



Non-destructive characterization techniques for battery performance and life-cycle assessment

Charlotte Gervillié-Mouravieff¹, Wurigumula Bao², Daniel A. Steingart^{3,4} & Ying Shirley Meng^{1,2}  

Abstract

Improving the performance and efficiency of batteries is key to enabling the broader adoption of electric vehicles and the effective use of intermittent renewable energy sources. However, this enhancement demands a more comprehensive understanding and improved surveillance of the essential mechanisms that control battery functionality over their entire lifespan. Unfortunately, from the moment batteries are sealed until their end of life, they remain a ‘black box’, and our knowledge of the health status of a commercial battery is limited to current (I), voltage (V), temperature (T) and impedance (R) measurements, at the cell or even module level during use, leading to an over-reliance on insufficient data to establish conservative safety margins and a systematic under-utilization of cells and batteries. Although the field of operando characterization is not new, the emergence of techniques capable of tracking commercial battery properties under realistic conditions has unlocked a trove of chemical, thermal and mechanical data that have the potential to revolutionize the development and utilization strategies of both new and used lithium-ion devices. In this Review, we examine the latest advances in non-destructive characterization techniques, including electrical sensors, optical fibres, acoustic transducers, X-ray-based imaging and thermal imaging (infrared camera or calorimetry), and their potential to improve our comprehension of degradation mechanisms, reduce time and cost, and enhance battery performance throughout their three main life stages: during the manufacturing process, during their utilization and, finally, at the end of their life.

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¹Department of NanoEngineering, University of California, San Diego, La Jolla, CA, USA. ²Pritzker School of Molecular Engineering, University of Chicago, Chicago, IL, USA. ³Department of Chemical Engineering, Columbia University, New York, NY, USA. ⁴Columbia Electrochemical Energy Center, Columbia University, New York, NY, USA.

✉e-mail: shirleymeng@uchicago.edu

Key points

- Non-destructive techniques capable of tracking commercial battery properties under realistic conditions have unlocked chemical, thermal and mechanical data with the potential to accelerate and optimize the development and utilization strategies of lithium-ion devices, both new and used.
- Before use, battery assembly wetting and formation cycles should be carefully monitored using imaging or advanced electrochemical techniques to reduce the scrap rate. This could be achieved using tomography, acoustic imaging or spectroscopic characterization.
- During use, thermal and mechanical phenomena at the cell level could contribute to enhance the battery management system (BMS). In this context, electrical and optical sensors offer large versatility of shape, sensitivity and accuracy.
- After use, accurate evaluation of battery degradation at the cell level and determination of their true end-of-life status is crucial for second-life applications. To preserve battery integrity, acoustic and thermographic imaging appear promising techniques.

Introduction

Non-destructive characterization techniques enable crucial insights while preserving the integrity of examined objects. Similar to non-invasive medical screening detecting various health conditions without harming the body, non-destructive characterization of batteries can provide critical data for optimizing performance and longevity without compromising the battery's structural integrity. Although the development of battery health records is vitally important¹, achieving this goal is not without its challenges. Batteries involve dynamic electrochemical and chemical reactions, and electronic and ionic conductivity limitations. Furthermore, the presence of electrode–electrolyte interface instability, lithium plating, cathode and anode degradation, and electrolyte decomposition has a considerable effect on battery performance. Hence, understanding and mitigating battery degradation mechanisms is crucial to enhance battery performance and ensure long-term durability.

Several characterization techniques have been developed to better understand battery degradation. Spectroscopic techniques (X-ray diffraction, Raman spectroscopy, Fourier transform infrared spectroscopy, ultraviolet–visible spectroscopy), nuclear magnetic resonance, microscopy techniques (scanning electron microscopy, transmission electron microscopy, atomic force microscopy), thermal techniques (differential scanning calorimetry, thermogravimetric analysis) and gas analysis (gas chromatography, mass spectrometry) allow to probe fundamental properties of batteries and optimize their behaviour under different conditions^{2–4}. However, most of these techniques require the disassembly (or ‘teardown’) of the cell for post-mortem characterization. Therefore, a predictive understanding of battery behaviour is missing owing to the lack of real-time information, potential sample alteration and inability to capture global and transient phenomena. Some laboratory-scale cell designs have enabled characterization to be conducted in situ with the cell in its fully assembled state^{5–8}. Nevertheless, most of those laboratory-scale cells utilize lithium metal as a counter/reference electrode in flooded electrolyte conditions, in which

the intricate reaction mechanisms between the electrolyte, anode and cathode⁹ are often overlooked. Understanding the evolution of commercial cells is imperative not only to maximize their performance and lifespan but also to evaluate their end of life for second-life applications or recycling purposes. However, the challenge becomes more considerable for the industry, as from the moment batteries are sealed until their end of life they remain a ‘black box’. Therefore, the current knowledge of their health status is limited to current (I), voltage (V), impedance (R) and, sometimes, temperature (T) measurements, at the cell or even module level when in use (Fig. 1). This lack of comprehensive knowledge leads to time-consuming trial and error development processes, an over-reliance on imperfect data to establish safety margins and a systematic under-utilization of cells and batteries. Consequently, battery conditioning, which includes processes such as electrolyte wetting and interface formation, can take up to 3 weeks and contribute to 48% of the entire manufacturing cost of a battery¹⁰. Despite this extended and intricate process, 5–10% of production capacity still ends up as production scrap¹¹. Another unfortunate consequence is the premature disposal of batteries considered ‘dead’ when they have lost 20% of their initial capacity while abandoning the remaining 80% as waste.

Techniques capable of tracking commercial battery properties under realistic conditions have unlocked a trove of chemical, thermal and mechanical data with the potential to accelerate and optimize the development and utilization strategies of lithium-ion devices, both new and used^{12,13}. In this Review, we examine the latest advances in non-destructive characterization techniques, highlighting their crucial role in the in-depth understanding of battery life during three main life stages: the manufacturing process, usage and the end of life. We then showcase the latest technological innovations and provide a fresh perspective on how these techniques can be applied to gain comprehensive insights into the entire life cycle of batteries (Fig. 1). As batteries progress through these stages, the type of integration and the monitored parameters evolve, necessitating introduction of appropriate characterizations. For the battery manufacturing process, we explore characterization techniques that can detect manufacturing defects, observe wetting phenomena and assess interface formations. At the ‘in use or online’ integration stage, we provide an overview of sensors that could seamlessly integrate into electrical vehicle battery packs, presenting novel safety and performance thresholds in the battery management system (BMS), focusing on optical fibre sensing. In the context of the end of life, we discuss the importance of characterizing this relatively unexplored state for the purposes of recycling and second-life applications. Finally, we discuss the potential of combining these techniques to drive ‘battery passport’ initiatives, which can improve battery data analysis and traceability and contribute to the advancement of new sustainable battery technologies (sodium-ion, solid-state batteries).

Non-destructive characterizations for battery manufacturing

Battery manufacturing has a crucial role in achieving optimum performance and longevity. From electrode production to cell assembly and battery electrochemistry activation, all steps of battery production have notable effects on the cell's electrochemical properties. As the manufacturing process advances, each step becomes more critical as it retains the value of the previous steps. Thus, rejecting cells just after the manufacturing process results in considerable time and money losses.

