

Investigation of Multi-reaction Evolution in Lithium-ion Batteries under Fast Charging via Non-destructive Multi-sensor

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Executive Summary

Fast charging is considered as a promising protocol for raising the charging efficiency of electric vehicles. However, high currents applied to lithium-ion batteries inevitably exacerbate unwanted side reactions and accelerate the degradation. Enhanced insights into ongoing processes inside lithium-ion batteries are crucial for guiding optimization of battery manufacturing and charging protocols. In this work, the multiple reactions such as Li plating, gas generation under different levels of fast-charging protocols are comprehensively monitored and investigated through combination of multiple non-destructive sensors. Based on the proposed indicators for quantitative characterization, reversible and irreversible Li plating evolution are analyzed. The advanced ultrasonic diagnostics are utilized to conduct the analysis on gas evolution throughout the lifecycle. This work offers an in-depth and extended understanding of multiple reactions inside batteries, contributing to guiding further developments of battery fast-charging technologies.

Keywords: Electric Vehicles, Smart charging, Batteries, Battery Management System, Energy storage systems

1 Introduction

Electric vehicles (EVs) are generally considered to play a vital role in addressing climate change and energy security issues. However, their large-scale deployment is still held back by the anxiety in charging duration and range. A growing consensus around the world is that fast charging provides a strategic opportunity to release range anxiety and cut down the charging time [1,2,3,4]. Despite the potential benefits, fast-charging is faced with challenges in terms of battery durability and safety[5,6,7].

High rates applied to lithium-ion batteries unavoidably aggravate various side reactions, cut down the lifetime and even pose potential safety risks. One of the mostly discussed issues is the unwanted Li-plating, a parasitic reaction on the anodes that competes with lithium intercalation. Li plating can be triggered during fast-charging as a result of the high anode overpotential, which arises from a combination of ohmic resistance, charge transfer kinetics and mass transport limitations[8,9]. The plated lithium metal will react with electrolyte to thicken solid-electrolyte-interface (SEI) layer or isolated dead lithium, directly resulting

in loss of active Li, consumption of the electrolyte and gas evolution inside the battery. Thus, fast-charging batteries easily suffer from capacity fade and internal resistance increase [5,8]. Li plating will also have impacts on battery safety performance. Batteries with Li metal plated at anode were found to show a much lower onset temperature of exothermic reactions and greater heat generation between 50°C and 100°C compared to fresh ones [10,11]. Moreover, accumulative metallic lithium can grow dendritically and pierce separator to cause internal short circuit [12,13], which can initiate thermal runaway. Besides, other side reactions, such as gas generation, mechanical degradation, and heat generation, can also lead to degradation in capacity, power performance and potential safety hazard [14,15,16]. Hence, it is vital and essential to comprehensively understand and monitor the multiple reactions within the whole lifecycle to ensure safe and reliable fast-charging.

To date considerable efforts have been devoted to monitoring Li plating on the anodes. Examples include spectroscopy techniques[17,18,19,20], surface microscopy [21,22], thermal methods [23,24] and reference electrode method [25]. While these methods have provided valuable insights, they often require either bulky auxiliary equipment or invasive probes inside batteries, which limit their applicability within EVs during operation. Thus, there is a strong need for operando electrochemical analysis, which can detect Li plating without the help of external equipment. In general, part of the plated lithium will be reversible, which is still in electrical contact with the anode. The reversible lithium can undergo charge transfer reaction into electrolyte and then re-intercalate into anode. The whole process is known as the lithium stripping process. The stripping of plated Li will result in a minimum in the differential voltage curve during the rest or discharge period after charging, which can be an effective and convenient indicator for Li plating detection [26,27]. Although some researches have been conducted on Li plating monitoring based on differential analysis of the open circuit voltage (OCV) after charging[26,27], the evolution of the indicator of Li plating during extended fast-charge cycling has not been further tracked. There is also a lack of quantitative and systematic understanding of the Li plating process at different degradation states. Regarding gas detection, widely used methods such as gas chromatography-mass spectrometry (GC-MS) and fourier transform-infrared spectroscopy (FT-IR) can not be employed in situ [28,29]. Besides, in-situ characterization methods usually require huge modifications of batteries, which are limited for short-term gas production analysis. There is a demand for convenient and sustainable characterization methods to investigate gas-related internal changes inside batteries, which can impact battery safety. The volumetric change of batteries related to the expansion and contraction of electrode active materials during cycling is a promising metric for battery diagnostics. The mechanical expansion of electrodes translates to battery surface more rapidly than the heat transfer to battery surface, providing a real-time look into battery electrochemical and mechanical state. Recent studies have used a series of methods to explore volume changes at the electrode and battery level [30,31,32]. Most of these previous studies, however, lack in an investigation on direct correlations between battery mechanical deformation and electrochemical reactions. Besides, the evolution of battery mechanical characteristics during extended fast-charge cycling has not been extensively studied.

