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Analysis Of Battery Cells Via Computer Tomography: Possibilities And Technical Limits Of Detectability Regarding Their Function And Safety Critical Failures

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Abstract

The development of electric vehicles (battery electric vehicles [BEV] and light electric vehicles [LEV]) is characterised by exponentially increasing requirements in terms of the range of functions, functionality, reliability and safety. This leads to a sharp increase in component functionality and component interactions within the product. Therefore, ensuring function and safety-critical characteristics is of enormous importance. This is particularly evident with regard to the battery system respectively to the battery cells. Both production-related deviations/errors and changes/errors during the period of use can cause critical cell conditions, which can lead to thermal runaway of the cells. Computed tomography (CT) is particularly important for analysing internal defects in the micrometer range. CT is ideal for detecting function- and safety-critical defects in lithium-ion cells. This applies both to the final test in the cell manufacturing process (before 1st use) and to the testing of cells already in use regarding the 2nd use. This paper shows the results of a research feasibility study on the analysis of function- and safety-critical faults in new and used lithium-ion cells with the aid of computer tomography (CT). The here presented analytics were carried out using a diondo d2 micro-CT system. Cells of the type 18650 Li-Ion battery from Panasonic (NCR-18650B) were used as reference test objects. Finally, the results are compared with the findings of other authors.

Keywords: battery cells, computer tomography, functional- and safety related failures, degradation, long-term reliability

1. Introduction

This paper focuses on the analysis of function- and safety-critical faults in lithium-ion cells with the aid of computer tomography (CT). The aim is to analyze possibilities and technical limits of detectability of function- and safety-critical cell faults in the micrometer range, which can reduce the runtime reliability or cause a risk. The results of a feasibility study are presented, in which representative cell types were analysed in several test series via CT. The spectrum of defects to be analysed includes both production-related deviations/defects and usage-related changes/defects. Section 1.1 explains the basic principles for the use of battery cells in Battery Electric Vehicles (BEV) / Light Electric Vehicles (LEV). Section 1.2 then outlines the possibilities for cell testing with respect to reliability and safety critical failures. Section 1.3 provides an overview of the basics and possible applications of computer tomography (CT) for analysing cells. Chapter 2 presents the research objectives in detail. Chapter 3 focuses on the implementation of the feasibility study for analysing representative cells and function- and safety-critical fault spectra. Chapter 4 shows the main results of the feasibility study and concludes with a comparison of the findings with other authors.

1.1. Battery electric vehicles, cell use

The development of electric vehicles (battery electric vehicles [BEV] and light electric vehicles [LEV]) is characterized by exponentially increasing requirements in terms of the range of functions, functionality, reliability, and safety. This leads to a sharp increase in component functionality and component interactions within the product. Therefore, ensuring function and safety-critical characteristics is of enormous importance. In the case of electric vehicles, this is particularly evident in the battery system. In BEVs, a battery system basically consists of cells that are assembled within sub-modules, several of which make up the entire battery module. In contrast, LEVs usually dispense with the sub-module structure and the interconnected cells directly represent the battery module.

The battery module is controlled by the battery management system (BMS): The BMS is used, among other things, to ensure long-term reliability, protect against deep discharge and detect malfunctions.

The flawless functioning of this complex system consisting of battery module and BMS requires highly reliable individual components. This applies in particular to the cells, of which there are several dozen (LEV: e-scooter or cargo bike) or thousands (BEV: e.g. TESLA Roadster with 6831 cells (Mojumder et al., 2022)), depending on the product type. Cells must therefore be tested for functional and safety-critical faults both during new production for 1st use and during potential reuse for 2nd use.

1.2. Cell testing with regard to reliability and safety critical failure spectrum

There are various procedures that are currently used for cell testing regarding the current battery status, the main ones being:

- Coulomb Counting (CC);
- Electrochemical Impedance Spectroscopy (EIS) test;
- Hybrid pulse power characterization (HPPC) test.

In these direct measurement procedures, physical measured variables or status indicators are recorded directly in order to draw conclusions about the current battery status. The efficiency of these methods varies in terms of the required effort and accuracy. However, these methods focus on cell function and cell health, none of these methods allow the inside of the cell to be analysed regarding possible internal functional or safety-critical faults that can cause unexpected problems in further use. Production-related faults include, among others:

- Anode/cathode geometry;
- Faulty Package: Positioning and spacing leads to short circuit;
- Interconnection points.