Before manufacturing, characterization of the material quality is required. Techniques such as inductively coupled plasma optical

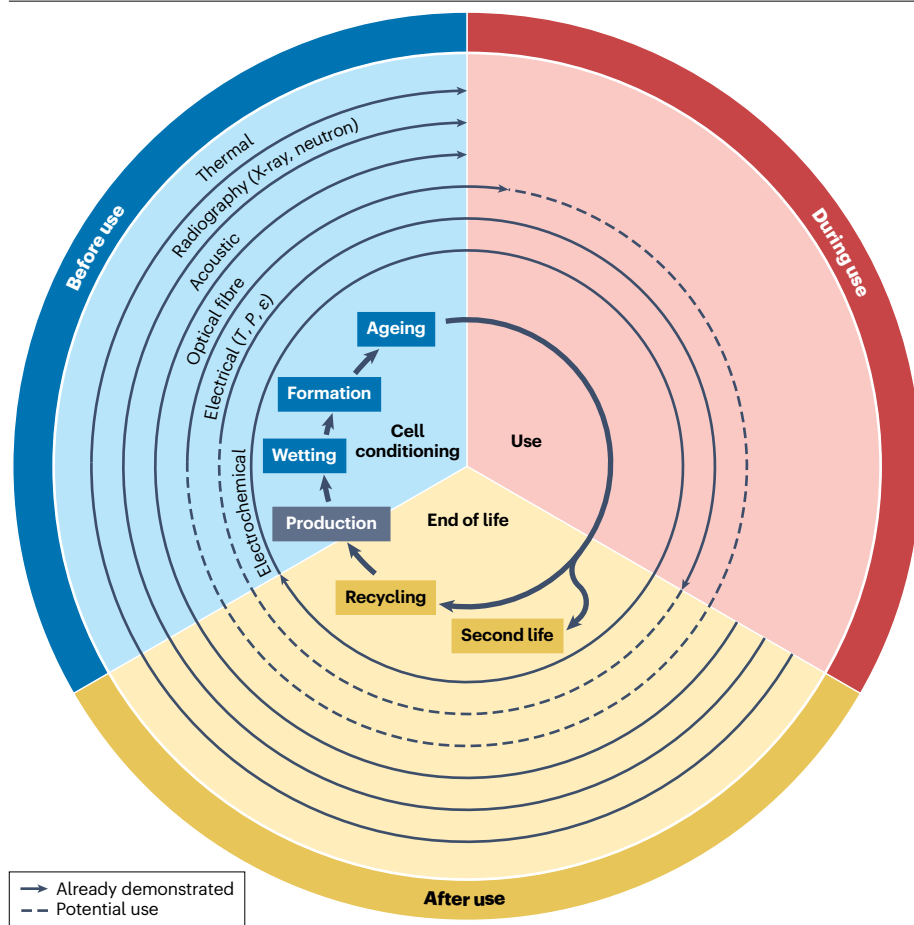


Fig. 1 | Road map of non-destructive characterization techniques for comprehensive commercial battery life-cycle analysis. Non-destructive characterization being used for commercial batteries. Solid line indicates that published research has utilized the technique to characterize a particular stage of battery life: before use, during use or after use. Dashed line represents our perspective on potential future applications of the technique. Absence of a line means that the technique cannot be used in this part of the battery's life.

emission, optical microscopy or scanning electron microscopy and energy-dispersive spectroscopy are commonly used to help identify the impurity concentration in the materials¹⁴. Throughout the material production process, quality assurance and quality control for both materials and electrodes are needed. This includes morphology and structural parameter characterizations that are realized through various techniques, including scanning electron microscopy, X-ray diffraction, profilometry or laser scanning. Additionally, the distribution of chemical species in the electrode can be examined using vibrational spectroscopies such as Fourier transform infrared spectroscopy and Raman spectroscopy^{15,16}.

However, once the cell assembly is completed, the jelly roll is enclosed within a bag or can, rendering its observation impossible. Furthermore, when the wetting is completed, batteries are hermetically closed and, essentially, become a 'black box'. From this moment, quality control relies on electrochemical performance metrics such as electrochemical impedance spectroscopy (EIS), the open circuit voltage and capacity^{17,18}. Unfortunately, these parameters only offer average values for the entire cells and lack detailed imaging and chemical data that could uncover the origins of defects or the battery's dynamic behaviour. Moreover, most electrochemical techniques require sophisticated instrumentation, are characterized by slow data acquisition times, and require expertise in electrochemistry and data fitting techniques^{19,20}. Consequently, even cells passing quality control may

exhibit post-production variations in capacity ageing trends and safety issues^{21,22}. The large number of variables during the conditioning process and limited knowledge about their aggregated effect on the battery properties necessitate parameterization through experimentation, which is highly time-consuming and can result in lengthy wetting and ageing processes. For instance, wetting and formation alone can take up to 3 weeks and contribute to 48% of the entire manufacturing cost¹⁰. Additionally, despite the efforts made to optimize the conditioning process, the lack of understanding of the different steps still leads to 5–10% of production capacity becoming production scrap¹¹. Therefore, non-destructive characterizations are vital for enhancing efficiency, reducing costs and minimizing scrap rates associated with battery production.

Battery assembly and packaging

Once the electrode manufacturing is completed, the anode and cathode are assembled in a dry room into an electrode stack. There are a multitude of folding and stacking techniques to precisely align anode and cathode electrodes with separators between each layer. During this process, it is crucial for the anode to overhang the cathode on all sides to prevent lithium dendrite formation. Additionally, the separator must ensure thorough coverage of the electrodes, without wrinkling or tearing to prevent defects such as holes, tears or misfolded electrodes. The final step in electrode stack assembly involves welding the external tab to the electrode foil tabs, using ultrasonic or laser techniques²³.