In this study, the multiple reactions inside the batteries under fast charging are systematically monitored and investigated through the application of multiple non-destructive sensors. Indicators for quantitative characterization of reversible and irreversible Li plating are proposed, based on which analysis on evolution of reversible and irreversible Li plating is conducted. Gas evolution analysis is performed via the advanced ultrasonic diagnostics within the whole lifetime. The evolution of battery electrochemical-thermal-mechanical characteristics during extended fast-charge cycling is also investigated. Based on the comprehensive analysis of multi-dimensional signals, multiple reactions show strong dependence on the level of fast charging.

2 Experiments

2.1 Cell cycling tests

In this work, commercial $\text{Li}(\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2})\text{O}_2$ (NCM622)/graphite pouch batteries with a nominal capacity of 1 Ah were employed. The discharging and charging cut-off voltage are specified as 3 V and 4.2 V by the manufacturer, respectively. Before the cyclic tests, a reference performance test (RPT) at 25 °C was conducted to measure the actual capacity of each tested battery. During RPT, the batteries were cycled three times under 1/2 C between 3 and 4.2 V using a constant current - constant voltage (CC-CV) charge and 1 C CC discharge protocol. The cutoff current during the CV charging step was 1/20 C. Multi-battery parallel fast charging tests were then performed for three different charging C-rates (3 C, 3.5 C and 4 C). As listed in Table 1, each cycle comprised a CC charging followed by 10 min rest, a CC-CV charging down to C/20, 30 min rest, a 1 C CC discharging down to 3V and 30 min rest before the next cycle. All cyclic tests were conducted in the thermal chamber at 25 °C to eliminate the effects of the fluctuations in ambient temperature. During the tests, electrical signals (voltage, current, electric quantity) were recorded at the time interval of 1 s using NEWARE battery cycler (CT-4008Tn-5V12A). Besides, a K type thermocouple is attached to the center of each cell surface to monitor the surface temperature per second.

Table 1: Profile for the fast charging tests

| Step no. | Step name | Duration | Condition |
|----------|--------------|----------|--|
| 1 | CC charge | | 3C (3.5C, 4C) until 4.2V |
| 2 | Rest | 10 min | |
| 3 | CCCV charge | | 3C (3.5C, 4C) until 4.2V; then 4.2V until $I < C/20$ |
| 4 | Rest | 30 min | |
| 5 | CC discharge | | 1C until 3V |
| 6 | Rest | 30 min | |

2.2 Strain measurements

In order to track the battery volume change during fast charging tests, the strain gauge was properly adhered to the center of each pouch battery surface before the cyclic tests, as illustrated in Fig.1. Please note that prior to mounting the strain gauges, the surface of each battery was cleaned up for effective adhesion of the sensors. The CC-33A adhesive from KYOWA was used to bond strain gauges, which cures at room temperature and can be used in environments of up to 80 °C. The strain gauges used in the experiments were configured in a quarter bridge configuration, i.e., each strain gage acting as one of the four resistors in a Wheatstone Bridge. An excitation voltage is provided across the bridge to conduct measurements[33,34]. Note that the strain is expressed in $\mu\epsilon$, which corresponds to a strain of 1 $\mu\text{m}/\text{m}$.

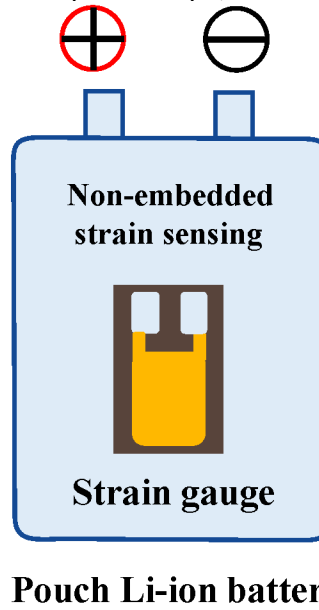


Figure 1: Experimental setup for pouch battery strain measurements

2.3 Ultrasonic scanning tests

To investigate the gassing behavior of the pouch batteries in detail, ultrasonic imaging technique is also employed. During the ultrasonic tests, a pair of ultrasonic focusing transducers were positioned one on each side of the pouch battery. Transducers and batteries were immersed in low-viscosity silicone oil, an ultrasonic coupling agent. The transducers were installed on a 2-dimensional motion system with a precision of 0.5 mm so that scanning and imaging could be performed. The driving source used in this work was a 350 V pulse signal with a pulse width of 250 ns, matching well with the ultrasonic transducers used. The schematic diagram of the path of the focused ultrasonic beam is presented as Fig.2. The waveform of the transmitted signal was recorded with a data collecting card. The peak-to-peak values (P-P value) of received transmitted waves were converted into a color scale to make the pseudo color image[35,36].