Figure 1 provides a schematic overview of typical examples of battery cell failures that may be related to the different phases of a cell's failure behaviour during its use phase. While failures due to degradation often only occur towards the end of the battery cell's life, defects introduced during production can cause premature failures by providing the basis for rapidly progressing degradation. The following faults, among others, play a decisive role in the utilisation phase:

- Solid electrolyte interphase (SEI) growth on the anode surface;
- SEI degradation;
- Deposits on the separator (dendrite growth);
- Gas development;
- Lithium plating.

Among other things, the faults mentioned can lead to short circuits (e.g. when a dendrite punctures the separator between the electrodes) and thus to thermal runaway and the death of the battery. In the event of a thermal runaway, there are often only limited options for bringing the resulting vehicle fire under control. The result is a controlled burning of the entire product - without partial fire extinguishing for the purpose of product rescue.

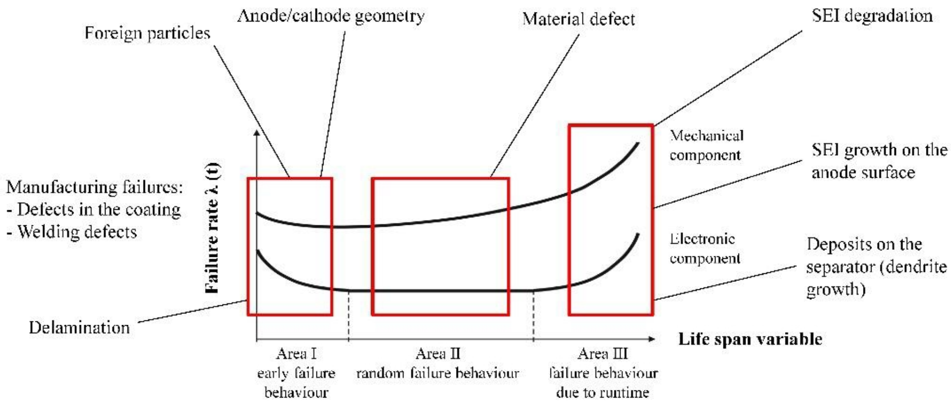

Fig. 1. Typical examples of battery cell failures possibly related to the different phases of cell failure behaviour in the use phase, based on (Bracke, 2024).

1.3. Computed tomography (CT)

Computed tomography (CT) is of particular importance when analysing faults. CT has been used for decades in the aerospace industry, automotive engineering, apparatus/plant engineering and electrical engineering to detect and analyze especially functional- and safety-critical faults; cf. (Pfeifer, 2001) and (Schmitt and Dietrich, 2023). Almost complete geometry measurements (external/internal contours) can be carried out mostly independently of the component structure (with some limitations regarding the material). In addition, internal defects can be detected down to one micrometre for small samples and up to ten micrometres for bigger and more complex test objects (e.g. battery cells). The measurement options and defect analysis depend largely on the CT set-up and the test procedure. If a CT with flat panel detector is used, the CT analysis is carried out as follows:

1) The test object is positioned and fixed on the manipulator of the industrial CT system;
2) approx. 2,000 - 3,000 equidistant X-ray images (so-called „projections“) are taken with the flat-panel detector over an angle of rotation of 360°;
3) reconstructed the volume of the battery cell on the basis of the previously recorded projections.

It should be noted that the positioning of the test object on the manipulator (movable turntable), scattered radiation, beam source/detector have a significant influence on the precision and quality of the component analysis; see also (Schmitt and Dietrich, 2023) and (Marxer et al., 2021).

CT is ideal for detecting function- and safety-critical defects (cf. safety-critical failure spectra in Section 1.2) in lithium-ion cells. This applies both to the final inspection in the cell manufacturing process (before 1st use) and to the inspection of cells already in use regarding 2nd use.

2. Goals of research study

The overall aim of the research feasibility study is to determine the possibilities and limitations of using computer tomography to detect function- and safety-critical faults in lithium-ion batteries. The following sub-goals are being pursued:

1) Development of a CT measurements procedure for the detection of function/safety-critical cell faults.
2) Detection of production-related deviations/defects before 1st use and changes/defects in used cells regarding 2nd use.
3) Analysis of the representative cell types: New manufactured cell (before 1st use) versus used cell (after 1st use).