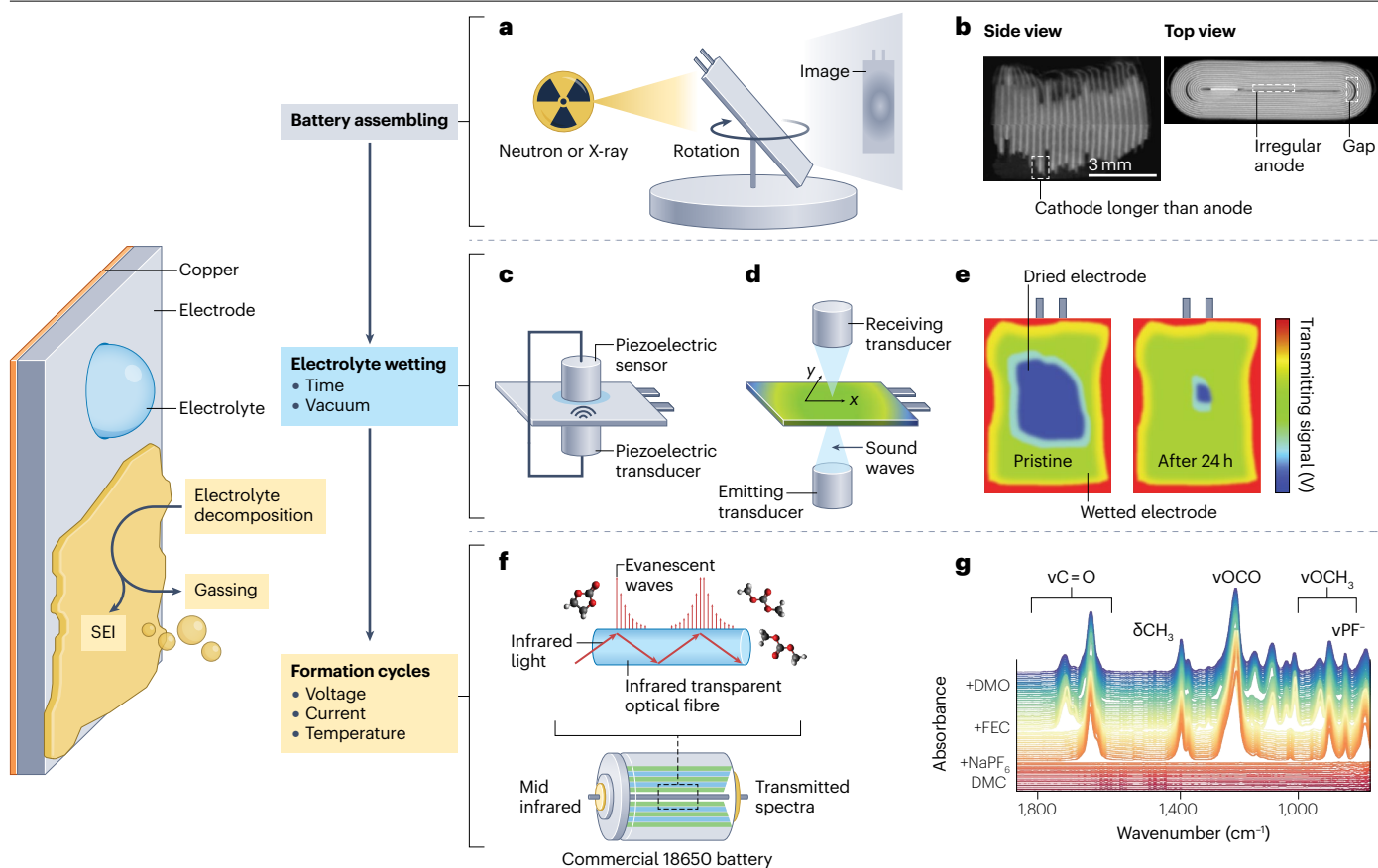


Fig. 2 | Non-destructive characterization to understand the battery manufacturing process, including battery assembling, battery wetting and formation cycles. a, b, Battery assembling. **a,** The battery tomography characterization principle. **b,** Computed tomography cross-section images taken from a pouch cell after assembly, highlighting defects in the electrode alignment. **c–e,** Electrolyte wetting. **c,** Battery acoustic characterization. **d,** Battery acoustic imaging characterization. **e,** Acoustic images of the wetting process. **f,** Infrared fibre evanescent wave within chalcogenide glass fibre principle placed in a 18650 cell. **g,** Infrared fibre evanescent wave absorbance spectra evolution during the addition of different electrolyte solutions. Panel **b** is reprinted from ref. 25, CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>); panel **e** is adapted with permission from ref. 39, Elsevier; panel **g** is adapted from ref. 50, Springer Nature Limited.

f, g, Formation cycles. **f,** Infrared fibre evanescent wave within chalcogenide glass fibre principle placed in a 18650 cell. **g,** Infrared fibre evanescent wave absorbance spectra evolution during the addition of different electrolyte solutions. Panel **b** is reprinted from ref. 25, CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>); panel **e** is adapted with permission from ref. 39, Elsevier; panel **g** is adapted from ref. 50, Springer Nature Limited.

The cell is then encased in pre-shaped packaging, sealed on all but one edge for electrolyte filling. At this stage, precise characterization of cell failures can help sort the cells and save the cost of electrolyte injection. The high-potential test in battery cell production is a traditional quality control procedure, where battery cells are subjected to high voltages to identify any separator defects or weaknesses, ensuring the safety and reliability of the battery²⁴. This test helps roughly sort cells by detecting short circuits. Computed tomography is a spatially non-destructive method that combines a series of X-ray measurements taken from different angles to produce detailed cross-sectional images, allowing to quantify material properties and to detect battery internal structural features, thus facilitating the identification of potential failures (Fig. 2a). It has been demonstrated that despite capacity and impedance measurements being in line with the manufacturers' specifications for commercial cells, computed tomography images reveal quality concerns, such as electrode misalignment and gaps in the winding structure²⁵ (Fig. 2b). Furthermore, various defects could be observed in 18650 and pouch cells using computed tomography, such as welding burrs, overlapping of electrode tabs and deflected

electrodes²². As conventional electrochemical methods based on electrical testing of batteries are limited in terms of information they can provide, computed tomography can be utilized both by industry and by academia to assess the impact of new processes on assembly and packaging quality without disassembly²⁶.

Battery wetting

After packaging, the electrolyte filling process in battery manufacturing comprises two phases: injection and wetting to introduce the appropriate amount of electrolyte into the cell, ensuring complete wetting of all pores in the cell materials to ensure even current distribution. If the electrolyte to material ratio is too high, this can reduce energy density, increase cell cost and increase safety liability owing to volatility of the electrolyte. Insufficient wetting leads to reduced capacity, rapid capacity decay, low power capability and non-uniform solid–electrolyte interphase (SEI) layer growth during formation²⁷. Despite the widespread recognition of potential time and cost savings for the industry, academic and patent literature on electrolyte filling is currently insufficient to optimize the process.

To assess battery wetting, electrochemical characterization techniques are commonly used. For example, high-frequency ($>10^5$ Hz) resistance measured by EIS is directly related to the Li^+ diffusion within the electrolyte through the pores of electrodes and separators, and could be directly linked to the wetting state of the cell²⁸. Additionally, open circuit voltage and electrical contact resistance measurements have been used to correlate the wetting state with resistance change as the electrolyte fluid allows ion exchange of the cell components^{29,30}. X-ray or neutron sources have been used to obtain information on non-homogeneous areas. Several studies have provided information on the effect of the cell design, electrode porosity and electrolyte, and helped optimize the wetting process^{31–33}. Neutron radiography, for instance, has revealed the impact of directional flow of electrolyte liquid within the electrode assembly, creating gas entrapments³⁴. However, these techniques often require expensive equipment and may only address specific issues. Moreover, X-ray imaging requires a contrast agent which limits their utilization for in-line measurements³⁵. A complement to previous methods, acoustic methods offer an inexpensive and high-quality visualization using sound waves. Acoustic sensing can be either passive³⁶ (measurements of the acoustic wave generated by the environment) or active³⁷ (by emitting sound waves and analysing their propagation (Fig. 2c)). Notably, ultrasound exhibits higher attenuation in electrodes or separators that are not adequately wetted, making acoustic sensing a preferred method for wetting monitoring³⁸. For example, an ultrasonic battery scanner has been employed to achieve 2D mapping of a commercial battery, enabling the visualization of electrolyte wetting, drying and gas formation during cycling³⁹ (Fig. 2d,e). This imaging technique enables rapid determination of the minimum required electrolyte injection volume and wetting time, facilitating the optimization of battery manufacturing processes.

