected by the magnitude of the amplitude; (b) tomography image collected from different materials in Li-metal batteries; (c) the corresponding contrast absorption values in (b); (d–f) x-ray transmitted abilities corresponding to the different kinds of Li dendritic morphologies in the in situ x-ray tomography experiments: (d) needle-like, (e) mixture of needle-like and island-like, (f) island-like.

mitigating risks necessitates comprehending the influence of charge–discharge rates on the electrode structure throughout the battery's lifespan. Cai et al. presented the pioneering application of x-ray CT technique for material parameter analysis in rectangular lithium iron phosphate batteries. The principles of electrode and material identification by CT technology are elucidated, while a detailed summary is provided on the method of tomographic images analysis based on sinusoidal function. Finally, the paper discusses the application of extracting material content and distribution from tomographic images (Fig. 6).[36]

Wu et al. were dedicated to mitigating the impact of CT imaging time on the charge and discharge processes by implementing a sparse view scanning strategy on

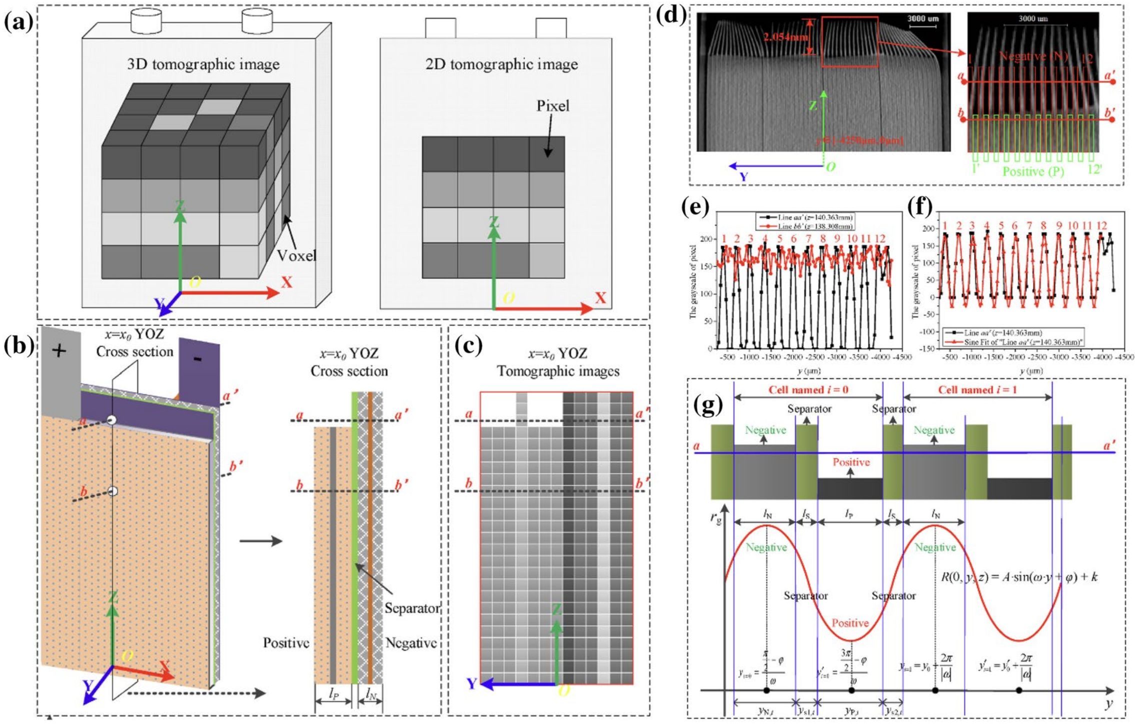


Fig. 6 (a) Schematic of 3D dataset; (b) schematic of the cross-section of the cell; (c) schematic of the YOZ tomographic image of the cell; (d) the projection interval of positive and negative tomographic images on the *G* (*0, y, z*); (e) the grayscales versus the *y* coordinates of the pixels on the line *aa'* and line *bb'*; (f) the fitted sine curve *R* (*x*, *y*, *z*) on the line *aa';* (g) schematic of the projected position of two cells in the YOZ plane on the fitted sine curve.

traditional CT for rapid imaging. Additionally, they propose an SVRNet model to obtain satisfactory images from sparse projections (Fig. 7a),[37] which outperforms other algorithms in terms of reconstruction results. Furthermore, their findings indicate that, during cycling, the deformation of positive and negative electrodes gradually increases while the displacement and strain distribution within the battery's electrodes become uneven (Fig. 7b and c).

