In Situ Mapping of State of Charge of Lithium-ion Battery by Quasi-Static Components of Guided Wave

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ABSTRACT

Accurately probing the interaction mechanism between acoustic signals and the state of charge (SOC) in lithium-ion batteries (LIBs) is challenging. This study introduces a novel method based on the quasi-static component of nonlinear guided waves (QSC-NGW) to reveal the evolution of electrode materials during LIB charging and discharging. Utilizing an empirical formula and phase-reversal technique, the method successfully captures high signal-to-noise ratio (SNR) QSC-NGW signals within LIBs. This approach effectively establishes a mapping relationship between the time-frequency domain eigenparameters of the signal and the battery's SOC. Subsequent in-situ characterization techniques, including scanning electron microscopy (SEM), enabled the observation of lithium-ion deintercalation processes within the electrodes. This facilitated precise identification of electrode surface structural evolution and direct extraction of mechanical property variations. These findings corroborate the efficacy of the QSC-NGW method. Compared to conventional linear acoustic techniques for extracting battery state parameters, the QSC-NGW method exhibits superior directivity and strong interpretability. Crucially, it eliminates the need for prior prediction of internal battery material parameters or the construction of complex guided wave dispersion models. The integration of the QSC-NGW method with in-situ characterization effectively captures SOC changes during cycling, demonstrating its broad application potential.

INTRODUCTION

Lithium-ion batteries (LIBs) have emerged as the predominant energy storage solution for new energy vehicles and portable electronic devices, owing to their superior specific energy density, exceptional cycle life, and wide operational temperature tolerance [1-3]. The strategic design of nickel-cobalt-manganese (NCM) ternary cathode materials, based on the synergistic effects of transition metal components, has significantly enhanced lattice stability and ion transport efficiency within electrode structures [4]. Nevertheless, establishing dynamic correlations between state of charge (SOC) monitoring and material evolution processes remains a critical challenge in current ternary system research. Conventional characterization techniques can be categorized into destructive methods (including structural analysis via atom probe

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tomography (APT) [5] and compositional characterization through X-ray photoelectron spectroscopy (XPS) [6]) and non-destructive approaches (encompassing electrochemical testing [7] and magnetic resonance techniques [8]). Although these methodologies provide multidimensional data for deciphering battery degradation mechanisms, they generally suffer from limitations associated with sophisticated instrumentation requirements and complex operational procedures. Consequently, the development of cost-effective, time-efficient testing methods with well-defined mechanistic interpretations presents substantial research significance.

Acoustic nondestructive characterization techniques have recently emerged as a cutting-edge research frontier in LIB state monitoring, driven by their real-time response characteristics and multiscale detection capabilities [9]. This methodological domain has witnessed continuous expansion of technical pathways over the past decade [10-15]. Current investigations predominantly focus on linear acoustic theoretical frameworks, which rely on the fundamental assumption of linear constitutive relationships derived from elastic deformation in homogeneous media. However, practical applications encounter inherent complexities arising from structural heterogeneities within battery systems, including multiphase composite media and lattice distortion effects, which inevitably induce nonlinear response phenomena during stress wave propagation. Notably, existing acoustic testing systems —whether employing linear or nonlinear theoretical models—face a fundamental dilemma in frequency selection: High-frequency acoustic waves demonstrate superior sensitivity for microdefect identification but suffer from substantial attenuation, while low-frequency signals exhibit enhanced penetration depth at the expense of compromised micron-scale resolution. This fundamental limitation poses a significant barrier to the engineering implementation of acoustic testing methodologies.