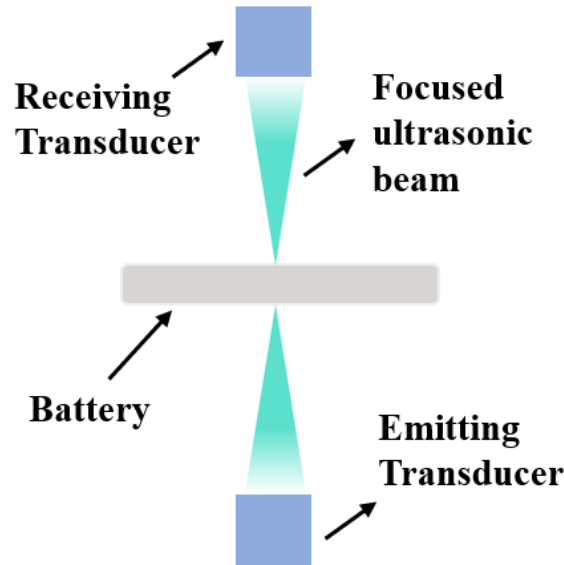


Figure 2: Illustration of the principle of the ultrasonic scanning instrument

3 Results and discussions

3.1 Analysis on evolution of reversible and irreversible Li plating via electrical signals

One complete cycling curve under each fast charging rate is shown in Fig.3, which includes charging, discharging, and relaxation processes. The duration of each charging stage varies with different fast charging rates. And the duration of relaxation I and II are fixedly set at 10 and 30 minutes for all rates.