Battery formation cycles

At the end of the conditioning, battery formation is one of the most crucial and closely guarded processes in the manufacturing of batteries, particularly for lithium-ion batteries. It consists of the initial charging and discharging cycles that a battery undergoes before it is ready for use. In the formation process, the anode and cathode interact with the electrolyte to create a SEI layer on the anode surface. The formation protocols are intricate and require precise control of temperature, voltage and charging rate to quickly produce a SEI that is uniform, electrically insulating, and that facilitate ion conduction⁴⁰. The SEI has a pivotal role in battery performance as it acts as a protective barrier, preventing further reaction between the anode and the electrolyte, thus reducing capacity drop and enhancing the overall capacity, efficiency and cycle life^{41,42}. The complexity of SEI formation mechanism is the biggest challenge for the development of new formation protocols that are optimized, more reliable and suitable for different active materials and electrolytes.

In industrial practice, electrochemical techniques are commonly used to monitor the cell response during formation protocols, particularly by analysing the first EIS spectrum semicircle, ascribed to the impedance of the SEI layer, which enables tracking of interface growth. To gain deeper insights into the mechanism of the battery formation, the gas generated due to electrolyte decomposition can be monitored. In this context, multiple approaches have been adopted to detect gas generation through imaging techniques, internal pressure assessments or external measurements of volume to gauge cell swelling. It is important to note that cell swelling does not arise solely from gas

generation but is also influenced by the expansion and contraction of the electrode material and the growth of interfaces.

For example, electrochemical dilatometry and X-ray computed tomography enable non-destructive probing of gas generation⁴³. For large-scale applications, operando pressure measurements with strain gauges mounted on cells are particularly suitable for tracking volumetric expansion during cycling⁴⁴. The effectiveness of this technique is evidenced by its ability to accurately monitor SEI growth and effectively decoupling gassing from swelling due to the SEI thickening on the negative electrode⁴⁵. Moreover, acoustic sensing allows to observe gas generation and SEI formation based on the distinct propagation of acoustic waves as a function of materials' gas, porosity and mechanical properties. An acoustic transducer provides the ability of acoustic measurements to track operando gas generated during formation as well as silicon passivation⁴⁶ (Fig. 2c).

Recently, optical fibre sensors have been successfully integrated into batteries for thermal and mechanical monitoring¹². Among them, fibre Bragg grating (FBG) sensors use periodic changes in an optical fibre's refractive index to reflect specific wavelengths, allowing them to measure various environmental factors such as temperature, strain and pressure within the cell. The sensitivities to these factors can be decoupled through careful design of implantation, packaging or sensor combinations. For example, a microstructured optical fibre with an air-hole pattern design is highly sensitive to hydraulic pressure. Therefore, one study has demonstrated the correlation between pressure increase and thermal events measured by FBG sensors during the first charge of 18650 cells that are ascribed to electrolyte decomposition. From the FBG sensor measurements, heat could be calculated, and the cascade reactions leading to SEI could be observed with this technique⁴⁷.

However, albeit valuable, these strategies cannot capture processes at the molecular levels occurring in the battery and the cascade of chemical reactions underlying the formation of the SEI. To close this gap, tilted FBG sensors can be used to measure the refractive index variation in the electrolyte during the formation cycles. Large refractive index variations can be attributed to unstable electrolytes, providing a preliminary understanding of their stability, but more detailed information is required to understand the underlying chemical changes causing these variations⁴⁸. To address the limitations of the refractive index sensors, photonic crystal fibres – commonly employed in developing biosensors for molecular recognition – have been used in combination with microfluidics to monitor the evolution of an electrolyte by Raman spectroscopy during cycling in battery systems. However, this approach relies on an external pump to extract small amounts of electrolyte, limiting continuous monitoring and the ability to track changes in the electrode materials⁴⁹. In another study, infrared fibre evanescent wave spectroscopy has been integrated in commercial 18650 sodium-ion cells (Fig. 2f). Fibre evanescent wave spectroscopy utilizes the interaction between guided light in an optical fibre and the evanescent field extending beyond its core to analyse the properties of substances or molecules near the surface of the fibre. Using infrared transparent chalcogenide fibres, electrolyte stability could be assessed in operando with excellent sensitivity, identifying the nature of electrolyte decomposition products and ion solvation dynamics as a function of voltage and current (Fig. 2g). Furthermore, embedding the fibre directly in the material enables the tracking of lithium uptake–removal processes in positive electrode materials using infrared spectroscopy⁵⁰.

The introduction of non-destructive battery characterization methods has the potential to improve the quality control of battery

manufacturing processes, facilitating the identification of defects at every stage. In addition, the knowledge gained from these techniques could enable rapid evaluation of new electrolytes and additives, facilitating the development of more effective formation protocols, ultimately resulting in higher production yields, improved cell quality and reduced costs. We anticipate rapid integration of acoustic monitoring, whereas in the long term, facilitated by synergistic efforts from different fields, infrared fibre or similar spectroscopic technologies could be integrated to optimize formation protocols.

Non-destructive characterization techniques in electrical vehicles

Electrical vehicles require batteries with improved performance and safety. Currently, to meet the requirements for mileage and traction power, battery packs must contain hundreds or thousands of single cells in series and parallel connections (Fig. 3a). Among those individual cells, variations in capacities, voltages, internal resistances and ageing effects can lead to system failures. These inconsistencies, arising from impacts, vibrations, temperature fluctuations or internal defects from manufacturing, result in degraded battery performance and excess heat generation during operation. Therefore, the BMS monitors and regulates the charging, discharging, temperature and overall health to ensure safe and efficient battery operation. The current BMSs rely on imprecise measurements of current, voltage and external temperature at the battery pack level, which cannot detect individual cell degradation⁵¹ (Fig. 3a).

Considerable efforts have been directed towards enhancing the BMS through modelling techniques for battery state and thermal evaluation and prediction⁵¹, including machine learning approaches for more accurate prediction of battery behaviour and health⁵². Additionally, recent advancements in onboard and online impedance spectroscopy have shown much promise, as the impedance spectrum of a battery cell at a given instant depends on the state of charge, as well as on other physical and chemical properties of the cell, such as ageing, and chemical degradation effects^{53,54}. However, it is essential to recognize that impedance is defined for time-invariant systems. Consequently, it cannot identify transient phenomena associated with battery degradation.

Notably, mechanical stress and temperature variations can serve as early indicators of chemical or structural defects and could adversely impact battery safety. Consequently, these parameters have to be carefully monitored to improve BMS and overall safety. Herein described below are sensors that hold the potential to monitor mechanical and thermal evaluations of cells within the battery pack.

Thermal management

Temperature variation is one of the main factors affecting battery degradation. High temperatures accelerate the electrolyte decomposition and SEI growth, leading to increased internal resistance, capacity degradation and higher heat generation. Conversely, lower temperatures can induce lithium metal deposition, potentially causing internal short circuits. Accurate temperature monitoring is essential to detect such occurrences. Present BMSs commonly integrate thermocouple sensors into the battery pack or specific cells to monitor the surface temperature (Fig. 3b). These sensors are easy to use, compact and robust⁵⁵. However, they are inferior in their accuracy and sensitivity with respect to alternative temperature measurement techniques. For instance, micro temperature sensors demonstrate higher precision and sensitivity in temperature measurements within a confined area⁵⁶ (Fig. 3c). Nevertheless, these micro-sensors are more fragile,

susceptible to damage owing to their size and more prone to electrical noise interference as opposed to thermocouples.