Addressing the technical constraints of acoustic nondestructive testing in LIBs, the quasi-static component of nonlinear guided wave (QSC-NGW) testing method has emerged as a novel research direction for material property assessment, capitalizing on its directional propagation characteristics and explainability [16-18]. The physical foundation of this technique lies in the nonlinear wave-matter interaction phenomenon: When guided waves with specific frequency components propagate through nonlinear media, they generate quasi-static components characterized by temporal width synchronization with incident wave envelopes and near-zero carrier frequencies. To validate the innovative potential of this methodology in SOC monitoring and electrode state evaluation, this study pioneers the establishment of a synergistic analysis framework integrating QSC-NGW testing with in situ characterization. This approach systematically elucidates the coupled evolution patterns between SOC variation and electrode structural transformation in nickel-cobalt-manganese lithium-ion battery (NCM-LIB) systems during cycling. This paper consists of three main parts: (I) The methodology describes the generation of QSC-NGW and the experimental system; (II) The acoustic experimental results describe the evolution of QSC-NGW during the NCM-LIB cycling process; (III) The in situ characterization describes the evolution of the electrode during the cycling process.

METHODOLOGY

QSC-NGW generation

As the primary wave propagates through a medium, it induces a static pulse component characterized by zero carrier frequency and temporal duration matching the envelope width of the primary wave. The structural configuration of NCM-LIBs cells resembles plate-like systems, enabling their characterization via nonlinear mixing theory. According to this theoretical framework, the amplitude of nonlinear mixing wave can be mathematically described as follows:

$$A_m = \frac{(f_{vol}^{-} + f_{surf}^{-})}{4P_{mm}} \frac{\sin\left[\left(k_a \pm k_b - k_n\right)x/2\right]}{\left(k_a \pm k_b - k_n\right)/2} \tag{1}$$

where A_m represents the amplitude of the nonlinear hybrid wave, f_{vol} corresponds to volumetric power flux, and f_{surf} indicates surface-bound power flux. Here, P_{mm} defines the mean power flux density, while k_a and k_b denote wave numbers of primary waves, with k_n specifying the wave number at a target angular frequency. The emergence of cumulative mixing harmonics requires simultaneous fulfillment of the phase matching criterion ($k_n = k_a \pm k_b$) and nonzero energy flux condition ($f_{vol} + f_{surf} \neq 0$). Notably, the second-order difference-frequency component asymptotically approaches zero under such configurations, leading to the following parametric expression for hybrid harmonic amplitude:

$$A_m = \frac{(f_{vol}^{-} + f_{surf}^{-})}{4P_{mm}} x \tag{2}$$

Conversely, the quasi-static constituent manifests as a self-interaction-induced difference-frequency component that inherently fulfills phase synchronization requirements.

Experimental system construction

To effectively capture the dynamic characteristics of QSC-NGW in NCM-LIBs, we developed a dedicated testing platform. This configuration integrates three core subsystems: (1) An integrated control module synchronizing signal generation with charge-discharge cycling protocols, (2) A high-speed data acquisition unit employing a digital oscilloscope with 250 MS/s sampling rate for precise QSC-NGW waveform characterization, and (3) The transducer array comprises a 5 MHz broadband piezoelectric transducer and a 0.25 MHz low-frequency resonant probe, strategically positioned 75 mm apart on the NCM-LIB surface. Interface coupling optimization was achieved using a commercial acoustic couplant (Olympus SWC-2). A schematic diagram of the NCM-LIB sample used in the experiment is shown in Figure 1.