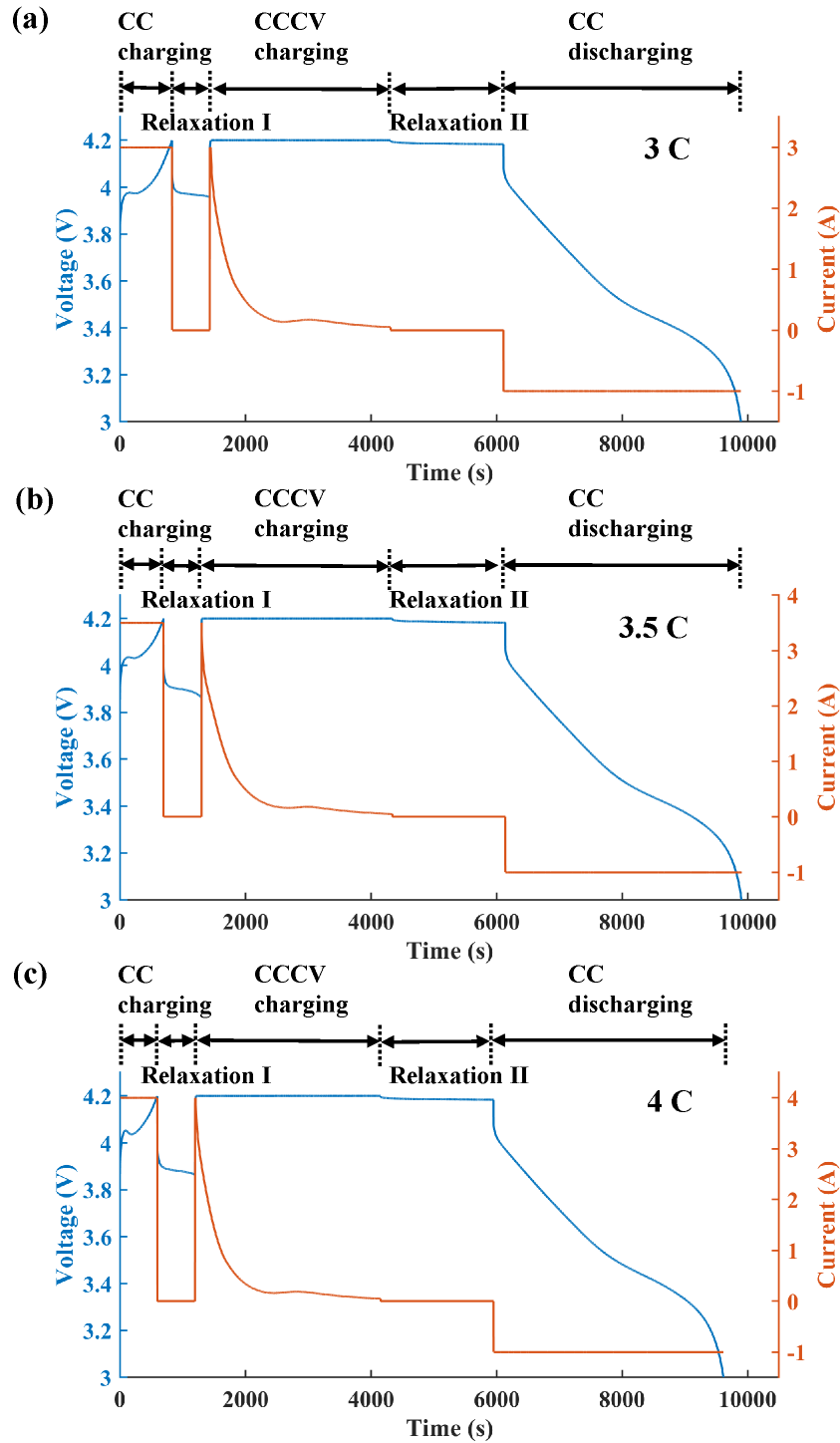


Figure 3: Voltage and current during the first cycle under different fast charging C-rates (a) 3 C (b) 3.5 C (c) 4 C

To investigate the evolution of reversible Li plating, non-destructive characterization is necessary for identifying the reversible Li plating. In this study, differential analysis of the relaxation voltage with respect to time is employed to non-destructively detect Li plating inside batteries [37, 38, 39,40]. Sequentially extract all voltage data at the stage of relaxation I during the degradation and then acquire differential voltage curves through differential processing.

Fig.4 exhibits the battery voltage ((a) (c) (e)) and differential voltage profiles ((b) (d) (f)) during the relaxation I period after different levels of fast charging. Distinct voltage plateau, which has been considered as a characteristic of reversible Li stripping after plating, can be found in the battery relaxation voltage profiles (Fig.4 (a) (c) (e)) after all charging rates in several initial cycles. Besides, according to the

literature [39, 40], the local minima in the differential voltage profile indicates the end of reversible Li stripping process. Here, define the occurrence time of the local minima in each differential voltage profile as t_{min} . Moreover, t_{min} has been reported to correlate to the fully stripping of the reversible lithium and exhibit a good linear relationship with the total amount of reversible lithium when the ambient temperature is constant [41]. Thus, t_{min} can be utilized as an in-situ quantitative indicator for the amount of cyclic reversible Li plating. It can be observed that the occurrence time of the local minima in each differential voltage becomes closer to zero, whatever the charging rate. The shape of differential voltage curve evolves during extended fast charging cycles, and the local minima of the differential voltage even disappears at the later stage of aging. In other words, the amount of the cyclic reversible Li plating gradually decreases with degradation, which results in a shorter period of time for re-intercalation of plated Li into the graphite anode during the relaxation period. It can be inferred that this drop can originate from the accumulation of loss of lithium inventory (LLI).

