Optimization of Electrical Current for Operando Neutron Depth Profiling: A Preliminary Study

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1. Introduction

With increasing demand for safe, reliable, and high-performance batteries in microelectronic devices, all-solid-state batteries (ASSBs) have recently gained attention as a promising alternative to traditional lithium-ion batteries. Because ASSBs use solid electrolytes instead of liquid electrolytes, they have better fire resistance [1,2] and can achieve higher voltages [2]. However, repeated cycling often leads to the formation of lithium immobilization layers at the electrolyte–anode interface. In some cases, this also causes anode materials to become trapped within the solid electrolyte, resulting in capacity fade or internal short circuits.

Various techniques, including XPS, SEM, and SIMS, have been employed to investigate these interface-related issues; however, they typically require destructive sample preparation, potentially altering the battery's pristine state. Such destructive sample preparation methods may distort interfacial reactions and introduce unintended contaminants, complicating the accurate observation of intrinsic electrochemical behaviors [3]. In contrast, neutron depth profiling (NDP) is a non-destructive method that can probe lithium distribution in batteries while preserving their pristine electrochemical state.

In this preliminary work, we explore battery degradation phenomena in lithium-ion cells via battery cycling to inform the design of an operando NDP system. Our results will guide the selection of optimal current conditions for operando NDP measurements.

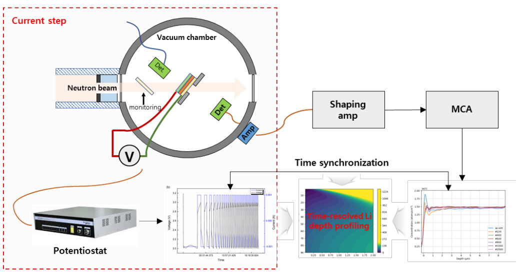


Fig. 1. Schematic figure of KAERI *operando* NDP system.

2. Methods

Given the limited commercial availability of ASSBs, we utilized conventional liquid-electrolyte lithium-ion cells as surrogate models. These cells undergo similar degradation processes under cycling, enabling us to investigate key failure modes relevant to ASSBs [4]. We specifically chose lithium-ion systems because NDP is uniquely suited to track lithium migration within the cell.

Each cell was connected to a potentiostat (WPG100E, WonATech), and cycled while monitoring voltage and current in real time. Cycling protocols varied by battery type, but all were subjected to at least 30 charge–discharge cycles (Fig. 2). Table 1 summarizes the key specifications and cycling conditions for each battery.

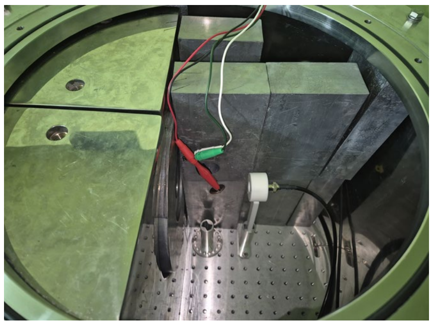


Fig. 2. Cycling environment of Li-ion battery. The battery is positioned in the NDP vacuum chamber.

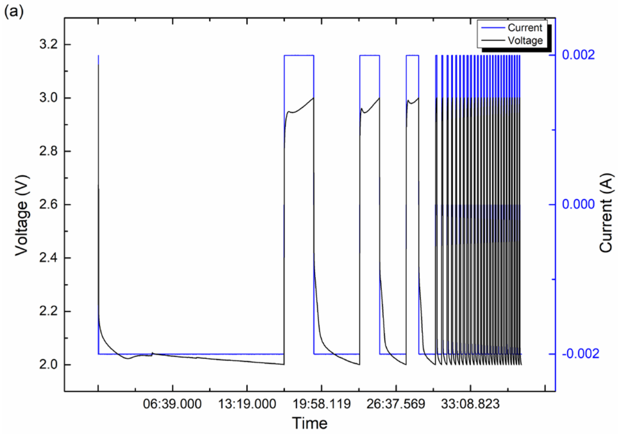
Table I: Specification and cycling conditions of prepared batteries.