FBG sensors provide real-time temperature readings with a sensitivity of 0.1 °C, are resistant to electromagnetic interference, are compact and are suitable for challenging environments owing to their dielectric characteristics. Furthermore, the integration of multiple FBG sensors into a single optical fibre, where each sensor reflects light at a specific wavelength, facilitates monitoring of separate cells and positions within a battery pack (Fig. 3d). However, surface measurements cannot precisely capture temperature fluctuations within the cell. Indeed, studies utilizing FBG sensors revealed that temperature discrepancies between the cell interior and surface ranged from 0.2 °C to 5.4 °C for 18650 cells cycled at rates from C/10 to 10C (ref. 47). Furthermore, variations of up to 2 °C were evident within the cell⁵⁷ (Fig. 3e). Furthermore, thermal measurements at different locations within and around the cell quantify heat generation during operation. For instance, fluctuations in generated heat, measured by FBG sensors, show remarkable sensitivity to variations in current applied to the cells within operational protocols such as the Worldwide Harmonized Light Vehicles Test Procedure (WLTP)⁵⁸ (Fig. 3f). Additionally, recent work has demonstrated the capability of optical fibre sensors to monitor the internal temperature and pressure of lithium-ion 18650 cells prior to and during thermal runaway⁵⁹. This technique, which identifies early safety warnings through slope changes in temperature and pressure differential curves before thermal runaway, presents a pathway for enhanced accuracy in predicting and managing cell thermal behaviour.

Mechanical management

Changes in electrode volume, electrolyte decomposition resulting in gas formation or mechanical abuse to the battery pack can cause defects and thermal runaway. Consequently, mechanical cell management is as important as electrical and thermal management. It has been demonstrated that a simple set-up with a load cell sensor positioned between a cell and a plate could enable the measurement of reversible and irreversible volume expansion in lithium-ion pouch cells (Fig. 3g). Notably, a direct relationship was established between irreversible pressure increase and performance degradation, along with a polarization increase, confirming the need for pressure monitoring⁴⁵. In the same study, the performance of cells containing SiO, silicon alloy or carbon-coated nano silicon electrodes has been investigated and substantial volume expansion of the silicon-based electrode within pouch cells has been observed⁶⁰. For large lithium-ion pouch cell measurements, General Electric has developed its own eddy current sensor (Fig. 3h). The device utilizes electromagnetic induction to measure changes in the size or shape of conductive objects, detecting expansions or contractions due to mechanical stress⁶¹. Additionally, by attaching a FBG sensor to the surface of a 18650 can, measurements of the battery state of charge in real time can be realized⁶² (Fig. 3i). Similar results can be obtained with carbon nanotube sensors attached to the surface of a pouch bag⁶³ (Fig. 3i). Reducing the sensor dimensions permits mechanical measurements to be undertaken directly within the cell⁶⁴. By using thin film strain gauges positioned at various locations, the internal strain distribution within the cell can be studied^{65,66} (Fig. 3j). Electrode-scale measurements have been conducted by optical fibres inserted into the electrodes (Fig. 3k) for monitoring internal stress fluctuations, revealing nano and micro silicon behaviour within the electrode during cycling⁶⁷. Lastly, it is important to note that the versatility of optical fibres opens up a wide range of sensing opportunities; manipulating the fibre's microstructure can enhance sensitivity to pressure⁶⁸ (Fig. 3l).

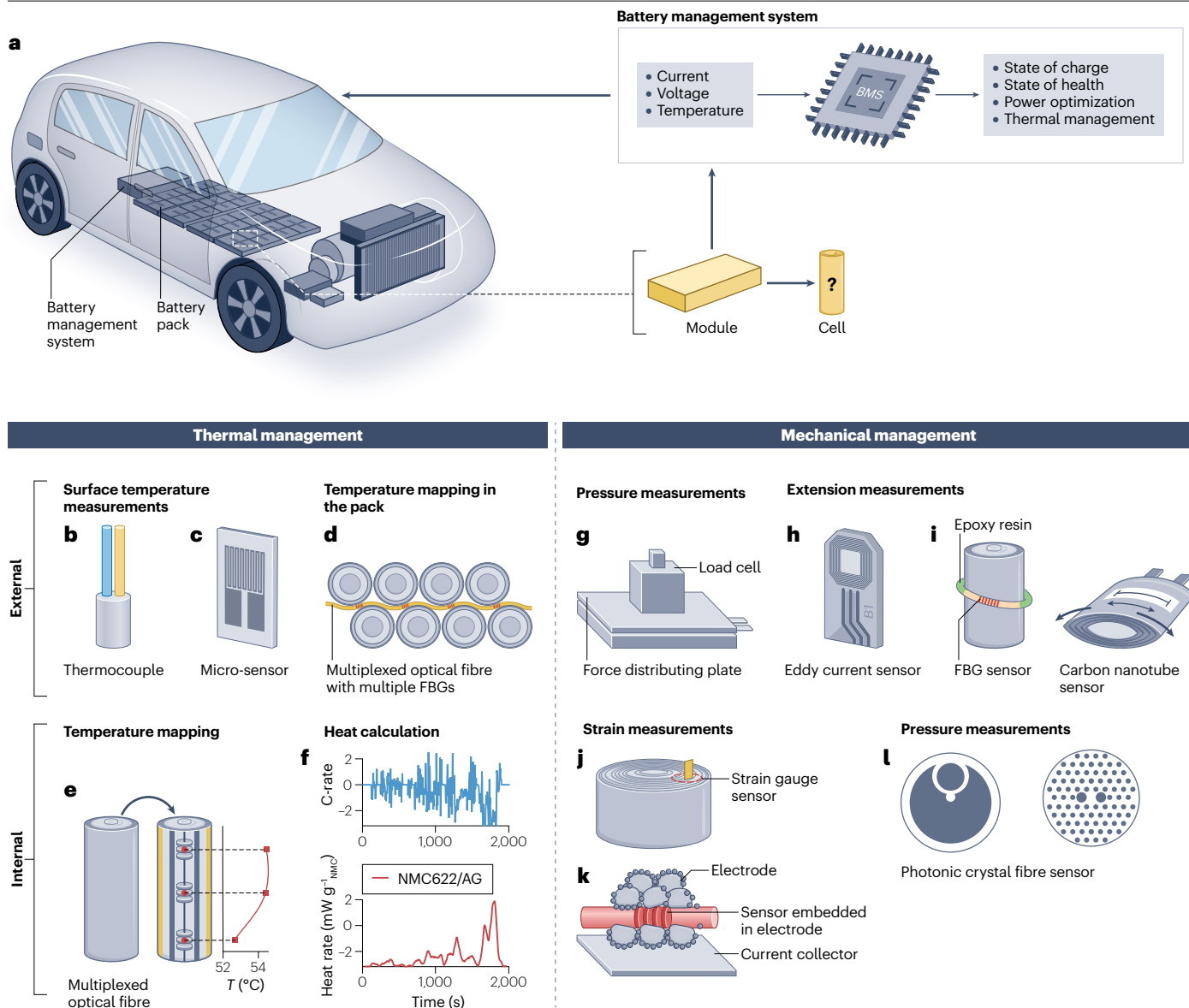


Fig. 3 | Non-destructive characterization opportunities for electric vehicles. **a**, Challenge for battery characterization in electric vehicles. Currently, performance management is governed by the battery management system (BMS) which relies on a subset of temperature (T) and voltage (V) measurements, at the module level only, to estimate state of charge and state of health, and to optimize power and thermal management. **b–f**, Thermal management. **b, c**, Temperature sensors for external cell measurements⁵⁶. **d**, Optical fibre with multiplexed fibre Bragg grating (FBG) inscribed in it to measure cell temperature in a module. **e**, Internal temperature mapping with multiplexed FBG sensor. **f**, Heat calculated

