

Neutron Depth Profiling for Investigating Lithium Distribution in Thin Film Li-ion Cells

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1. Introduction

There has been the increasing emergence of micro electronic devices such as smart watches, rings, and wireless earphones. Along with this phenomenon, demand for micro Li-ion cells has also increased. Due to the small size of micro Li-ion cells, they must have high energy density in order to achieve high power capacity. Although much progress has been made in developing battery components(e.g., electrolytes, electrodes) to achieve high energy density, improvements to the cell themselves have been less significant [1]. For example, issues such as the instability of cell interfaces affect not only cell capacity but also safety through various side reactions that occur at the interface. In order to analyze various side reactions occurring in Li-ion cells(e.g., dendrite growth, solid-electrolyte interphase) in bulk, non-destructive analysis techniques should be utilized. Among them, Neutron Depth Profiling (NDP) has recently been actively studied due to its ability to analyze the distribution of Li within cell [2-3]. In this study, we investigated whether it is feasible to analyze the Li distribution in thin-film Li-ion cells using a KAERI-NDP system.

2. Methods

2.1 Neutron Depth Profiling (NDP)

NDP is a for quantitatively analyzing the concentration distribution of specific light elements as a function of depth. When a neutron induces a nuclear reaction with a light element, charged particles are emitted with specific, well-defined initial kinetic energies. As these particles travel through the material, they continuously lose energy. By measuring the final residual energy of the particles that exit the sample, the depth from which they originated can be determined. The most common elements analyzed by NDP are boron and lithium. Their reaction with neutrons is as follows:

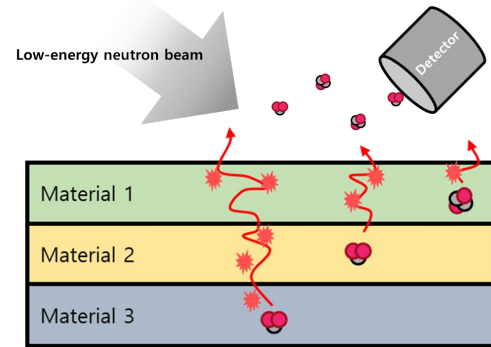
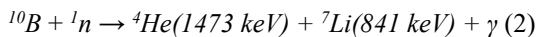
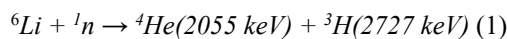


Fig. 1. A schematic figure of the principles of NDP analysis.

2.2 Thin-Film Li-ion cell Configuration

We promoted cooperation with ‘Electronic and Hybrid Materials Research Center’ at KIST in order to analyze cells that are actually being researched and developed. The cell provided by KIST is a thin-film type Li-ion cell. The cell was made by depositing cell components onto a glass plate. The detailed specification for the cell is presented in Figure 2. Based on the information provided by KIST, the total thickness of the sample is 1.472 μm .

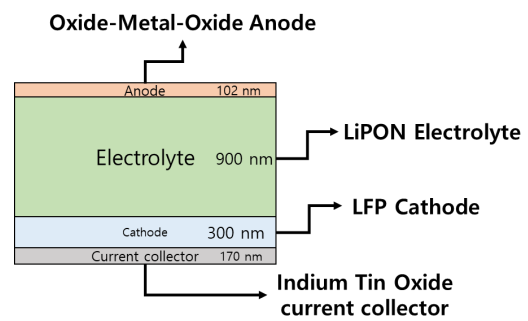


Fig. 2. Component materials and thickness of cell.

2.3 Experimental setup

In our NDP analysis, we used a vacuum chamber maintained at approximately 10^{-2} mbar for charged particle detection. Thin boron film and two ion-implanted Si detectors, which are for NDP measurement and neutron flux monitoring, are placed inside the chamber. Detectors have a 150 mm^2 active area and 100 μm depletion depth. Also, we used a PTFE sample mask and support to hold the sample in place. The PTFE

sample holder was positioned at a 30° angle to the detector.