The here presented research feasibility study was carried out using a diondo d2 micro-CT system, which will be further described in Section 3.2. A sample of four cells of the type 18650 Li-Ion battery from Panasonic (NCR-18650B) were used as reference test objects. These battery cells were aged to a capacitive state of health (SOH_c) of just under 30 %. New battery cells were also measured with the CT system to compare changes/defects in the cells.

3. Case Study: Computer tomography analysis of new and aged battery cells

This chapter presents the case study "CT analysis of battery cells" carried out in this paper. For this purpose, the fundamentals of the battery cell are first presented with a focus on the cylindrical cell investigated here; furthermore the expected error spectra from production and degradation due to the use of the battery cell are presented (cf. Section 3.1). Finally, the CT system and the associated measurement process of the battery cells are analysed, cf. Section 3.2.

3.1. Fundamentals on battery cells

There are several battery cell designs that play an important role in the context of electromobility, including cylindrical cells, prismatic cells and pouch cells. Each design has its advantages and disadvantages depending on the specific requirements of the electric vehicle, with some manufacturers opting for cylindrical cells and others for prismatic cells. Cylindrical cells offer good heat dissipation due to their round diameter and are space saving. Prismatic cells, with their rectangular shape, can be easily arranged into compact modules. Pouch cells, on the other hand, are flat and flexible pocket cells without a hard casing, making them flexible in use (e.g. additionally in smartphones and notebooks). This case study initially focusses on the cylindrical cells; the findings of this work can subsequently be transferred to other designs, as the basic layer structure of the lithium-ion cell is identical over all designs.

3.1.1. Cylindrical battery

In the cylindrical cell, the individual layers consisting of anode with current collector, cathode with current collector and separator are wound around a battery core, which is designed to maintain structural integrity (the so-called jelly roll) and is protected from environmental influences by an outer metal shell. Figure 2 shows the schematic layered structure of the cylindrical cell, with two anodes or cathodes adjacent to each current collector. Cylindrical battery cells are for example currently used in the TESLA Roadster (see above), in the TESLA Model S with 7104 cells (Sharma et al., 2019), in the Rimac Nevera with 6960 cells (Mojumder et al., 2022) or in many LEVs such as e-scooters, and therefore have a wide range of applications. Depending on the application, the cells are screwed, welded and glued together to form sub-modules or even the complete battery modules, allowing different degrees of disassembly. Due to their round shape, gaps are created between the cells during assembly, which are ideal for cooling the system.

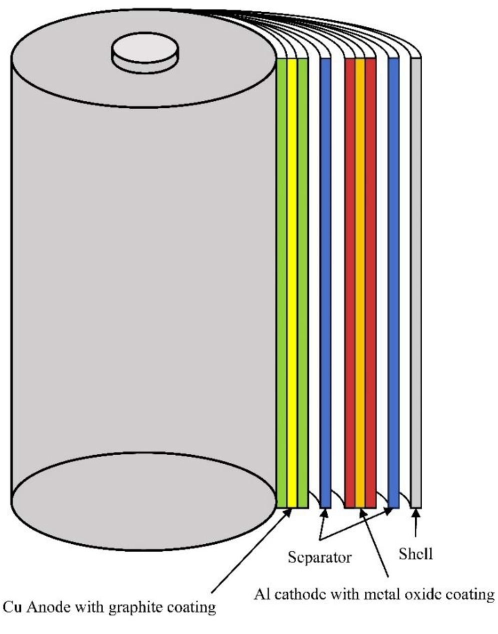

Fig. 2. Schematic structure of a cylindrical cell with the different layers (cf. Köllner, 2023).

3.1.2. Types of defects

The CT examinations are intended to detect as many different types of defects in the battery cells as possible safely and non-destructively. CT is therefore suitable for analysing new battery cells coming out of production as well as cells that have already been aged in order to assess both the current condition of the battery cell and the risk of function- and safety-critical changes. The errors in the production include, for example, errors in the placement position or deviations in the number and sequence of cathodes (faulty package: Positioning and spacing leads to short circuit), anodes and separators in the stack. Furthermore foreign materials and contaminants entering the cell during the production process, poor electrode connection or faulty welding of the contact tab, and generally insufficient quality in assembly. In the following, the underlying degradation mechanisms and different types of defects and their detection via µ-CT examinations and are presented below.