Electrodes located near the center exhibit more bending than those closer to the edge. The circulation rate and number of cycles significantly affect internal structure displacement and strain in LIBs with high circulation rates having a greater effect on deformation; therefore, it is best to avoid high current charging or discharging to slow capacity decline and reduce deformation.[37]

The battery manufacturing process is a critical determinant of LIB performance. In the assembly process, issues such as misalignment and pole plate deflection are inevitable during winding and sealing procedures. Enhancing the quality assurance of LIB production holds paramount importance for the further advancement of the electric vehicle market and its diverse applications. Dayani et al. employed laboratory and synchrotron x-ray computed tomography (XCT and SXCT, respectively) to conduct microscopic in situ investigations on commercially available LIBs. The aim of their study was to offer comprehensive guidance for selecting the most suitable imaging devices and parameters to address scientific inquiries related to LIBs, including quality management during production processes, safety state diagnosis, and evaluation of secondary life possibilities.[38] Figure 8 illustrates a comparison between two batteries for SXCT

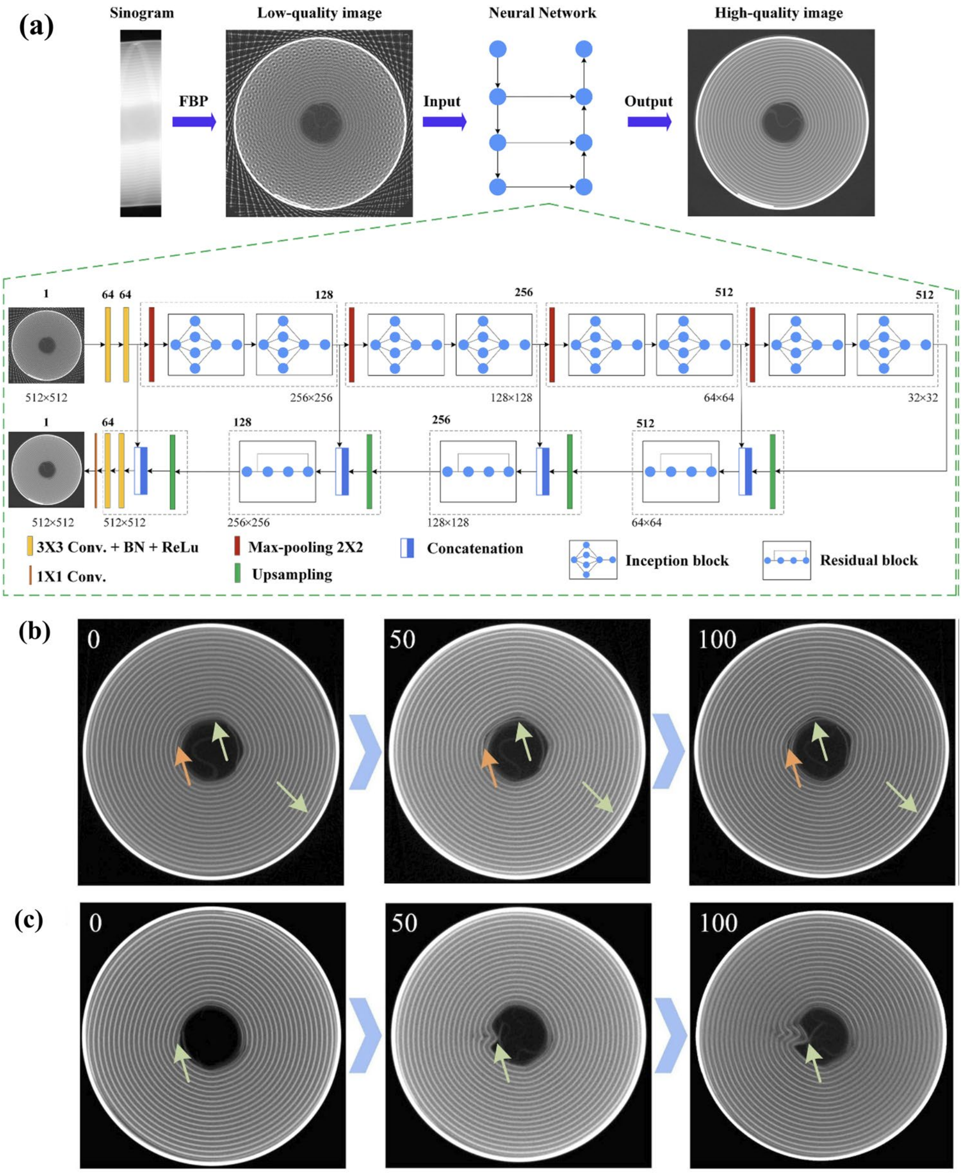


Fig. 7 (a) Architectures of the sparse-view reconstruction network (SVRNet) model. Radial cross-sectional view of fresh battery (cycled 0th) and after 50 and 100 cycles at (b) 1C and (c) 2C. With the increase of cycles, the electrodes gradually bend and the gaps gradually increase as indicated by the arrows. The electrode deformation is more serious at the 2C rate.

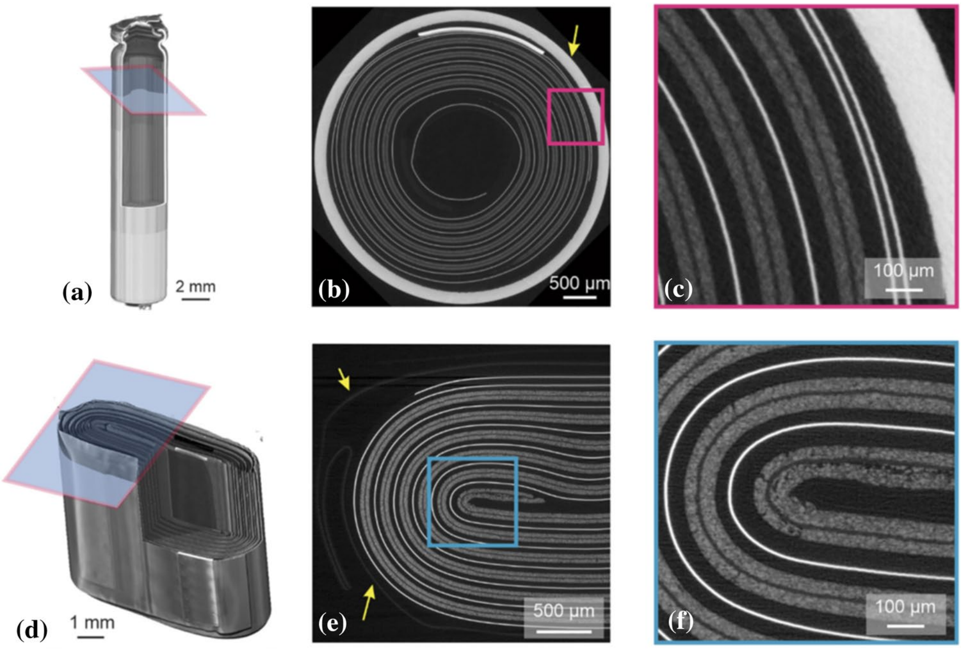


Fig. 8 XCT of a small cylindrical lithium-ion battery: (a) 3D rendered view of the 60-mAh cylindrical cell (Lab XCT data); (b) slice of SXCT reconstruction of the same cell; the *boxed section* is enlarged in (c); (d) 3D rendered view of the 30-mAh small pouch cell (XCT data); (e) SXCT reconstruction of the pouch cell; (f) enlargement of the marked section in (e). (b and e) measured at the BAMline: beam energy = 42 kV, 3200 projections, 0.5 s exposure time, 3.6 μm effective pixel size.