from FBG sensing during the Worldwide Harmonized Light Vehicles Test Procedure (WLTP) of NMC-based cells. **g–l**, Mechanical management. **g**, Load cell for external pressure measurements⁴⁵. **h**, Eddy current sensor⁶¹. **i**, External strain measurements with FBG sensors⁹⁹ and carbon nanotube sensors. **j**, Strain gauge sensor integrated into a 18650 cell. **k**, FBG sensor embedded in electrode for internal strain measurements⁶⁷. **l**, Photonic crystal fibre for pressure measurements inside cells^{47,68}. Panel **f** is adapted with permission from ref. 58, Elsevier; panel **h** is reprinted with permission from ref. 61, ASME; panel **l** (left) is adapted from ref. 47, Springer Nature Limited.

To enhance BMSs, the integration of temperature and mechanical sensors, either inside or outside the cell, offers an opportunity to enable early monitoring of degradation. These data could allow battery manufacturers to take preventive measures such as repair or replacement of a battery, reducing potential safety risks for users. Furthermore, these measurements could also extend batteries life by providing accurate assessments of their state of health.

Characterizations of batteries for end of life or second-life options

Evaluation of retired batteries for secondary application

Recently, the electrical vehicle sector has witnessed the emergence of a second-life market for batteries, where retired electrical vehicle batteries constitute the initial phase preceding the eventual recycling and recovery of materials. The battery pack is considered to have reached

its end of life when its capacity falls below 80% of the initial capacity within the domain of high energy-density electrical vehicles. To take advantage of the remaining capacity, repurposing those batteries in less power-intensive applications, including energy storage systems and low-power devices, has gained increasing interest. Especially because data-driven battery degradation models with anticipated cycling conditions show that second-life batteries with 70–80% of their initial capacity can last more than 10 years for certain applications⁶⁹.

However, degradation phenomena during battery ageing are poorly understood and can impact battery safety. For instance, under certain cycling conditions, lithium ions can be reduced to lithium metal plating on the surface of the graphite anode, forming uneven and dendritic lithium deposits that may cause internal short circuits and safety hazards. Additionally, other phenomena such as gas generation, cathode cracking, electrode delamination, binder decomposition and current collector corrosion also impact battery ageing and safety. In addition, cells within the same battery pack frequently demonstrate performance disparities, the worst cell in a series-connected string determining the end of life. As a result, accurate evaluation of battery degradation at the cell level and determination of their true end-of-life status within second-life applications is essential. To this end, battery packs have to be disassembled in order to “screen, sort and classify” the cells⁷⁰ (Fig. 4a). These steps aim to identify cells that do not meet the requirements for second-life applications so as to regroup batteries with comparable degradation levels and similar electrochemical behaviours⁷¹.

Electrochemical performance evaluation

To quantify battery degradation, electrochemical tests are typically conducted, including open circuit voltage, internal resistance and capacity measurements. Among them, incremental capacity (DV-IC) analysis can be used to assess the health and performance of a battery⁷². Additionally, EIS can be used to measure a battery’s ohmic resistance, charge transfer resistance, diffusion, electrode degradation and state of health⁷³. However, interpreting the EIS data requires complex algorithms, either model-based, using equivalent circuits, or data-driven⁷⁴. However, the intricate nature of battery degradation and the diversity of materials and electrolytes within battery systems limit the applicability of a universal degradation model, leading to ambiguities about battery ageing⁷⁵. Finally, as previously mentioned, EIS and electrochemical tests in general give averaged properties of the entire cell, potentially neglecting safety risks arising from local defects. Especially during the cycling of large-size batteries, variations of voltage, current density and local changes in the state of charge can lead to non-uniform local heat generation under different operating conditions, resulting in non-uniform ageing and increased local resistance over the battery surface as well as localized hot spots and severe temperature gradients.

Imaging techniques

Advanced non-destructive and non-invasive techniques are required to characterize battery health. In this case, however, electrical or fibre-based sensors cannot be used. Imaging techniques such as X-ray

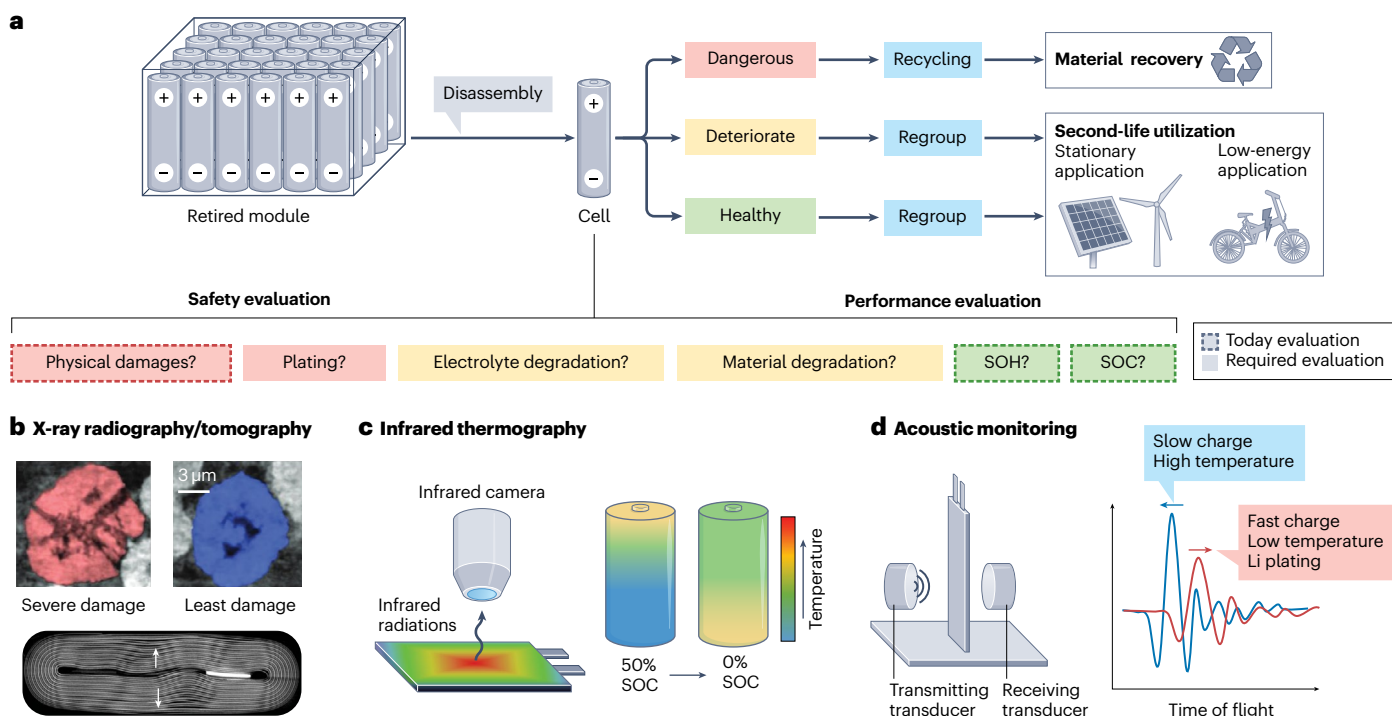


Fig. 4 | Characterization of batteries for end-of-life or second-life options.