The cause of battery ageing and degradation lies in the chemical processes within the battery cell during the usage. Various processes lead to a loss of active material or active lithium over time. The development of the solid electrolyte interface (SEI), which forms as a passive layer between the anode and electrolyte, plays a decisive role in this process. This layer is formed during the initial cycles of the battery (also known as formation cycling) and inhibits the decomposition of the electrolyte and possible corrosion of the anode. The formation of the SEI leads to a loss of active material and thus to a decrease in capacity (Vetter et al., 2005; Agubra and Fergus, 2013; Barré et al., 2013). In addition, during intercalation or deintercalation, i.e. the storage and removal of lithium ions in the respective electrodes during cyclisation, microcracks can occur in the active materials, which can interrupt the connection between the active material and the current collector (Vetter et al., 2005; Wang et al., 2005; Ohzuku et al., 1993; Fischhaber et al., 2016). Another relevant ageing mechanism is the deposition of metallic lithium, also known as lithium plating, which causes an accelerated decrease in capacitance and explains the non-linear strongly decreasing ageing behaviour at the end of the battery life (Broussely et al., 2005; Ecker et al., 2014; Fischhaber et al., 2016). The progression of lithium plating and growth of the SEI can lead to penetration of the electrodes and the separator pores, which in the best case can lead to a reduction in the available active surface area. In the worst case, with complete penetration of the separator, to a short circuit and subsequent thermal runaway (Vetter et al., 2005; Bitzer and Gruhle, 2014; Waldmann et al., 2014; Fischhaber et al., 2016).

Anode overhang: Locally increased current densities at the edges of the anodes can lead to the formation of dendrites and thus to lithium-plating. For this reason, an overhang of the anodes is provided for in the cell design. In production, the sheets of anodes, cathodes and separators can be manufactured relatively precisely. However, during stacking of the sheets or subsequent calendaring, a translational or rotational offset of the layers can occur in relation to each other, which leads to a varying overhang, kinked electrode sheets or ablation. CT examinations and subsequent measurements can be used to characterize the anode overhang and thus identify possible risk.

Foreign material particles: The entry of particles from the production environment poses a very high risk to the battery cells. If particles get between the stacks of anodes, separators and cathodes, a separator can be damaged. Conductive particles can be the starting point for the formation of dendrites. In both cases, short circuits in the affected cell may occur during subsequent operation. The foreign material particles often consist of the materials found in cell production, which usually include aluminium, copper, stainless steel, NMC material from cathode coating, and plastics. The particles can be formed by abrasive processes of the tools or because of the mechanical separation of the belts. Particles with a size of about 50-100 µm are to be seen as critical.

Delamination and defects in the coatings: As a result of process-related influences such as the parameters during drying or possible contamination of the metallic surfaces of anode and cathode sheets, delamination of the coatings can occur, which can be intensified by forming processes or thermal cycling in subsequent operation.

Cracks in the NMC coating of the cathode: In production, cracks in the NMC coating of the cathode can occur due to drying processes of the wet-deposited coating or as a result of forming processes after drying. Cracks can also occur because of cyclic loads during charge/discharge cycles or because of thermal expansion during the subsequent operation of the cell.

Welding defects in the contact tabs: The contact tabs of the battery cell are crucial for the electrical connection of the individual layers in the cell. Welding defects can lead to an increase in electrical resistance and thus to heat problems due to electrical power loss. Mechanical stress caused by vibration or mechanical shocks can lead to cracks or fractures in the faulty welded joints. In the worst case, short circuits can occur due to faults in the welded joints, which can lead to increased current flow and overheating.

3.2. Computer tomography analysis of battery cells

The automated manufacturing of battery cells is a complex process and requires careful monitoring and control during production. Even minor deviations from the planned design of the cell can have a significant impact on its service life and, in the worst case, lead to electrical short circuits with the risk of a thermal runaway of the battery during the aging process or in subsequent field use.

In addition to inline sensor technology such as thickness measurements of the coatings of cathode and anode, optical inspection of the surfaces of the cell components for the detection of foreign bodies, impurities or scratches, as well as various electrical tests, industrial CT is increasingly being used for non-destructive testing of battery cells and modules.