measurements, employing identical scanning parameters. A cylindrical battery featuring a robust metal shell exhibits reduced resolution in the cathode active material compared to a soft-pack battery with a thin shell composed of pliable material (refer to Fig. 8e). The diminished level of detail can be attributed to the high x-ray absorption caused by the thick metal casing, which significantly attenuates x-ray photons. Consequently, this highly attenuated x-ray beam displays lower sensitivity towards weakly attenuated cathode particles. While higher energy levels mitigate casing attenuation and also affect other materials, this decreased attenuation proves detrimental due to the resulting lower contrast. Thus, elevating the x-ray energy does not entirely resolve the challenge of simultaneous imaging of hard and soft materials. In summary, for high-resolution XCT analysis of commercial LIBs, utilizing soft-shell materials in small-scale battery production is more suitable.

Luciana et al. compared two nondestructive testing methods: scanning acoustic microscopy (SAM) and x-ray computed tomography (CT), for three-dimensional visualization of defects at various depths within soft-pack batteries. A soft-pack battery with eight electrodes was utilized in the study (Fig. 9).[22] Utilizing a 15-MHz transducer in reflection mode, SAM is capable of detecting defects up to a depth of 4 electrodes within a span of 2 minutes, exhibiting a lateral resolution of 150 μm. On the other hand, CT could identify defects across all eight stacked electrodes in the soft-pack battery; however, it required approximately 12.5 hours for measurement. Consequently, these two methods can complement each other in detecting post-production defects within thin soft-pack batteries as a final test on the production line or for repurposing single batteries. Both approaches possess potential to expand the range of non-destructive quality assurance testing in lithium-ion battery production, thereby enhancing safety and productivity within battery technology.

Module Evaluation

Macroscopic imaging of battery modules is crucial for capturing design parameters and evaluating performance and safety issues arising from mechanical and electrochemical instability during the charge–discharge cycle. Furthermore, a single faulty electronic component can lead to the failure of the entire battery module. Once in use, after numerous charge cycles, electrode deformation may occur (resulting in increased gap between the electrode and the active material), leading to delamination of the electrode material from the collector. This delamination hinders electron conduction, causing a drop in voltage across the electrode. XCT can be employed as a non-destructive method to examine these potential problems or defects, assess battery component integrity, and prevent deterioration and safety hazards associated with rechargeable LIBs. Figure 10 shows an example where various components of battery cells and modules are examined using images obtained from a tablet-based CT system (Metrotom 1500; Zeiss), highlighting common macroscopic features and deficiencies relevant to quality assessment. Simultaneously, Fig. 11 illustrates the CT plan and three-dimensional (3D) modeling diagram of the lithium battery module from various perspectives. It is evident that, in certain instances, the integration of non-destructive