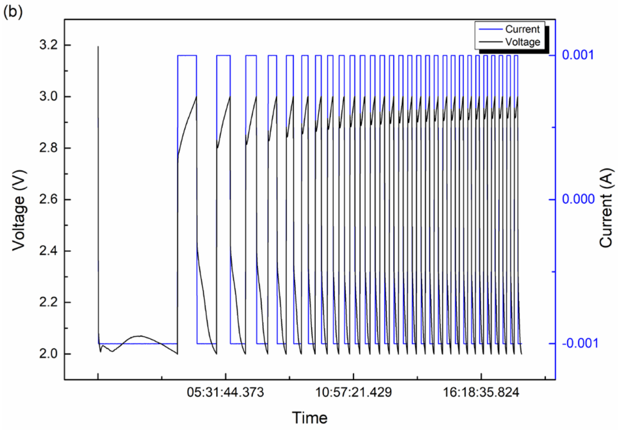
Brand	MAXELL	SANYO	Panasonic
Name	ML1220-T10	ML1220-TT2	VL2330
Nominal voltage	3 V	3 V	3 V
Nominal capacity	18 mAh	14 mAh	50 mAh
Cathode Material	Manganese Dioxide	Manganese Dioxide	LiV_2O_5
Current	2 mA	1 mA	5 mA

Cutoff Voltage	3 V/2 V	3 V/2 V	3 V/2 V

3. Result & Discussion

As shown in Fig. 3, battery capacities exhibited marked degradation over repeated cycles, evidenced by progressively shorter charge–discharge intervals. Higher current densities accelerated capacity loss more dramatically than lower currents, as illustrated by the discharge capacity curves normalized to the initial capacity (Fig. 4). Notably, the most significant reduction in capacity was observed during the initial few cycles. For instance, the cell cycled at 5 mA retained only 1.81% of its initial capacity after just two cycles, whereas those cycled at lower currents (2 mA and 1 mA) retained approximately 24% under identical conditions.





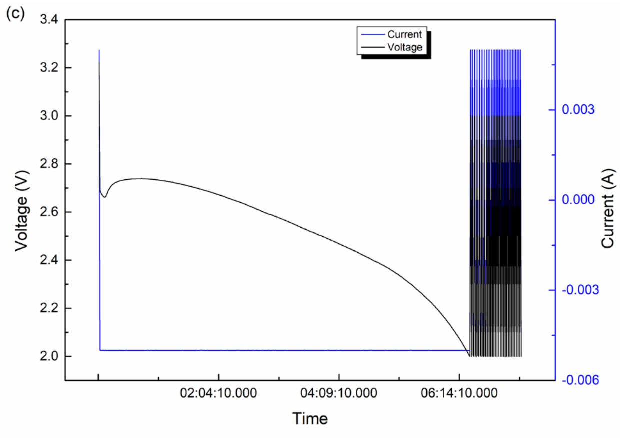


Fig. 3. Voltage and current profiles of a) ML1220-T10, b) ML1220-TT2, c) VL2330.

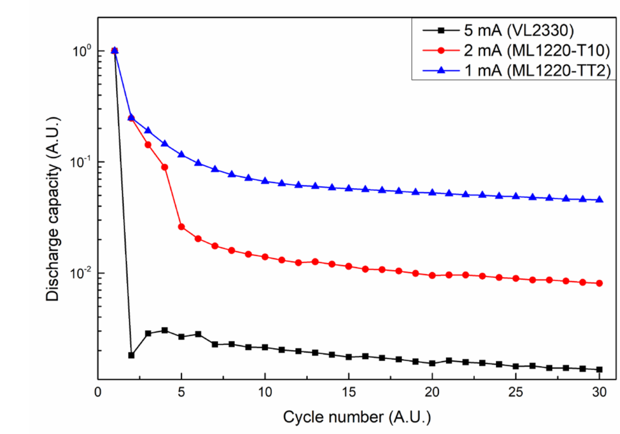


Fig. 4. Discharge capacity evolution of a battery cycled under different current conditions.

These findings underscore the importance of selecting appropriate current densities for operando NDP analysis. While higher currents can reveal degradation more rapidly, they may obscure subtler trends; conversely, lower currents provide clearer insights into gradual changes but demand longer measurement periods. Indeed, previous study on ASSB using operando NDP employed currents below 1 mA, enabling longer cycling and more detailed degradation analysis [5]. In light of these considerations, we will leverage our present results to refine the current protocols for our planned operando NDP system.

4. Conclusions

In this preliminary study, we employed battery cycling to investigate how different cycling currents influence the degradation of lithium-ion batteries. Our findings underscore that careful selection of current density is essential for precisely characterizing degradation mechanisms using operando NDP. Building on these insights, we are developing an operando NDP system that will simultaneously record NDP spectra and electrochemical data for a more comprehensive understanding of battery aging phenomena.

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