3.2.1. Computer tomography (CT) system

In recent years, industrial CT has established itself as a non-destructive testing technique for inspection, quality control and failure analysis in many industrial sectors (cf. De Chiffre et al., 2014; Wevers et al., 2018; Buratti et al., 2018). In battery production, high-performance CT systems are currently used primarily to test individual battery cells during production or to identify the root cause in case of failures. CT enables a detailed three-dimensional representation of the internal structure of the battery cells and thus a precise localization and characterization of any production-related defects and impurities; cf. also (Schmitt and Dietrich, 2023) and (Marxer et al., 2021).

The latest developments in CT technology, such as photon-counting X-ray detectors in combination with Metaljet X-ray sources, will enable the realization of high-speed CT systems for 100 % inline testing of battery cells every second during production. The use of appropriate systems will lead to significantly improved efficiency in quality control and thus enable a faster time to market for battery cells. The characteristics to be detected in the battery cells have dimensions starting from approx. 50 µm in diameter. Their reliable detection requires high spatial resolution in the CT scan and at the same time sufficient X-ray transmission of the sometimes highly absorbent materials of the battery cells, including copper, nickel, manganese and cobalt. For this reason, high-performance X-ray sources with a very small focal spot size as well as flat-panel X-ray detectors with high efficiency are used as main components of the CT systems.

For the CT examinations of the battery cells described here, a µ-CT system based on a granite manipulator with transmission X-ray source (manufacturer: XRAY WorX, model: 240 TCHE Plus, energy range: 50 – 240 keV, focus size: 0.9 µm) and flat-panel X-ray detector (manufacturer: Varex, model: 4343DXI, pixel matrix: 3k x 3k, pixel size: 139 µm) is used (cf. Figure 3).

Fig. 3. Used µ-CT system with transmission X-ray source and flat-panel detector (Source: diondo GmbH).

3.2.2. Measurement process

To perform the CT examination, the battery cell is fixed in a suitable holder on the manipulator of the CT system to ensure a stable position during the measurement. After alignment of the sample in the X-ray beam, up to 3,000 X-ray images (so-called "projections") of the cell are taken at a full rotation of 360°, which are then calculated into volume using the mathematical method of 3D cone beam reconstruction. Based on the 3D data generated in this way, a detailed analysis of the various defects and other characteristics in the battery cell is carried out.

4. Results

This chapter considers the results of the case study "CT analysis of battery cells". For this purpose, in Section 4.1 the production related failures, while in Section 4.2 the failures from the usage phase of the battery cells using new and aged batteries are analysed.

4.1. Analysis regarding production related failures

The CT analysis of new battery cells for the investigation of defects from production shows clear characteristics. Some of these were also carried out on pouch cells, and the defects shown here can also be found in the cylindrical battery cells due to a similar layer structure and materials. In Figure 4 and 5 the different types of detected defects in the cells are shown. On the left side of Figure 4, an example of a particle with foreign material in the anode of the battery cell is shown and is marked in red. Due to the deviating density, a foreign particle can be easily observed in the vicinity of the anode. On the right side an enlarged view of the NMC coating of the cathode is shown, with fractures that occurred during the manufacture of the battery cell clearly visible in red.

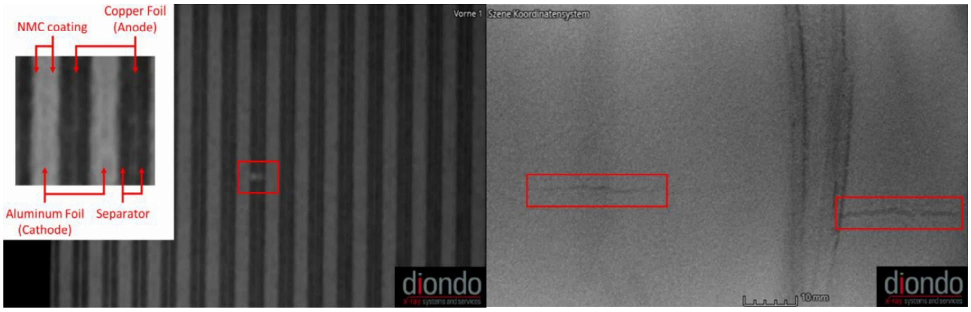

Fig. 4. (left) Particles of foreign material in a battery cell; (right) Cracks in the NMC coating of the cathode.