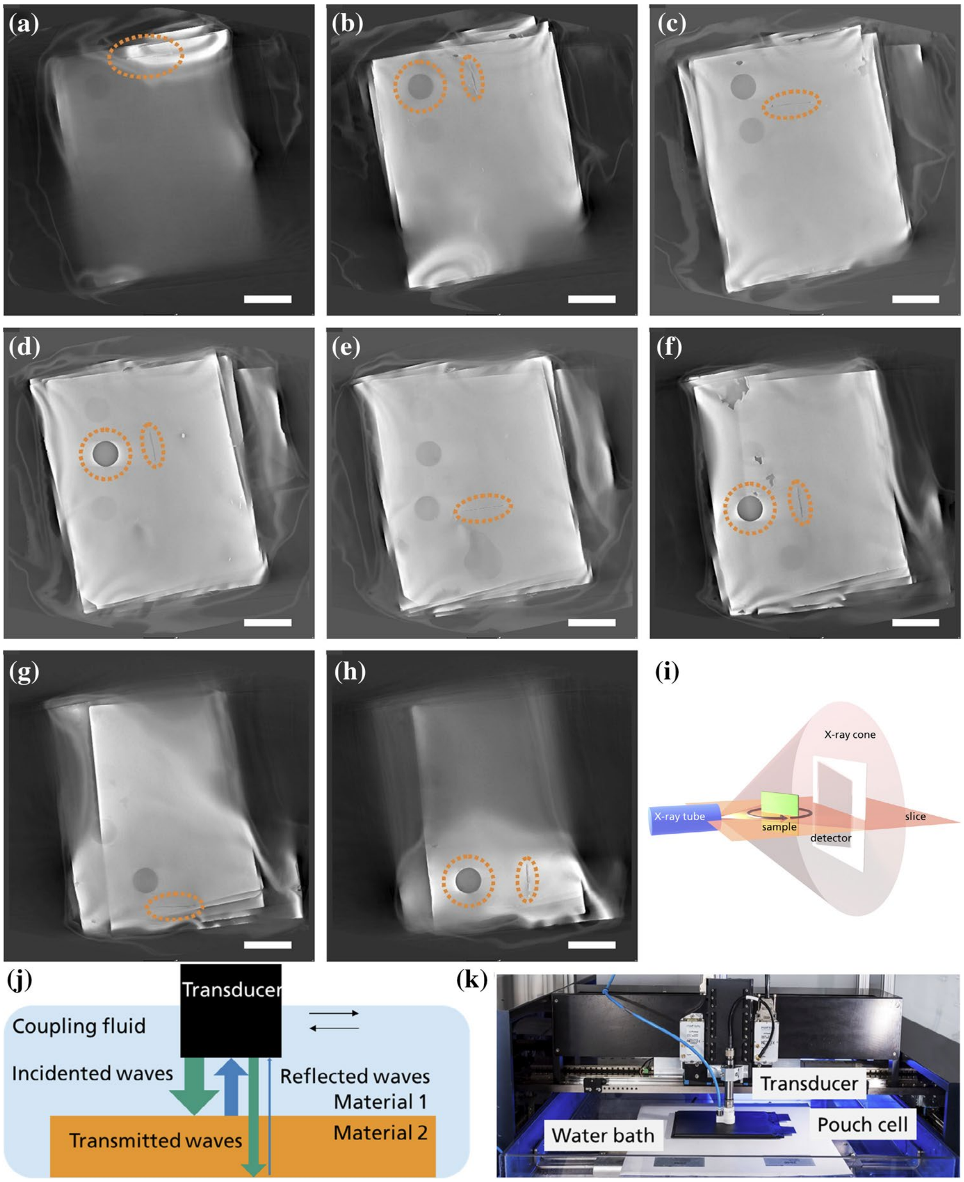


Fig. 9 CT images of the dummy pouch cell showing damaged cathode sheets with resolution of 22 μm. The image planes of (a–h) are parallel and have been collectively aligned to show large areas of the cathode sheets. (I) CT measurement setup in 3D showing the rotating sample in one rotation angle and a selected slice for construction. Schematic diagram of SAM in (j) reflection mode showing the partial reflection at the interface of materials 1 and 2. (k) photo of the equipment used in this study.