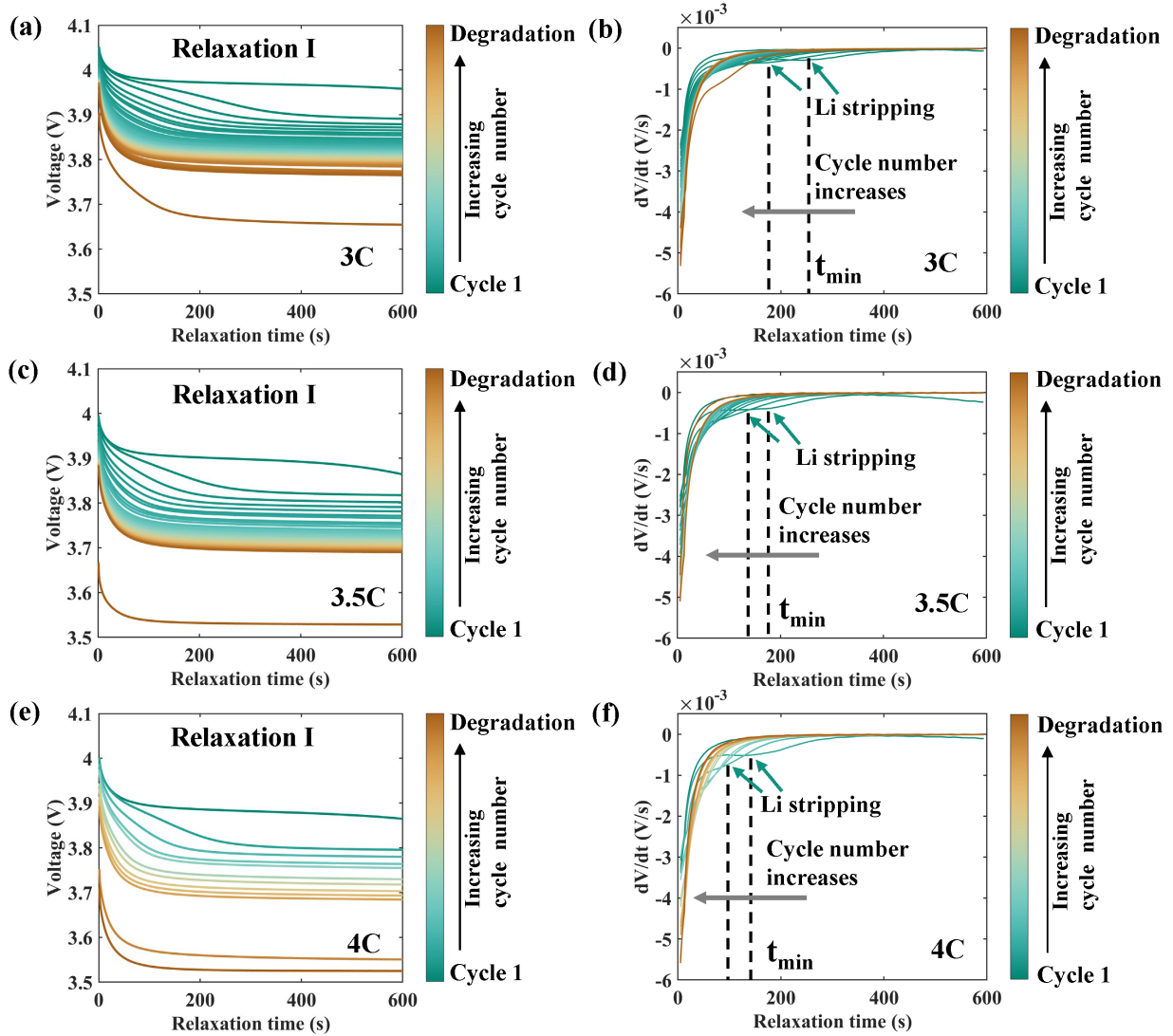


Figure 4: Voltage and differential voltages during relaxation I after fast charging

Generally, among all irreversible side reactions, irreversible Li plating on the anode significantly accelerates capacity decay, becoming the primary failure mode of batteries under fast charging [42]. Furthermore, though as another key side reaction, the growth of SEI film is positively correlated with the ambient temperature and exposure time and far from severe under room temperature [43,44]. Therefore, it is reasonable to assume that irreversible Li plating contributes mostly to capacity loss during the early fast charging cycles under 25°C. Extract cyclic capacity loss as one of indicators of the cyclic irreversible Li plating amount. Meantime, coulombic efficiency (CE, the ratio of discharge capacity over charge capacity in a battery) is also used as an indicator of the irreversible Li plating amount in each cycle since its decline

directly results from the consumption of active lithium ions[45].

Fig.5 displays the cyclic capacity loss and CE evolution with t_{min} during the early cycles. Specifically, with cycle number increasing, cyclic capacity loss decreases as t_{min} does and CE rises up. It is clear that cyclic irreversible Li plating amount gradually drops during extended fast-charge cycling as reversible Li plating amount does, which can be attributed to a reduction in the accessible Li inventory within the cell, which reduces the driving force for continued plating in later cycles. Moreover, both indicators of irreversible Li plating amount exhibit linear relationships with the that of amount of reversible Li plating under three fast charging conditions, offering a new quantitative understanding of Li plating reactions.

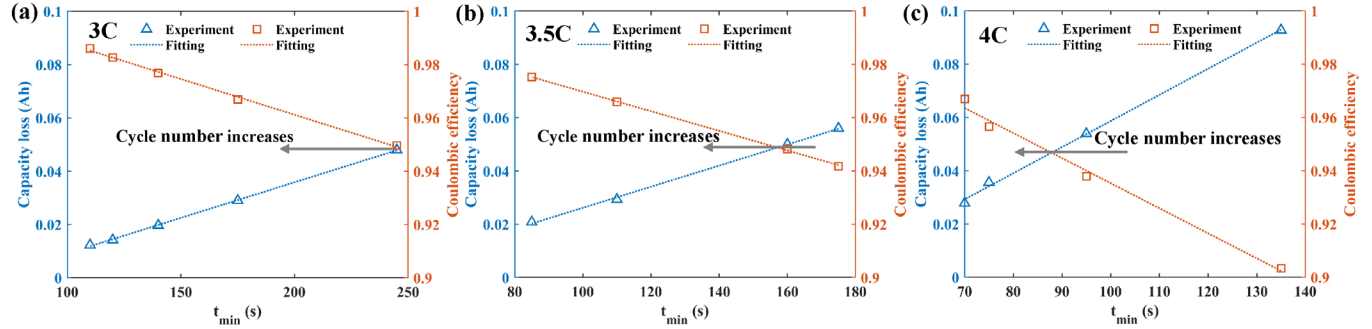


Figure 5: Capacity loss and coulomb efficiency evolution with t_{min} (a) 3 C (b) 3.5 C (c) 4 C