a, Steps for retired battery second-life or end-of-life application. Battery pack and module are disassembled, screened and sorted depending on their remaining performances. Performances are evaluated using properties (surrounded by dotted line). However, all properties cited in the middle part of the figure should be evaluated to ensure maximal safety and performance of the second-life application. **b**, Phenomena observed with X-ray imaging, including electrode

ageing and cell swelling. **c**, Infrared thermography principle and example of infrared images of 18650 cells depending on their state of charge (SOC)⁷⁹. **d**, Acoustic scanning principle and plating effect on time-of-flight signal. SOH, state of health. Panel **b** (top) is reprinted with permission from ref. 76, Wiley; panel **b** (bottom) is reprinted from ref. 77, CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0>).

imaging, infrared thermography and acoustics can provide rapid evaluation of the physical and mechanical properties of the cells. These techniques provide information about various parameters such as temperature, electrolyte degradation and gassing issues, structural deformation and active material degradation. X-ray imaging is an extremely useful technique to study battery degradation phenomena, allowing, for example, to observe damage at the particle scale⁷⁶ (Fig. 4b, top). For example, in one study, synchrotron-based computed tomography techniques was used for imaging the internal morphology deformation of a LiCoO₂/graphite jelly roll pouch cell caused by gas evolution under abuse conditions⁷⁷ (Fig. 4b). The non-uniform expansion of the pouch cell jelly roll was observed with the most notable expansion detected in regions where the jelly roll had been deformed prior to abuse, suggesting that the stack pressure in these distorted areas is lower compared with the rest of the assembly. Alternatively, non-destructive computed tomography measurements using X-ray and neutron techniques can serve as powerful instruments for understanding battery degradation at different scales. However, the prohibitive costs and extensive duration of these experiments hinder their widespread industrial application.

Infrared thermography is a technique that can capture and visualize spatially non-uniform temperature variations in objects or surfaces using infrared radiation. In particular, infrared thermography of cells during cycling shows that the hottest spot in the cell initially lies near the positive tab and progresses towards the centre as discharge progresses. These findings are consistent across 18650 and pouch cells^{78,79} (Fig. 4c). Additionally, infrared can be applied to demonstrate the orientation effect due to the gravity effect on large-pouch cell ageing and thermal behaviour. The cells aged in a flat orientation retained higher capacity than those aged in a rotated orientation, which guides that the same orientation cells should be packed together for the secondary application to prevent the non-uniform ageing behaviour⁸⁰. Precise infrared thermography measurements require costly infrared cameras and control of the environments, but the technique offers non-contact, rapid and versatile temperature measurements, making it a valuable option for battery end-of-life characterization.

Acoustic measurements offer non-destructive, high-resolution and real-time inspection capabilities. For instance, acoustic imaging was found effective in evaluating electrolyte consumption and gas formation in a NMC532/graphite pouch cell³⁹. Ultrasonic imaging reveals that the cells are no longer well-wetted after 2 years of testing at 55 °C, which shows the real end of life of the battery. Additionally, spatially resolved operando acoustic scanning of commercial pouch cells with a scanning time of only 2 min has been realized. In this custom design, the rate of electrolyte consumption during cycling along with spatial variations in interfacial roughening could be observed (Fig. 4d, left).

Metallic lithium diagnostics

Monitoring the amount of metallic lithium in the battery is crucial for the safety of used batteries. In commercial lithium-ion batteries, lithium plating occurs when the battery is cycled at a high C-rate or low temperature. It was previously demonstrated that lithium plating is the key reason for the increased risk of battery thermal runaway after ageing⁸¹. In particular, nanosized lithium metal and dendrite morphology formed upon ageing can lead to internal short circuits, electrolyte decomposition and exothermic reactions that can cause uncontrolled heat generation and escalating feedback mechanisms that intensify the reaction rates.

In several studies, non-destructive in situ nuclear magnetic resonance has been used to quantify the inactive metallic lithium in LiFePO₄/anode-free cells and observe the corrosion behaviour of the lithium metal during storage⁸². Another approach is to apply non-destructive electron paramagnetic resonance imaging to monitor the uniformity of lithium plating and its influence on the evolution of the inactive metallic lithium in LiCoO₂/anode-free pouch cells⁸³. Acoustic characterization is a scalable, non-destructive and operando technique that can be utilized to detect lithium metal plating in commercial lithium-ion batteries. It has been demonstrated that such measurements can reveal lithium metal plating within commercial-scale LiCoO₂/graphite pouch cells during operation⁸⁴ (Fig. 4d, right). This method is instrumental in establishing a correlation between lithium metal plating, current rate and operational temperature as it provides important insights into the plating dynamics⁴⁶.

The successful screening and sorting of batteries for second life depends on the capability to understand the degradation mechanisms. Achieving high quality control is essential for the repurposing of batteries in line with safety standards. Acoustic monitoring is a particularly promising technology for battery end-of-life assessment because it provides information on wetting, gas generation and lithium plating. However, for the widespread adoption of acoustic monitoring for end-of-life characterization, quick tests that reliably assess the health of a battery with an unknown history have to be demonstrated.

Outlook

Challenges of non-destructive characterizations

Throughout their life cycle, batteries experience electrochemical and chemical side reactions such as SEI formation, electrolyte decomposition, lithium plating, dendrite formation and physical structural changes including particle cracking, fragmentation and delamination. Non-destructive characterization of batteries is essential to provide real-time monitoring of their behaviour during usage, enabling optimization, safety and longevity improvements while supporting second-life applications and recycling efforts. However, non-destructive battery characterization comes with its own set of challenges and multiple requirements are necessary for adoption:

- Cell-level monitoring: to provide more precise information about the individual cell's degradation, monitoring should be done at the cell level and not at the pack level.
- Operando evaluation: the techniques must monitor, in real time and under operational conditions, battery functional behaviour during actual use – for instance, EIS cannot be considered as an operando technique.
- Cost: price of the non-destructive characterization is a key parameter for implementation and can be an obstacle to adopting costly techniques such as X-ray tomography.
- Accuracy of measurement and sensitivity to external influences: at which precision a phenomenon can be monitored and how external factors (temperature or vibrations) can impact the measurement.
- Integration into the production line or electrical vehicles.