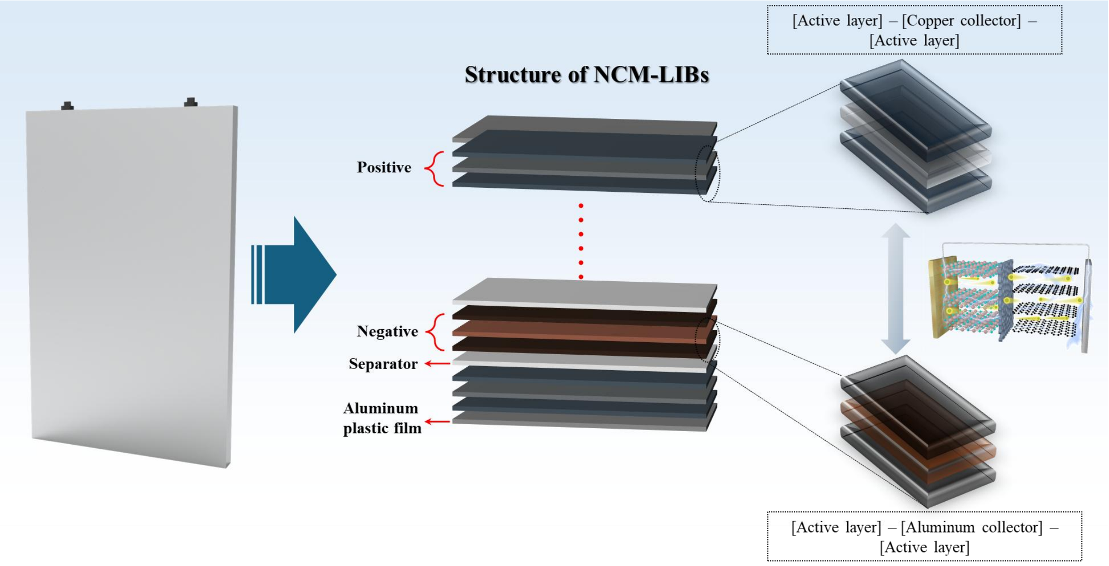

Figure 1: Schematic diagram of NCM-LIB.

EXPERIMENTAL RESULTS ON THE QSC-NGW OF NCM-LIBS

To obtain the propagation characteristics of QSC-NGW during the LIB cycling process, we extracted the QSC-NGW time domain signals from NCM-LIBs at various SOC, as illustrated in Figure 2. The black solid line represents the QSC-NGW signal received at full charge, and the blue solid line represents the signal received at empty charge. As the discharge process progresses, the wave packet shifts to the right. This indicates that the QSC-NGW duration increases during discharge. Concurrently, we observed that the magnitude first decreases and then increases.

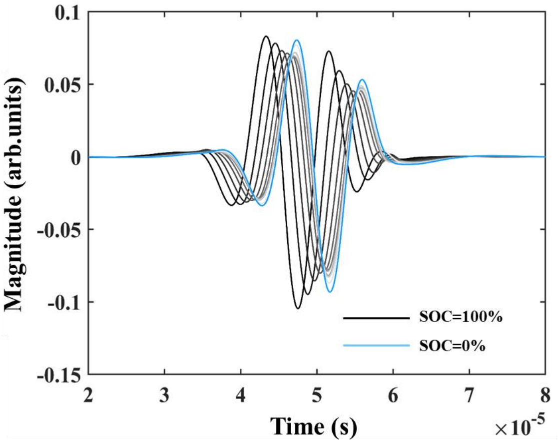

Figure 2: QSC-NGW signals received under different SOC.

Subsequently, we compared the results of the QSC-NGW method with those of the conventional guided wave method and the quasi-static component of nonlinear bulk wave method, as shown in Figures 3 and 4. It can be seen that the QSC-NGW method has better consistency and stability than other methods.

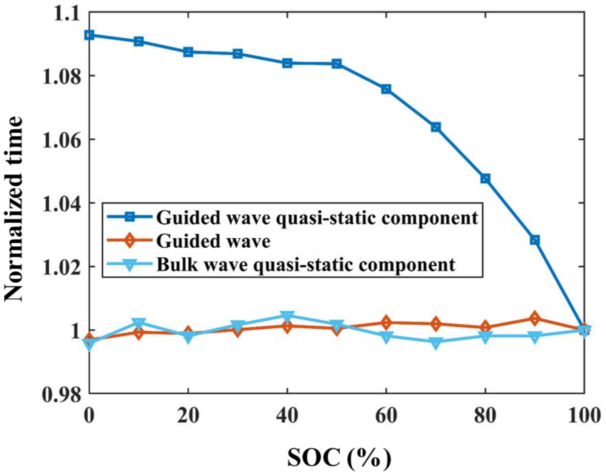

Figure 3: Comparison of different ultrasonic testing methods on time.

