

## Analysis of Lithium Reaction Distribution in Cross Sectional Electrode Using X-ray Micro-beam

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### 1 Introduction

Lithium ion batteries (LIBs) are used in consumer mobile electronics and expected to be widely applied in products such as energy storage systems and electric vehicles, that demand high capacity, long-life, improved safety, and low cost. The cathodes of conventional LIBs, however, have insufficient performance and stability under charging and discharging at high current rate. Since the charge and discharge reactions of LIB proceed by the transfer of lithium ions ( $\text{Li}^+$ ) between the cathode and anode,  $\text{Li}^+$  can transfer via three diffusion paths: 1) in the active material of the cathode and anode, 2) at the interface between the active material and the liquid electrolyte, and 3) in the liquid electrolyte. Some barriers suppressing the  $\text{Li}^+$  transfer in many paths lead to non-uniform reaction distributions in the cross section direction of electrodes. Moreover, the non-uniform reaction distributions may result in insufficient performance and stability, including reduced power output, underutilization of capacity, localized heat generation, and overcharge or overdischarge.

Therefore, we measured the  $\text{Li}^+$  reaction distribution in the cross section of the cathode by developing an operand-measurement with an X-ray micro-beam during the charge-discharge process.

### 2 Experiment

A LIB-cell for the cross sectional observation with an optical microscope was modified for the measurement of the X-ray micro-beam diffraction by using the cover plate with the X-ray transmissive window. The cathode active material was  $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$  (NCM). The X-ray diffraction measurements during the charge-discharge process were carried out using the X-ray micro-beam of 20  $\mu\text{m}$  size and two dimensional X-ray detector at the BL15A1 @ Photon Factory, Japan. During the X-ray irradiation points were repeatedly changed from the Al current collector side to the separator side by moving the LIB-cell, the diffracted X-ray images from each X-ray irradiation point were detected every 1 sec.

### 3 Results and Discussion

Figure 1 shows the fluorescence X-ray intensity of Ni (originating from NCM) and Cu vs. cross sectional position of the electrode. The X-ray irradiation points were #1 ~ #5, where #1 and #5 were the Al corrector and separator side, respectively.

Since the NCM[113] diffraction angle shifted toward higher (lower) angle during the charge (discharge) process, the [113] diffraction angle was the good index for the  $\text{Li}^+$  transfer reaction in the cathode. The change of

the NCM[113] diffraction angle at the separator side (#5) was larger than that at the Al corrector side (#1) during the discharge process in Fig. 2. This indicates that the  $\text{Li}^+$  transfer reaction in the cathode preferentially proceeds at the separator side compared with the Al corrector side.

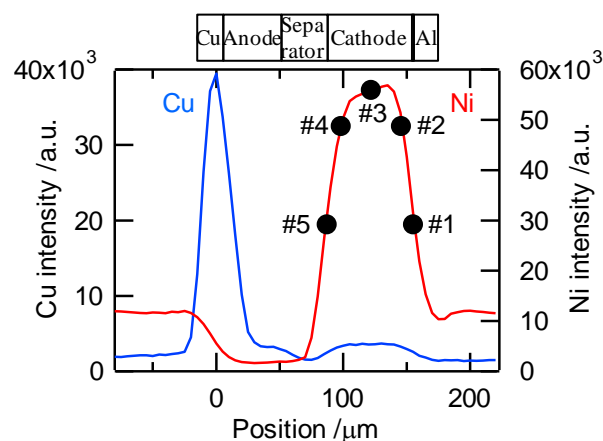


Fig.1 Fluorescence X-ray intensity of Ni and Cu vs. cross sectional position of the electrode.

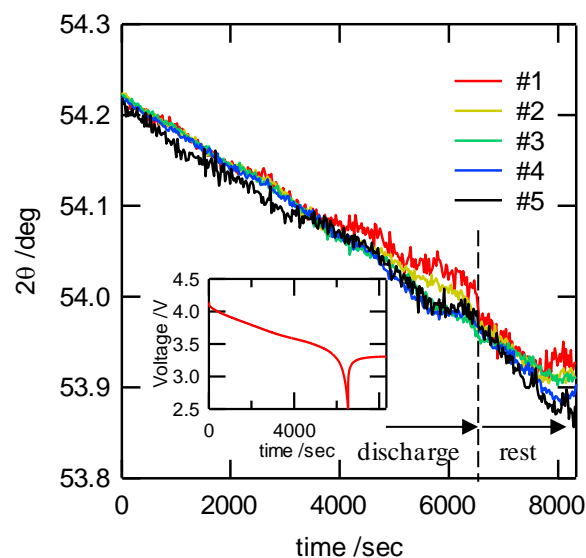


Fig.2 Time vs.  $2\theta$  angle of NCM[113] diffraction during discharge process. Inset is discharge curve.

### Acknowledgement

We thank Dr. H. Nitani and Y. Takeichi for their experimental supports.

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