3.2 Analysis on gas evolution via ultrasonic signals

An ultrasound wave is emitted from the focusing transducer on the one side, penetrates the cell and is received by another transducer on the other side. The ultrasonic transmission waveform at each position was recorded via progressive scanning. The P-P value of the ultrasonic transmission wave was transformed into colors from blue to green under the given scale (between 0 and 1250 mV) to create ultrasonic transmission images of the pouch battery. Fig.6 shows the ultrasonic transmission images of the NMC622/graphite pouch cells at different degradation stages under 3 charging rates.

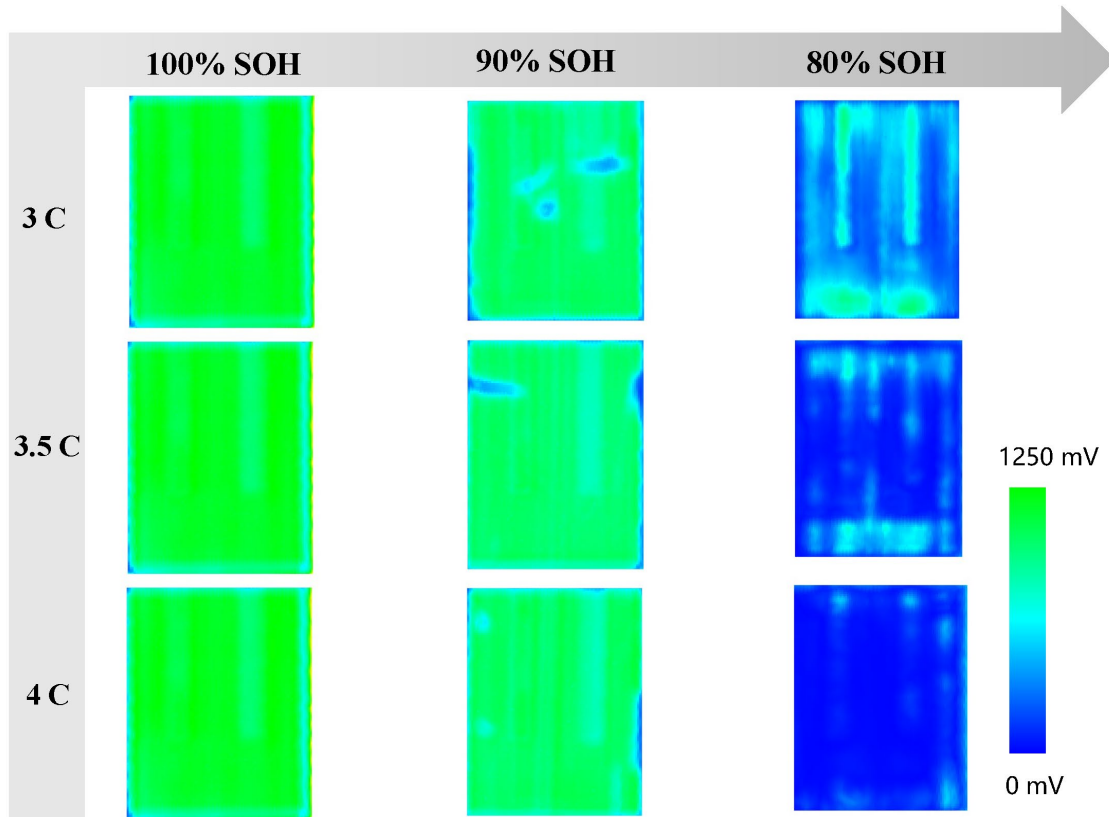


Figure 6: Comparison of ultrasonic diagnostic results at different degradation stages for gas evolution analysis

Ultrasonic waves are very sensitive to gas, even in small amounts[36]. When ultrasonic waves pass through the interface of two different media, the transmission and reflection ratios can be calculated by

Equations (1) and (2), respectively, where Z_{a1} and Z_{a2} are the acoustic impedances of the incident and emergent media, respectively[46].

$$\text{Transmission rate} = \frac{4Z_{a1} \cdot Z_{a2}}{(Z_{a1} + Z_{a2})^2} \quad (1)$$

$$\text{Reflection rate} = \frac{(Z_{a2} - Z_{a1})^2}{(Z_{a1} + Z_{a2})^2} \quad (2)$$

According to Equations (1) and (2), similar Z_{a1} and Z_{a2} lead to a higher ratio of transmission while different Z_{a1} and Z_{a2} lead to a higher ratio of reflection. Generally, the acoustic impedance of gas is about 0.0004 MRayl and the acoustic impedance of liquid and solid materials range from 1 to 50 MRayl. Such a great difference indicates that ultrasonic waves could lose more than 99.99% of transmitted intensity at the gas/solid interfaces. Thus, a small amount of gas can remarkably lower the ultrasonic transmissivity. The early stages of gassing and even the gassing onset cycle can be non-destructively monitored utilizing the ultrasonic technique[36].

