


実験報告書様式(一般利用課題・成果公開利用)

(※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。)

 <b>MLF Experimental Report</b>	提出日 Date of Report 2017/07/26
課題番号 Project No. 2016B0213 実験課題名 Title of experiment Detecting Li in a charged cathode of Li-ion battery by muonic x-ray 実験責任者名 Name of principal investigator Izumi Umegaki 所属 Affiliation Toyota Central Research and Development Laboratories, Inc.	装置責任者 Name of responsible person Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) D2 実施日 Date of Experiment 2017/02/19-2017/02/21

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)  
 Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.
(1) A cathode sheet removed a test pouch Li-ion cell after the first cycle of an electrochemical measurement (2) An as prepared cathode sheet  After the first cycle of an electrochemical measurement, Li is lost in a cathode. The amount of lost Li is equivalent to the non reversible capacity of a Li-ion battery. The Li content of the cathode after the first cycle was analyzed by ICP-OES technique (CIROS 120EOP, Rigaku).

2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。)
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.
We have performed an elemental analysis with muonic x-ray ( $\mu$ XEA) on 100 $\mu$ m thick cathode sheets. By selecting the momentum of incident negative muon as 20 MeV/c, the position that the muon stops at can be adjusted in the sample. The sample was hold by an Al holder so as to keep the sample facing towards to the beamline. The sample and the holder were installed in an Al chamber. The Al chamber was evacuated down to $10^{-4}$ Pa before opening the gate valve. Ge semiconductor detectors were arranged around the Al chamber. The muonic x-ray was detected with synchronizing with the frequency of 25 Hz of the muon pulse in J-PARC.

## 2. 実験方法及び結果(つづき) Experimental method and results (continued)

Figure 1 shows the muonic x-ray energy spectrum for the as-prepared cathode sheet (before) and the cathode removed from a charged pouch test Li-ion cell (after) with the momentum 20 MeV/c. The value of the momentum was calculated by the PHITS program [1], and it was also confirmed that most of peaks observed with the momentum of 20MeV/c were assigned as Li, Ni, Co, Mn, and O from the cathode material. The peaks of Al from the sample holder and the sample chamber were also observed.

The intensity of each peak was calculated by Covell's method [2]. Table 1 summarized the intensity normalized by the intensity of the peak of Mn observed at 49 keV. It is found that most peaks are almost the same intense except a peak at 19keV, which is assigned as C and Li. It is noted that it is difficult to distinguish these two peaks by a conventional Ge detector because of small difference in energy. The difference of the peak of C+Li originates from the Li loss due to the first cycle of an electrochemical measurement. The Li content of the cathode from the charged battery was obtained as  $x=0.85$  in  $\text{Li}_x(\text{NiCoMn})\text{O}_2$  by the ICP-OES. Therefore, we have concluded that the 15% of Li loss in a cathode is detectable by  $\mu\text{XEA}$ .

### 【Reference】

[1] T. Sato *et al.*, Nucl. Sci. Technol. **50**, (2013) 913–923.

[2] D. F. Covell, Analytical Chemistry **31**, (1959) 1785.

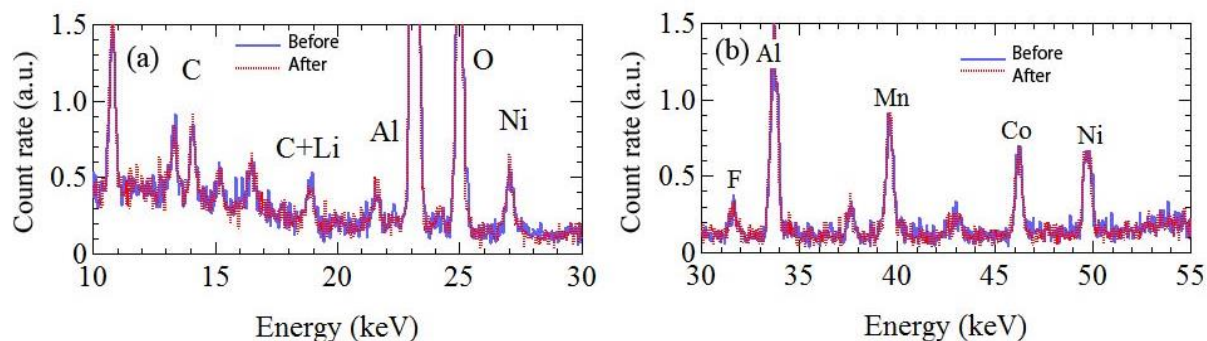


Figure 1 The  $\mu\text{XEA}$  spectra obtained for the test pouch Li-ion cell in the energy range of (a) from 10 to 30 keV, and (b) from 30 to 55 keV. The vertical axis is normalized by the intensity of the peak of Mn at 39 keV.

Table 1 Comparison of the intensity of main peaks for the cathode sheets before and after the first cycle of the electrochemical method. The intensity is normalized by the intensity of the peak of Mn.

	C 14keV	C+Li 19eV	O 25keV	Al 33keV	Mn 40keV	Co 46keV	Ni 49keV
Before	0.55	0.44	2.58	1.83	1	0.79	0.92
After	0.53	0.28	2.34	1.67	1	0.77	0.89