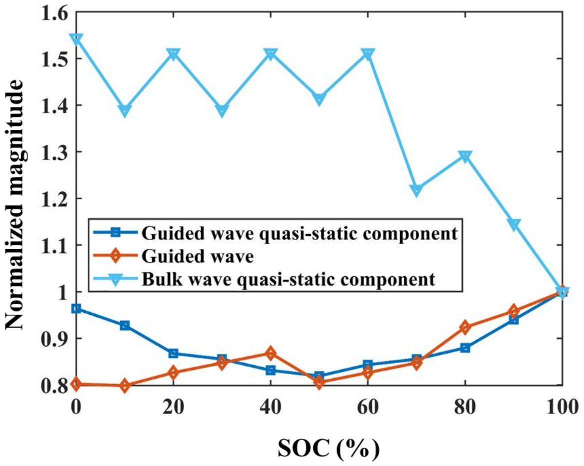

Figure 4: Comparison of different ultrasonic testing methods on magnitude.

EXPERIMENTAL RESULTS ON THE IN SITU CHARACTERIZATION

In the results of the QSC-NGW acoustic experiment, we discovered an interesting phenomenon in addition to the time exhibiting a relatively linear trend with changes in the SOC: the amplitude first decreased and then increased. Under consistent coupling conditions, Ladpli speculated that this phenomenon was caused by changes in lattice strain resulting from the insertion and extraction of lithium ions within the electrode [19]. To investigate this phenomenon, we characterized the cathode material in situ at different SOC. The SEM and AFM results are shown in Figures 5 and 6, respectively.

When NCM-LIBs are highly charged, lithium ions are extracted from the cathode. At this point, the NCM cathode surface is relatively porous, and the particles are close together. As the SOC decreases, the porosity of the NCM cathode surface gradually decreases while the distance between particles gradually increases due to lithium ion insertion. Concurrently, as the SOC decreases, the elastic modulus of the NCM electrode surface increases slightly due to lithium ion insertion.

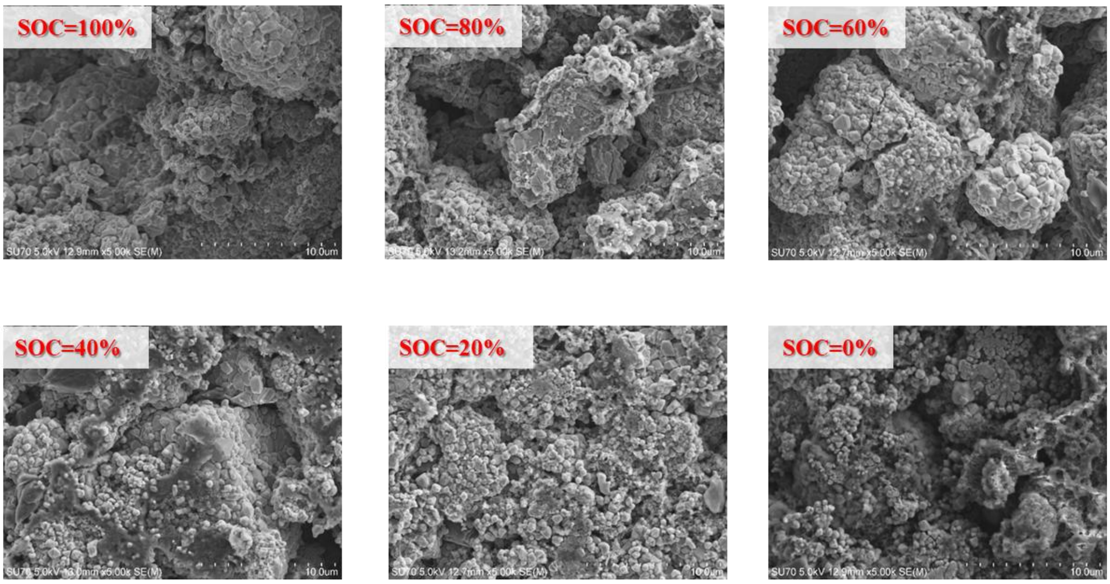

Figure 5: SEM results of NCM cathode under different SOC.