For 100% state of health (SOH), almost all areas of the battery exhibit green as shown in Fig.6, indicating that there is little gas inside the battery. When normalized capacity fades to 90%, each battery starts to exhibit light blue and even deep blue in several scattered areas. These blue areas directly demonstrate that gas begins to form. Since electrolyte decomposition and the growth of SEI film are far from severe in early cycles at the temperature of 25°C, it can be inferred that the gas mostly results from the secondary reactions of deposited Li under fast charging. Further, the ultrasonic transmission images showing large area of blue at 80% SOH indicate that plated Li continually reacts with electrolyte and consequently leads to gas generation and accumulation. This side reaction directly results in irreversible LLI and capacity loss. Besides, it is worth noting that under harsh charging circumstances like higher C-rates, both Li plating and the secondary reaction of deposited Li with the electrolyte progress more severely. As one of the most terrible safety issue in Li-ion batteries, Li plating generates a series of parasitic reactions with conventional organic liquid electrolytes, which may be the fuse for thermal runaway. Hence, with the advantage of exploring the evolution process of gas generation, the ultrasonic diagnosis will be vital for guiding safe fast-charging procedures in the future.

3.3 Analysis on evolution of electrochemical-thermal-mechanical characteristics

Electrode materials naturally expand/shrink during battery cycling. When a battery is charged, the graphite anode expands ~13.1% in volume while the lithium nickel manganese cobalt oxide (NMC) cathode materials contract for 3% in volume[47]. Since deformation of graphite anode significantly exceeds that of NMC cathode, the battery strain is inherently dominated by the graphite anode and can potentially reflect the corresponding electrochemical processes[47]. Meantime, battery operando temperature rise is strongly influenced by the charging protocol, thus it should be monitored for thermal safety management during high-rate applications.

Fig.7 exhibits battery strain, voltage and temperature profiles (including charging, discharging, and relaxation processes) of three fast-charging protocols at different degradation states. For all batteries, the strain, voltage and temperature increase during the initial CC charging, and reach the local maxima around the end of CC period. The local maximum of strain can be attributed to Li plating and thermal expansion, as both strain and temperature drop down during the following relaxation. Through this relaxation, the processes of Li stripping trigger the plateaus in the voltage profiles and partly contribute to the strain relaxation. Local maxima can also be observed in strain curves at the second charging stage. These local maxima are mainly related with the sudden temperature rise, since CV charging mode almost covers this period and there is little Li plating. Interestingly, the strain remains stable or decreases slightly despite continuous Li intercalation during a period of CV charging, and the same agrees well with the results reported in the literatures[48,49]. The reason is that graphite undergoes a phase transformation without volume expansion from LiC_{24} phase (Stage 2L) to LiC_{12} phase (Stage 2) [50], while NMC cathode experiences a volume contraction simultaneously [51]. Similarly, distinct plateaus can be observed in strain profiles during CC discharging between 6000 and 8000 s, where battery strain drops little or even increases slightly. Graphite does not contract through the phase transformation period, but meanwhile the cathode material swells with a decrease in battery state of charge (SOC). For both Li intercalation and deintercalation processes, there are also plateaus in temperature profiles during the phase transformation period of graphite anode. Furthermore, all batteries differ from each other in external characteristics, especially in the mechanical characteristic as shown in Fig.7. It reflects that internal multiple reactions show strong dependence on the level of fast charging.