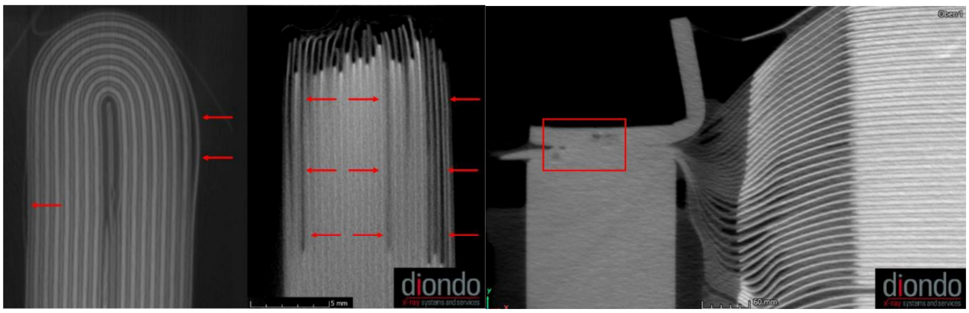

Fig. 5. (left) Delamination and defects in the coating; (right) Welding defects in the contact tabs of the battery cells.

The left-hand side of Figure 5 shows delaminations and defects in the coating of battery cells, which – as already described – are caused by various influences during production, such as contamination of the surfaces. This can be the starting point for increased swelling of the surrounding layers during use of the battery cell. On the right side an example for a welding defect in the contact tabs is shown, which can increase the electrical resistance and decrease the efficiency of the battery cell during the usage.

4.2. Analysis of new and used cells regarding runtime related failures

A direct comparison of the CT image of two different cells (a new and an aged battery cell with 30 % SOH_c) – as shown in Figure 6 – reveals clear differences between the cells. However, the comparability is limited in this work, due to the fact that two different cells with different ageing states were used. This means that at this point this is only an indication of possible fault types but no clear evidence for the different ageing effects; the types of faults found can be production- or age-related.

The observed defects in the measured aged cell are more common towards the core than at the edges. Particularly noticeable are the cracks in the cathodes, shown in red. This could be caused by the disintegration of the lithium particles in the cathode or by locally increased delamination of the cathode material. Another cause could be the drying process of the cathode coating during the production, which has either been present since the beginning of the battery life or was aggravated during ageing. The aged cell also shows increased deformation of the anode current collector, as shown in the green area. This can be due either to a manufacturing defect in the cell or to internal changes during the usage. Local swelling of the surrounding anode layers - indicated by the green arrow - due to local growth of SEI layers or dendrite growth, speaks in favour of the second reason. In both cases, the separator that spatially and electrochemically separates the two electrodes is compromised, increasing the risk of short-circuits and thermal runaway in the long term.

The advanced age of the batteries (remaining approx. 30 % SOH_c) and some of the observed failures, including the severe swelling of the anode layers or the cracking in the cathode layers, possibly caused by local growth of SEI layers or dendrite growth, indicate an overall increase in function- and safety-critical risks. Further utilisation of the battery cells could mean cell death in the form of thermal runaway. These degradation mechanisms may have been initiated by the above-mentioned production errors and accelerate certain mechanisms or cause them in the first place. Other batteries in the test series showed similar results and were not analysed further.