Considering the most suitable method requires balance between all these parameters and successfully implementing these techniques will require collaborative efforts from various fields. For conditioning and end-of-life characterization, the optimized ratio between experiment time and resolution will need to be found. Integrating sensors into electrical vehicles requires defining the number, location and

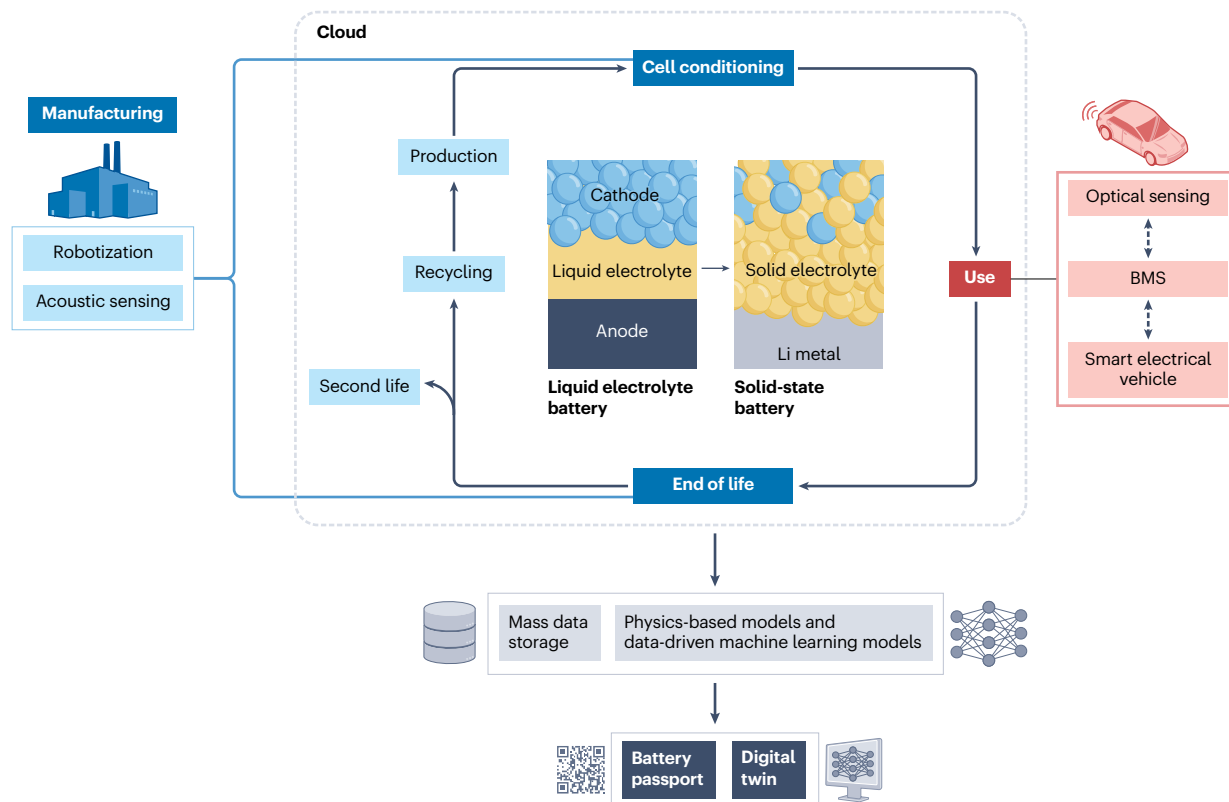


Fig. 5 | Towards battery traceability. Integration of non-destructive characterization for battery life-cycle assessment. Acoustic and optical sensing techniques are suggested to image and measure degradation phenomena occurring throughout conditioning, usage and end-of-life stages. The Integration

of these characterization data into physics-based models or data-driven machine learning models can enhance battery digital twins, facilitating the adoption of battery passports. BMS, battery management system.

positioning of these sensors around or inside the battery cell, while addressing the critical challenge of wiring and seamless integration into the system. Furthermore, careful consideration must be given to acquiring, storing and processing sensor data through the BMS. In this context, acoustic monitoring emerges as a promising technique for non-destructive battery characterization due to its versatility, cost-effectiveness and ability to assess critical battery properties such as wetting, SEI formation and dead lithium, without compromising the structural integrity of the battery. On the other hand, optical fibres are an ideal choice for battery monitoring during usage, thanks to their compact size and high sensitivity to mechanical and thermal variations⁸⁵. Figure 5 shows the complementarity of acoustic and optical fibre sensing.

Digital twin and advanced battery modelling. For short-term applications, non-destructive characterization has a key role in refining digital twins. Battery digital twins are designed to replicate the behaviour and performance of a physical battery through real-time data and predictive modelling, enabling precise monitoring and optimization of battery systems. Integrating new data will refine those models and allows for precise behaviour assessment. As machine learning grows in every domain, the convergence of this field with non-destructive characterization techniques presents a timely and promising opportunity to elevate our understanding and management of battery behaviour and

performance^{86,87}. The acquisition of realistic experimental data during battery operation has the potential to drive the development of precise machine learning and deep learning algorithms, further empowered by hardware advancements such as significant GPU improvements, enabling accurate predictions of battery degradation and remaining lifespan. Already, data-driven regression models using machine learning have been used to predict the state of charge of a battery based on FBG sensor data, achieving a supervised machine learning model accuracy of 99.95%⁸⁸. Studies using various models have verified the efficacy of sensor data in accurately predicting not only the state of charge of a battery but also its state of health^{62,89–91}.

Battery next generation. Non-destructive characterization will also have a crucial role in advancing the development and fostering the global adoption of the next generation of battery. For instance, lithium metal cells, being considered as the next-generation battery (whether liquid or solid), possess a lithium reservoir at the anode that hides performance degradation on capacity curves⁹². Furthermore, the continuous formation of highly reactive dead lithium at the anode during cycling underscores the importance of developing new characterization techniques that can monitor the health of those cells. In parallel, solid-state batteries present improved safety and higher energy density compared with conventional lithium-ion cells. However, the effects of stack pressure and chemomechanical

volume changes within solid-state batteries can significantly impact cell performance^{93,94}. Studies that use acoustic or optical sensing have already demonstrated the feasibility of characterizing the mechanical properties of these cells^{67,95}. These studies are just the starting point, and we anticipate that the evolution of the next-generation batteries will, and must, be accompanied by advancements in non-destructive characterization techniques.

Battery passport legislation. In Europe, battery life-cycle monitoring is already becoming a reality. Recent adoption of new regulations by the European Council aims to strengthen sustainability rules for batteries include the mandatory implementation of a battery passport for all electrical vehicle batteries^{96,97}. These measures have the objective to promote circular economy through end-of-life requirements and to ensure improved battery and safe working conditions, while addressing fair competition and consumer information. These initiatives encourage collaboration between academia and industry to develop new non-destructive characterization techniques for commercial cells but also for new chemistry⁹⁸.

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Author contributions

All the authors contributed substantially to discussion of the content. C.G.-M. wrote the article. All the authors reviewed and/or edited the manuscript before submission.

Competing interests

The authors have no competing interests to declare.

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