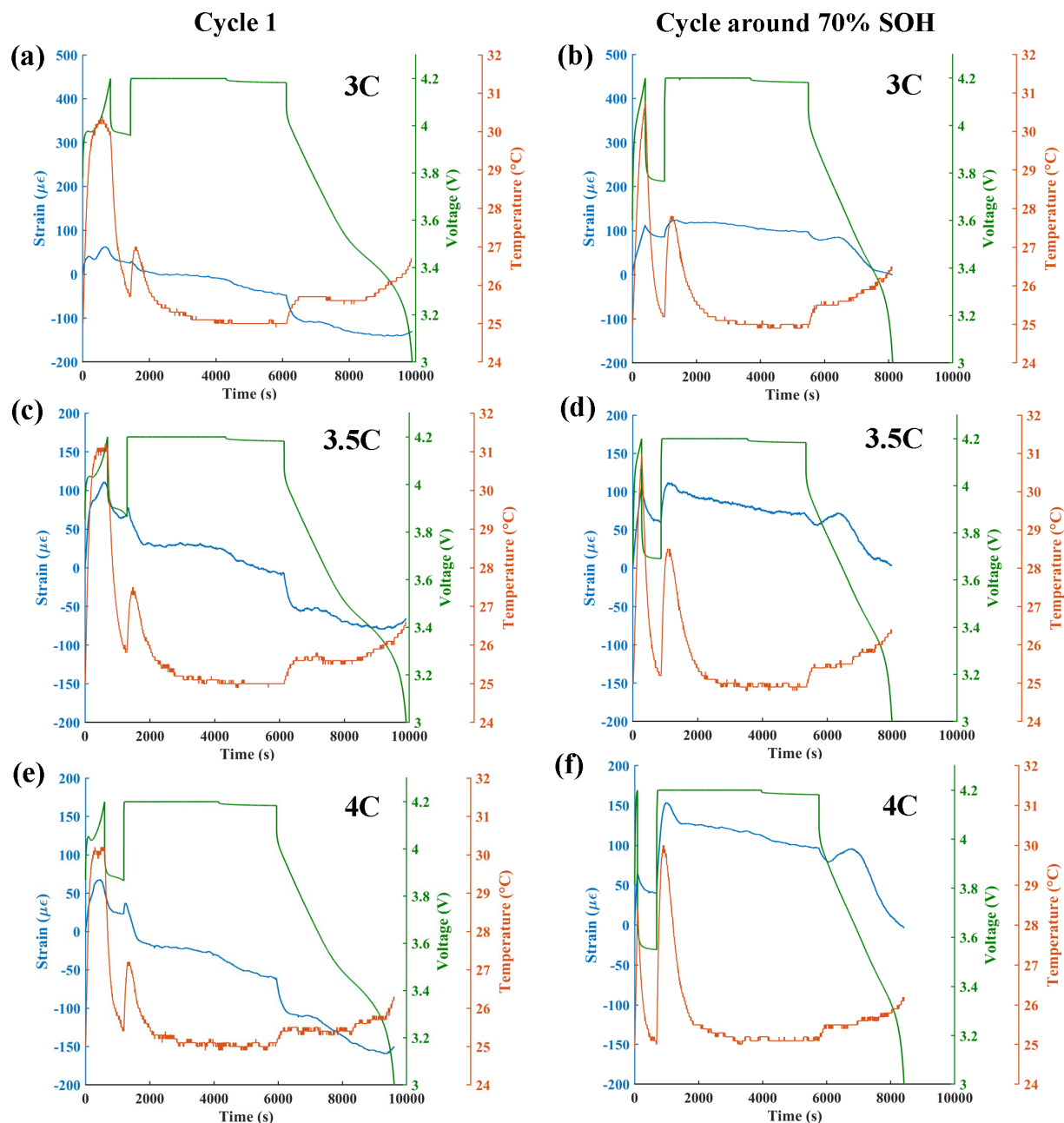


Figure 7: Voltage, temperature and strain evolution under different levels of fast-charging protocols

With degradation, battery strain, voltage and temperature increase reach the local maxima earlier during the initial CC charging. The shortening of CC charging stage resulting from the increase in internal resistance is mainly responsible for the evolution. Meantime, there are distinct changes in local maxima of temperature profiles, which is related to both resistance and electrochemical processes. Nevertheless, among the three signals, strain profile shows the most significant changes between different stages due to the internal multiple reactions triggered by fast charging. Therefore, the mechanical strain shows significant potential for guiding the optimization of fast charging protocols during the whole lifecycle with the ability to in-situ monitor multi-reaction evolution.

4 Conclusion

In this work, multiple reactions inside cells under fast charging are comprehensively investigated through combination of multiple non-destructive sensors. With the proposed indicators, reversible and irreversible Li plating evolution are quantitatively analyzed. Gas evolution analysis is performed throughout the lifecycle based on the advanced ultrasonic tests. Analysis on evolution of battery

electrochemical-thermal-mechanical characteristics is performed. Internal multiple reactions exhibit strong dependence on the level of fast charging as evidenced by analysis of multi-dimensional signals. This work offers a multidimensional view to explore internal electrochemical reactions and even the health of batteries and hence enables developing a new battery management framework. And our future research will focus on the optimization of charging procedures for safe and health-care fast-charging protocols during the whole lifecycle with the assistance of multiple non-destructive sensors.

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