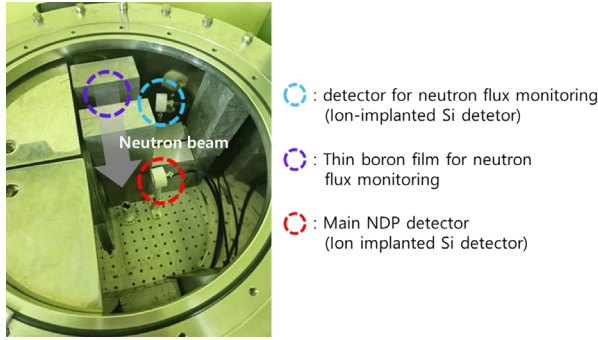


Fig. 3. Structure of KAERI-NDP vacuum chamber.

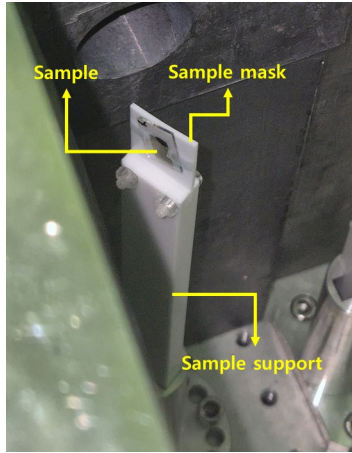


Fig. 4. Figure of sample fixed in neutron irradiation position.

At the time of the experiment, HANARO was operating at an output of 27 MW. The neutron flux at this output corresponds to 1.53×10^8 neutrons/cm²·s. The experiment lasted for 18,000 seconds.

In NDP analysis, α particles have higher depth resolution but a short range, which makes it difficult to analyze thick samples. Since the sample thickness was about 1 ~ 2 μ m, we decided to utilize α particles for analysis.

2.4 Residual energy to depth conversion

In order to convert the energy information of α particles into depth information, their residual energy upon reaching the sample surface must be determined. We calculated this using a TRIM simulation, starting from the cathode current collector and proceeding at intervals of 0.05 μ m up to a thickness of 1.3 μ m. The resulting data were then fitted to the experimental formula derived by J.K. Shultis, as shown below [4]:

$$x(E) = a + bE + cE^{1.5} + dE^3 + ce^{-E} \quad (3)$$

In this equation, $x(E)$ is the depth in (μ m), E is the residual energy (MeV), and the coefficients (a, b, c, d) are determined from the fit. Our fitting result is shown in Figure 5, providing the final conversion formula for our analysis.

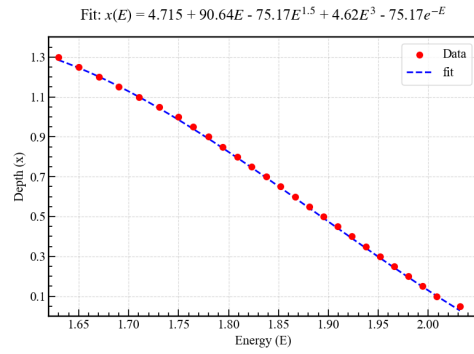


Fig. 5. Data acquired from simulation and fitting result.

3. Result & Discussion

The final NDP spectrum is consistent with the cell's layer structure. Pronounced changes in the count rate occur at depths that match the interfaces specified by KIST. However, due to the limited depth resolution, the measured interface is broadened, which can make lithium appear to extend into the anode. Consequently, the slope of the spectrum is steep on the anode side and relatively flat on the cathode side. This is because the depth resolution of NDP decreases as the deeper the sample is analyzed.

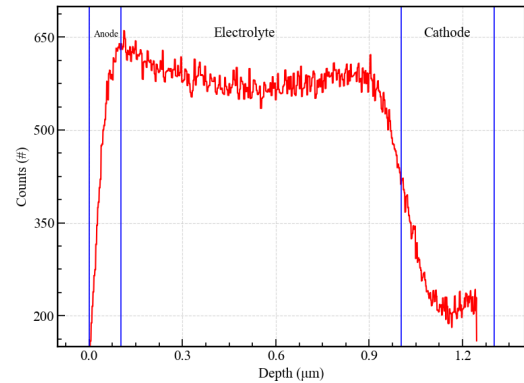


Fig. 6. Depth transformed NDP spectrum.

In this study, we were able to confirm the distribution of Li, consistent with the cell's structure. This result establishes a clear framework for future system improvements. However, the current analysis is qualitative, lacking a quantitative evaluation of Li concentration. To address this limitation, we are fabricating a new sample mask tailored to the cell's geometry. This will enable a direct relative analysis of our sample against a SRM-2137, allowing for precise quantification in our future work.

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