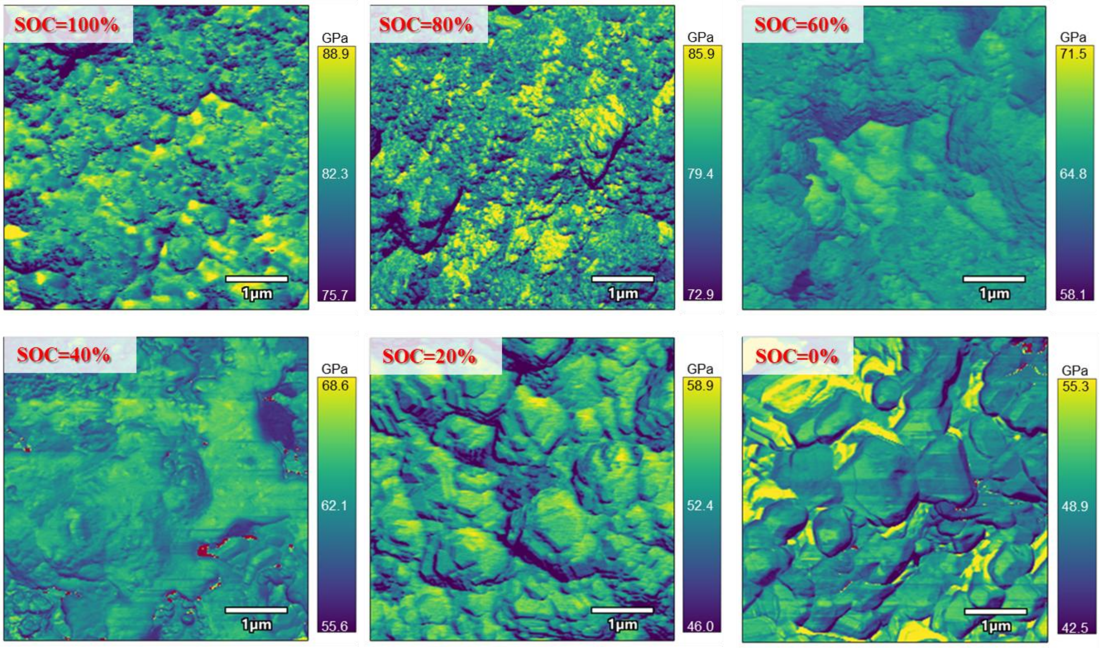

Figure 6: AFM results of NCM cathode under different SOC.

CONCLUSION

This study introduces a novel QSC-NGW method for SOC assessment in NCM-LIBs. Leveraging its strong directivity, we establish mappings between acoustic parameters, SOC, and cathode micro-properties. In-situ analyses reveal QSC-NGW time variations correlate with electrode density/roughness evolution, while magnitude changes reflect lattice contraction/expansion linked to NCM structural shifts. Compared to linear guided/bulk waves and nonlinear bulk wave quasi-static components, QSC-NGW demonstrates superior directivity and interpretability in attenuative battery media. The method simplifies signal characterization and provides an in-situ ultrasonic assessment framework. Future work should extend its applicability across NCM-LIB service conditions and other battery architectures.

ACKNOWLEDGEMENTS

This work was supported by the National Natural Science Foundation of China (Grants Nos.: 12425412 and 12134002). It also supported by the project of National Science Foundation of Fujian Province of China (Grant No.:2022J06007) and the "Fujian Province Outstanding Postdoctoral Funding Program".

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