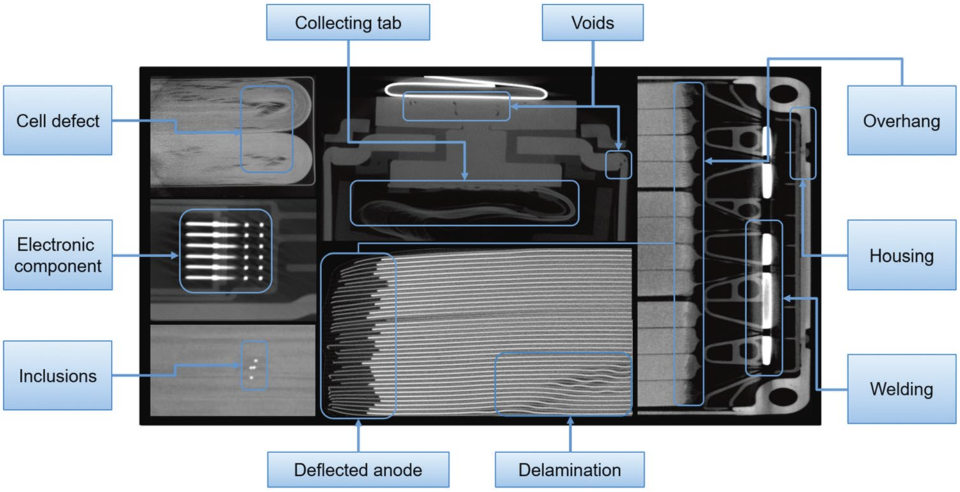


Fig. 10 XCT inspection of cell and module components highlighting macro-scale features relevant to battery quality inspection (images zoomed-in at different scales).

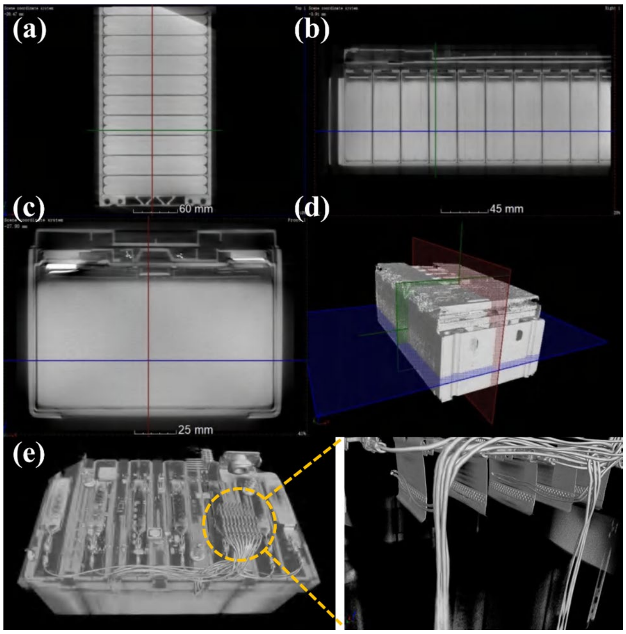


Fig. 11 Battery module test of LIBs: (a) *X* direction, (b) *Y* direction, (c) *Z* direction, (d) 3D diagram, (e) module internal wiring harness street,

testing techniques such as CT can yield sufficient detail to elucidate the multi-scale behavior of the battery module.

Conclusions

With the continuous advancement of new energy detection technologies and the imperative to ensure the safety of energy storage systems while minimizing costs, it becomes essential to integrate emerging battery characterization technologies into the relevant measurement/inspection workflow. The utilization of industrial CT presents numerous advantages compare to conventional measurement technologies, encompassing the capacity to conduct component measurements of intricate internal structures within cells by means of high-density information in a non-contact and non-destructive manner, offering morphological insights into electrode materials and structures at various scales, ranging from macroscopic characteristics of battery packs (hundreds of millimeters) to microscopic features at the particle level (tens of nanometers). Different problems, including electrode misalignment, anode overhang, separator integrity, tab weld quality, foreign material detection, electrolyte distribution, internal short circuits, cell and module assembly, density variations, porosity in electrodes, structural integrity of casing, and thermal runaway precursors, can be detected by CT. This paper comprehensively reviews the CT detection technology to ensure the overall structure of the battery on the basis of its internal materials, cells, battery modules and other aspects of comprehensive detection to accomplish the objective of quality control and risk reduction in LIBs. Moreover, the non-destructive imaging capability of CT facilitates the analysis of energy efficiency decline, battery aging effects, and the identification of causes behind battery degradation and failure after multiple charge and discharge cycles. This aids in optimizing battery design, assessing quality, and holds promising prospects for application in battery safety detection and fault analysis.

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Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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