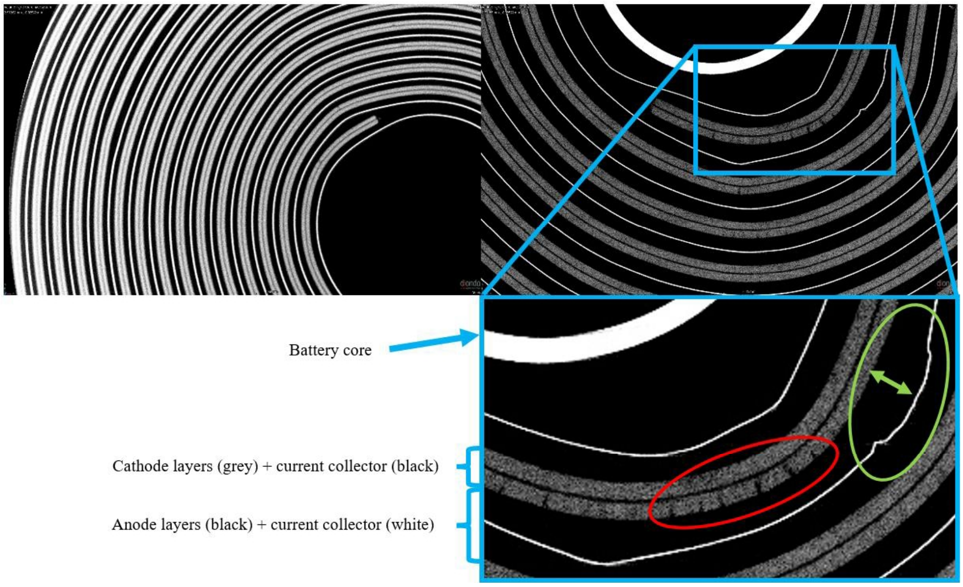

Fig. 6. Enlarged CT cross-sectional images of battery cells. Top left the new cell, top right the aged cell. Below is the enlarged view of the blue section. Points of interest are circled in green and red.

5. Comparison: Findings of other authors

CT analysis of battery cells has become increasingly popular with the growing importance of non-destructive testing methods for the analysis of battery cells used in electromobility for the initial usage in the 1st life and with a view to further usage in a 2nd life application. Therefore, the use of CT to examine cells is part of several works that are briefly listed here. The ageing of battery cells was for example investigated in (Rahe et al., 2019). In this work, cells were examined post-mortem, i.e. after the end of use or death of the battery, with a nano CT, whereby cells with a SOH_c of 80 % were analysed. The occurring effects, such as the swelling of the cathode or the breaking up of the lithium particles, are consistent with the degradation mechanisms occurring in our work. In (Taiwo et al., 2017), battery cells with a loss of 51 % SOH_c were analysed with the use of CT during ageing (in-situ). The evolution of lithium-based microstructures, such as the growing SEI or dendrites, was explicitly investigated and show to be critical degradation mechanisms regarding the risk of a thermal runaway long-term. In the work of (Wu et al., 2018) and (Kong et al., 2022), production errors occurring in lithium-ion batteries were investigated using CT, with (Wu et al., 2018) also analysing these errors as the cause of problems during the usage phase, comparing them to the results of used cells (post-mortem). Other studies investigate the degradation of lithium-ion batteries at a much deeper level. In (Frisco et al., 2016) a nano-resolution CT is used to investigate anode degradation and pore morphological changes. The Nano-CT technology for high-resolution analysis is also used in (Goldbach et al., 2023). Here, electron microscope images and nano-CT images are compared under laboratory conditions with a 3D reconstruction of new and aged cells regarding structural differences over time. In (Yusuf et al., 2022), a combined investigation method consisting of simultaneous neutron and X-ray imaging is used as the basis for the investigation of fast-charged lithium-ion batteries in an ex-situ environment. Although these studies provide a deeper insight into the degradation of battery cells, they don't (yet) have a real application character in an industrial context of battery testing due to laboratory-related high precision and therefore poor economic efficiency (cost and time efficiency).

This paper sets itself apart and investigates how the various defects that arise during production and use can be detected and analysed in a standardised manner using a CT in context of a real world application. Production errors that can cause or exacerbate degradation mechanisms in the further usage are analysed here. The batteries were aged to a very low SOH_c, whereby the literature often only analyses the first life (up to 80 % SOH_c) or the death of the cell in the form of thermal runaway. This should lead to a standardised analysis of possible function- and safety-critical mechanisms that originate from production or use, which will be relevant for investigations regarding the first- and second-life of battery cells.

6. Summary

This case study shows that the detection and analysis of defects as small as ten micrometers are possible. Reliable detection of internal structures that may contain safety-critical defects is possible. The CT opens new perspectives compared to conventional condition analysis methods. It not only provides detailed information on the condition of the cell, but also gives additional insight into its internal structure and potential deviation/changes respectively internal failures. In contrast to conventional capacitance measurement, which can induce a certain degree of ageing, CT enables a precise assessment of the safety-related condition and the detection of degradation indicators.

CT makes a significant contribution to the detection of functional and safety-critical failures and to the substantiation of the long-term reliability of the cell. The question of cost-effectiveness, particularly regarding the inspection of used cells, as well as the time efficiency for analysis on such a scale remain unanswered at present.

Overall, this paper provides a first indication of the use of CT analysis of new and used cells to detect internal defects from the production and use of the cells, which may pose functional and safety critical risks for the use of the battery cell in a first and second life. With a larger study, these results can be further extended and standardised to make more precise statements over the state of the battery cells. Following this, the results can be analysed in a real application where other factors such as cost and time efficiency (e.g. inline-CT) are relevant.

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